H2Teesside Project

Planning Inspectorate Reference: EN070009

Land within the boroughs of Redcar and Cleveland and Stockton-on-Tees, Teesside and within the borough of Hartlepool, County Durham

Document Reference 8.41: Report to Inform Assessment of Air Quality Impacts on Teesmouth and Cleveland Coast SSSI

The Infrastructure Planning (Applications: Prescribed Forms and Procedure) Regulations 2009 - Regulation 5(2)(g)

Applicant: H2 Teesside Ltd

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1.0 INTRODUCTION

1.1 Overview

1.1.1 This Report to Inform Assessment of Air Quality Impacts on Teesmouth and Cleveland Coast Site of Special Scientific Interest (SSSI) has been prepared on behalf of the Applicant in response to comments from Natural England and the Examining Authority that it would be useful for the Applicant's position on impacts to the SSSI to be set out in one document [REP6a-019].

1.1.2 As the main discussion in Examination on impacts to the SSSI have focussed on Air Quality impacts, this note focusses on those impacts. In respect of other impacts, these are covered in the Ecology ES Chapter (APP-064) and the Applicant's analysis remains unchanged.

1.1.3 It should be noted that this report does not present any new air quality data. The data on which this report is based were presented in the Deadline 5 HRA report [REP5-095] and the updated ES Appendix 8A (please refer to the Technical Note: Updates to Air Quality and Traffic Cumulative Assessments (document 6.4.42 REP5-034).

1.2 Sensitivity of Teesmouth and Cleveland Coast SSSI

1.2.1 The main pollutants of concern for the Teesmouth and Cleveland Coast SSSI are oxides of nitrogen (NOx), ammonia (NH_3) and sulphur dioxide (SO_2). Ammonia can have a directly toxic effect upon vegetation, particularly at close distances to the source such as near road verges. NOx can also be toxic at very high concentrations (far above the annual average Critical Level). However, in particular, high levels of NOx and NH_3 are likely to increase the total nitrogen and acid deposition to soils, potentially leading to deleterious effects in resident ecosystems. For example, an increase in the total nitrogen deposition from the atmosphere is widely known to enhance soil fertility and to lead to eutrophication.

1.2.2 The Air Pollution Information System (APIS) forms the major source of information regarding the air quality impact pathway. It specifies a NOx concentration (Critical Level) for the protection of vegetation of 30 $\mu g/m^{-3}$. In addition, ecological studies have determined 'Critical Loads' for atmospheric nitrogen deposition (that is, NOx combined with ammonia NH_3).

1.2.3 Teesmouth and Cleveland Coast SSSI is designated for its calcareous dune habitats in addition to its bird interest. The assessment of impacts on bird interest is identical to that for the Teesmouth and Cleveland Coast SPA and Ramsar site presented in the Deadline 5 Report to Inform Appropriate Assessment [REP5-011] and reaches a conclusion of no likely significant effect for the same reasons. Therefore, this report focuses on the calcareous dunes, as requested by Natural England in their Deadline 5A response [REP5-065].

1.2.4 The Coatham Dunes within the Teesmouth and Cleveland Coast SSSI are immediately north of the Proposed Development Main Site. Therefore, air quality impacts on Coatham Dunes represent the worst-case air quality impacts of the Proposed Development on the SSSI.

1.2.5 The sand dunes which form part of the Teesmouth and Cleveland Coast SSSI are calcareous as demonstrated by the presence of calcareous vegetation on the dunes. As set out in Bobbink *et al* (2022) (Appendix A.1) surveys have indicated that calcareous, iron-rich dunes exhibit co-limitation of nitrogen and phosphorus and that phosphorus limitation is a factor in calcareous dunes and '*may lead to fewer botanical responses in calcareous dunes compared with acidic or decalcified dune sites'*. There is therefore a justification for considering that the lowest critical load of 5kgN/ha/yr is less appropriate than a slightly higher critical load of 10 kgN/ha/yr as was used on APIS for calcareous dune systems before the critical loads reported on APIS were updated in 2023.

1.2.6 The APIS Site Relevant Critical Load app contains columns to list if lichens or bryophytes are integral to any feature for which a site is designated, and for the SSSI they are either blank or it says 'no'. Nowhere does APIS currently indicate that lower plants are integral to the interest features of Teesmouth and Cleveland Coast SSSI. This is therefore the justification for using the higher critical level of 3µg/m^3 for ammonia.

2.0 LIKELY SIGNIFICANT EFFECTS

2.1 Construction

Atmospheric Pollution

2.1.1 There will be no construction period stack emissions. An assessment of plans and projects likely to generate road traffic emissions to air which are capable of affecting the SSSI has been completed (See Annex G of the Report to inform HRA (CR1-023) as also re-submitted at Deadline 6A (REP6a-010), following Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations. Natural England guidance (Appendix A.2) identifies that traffic exhaust emissions are only relevant to ecological receptors located within 200m of the source. Further details on the assessment of cumulative road traffic emissions impacts using the NAE001 Methodology are included in Annex G.

2.1.2 Step 1 of Annex G shows that seven links that will be used by construction traffic are within 200m of the Teesmouth and Cleveland Coast SPA and thus SSSI. Specifically:

- Road link 1 (A1085 Trunk Road, 100 m east of Ennis Road) and road link 10 (Unnamed Road, 725 m east of A178 Seaton Carew Road) are 5 m from the SSSI.
- Road link 9 (A178 Seaton Carew Road, 535 m north of Huntsman Drive) is 3 m from the SSSI.
- Road link 10 (Unnamed Road, 725m east of A178 Seaton Carew Road) is 5 m from the SSSI.

- Road link 14 (A1185 west of A178 Seaton Carew Road) is 20 m from the SSSI.
- Road link 8 (A1046 Port Clarence Road, 20 m north of Beech Terrace) is 32 m from the SSSI.
- Road link 3 (A1042 Kirkleatham Lane) is 125 m from the SSSI.
- Road link 13 (B1275 Belasis Avenue) is 160 m from the SSSI.

2.1.3 Tables 8A-28 to 8A-31 of updated ES Appendix 8A in the Technical Note: Updates to Air Quality and Traffic Cumulative Assessments (6.4.42) [REP5-034] show that at no point will the total (including contributions from the H2Teesside project construction traffic and from cumulative developments as listed in the Updated Cumulative and Combined Effects Assessment [REP5-015], on top of the local background and baseline traffic) critical levels or loads for NOx, ammonia or acid deposition be exceeded within the SSSI during construction, except adjacent to the roadside of RE002 and RE007 where NOx will slightly exceed the critical level, being 33.4 μgm^{-3} and 32.1 μgm^{-3} respectively. At these receptors, the contribution from the H2Teesside project is below 1% of the critical level. At these concentrations NOx is only relevant as a source of nitrogen deposition.

2.1.4 The maximum nitrogen deposition on these road links due to the Proposed Development, as reported in Table 8A-23 of updated ES Appendix 8A [REP5-034], as submitted at Deadline 5, will be 0.2 kgN/ha/yr which is 2% of the critical load. Predicted deposition values decrease further away from the road, reaching 1% of the critical load at 10 m from the road and <1% at 20 m and further. Moreover, this impact will be temporary and will therefore not affect long-term nitrogen deposition. This is relevant because the critical load system for nitrogen deposition assumes decades of continuous exposure (World Health Organisation, 2000) (Appendix A.3). Caporn, et al., 2016 (Appendix A.4) specifically addresses this point in sections 2.2.1 and 5.1 stating that '*The current rate of N deposition is primarily a proxy for long-term cumulative N deposition. Thus we would not expect that a change in N deposition, either increasing or decreasing, would immediately change species richness or composition, but instead these would be gradually influenced by longer-term changes in N deposition'.*

2.1.5 In addition, atmospheric pollution predictions have been made for the peak construction year (one year, 2026, out of a five year programme). However, the amount of construction traffic, particularly of Heavy Goods Vehicles, will be substantially smaller outside of the peak year, as detailed in Appendix 15A [REP5-034].

2.2 Operational Period

<u>Atmospheric pollution</u>

2.2.1 This assessment strictly follows Natural England guidance (Appendix A.2). The discussion below focusses on stack emissions (from the operational period) rather than vehicle exhaust emissions. This is because there will be negligible operational vehicle emissions within 200m of sensitive parts of Teesmouth and Cleveland Coast SSSI.

2.2.2 The model outputs shown in the tables below were extracted from the air quality assessment as presented in Appendix 1A.0 Air Quality of the Change Application Report [CR1-045]. The data used in the dispersion model has been updated following the Changes as described in the Change Application Report.

Annual Average Oxides of Nitrogen NOx

2.2.3 For annual average NOx, likely significant effects cannot be mathematically dismissed for Teesmouth and Cleveland Coast SSSI. For the SSSI the impact is driven by the cumulative impact, with the contribution of the Proposed Development alone being at 1% of the critical level. (Table 4-8). It should be noted that the Environment Agency and Natural England have agreed that depositional impacts that are below 1% of the relevant critical load for a site can be regarded as likely to be insignificant. Guidance from the IAQM clarifies that the 1% threshold is not intended to be precise to a set number of decimal places but to the nearest whole number (paragraph 5.5.2.6 of Institute of Air Quality Management, 2020).

Table 4-8: Annual Mean Oxides of Nitrogen Concentrations within the Study Area

RECEPTOR	CONSERVATION DESIGNATION	ALONE PC (PROCESS CONTRIBUTION) (µg/m^{-3})	ALONE PC AS % OF CRITICAL LEVEL	IN COMBINATION PC (µg/m^{-3})	IN COMBINATION PC AS % OF CRITICAL LEVEL
OE1 – OE3 (worst case data reported)	Teesmouth & Cleveland Coast SSSI	0.3	1.1	3.5	11.7

24hr Oxides of Nitrogen NOx

2.2.4 It can be seen below (Table 4-9) that likely significant effects cannot be mathematically dismissed for Teesmouth and Cleveland Coast SSSI. For that SSSI the impact is driven by the cumulative impact, with the contribution of the Proposed Development alone being below 10% of the critical level.

Table 4-9: Maximum 24hr Oxides of Nitrogen Concentrations within the Study Area

RECEPTOR	CONSERVATION DESIGNATION	ALONE PC (µg/m^{-3})	ALONE PC AS % OF CRITICAL LEVEL	IN COMBINATION PC (µg/m^{-3})	IN COMBINATION PC AS % OF CRITICAL LEVEL
OE1 – OE3 (worst case data reported)	Teesmouth & Cleveland Coast SSSI	2.9	3.8	12.4	16.5

Ammonia

2.2.5 It can be seen below (Table 4-10) that likely significant effects from ammonia in atmosphere can be dismissed on Teesmouth and Cleveland Coast SSSI because notwithstanding any increase in ammonia concentrations either alone or cumulatively the critical level of 3 μgm^{-3} will not be exceeded being a maximum of 1.4 $\mu g/m^{-3}$.

Table 4-10: Annual Mean Ammonia Concentrations within the Study Area

RECEPTOR	CONSERVATION DESIGNATION	ALONE PC ($\mu g/m^{-3}$)	ALONE PC AS % OF CRITICAL LEVEL	IN COMBINATION PC ($\mu g/m^{-3}$)	IN COMBINATION PC AS % OF CRITICAL LEVEL
OE1 – OE3 (worst case data reported)	Teesmouth & Cleveland Coast SSSI	0.01	0.3	0.10	3.3

Nitrogen deposition

2.2.6 For annual average nitrogen, likely significant effects cannot be mathematically dismissed for Teesmouth and Cleveland Coast SSSI. For the SSSI the impact is driven by the cumulative impact, with the contribution of the Proposed Development alone being at 1% of the critical load (Table 4-11).

Table 4-11: Nitrogen deposition values within the Study Area

RECEPTOR	CONSERVATION DESIGNATION	CRITICAL LOAD USED	ALONE PC (KGN/HA/YR)	ALONE PC AS % OF CRITICAL LEVEL	IN COMBINATION PC (KGN/HA/YR)	IN COMBINATION PC AS % OF CRITICAL LEVEL
OE1 – OE3 (worst case data reported)	Teesmouth & Cleveland Coast SSSI	10[1]	0.11	1.1	1.0	10.1

Acid deposition

2.2.7 For annual average acid deposition, likely significant effects can be mathematically dismissed for Teesmouth and Cleveland Coast SSSI. While the cumulative impact exceeds 1% of the critical load, the contribution of the Proposed Development alone being less than 0.01 i.e. not visible in the modelling when reported to 2 decimal places to avoid false precision (Table 4-12). As such it is considered

[1] Critical load for calcareous dunes. Appropriate habitat for areas of greatest deposition.

reasonable to dismiss the contribution of the Proposed Development to the modelled cumulative impact as imperceptible.

Table 4-12: Acid deposition values within the Study Area

RECEPTOR	CONSERVATION DESIGNATION	ALONE PC (KEQ/HA/YR)	ALONE PC AS % OF CRITICAL LEVEL	IN COMBINATION PC (KEQ/HA/YR)	IN COMBINATION PC AS % OF CRITICAL LEVEL
OE1 – OE3 (worst case data reported)	Teesmouth & Cleveland Coast SSSI	< 0.01	0.2	0.132	1.5

2.3 Summary

2.3.1 Having assessed the likely significant effects of the Proposed Development with reference purely to exceedance (or otherwise) of the numerical screening criteria, two cumulative impacts could not be dismissed on purely mathematical grounds:

- NOx at Teesmouth and Cleveland Coast SSSI
- Nitrogen deposition at Teesmouth and Cleveland Coast SSSI

2.3.2 In both cases when considered alongside other plans and projects the insignificance screening thresholds were exceeded. However, they are not exceeded by the Proposed Development alone. When undertaking the assessment the ecological sensitivity of the interest features and how nitrogen deposition or NOx may affect them is key to the interpretation.

3.0 FURTHER ASSESSMENT

3.1.1 The long-term (operational) forecast nitrogen dose at the maximum point of impact within the Teesmouth and Cleveland Coast SSSI is at the threshold of insignificance of 1% of the critical load (being 1.1% of the critical load). It should be noted that The Environment Agency and Natural England have agreed that depositional impacts that are below 1% of the relevant critical load for a site can be regarded as likely to be insignificant.

3.1.2 Guidance from the IAQM clarifies that the 1% threshold is not intended to be precise to a set number of decimal places but to the nearest whole number (paragraph 5.5.2.6 of Institute of Air Quality Management, 2020) (Appendix A.5). The PEC will also exceed the critical load being a maximum of 12.99 kgN/ha/yr at Coatham Sands/Dunes and 13.89 kgN/ha/yr at North Gare Sands. This is due to the fact that current nitrogen deposition exceeds the critical load.

3.1.3 The SSSI was designated in 2015 when the background nitrogen dose to short vegetation according to APIS was 13.07 to 13.53 kgN/ha/yr at Coatham Sands/Dunes and North Gare Sands. Moreover, APIS shows that in the years prior to 2015 (prior to designation) the background nitrogen deposition dose to short vegetation was higher; for example, being 14.69 to 14.77 kgN/ha/yr in 2003 at Coatham Sands/Dunes and North Gare Sands. The calcareous dune habitat has thus developed and persisted in close proximity to an operational steel works and other industrial facilities when nitrogen deposition rates were considerably higher than the lower critical load of 10 kgN/ha/yr, or than is forecast to be the case under the 'in combination' assessment (13.89 kgN/ha/yr maximum).

3.1.4 Since total nitrogen deposition is forecast to remain on an improving trend even when growth is considered 'in combination' and would therefore remain below historic nitrogen deposition rates under which the habitat in question developed, no significant effect on the SSSI is expected.

3.1.5 Moreover, the habitat structure has been extensively changed due to slag deposition and movement from at least the 1940s to the early 2000s (Appendix A.6). Much of the dunes north of the Main Site (i.e. Coatham Dunes) have developed on slag deposits from the various historic industrial activities in that area (notably Warrenby Slag Works). In these decades N deposition will have been higher than it is now due to much higher NOx emissions from the former steelworks and other industrial emitters (and was certainly higher in 2003 than it is now according to APIS). For example, UK N deposition reduced from 465 kt N in 1990 to 278 kt N in 2017 (Appendix A.7).

3.1.6 Therefore, no likely significant effect will arise on Teesmouth and Cleveland Coast SSSI, based on the small contribution of the proposed project, the fact that nitrogen deposition is modelled to remain below historic levels (thus denoting a net improvement even when cumulative deposition is considered), and the fact that much of the dune interest developed when pollution levels were higher than at present.

3.1.7 The Applicant's view remains that if the total nitrogen deposition rate will remain lower with the Proposed Development consented (even allowing for other plans and projects) than it has been historically or when the SSSI vegetation interest generally developed, it cannot be argued that the scheme will be harming the interest of the SSSI, even by impeding restoration. That is particularly the case given the contribution of the Proposed Development is at the '1% of the upper critical load' level for being dismissed as imperceptible and is therefore not a significant contributor to overall nitrogen deposition.

3.1.8 Moreover, other factors such as management of recreational pressure are likely having a greater effect on limiting potential for restoration of the dune vegetation than air quality. Mitigation of nitrogen deposition from the Proposed Development has been embedded in the design including controlling emissions through process design and selection of appropriate stack heights to deliver effective dispersion of residual emissions. The Applicant understands that Natural England's concern is not with the emissions of the Proposed Development itself, but the cumulative emissions of a range of developments around Teesside. Even if it was not agreed by Natural England that no likely significant effect can be concluded, as the contribution of the Proposed Development to a cumulative impact is so small as to be imperceptible, the Applicant considers that it would not be appropriate for any additional mitigation to be applied to address the residual effects of the Proposed Development.

3.1.9 Notwithstanding the above project position, the Applicant generally notes (but emphasises that this does not affect the position it has stated above) that it is aware of and is contributing to wider strategic discussions regarding industrial development on Teesside working with the Environment Agency, Natural England and other parties to better understand the condition and resilience of the dune habitat when considering the reduction in industrial emissions across Teesside over many years and the potential new decarbonisation developments being progressed by various parties to align with the Track 1 status of the Teesside industrial cluster.

4.0 CONCLUSION

4.1.1 It is concluded that, following consideration of mitigation designed into the plant to reduce emissions, there would be no likely significant residual effect from the Proposed Development on Teesmouth and Cleveland Coast SSSI either alone or cumulatively with other projects. This is because:

- the residual contribution of H2Teesside to any cumulative nitrogen deposition impact is negligible and therefore directly addressing the residual contribution would likely convey a similarly negligible benefit to the SSSI;
- the SSSI vegetation developed at a time when nitrogen deposition rates were higher than they would be even with the Proposed Development in operation; and
- because the dunes at the most affected location (Coatham Dunes) are very significantly affected by recreational disturbance, and any benefit conveyed by reducing nitrogen deposition to the very small extent required to address the residual impact of the Proposed Development would likely be more than offset by the damage caused by continuing recreational disturbance and trampling.

APPENDIX A REFERENCED DOCUMENTS

A.1 Bobbink R, Loran C, Tomassen H. 2022. Review and Revision of Empirical Critical Loads of Nitrogen for Europe. Report for the German Environment Agency

TEXTE
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Review and revision of empirical critical loads of nitrogen for Europe

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Abstract

This report describes the scientific background and results from the review and revision of empirical critical loads of nitrogen that had been established for Europe in 2011 under the auspices of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention). In 2020, the Coordination Centre for Effects started a project under the LRTAP Convention to bring empirical critical loads up to date. New relevant information from studies (2010 - summer 2021) on the impacts of nitrogen on natural and semi-natural ecosystems was incorporated in the existing European database on empirical critical loads of N ($CL_{emp}N$). The current review and revision used for the first time gradient studies to evaluate and determine the $Cl_{emp}N$. The $CL_{emp}N$ were structured according to the updated classification used within the European Nature Information System (EUNIS).

Consensus on the results was reached in a UNECE expert workshop on empirical critical loads of nitrogen (26-28 October 2021, Berne, Switzerland), organised by the Swiss Federal Office for the Environment (BAFU), the Coordination Centre for Effects, and the B-WARE Research Centre. The results, as presented in Table 1 of the Executive Summary, show that in many cases the outer ranges of the empirical critical loads have decreased. The resulting 2021 European database includes both revised and newly established value ranges of $CL_{emp}N$ for each EUNIS class. The outcomes of this report are of major importance for the protection of N-sensitive natural and semi-natural ecosystems across Europe. This knowledge is used to support European policies to reduce air pollution.

Kurzzusammenfassung

Dieser Bericht beschreibt den wissenschaftlichen Hintergrund und die Ergebnisse der Überprüfung und Überarbeitung der empirischen Critical Loads für Stickstoff ($CL_{emp}N$), die 2011 im Rahmen des UNECE-Übereinkommens über weiträumige grenzüberschreitende Luftverunreinigung (Convention on Long-range Transboundary Air Pollution, LRTAP) für Europa festgelegt wurden. Im Jahr 2020 startete das Coordination Centre for Effects ein Projekt im Rahmen des LRTAP-Übereinkommens, um die empirischen Critical Loads auf den neuesten Stand zu bringen. Neue relevante Informationen aus Studien (2010 - Sommer 2021) zu den Auswirkungen von Stickstoff auf natürliche und naturnahe Ökosysteme wurden in die bestehende europäische Datenbank zu empirischen Critical Loads für N ($CL_{emp}N$) aufgenommen. Bei der aktuellen Überprüfung und Überarbeitung wurden zum ersten Mal Gradientenstudien zur Bewertung und Bestimmung der $CL_{emp}N$ herangezogen. Die $CL_{emp}N$ wurden gemäß der aktualisierten Klassifizierung des Europäischen Naturinformationssystems (EUNIS) strukturiert.

Ein Konsens über die Ergebnisse wurde in einem UNECE-Expertenworkshop zu empirischen Critical Loads für Stickstoff (26.-28. Oktober 2021, Bern, Schweiz) erzielt, der vom Schweizer Bundesamt für Umwelt (BAFU), dem Coordination Centre for Effects und dem Forschungszentrum B-WARE organisiert wurde. Die Ergebnisse, die in Tabelle 1 der Zusammenfassung dargestellt sind, zeigen, dass sich in vielen Fällen die äußeren Bereiche der empirischen Critical Loads verringert haben. Die sich daraus ergebende europäische Datenbank für 2021 enthält sowohl überarbeitete als auch neu festgelegte Wertebereiche für $CL_{emp}N$ für jede EUNIS-Klasse. Die Ergebnisse dieses Berichts sind von großer Bedeutung für den Schutz von N-empfindlichen natürlichen und naturnahen Ökosystemen in ganz Europa. Dieses Wissen wird zur Unterstützung der europäischen Politik zur Verringerung der Luftverschmutzung genutzt.

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Acknowledgement

This report is the result of an expert collaboration under the Working Group on Effects (WGE) of the UNECE Convention on Long-Range Transboundary Air Pollution (CLRTAP). Therefore, special thanks go to the involved experts from many institutions across Europe representing the International Cooperative Program (ICP) Forests, ICP Integrated Monitoring, ICP Modelling & Mapping, ICP Waters and ICP Vegetation of the Working Group on Effects.

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Summary

I. Introduction

- This report describes the scientific background and results from the review and revision of empirical critical loads of nitrogen ($CL_{emp}N$) as established for Europe in 2011 under the auspices of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention). In June 2020 the Coordination Centre for Effects (CCE) started a project under the LRTAP Convention to bring the empirical critical loads of nitrogen up to date. New relevant information on the impacts of nitrogen on natural and semi-natural ecosystems was incorporated.
- This report describes the scientific background and results from the review and revision of empirical critical loads of nitrogen ($CL_{emp}N$) as established for Europe in 2011 under the auspices of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention). In June 2020 the Coordination Centre for Effects (CCE) started a project under the LRTAP Convention to bring the empirical critical loads of nitrogen up to date. New relevant information on the impacts of nitrogen on natural and semi-natural ecosystems was incorporated.
- The workshop on the review and revision of empirical critical loads for Europe was held under the LRTAP Convention, in Berne, from 26 to 28 October 2021. The workshop was organised by the Coordination Centre for Effects (CCE) and hosted by the Swiss Federal Office for the Environment (BAFU).
- The workshop was attended by 37 participants from Austria, Canada, Finland, France, Germany, the Netherlands, Norway, Spain, Sweden, Switzerland, the United Kingdom, and the United States, and by representatives from the International Cooperative Programme (ICP) on ICP Waters, ICP Vegetation and ICP Modelling and Mapping. The secretariat to the Convention was not represented.
- The decision to organise the workshop was adopted at the 39th session of the Working Group on Effects (ECE/EB.AIR/144/Add.2), following recommendations from 35th session of the Task Force on Modelling and Mapping (2-4 April 2019) held as web-conference.
- The meeting was opened by Reto Meier on behalf of the BAFU and Alice James Casas as chair of the ICP Modelling and Mapping.

II. Objectives and structure of the workshop

- The objective of the workshop was the revision of the $CL_{emp}N$ for natural and semi-natural ecosystems, which were set at the last expert workshop held in Noordwijkerhout from 23 to 25 June 2010 (see Bobbink and Hettelingh, 2011), based on additional scientific information available for the period from 2010 to 2021. The workshop discussions were based on the new and updated background document that was sent to all participants prior to this workshop.
- Most of the European Nature Information System (EUNIS) classes, which were addressed in the present revision, have changed since the last update by Bobbink and Hettelingh (2011): marine habitats (EUNIS class MA, formerly A), coastal habitats (EUNIS class N, formerly B),

inland surface waters (EUNIS class C), mires, bogs and fens (EUNIS class Q, formerly D), grasslands and lands dominated by forbs, mosses or lichens (EUNIS class R, formerly E), heathland, scrubland and tundra (EUNIS class S, formerly F), woodland, forest and other wooded land (EUNIS class T, formerly G).

- An international team of 43 scientists prepared the background document for the workshop in the period from June 2020 to September 2021, after a web-based kick-off meeting in June 2020:
  - EUNIS class MA – Roland Bobbink
  - EUNIS class N – Laurence Jones, Emiel Brouwer and Eva Remke
  - EUNIS class C – Heleen De Wit, Roland Bobbink, Christin Loran, Linda May and Jan-Erik Thrane
  - EUNIS class Q - Chris Field, Julian Aherne, Roland Bobbink and Hilde Tomassen
  - EUNIS class R - Carly Stevens, Rocio Alonso, Vegar Bakkestuen, Erika Hiltbrunner and Lukas Kohli
  - EUNIS class S - Leon Van den Berg, Julian Aherne, Andrea Britton, Simon Caporn, Héctor García Gómez and Liv Guri Velle
  - EUNIS class T – Sabine Braun, Rocio Alonso, Frank Ashwood, Tomáš Chuman, Lucienne De Witte, Thomas Dirnböck, Per Erik Karlsson, Sirkku Manninen, Michael Perring, Hans Tömmervik, Simon Tresch, Liisa Ukonmaanaho and Elena Vanguelova
  - Chapter 10 "Use of empirical critical loads of nitrogen in risk assessment and nature protection" – Markus Geupel, Khalid Aazem, Sabine Augustin, Jesper Bak, Alice James Casas, Laurence Jones, Christin Loran, Reto Meier, Anne-Katrin Prescher, Thomas Scheuschner, Axel Ssymank and Susan Zappala
- The background document was, after the internal review round, reviewed by Sabine Augustin, Ariel Bergamini, Leonor Calvo Galván, Tara Greaver, Kevin Hicks, Raúl Ochoa-Huesa, Tonje I. Økland and Jan Roelofs.
- The working groups exchanged their progress in short plenary sessions. Results, conclusions, and recommendations were discussed and summarised in a final plenary session chaired by Christin Loran.

III. Conclusions

- Statistically and biologically significant outcomes of field addition experiments were the basis for the assessment of the $CL_{emp}N$. Only studies which have independent N treatments and realistic N loads, and durations were used for updating and refining $CL_{emp}N$ values.
- In the present review and revision period an increasing number of gradient studies on atmospheric N deposition have been published in several EUNIS habitat types and proved to be useful for evaluation and setting of the $Cl_{emp}N$.
- Studies with higher N additions or shorter experimental periods were only interpreted with respect to the understanding of effects mechanisms, possible N limitation or sensitivity of the system. The methods used in these studies were carefully scrutinised to identify factors

related to the experimental design or data analysis, which may constrain their use in assessing $CL_{emp}N$ ranges.

- $CL_{emp}N$ were agreed on for a range of N deposition values for all treated EUNIS classes (level 2 or 3). New results regarding nitrogen effects in surface waters could be included based on activities presented by the ICP Waters. Novel findings for some Mediterranean habitats could be adopted as well.
- The new Chapter 10 "Use of $CL_{emp}N$ in risk assessment and nature protection" was presented and discussed. It contains a selection of examples of the use of $CL_{emp}N$ on different scales and in different European countries to provide guidance to practitioners and policy makers on how $CL_{emp}N$ can be used in practice.
- The assessment of the reliability of the $CL_{emp}N$ ranges was continued from the last update in 2011; distinguishing between 'reliable', 'quite reliable' and 'expert judgement' symbolised by ##, # and (#), respectively.
- $CL_{emp}N$ ranges resulting from the reviewing and revising procedure were agreed by consensus at the workshop, as summarised in Table 1. In more than 40% of the EUNIS types presented, the lower value of the range had become lower than in 2011. The same applies for the upper value. In only one case did the upper value increase.

IV. Recommendations

- More research and data are required to establish a $CL_{emp}N$ for the following ecosystems: several grasslands and steppe meadows; all Mediterranean vegetation types; wet (swamp) forests; many mires and fens and several coastal habitats; in addition, more research is needed for all distinguished EUNIS habitat types that have an 'expert' judgement rating.
- Impacts of N enrichment in (sensitive) freshwater and shallow marine ecosystems (including coastal waters) need further research.
- More well-designed gradient studies with both (very) low and high N loads are needed, especially in EUNIS habitat types that are hardly investigated. Furthermore, combining the results of both experimental and gradients studies increases the reliability of the $CL_{emp}N$.
- More research is needed on the differential effects of the deposited N forms (NO_x or NH_y) to be able to determine the critical loads for oxidised and reduced nitrogen separately in the future.
- To refine the current $CL_{emp}N$, long-term experiments (10-20 years) with a high N addition frequency of between 5 and 50 kg N ha^{-1} yr^{-1} in regions with low background deposition are crucial. This would increase the reliability of the derived $CL_{emp}N$ if the lowest treatment level does not exceed the current critical load.
- Climate change and nitrogen deposition are likely to have strong interactive effects on ecosystem functioning, with climate change altering ecosystem responses to nitrogen deposition and vice versa. More experimental studies are needed to investigate these interactions, as well as more gradient studies that explicitly examine the impacts of nitrogen deposition in combination with climatic gradients.
- In conclusion, it is crucial to understand the long-term effects of increased N deposition on ecosystem processes in a representative range of ecosystems. It is therefore important to

quantify the effects of N deposition by manipulating N inputs in long-term ecosystem studies in both pristine and affected areas. These data in combination with gradient studies are essential to validate critical loads and develop robust dynamic ecosystem models and/or multiple correlative species models that are reliable enough to use in calculating $CL_{emp}N$ for natural and semi-natural ecosystems, and to predict natural recovery rates for N-affected systems.

Table 1. Overview of empirical N critical loads (kg N ha^{-1} yr^{-1}) to natural and semi-natural ecosystems (column 1), classified according to EUNIS (column 2), as established in 2011 (column 3), and as revised in 2022 (column 4). The reliability is indicated by ## reliable; # quite reliable and (#) expert judgement (column 5). Column 6 provides a selection of effects that may occur when critical loads are exceeded. Finally, changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Marine habitats (MA)					
Atlantic upper-mid salt marshes	MA223	20-30	**10-20**	(#)	Increase in dominance of graminoids; decline positive indicator species
Atlantic mid-low salt marshes	MA224	20-30	**10-20**	(#)	Increase in late successional species; decline positive indicator species
Atlantic pioneer salt marshes	MA225	20-30	20-30	(#)	Increase in late successional species; increase in productivity species
Coastal habitat (N)					
Shifting coastal dunes	N13, N14	10-20	10-20	#	Biomass increase; increased N leaching; reduced root biomass
Coastal dune grasslands (grey dunes)	N15	8-15	**5**-15	##	Increased biomass and cover of graminoids and mesophilic forbs; decrease in oligotrophic species including lichens; increased tissue N; increased N leaching; soil acidification
Coastal dune heaths	N18, N19	10-20	10-**15**	#	Increased plant production; increased N leaching; accelerated succession; typical lichen C:N decrease; increased yearly increment *Calluna*
Moist and wet dune slacks	N1H	10-20	**5-15**	#	Increased cover of graminoids and mesophilic forbs; decrease in oligotrophic species; increased Ellenberg N
Dune-slack pools (freshwater aquatic communities of	N1H1, N1J1	10-20	10-20	(#)	Increased biomass and rate of succession

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
permanent Atlantic and Baltic or Mediterranean and Black Sea dune-slack water bodies)					
Inland surface water habitats (C) [a]					
Permanent oligotrophic lakes, ponds and pools (including soft-water lakes)	C1.1	3-10	**2**-10 [b]	##	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P; shifts in macrophyte community
Alpine and sub-Arctic clear water lakes	C1.1		**2-4**	**##**	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Boreal clear water lakes	C1.1		**3-6**	**##**	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Atlantic soft water bodies	C1.1, elements C1.2	3-10	**5**-10	##	Change in species composition of macrophyte communities
Permanent dystrophic lakes, ponds and pools	C1.4	3-10	**5**-10 [c]	(#)	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Mire, bog and fen habitats (Q)					
Raised and blanket bogs	Q1	5-10	5-10	##	Increase in vascular plants; decrease in bryophytes; altered growth and species composition of bryophytes; increased N in peat and peat water
Valley mires, poor fens and transition mires	Q2	10-15	**5**-15	**##**	Increase in sedges and vascular plants; negative effects on bryophytes
Palsa and polygon mires	Q3		**3-10**	**(#)**	Increase in graminoids, tissue N concentrations and decomposition rate
Rich fens	Q41-Q44	15-30	15-**25**	**#**	Increase in tall vascular plants (especially graminoids); decrease in bryophytes
Arctic-alpine rich fens	Q45	15-25	15-25	(#)	Increase in vascular plants; decrease in bryophytes

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Grasslands and tall forb habitats (R)					
Semi-dry Perennial calcareous grassland (basic meadow steppe)	R1A	15-25	**10-20**	##	Increase in tall grasses; decline in diversity; change in species composition; increased mineralisation; N leaching; surface acidification
Mediterranean closely grazed dry grasslands or Mediterranean tall perennial dry grassland or Mediterranean annual-rich dry grassland	R1D or R1E or R1F	15-25	**5-15**	(#)	Increased production; dominance by graminoids; changes to soil crusts; changes to soil nutrient cycling
Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta*	R1M	10-15	**6-10**	##	Increase in graminoids; decline of typical species; decrease in total species richness
Oceanic to subcontinental inland sand grassland on dry acid and neutral soils or Inland sanddrift and dune with siliceous grassland	R1P or R1Q	8-15	**5**-15	(#)	Decrease in lichens; increase in biomass
Low and medium altitude hay meadows	R22	20-30	**10-20**	(#)	Increase in tall grasses; decrease in diversity; decline of typical species
Mountain hay meadows	R23	10-20	10-**15**	**#**	Increase in nitrophilous graminoids; changes in diversity; decline of typical species
Moist or wet mesotrophic to eutrophic hay meadow	R35	15-25	15-25	(#)	Increase in tall graminoids; decreased diversity; decrease in bryophytes
Temperate and boreal moist and wet oligotrophic grasslands	R37	10-20	10-20	#	Increase in tall graminoids; decreased diversity; decrease in bryophytes

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
▶ Moss and lichen dominated mountain summits	(Earlier E4.2)	5-10	5-10	#	Change in species composition; effects on bryophytes or lichens
▶ Temperate acidophilous alpine grasslands	R43	5-10	5-10	#	Changes in species composition; increase in plant production
Arctic-alpine calcareous grassland	R44	5-10	5-10	#	Changes in species composition; increase in plant production
Heathland, scrub and tundra habitats (S)					
Tundra	S1	3-5	3-5 [d]	#	Changes in biomass; physiological effects; changes in bryophyte species composition; decrease in lichen species richness
Arctic, alpine and subalpine scrub habitats	S2	5-15	5-**10** [d]	#	Decline in lichens; bryophytes and evergreen shrubs
Lowland to montane temperate and submediterranean *Juniperus* scrub	S31		**5-15**	**(#)**	Shift in vegetation community composition; reduced seed viability
Northern wet heath	S411				
▶ U' Calluna-dominated wet heath (upland)	S411	10-20	**5–15** [e]	**##**	Decreased heather dominance; decline in lichens and mosses; increased N leaching
▶ 'L' Erica tetralix-dominated wet heath (lowland)	S411	10-20	**5-15** [e]	**##**	Transition from heather to grass dominance; decrease in heather cover; shift in vegetation community composition
Dry heaths	S42	10-20	**5-15** [e]	**##**	Transition from heather to grass dominance; decline in lichens; changes in plant biochemistry; increased sensitivity to abiotic stress

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Maquis, arborescent matorral and thermo-Mediterranean scrub	S5	20-30	**5-15**	(#)	Change in plant species richness and community composition; nitrate leaching; acidification of soil.
Garrigue	S6		**5-15**	**#**	Changes in species composition; decline in shrub cover; increased invasion of annual herbs
Forest habitats (T)					
Broadleaved deciduous forest	T1	10-20	10-**15**	##	Changes in soil processes; nutrient imbalance; altered composition mycorrhiza and ground vegetation
Fagus forest on non-acid and acid soils	T17, T18	10-20	10-**15**	(#)	Changes in ground vegetation and mycorrhiza; nutrient imbalance; changes in soil fauna
Mediterranean *Fagus* forest on acid soils	T18		**10-15**	**(#)**	Annual height and volume tree growth; analogy to temperate *Fagus* forest
Acidophilous *Quercus* forest	T1B	10-15	10-15	(#)	Decrease in mycorrhiza; loss of epiphytic lichens and bryophytes; changes in ground vegetation
Carpinus and *Quercus* mesic deciduous forest	T1E	15-20	15-20	(#)	Changes in ground vegetation
Mediterranean evergreen *Quercus* forest	T21	10-20	10-**15**	(#)	NO_3 in soil water and streams
Coniferous forests	T3	5-15	**3**-15	##	Changes in soil processes; nutrient imbalance; altered composition mycorrhiza and ground vegetation; increase in mortality with drought
Temperate mountain *Picea* forest, Temperate mountain *Abies* forest	T31, T32	10-15	10-15	(#)	Decreased biomass of fine roots; nutrient imbalance; decrease in mycorrhiza; changed soil fauna

Ecosystem type	**EUNIS code**	**2011 kg N ha^{-1} yr^{-1}**	**2022 kg N ha^{-1} yr^{-1}**	**2022 reliability**	**Indication of exceedance**
Mediterranean mountain *Abies* forest	T33		**10-15**	**(#)**	Tree foliar stoichiometry; tree physiology; soil N losses
Temperate continental *Pinus sylvestris* forest	T35	5-15	5-15	#	Changes in ground vegetation and mycorrhiza; nutrient imbalances; increased N_2O and NO emissions
Mediterranean montane *Pinus sylvestris-Pinus nigra* forest	T37		**5-17**	**(#)**	Lichen chemistry and community changes in Mediterranean mixed-conifer forests in USA
Mediterranean lowland to submontane *Pinus* forest	T3A	3-15	**5-10**	(#)	Reduction in fine-root biomass; shift in lichen community
Dark taiga	T3F	5-10	**3-5** [f]	##	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation
Pinus sylvestris light taiga	T3G	5-10	**2-5** [f]	#	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation

a) The lower part of the $CL_{emp}N$ range should be applied for lakes in small catchments (with high lake to catchment ratios), because these are most exposed to atmospheric deposition, given that a relatively high fraction of their N inputs is deposited directly on the lakes and is not retained in the catchments. Similarly, the lower part of the range should be applied for lakes in catchments with thin soils, sparse vegetation and/or with a high proportion of bare rock.

b) This $CL_{emp}N$ should only be applied to oligotrophic waters with low alkalinity and with no significant agricultural or other human inputs. Apply the lower end of the range to clear-water sub-Arctic and alpine lakes, the middle range to boreal lakes and the higher end of the range to Atlantic soft waters.

c) This $CL_{emp}N$ should only be applied to waters with low alkalinity and with no significant agricultural or other direct human inputs. Apply the lower end of the range to boreal dystrophic lakes.

d) Use towards high end of range if phosphorus limited, and towards lower end if phosphorus is not limiting.

e) Use towards high end of range with high intensity management, and use towards lower end of range with low intensity management.

f) Mainly based on N deposition impacts on lichens and bryophytes.

Zusammenfassung

I. Einleitung

- Dieser Bericht beschreibt den wissenschaftlichen Hintergrund und die Ergebnisse der Überprüfung und Überarbeitung der empirischen Critical Loads für Stickstoff ($CL_{emp}N$), die 2011 im Rahmen des UNECE-Übereinkommens über weiträumige grenzüberschreitende Luftverunreinigung (Convention on Long-range Transboundary Air Pollution, LRTAP) für Europa festgelegt wurden. Im Juni 2020 startete das Coordination Centre for Effects (CCE) ein Projekt im Rahmen des LRTAP-Übereinkommens, um die empirischen Critical Loads für Stickstoff auf den neuesten Stand zu bringen. Es wurden neue relevante Informationen über die Auswirkungen von Stickstoff auf natürliche und naturnahe Ökosysteme berücksichtigt.
- Im Rahmen des LRTAP-Übereinkommens fand vom 26. bis 28. Oktober 2021 in Bern ein Workshop zur Überprüfung und Überarbeitung der empirischen Critical Loads für Europa statt. Der Workshop wurde vom Coordination Centre for Effects (CCE) organisiert und vom Schweizer Bundesamt für Umwelt (BAFU) ausgerichtet.
- Der Workshop wurde von 37 Teilnehmenden aus Österreich, Kanada, Finnland, Frankreich, Deutschland, den Niederlanden, Norwegen, Spanien, Schweden, der Schweiz, dem Vereinigten Königreich und den Vereinigten Staaten sowie von Vertretern des International Cooperative Programme (ICP) Waters, ICP Vegetation und ICP Modelling and Mapping besucht. Das Sekretariat der Konvention war nicht vertreten.
- Die Entscheidung, den Workshop zu organisieren, wurde auf der 39. Sitzung der Working Group on Effects (ECE/EB.AIR/144/Add.2) nach Empfehlungen der 35. Sitzung der Task Force Modelling and Mapping (2. - 4. April 2019) getroffen, die als Webkonferenz stattfand.
- Die Sitzung wurde von Reto Meier im Namen des BAFU und Alice James Casas als Vorsitzende des ICP Modelling and Mapping eröffnet.

II. Ziele und Struktur des Workshops

- Ziel des Workshops war die Überarbeitung der $CL_{emp}N$ für natürliche und naturnahe Ökosysteme, die auf dem letzten Expertenworkshop vom 23. bis 25. Juni 2010 in Noordwijkerhout festgelegt wurden (siehe Bobbink und Hettelingh, 2011), auf Grundlage zusätzlicher wissenschaftlicher Informationen für den Zeitraum von 2010 bis 2021. Die Diskussionen auf dem Workshop basierten auf dem neuen und aktualisierten Hintergrunddokument, das allen Teilnehmern vor dem Workshop zugesandt worden war.
- Die meisten Habitatklassen des Europäischen Naturinformationssystems (European Nature Information System, EUNIS), die in der vorliegenden Überarbeitung behandelt wurden, haben sich seit der letzten Aktualisierung durch Bobbink und Hettelingh (2011) geändert: Marine Lebensräume (EUNIS-Klasse MA, früher A), Küstenlebensräume (EUNIS-Klasse N, früher B), Binnengewässer (EUNIS-Klasse C), Moore und Sümpfe (EUNIS-Klasse Q, früher D), Grasland und von krautigen Pflanzen, Moosen oder Flechten dominierte Flächen (EUNIS-Klasse R, ehemals E), Heideland, Buschland und Tundra (EUNIS-Klasse S, ehemals F), Wälder, Forste und andere bewaldete Flächen (EUNIS-Klasse T, ehemals G).

- Ein internationales Team von 43 Expertinnen und Experten erstellte das Hintergrunddokument für den Workshop im Zeitraum von Juni 2020 bis September 2021, nach einem digitalen Kick-off-Meeting im Juni 2020:
  - EUNIS Klasse MA – Roland Bobbink
  - EUNIS-Klasse N – Laurence Jones, Emiel Brouwer und Eva Remke
  - EUNIS-Klasse C – Heleen De Wit, Roland Bobbink, Christin Loran, Linda May und Jan-Erik Thrane
  - EUNIS-Klasse Q - Chris Field, Julian Aherne, Roland Bobbink und Hilde Tomassen
  - EUNIS-Klasse R - Carly Stevens, Rocio Alonso, Vegar Bakkestuen, Erika Hiltbrunner und Lukas Kohli
  - EUNIS-Klasse S - Leon Van den Berg, Julian Aherne, Andrea Britton, Simon Caporn, Héctor García Gómez und Liv Guri Velle
  - EUNIS-Klasse T – Sabine Braun, Rocio Alonso, Frank Ashwood, Tomáš Chuman, Lucienne De Witte, Thomas Dirnböck, Per Erik Karlsson, Sirkku Manninen, Michael Perring, Hans Tömmervik, Simon Tresch, Liisa Ukonmaanaho und Elena Vanguelova
  - Kapitel 10 "Verwendung von empirischen Critical Loads für Stickstoff für Risikobewertung und Naturschutz" - Markus Geupel, Khalid Aazem, Sabine Augustin, Jesper Bak, Alice James Casas, Laurence Jones, Christin Loran, Reto Meier, Anne-Katrin Prescher, Thomas Scheuschner, Axel Ssymank und Susan Zappala
- Nach der internen Überprüfung, wurde das Hintergrundpapier überprüft durch Sabine Augustin, Ariel Bergamini, Leonor Calvo Galván, Tara Greaver, Kevin Hicks, Raúl Ochoa-Huesa, Tonje I. Økland und Jan Roelofs.
- Die Arbeitsgruppen tauschten sich in kurzen Sitzungen über ihre Fortschritte aus. Ergebnisse, Schlussfolgerungen und Empfehlungen wurden in einer abschließenden Vollversammlung unter dem Vorsitz von Christin Loran diskutiert und zusammengefasst.

III. Schlussfolgerungen

- Statistisch und biologisch signifikante Ergebnisse von Feldversuchen waren die Grundlage für die Bewertung der $CL_{emp}N$. Für die Aktualisierung und Verfeinerung der $CL_{emp}N$-Werte wurden nur Studien mit unabhängigen N-Behandlungen und realistischen N-Frachten und -Zeiträumen herangezogen.
- Im aktuell überprüften Zeitraum wurde eine zunehmende Anzahl von Gradientenstudien zur atmosphärischen N-Deposition in mehreren EUNIS-Lebensraumtypen veröffentlicht, die sich als nützlich für die Bewertung und Festlegung der $Cl_{emp}N$ erwiesen haben.
- Studien mit höheren N-Zugaben oder kürzeren Versuchszeiträumen wurden nur im Hinblick auf das Verständnis der Wirkungsmechanismen, einer möglichen N-Limitierung oder der Empfindlichkeit des Systems interpretiert. Die in diesen Studien angewandten Methoden wurden sorgfältig geprüft, um Faktoren des Versuchsaufbaus oder der Datenanalyse zu identifizieren, die ihre Verwendung bei der Bewertung der $CL_{emp}N$-Bereiche einschränken könnten.

- Für alle untersuchten EUNIS-Klassen (Stufe 2 oder 3) wurde eine Einigung über $CL_{emp}N$ für eine Reihe von N-Depositionswerten erzielt. Neue Ergebnisse zu den Auswirkungen von Stickstoff in Oberflächengewässern konnten auf der Grundlage der von ICP Waters vorgestellten Aktivitäten einbezogen werden. Neue Erkenntnisse für einige mediterrane Lebensräume konnten ebenfalls aufgenommen werden.
- Das neue Kapitel 10 "Verwendung von $CL_{emp}N$ in der Risikobewertung und im Naturschutz" wurde vorgestellt und diskutiert. Es enthält eine Auswahl von Beispielen für die Verwendung von $CL_{emp}N$ auf unterschiedlichen Skalen und in unterschiedlichen europäischen Ländern, um Fachleuten und politischen Entscheidungsträgern eine Anleitung zu geben, wie $CL_{emp}N$ in der Praxis eingesetzt werden kann.
- Die Bewertung der Zuverlässigkeit der $CL_{emp}N$-Bereiche wurde seit der letzten Aktualisierung im Jahr 2011 fortgesetzt, wobei zwischen "zuverlässig", "ziemlich zuverlässig" und "Expertenurteil" unterschieden wurde, symbolisiert jeweils durch ##, # bzw. (#).
- Die aus dem Überprüfungs- und Revisionsverfahren resultierenden $CL_{emp}N$-Bereiche wurden auf dem Workshop einvernehmlich festgelegt und sind in Tabelle 1 zusammengefasst. Bei mehr als 40 % der vorgestellten EUNIS-Typen war der untere Wert der Spanne niedriger als 2011. Das Gleiche gilt für den oberen Wert. Nur in einem Fall hat sich der obere Wert erhöht.

IV. Empfehlungen

- Für die folgenden Ökosysteme sind weitere Untersuchungen und Daten erforderlich, um ein CLempN zu erstellen: mehrere Gras- und Steppenlandschaften, alle mediterranen Vegetationstypen, feuchte (Sumpf-)Wälder, viele Moore und Flachmoore sowie mehrere Küstenlebensräume; darüber hinaus ist weitere Forschung für alle EUNIS-Lebensraumtypen erforderlich, die ein Experten-Rating haben.
- Die Auswirkungen der N-Anreicherung in (sensitiven) Süßwasser- und flachen Meeresökosystemen (einschließlich Küstengewässern) müssen weiter erforscht werden.
- Es werden mehr gut konzipierte Gradientenstudien mit sowohl (sehr) niedrigen als auch hohen N-Belastungen benötigt, insbesondere in EUNIS-Lebensraumtypen, die kaum untersucht werden. Darüber hinaus erhöht das Zusammenführen von Ergebnissen aus experimentellen und Gradientenstudien die Zuverlässigkeit der $CL_{emp}N$.
- Die unterschiedlichen Auswirkungen der abgelagerten N-Formen (NO_x oder NH_y) müssen weiter erforscht werden, um die Critical Loads für oxidierten und reduzierten Stickstoff in Zukunft getrennt bestimmen zu können.
- Zur Weiterentwicklung der aktuellen $CL_{emp}N$ sind Langzeitexperimente (10-20 Jahre) mit einer hohen N-Zugabehäufigkeit zwischen 5 und 50 kg N ha^{-1} yr^{-1} in Regionen mit geringer Hintergrunddeposition von entscheidender Bedeutung. Dies würde die Zuverlässigkeit der abgeleiteten CLempN erhöhen, wenn die niedrigste Behandlungsstufe den derzeitigen Critical Load nicht überschreitet.
- Der Klimawandel und die Stickstoffdeposition haben wahrscheinlich starke interaktive Auswirkungen auf die Funktionsweise von Ökosystemen, wobei der Klimawandel die Reaktionen der Ökosysteme auf die Stickstoffdeposition verändert und umgekehrt. Es werden mehr experimentelle Studien benötigt, um diese Wechselwirkungen zu untersuchen,

sowie mehr Gradientenstudien, die ausdrücklich die Auswirkungen der Stickstoffdeposition in Kombination mit klimatischen Gradienten untersuchen.

- Zusammenfassend lässt sich sagen, dass es von entscheidender Bedeutung ist, die langfristigen Auswirkungen einer erhöhten N-Deposition auf Ökosystemprozesse in einer repräsentativen Auswahl von Ökosystemen zu verstehen. Daher ist es wichtig, die Auswirkungen der N-Deposition durch manipulierte N-Einträge in langfristigen Ökosystemstudien sowohl in unberührten als auch in beeinträchtigten Gebieten zu quantifizieren. Diese Daten sind in Verbindung mit Gradientenstudien unerlässlich, um Critical Loads zu validieren und robuste dynamische Ökosystemmodelle und/oder Modelle für mehrere korrelierende Arten zu entwickeln, die zuverlässig genug sind, um $CL_{emp}N$ für natürliche und naturnahe Ökosysteme zu berechnen und natürliche Erholungsraten für N-belastete Systeme vorherzusagen.

Table 1. Überblick über die empirischen Critical Loads von Stickstoff (kg N ha^{-1} a^{-1}) für natürliche und naturnahe Ökosysteme (Spalte 1), klassifiziert nach EUNIS (Spalte 2), wie 2011 ermittelt (Spalte 3) und wie 2022 überarbeitet (Spalte 4). Die Verlässigkeit wird durch ## zuverlässig; # ziemlich zuverlässig und (#) Expertenurteil angegeben (Spalte 5). Spalte 6 zeigt eine Auswahl von Effekten, die bei Überschreitung der Critical Loads auftreten können. Änderungen im Vergleich zu 2011 sind als fettgedruckte Werte angegeben.

Ökosystemtyp	EUNIS Code	2011 kg N ha^{-1} a^{-1}	2022 kg N ha^{-1} a^{-1}	2022 Verlässlich-keit	Überschreitungsindikator
Marine Habitate (MA)					
Atlantische mäßig-stark salzbeeinflusste Wiesen	MA223	20-30	**10-20**	(#)	Zunahme der Dominanz von Graminoiden; Rückgang positiver Indikatorarten
Atlantische niedrige-mäßig salzbeeinflusste Wiesen	MA224	20-30	**10-20**	(#)	Zunahme von Arten später Sukzessionsstadien; Abnahme von positiven Indikatorarten
Atlantische Pionier salzbeeinflusste Wiese	MA225	20-30	20-30	(#)	Zunahme von Arten später Sukzessionsstadien; Zunahme von produktiven Arten
Küstenhabitate (N)					
Wanderdünen der Küsten	N13, N14	10-20	10-20	**#**	Zunahme der Biomasse; verstärkte N-Auswaschung; reduzierte Wurzelbiomasse
Stabile Küstendünen (graue Dünen)	N15	8-15	**5**-15	**##**	Erhöhte Biomasse und Bodenbedeckung durch Graminoiden und mesophilen Gräsern; Rückgang der oligotrophen Arten, einschließlich Flechten; erhöhter N-Gehalt im Gewebe; verstärkte N-Auswaschung; Bodenversauerung

Ökosystemtyp	**EUNIS Code**	**2011 kg N ha^{-1} a^{-1}**	**2022 kg N ha^{-1} a^{-1}**	**2022 Verlässlich-keit**	**Überschreitungsindikator**
Dünenheiden an der Küste	N18, N19	10-20	10-**15**	**#**	Erhöhte Pflanzenproduktion; verstärkte N-Auswaschung; beschleunigte Sukzession; typische C:N Abnahme in Flechten; erhöhter jährlicher *Calluna* Zuwachs
Feuchte und nasse Dünentümpel	N1H	10-20	**5-15**	**#**	Erhöhte Bodenbedeckung durch Graminoiden und mesophilen Gräsern; Abnahme von oligotrophen Arten; erhöhter Ellenberg N-Wert
Dünentümpel Wasserbecken (Aquatische Süßwassergemein-schaften von permanenten atlantischen und baltischen oder mediterranen und Schwarzmeer Dünentümpelgewässern)	N1H1, N1J1	10-20	10-20	(#)	Zunehmende Biomasse und Sukzessionsrate
Süßwasserhabitate (C) [a]					
Dauerhaft oligotrophe Stillgewässer (einschließlich Weichwasserseen)	C1.1	3-10	**2**-10 [b]	##	Erhöhte Algenproduktivität und Wechsel der Nährstofflimitierung für Phytoplankton von N auf P; Veränderungen der Makrophytengemeinschaft
Alpine und subarktische Klarwasserseen	C1.1		**2-4**	**##**	Erhöhte Algenproduktivität und Wechsel der Nährstofflimitierung für Phytoplankton von N auf P
Boreale Klarwasserseen	C1.1		**3-6**	**##**	Erhöhte Algenproduktivität und Wechsel der Nährstofflimitierung für Phytoplankton von N auf P
Atlantische weiche Gewässer	C1.1, Elemente C1.2	3-10	**5**-10	##	Veränderung der Artenzusammensetzung von Makrophytengemeinschaften
Dauerhaft dystrophe Stillgewässer	C1.4	**3-10**	**5**-10 [c]	(#)	Erhöhte Algenproduktivität und Wechsel der Nährstofflimitierung für Phytoplankton von N auf P
Sumpf- und Moorhabitate (Q)					

Ökosystemtyp	EUNIS Code	2011 kg N ha^{-1} a^{-1}	2022 kg N ha^{-1} a^{-1}	2022 Verlässlich-keit	Überschreitungsindikator
Hoch- und Deckenmoore	Q1	5-10	5-10	##	Zunahme der Gefäßpflanzen; Rückgang der Moose; verändertes Wachstum und veränderte Artenzusammensetzung von Moosen; erhöhter N-Gehalt in Torf und Torfwasser
Nährstoffärmere und nährstoffarme Niedermoore	Q2	10-15	**5**-15	**##**	Zunahme von Riedgräsern und anderen Gefäßpflanzen; negative Auswirkungen auf Bryophyten
Palsa- und Polygonmoore	Q3		**3-10**	**(#)**	Zunahme der Graminoiden, der N-Konzentration im Gewebe und der Zersetzungsrate
Nährstoffreiche Niedermoore	Q41-Q44	15-30	15-**25**	**#**	Zunahme hoher Gefäßpflanzen (insbesondere Graminoide); Rückgang von Moosen
Nährstoffreiche arktische und Berg-Flachmoore	Q45	15-25	15-25	(#)	Zunahme von Gefäßpflanzen; Rückgang von Moosen
Graslandhabitate (R)					
Kalkreicher Halbtrockenrasen (basische Wiesensteppe)	R1A	15-25	**10-20**	##	Zunahme der Hochgräser; Rückgang der Artenvielfalt; Veränderung der Artenzusammensetzung; verstärkte Mineralisation; N-Auswaschung; Oberflächenversauerund
Mediterraner, dicht beweideter Trockenrasen oder Mediterraner hoher mehrjähriger Trockenrasen oder Mediterraner jährlicher Trockenrasen	R1D oder R1E oder R1F	**15-25**	**5-15**	(#)	Erhöhte Produktion; Dominanz von Graminoiden; Veränderung der Bodenkruste; Veränderung des Nährstoffkreislaufs im Boden
Tief- bis Bergland, trockener bis mesischer Rasen, gewöhnlich dominiert von *Nardus stricta*	R1M	10-15	**6-10**	##	Zunahme der Graminoiden; Rückgang der typischen Arten; Rückgang des gesamten Artenreichtums
Ozeanischer bis subkontinentaler Binnen-Sandrasen auf trockenen sauren und neutralen Böden	R1P oder R1Q	8-15	**5**-15	(#)	Rückgang der Flechten; Zunahme der Biomasse

Ökosystemtyp	**EUNIS Code**	**2011 kg N ha^{-1} a^{-1}**	**2022 kg N ha^{-1} a^{-1}**	**2022 Verlässlich-keit**	**Überschreitungsindikator**
oder Sandverwehungen und Dünen mit silikatischem Grasland					
Mähwiesen tiefer und mittlerer Lagen	R22	20-30	**10-20**	(#)	Zunahme an Hochgräsern; Rückgang der Artenvielfalt; Rückgang typischer Arten
Bergmähwiesen	R23	10-20	10-**15**	**#**	Zunahme der nitrophilen Graminoiden; Veränderung der Artenvielfalt; Rückgang typischer Arten
Feuchte oder nasse mesotrophe bis eutrophe Mähwiesen	R35	15-25	15-25	(#)	Zunahme der hohen Graminoiden; geringere Artenvielfalt; Rückgang der Moose
Gemäßigte und boreale feuchte und nasse oligotrophe Rasen	R37	**10-20**	10-20	#	Zunahme der hohen Graminoiden; geringere Artenvielfalt; Rückgang der Moose
Moos- und flechtendominierte Berggipfel	**(Früher E4.2)**	5-10	5-10	#	Veränderungen in der Artenzusammensetzung; Auswirkungen auf Bryophyten oder Flechten
Gemäßigte acidophile alpine Rasen	R43	5-10	5-10	#	Veränderungen in der Artenzusammensetzung; Zunahme der Pflanzenproduktion
Arktisch-alpine kalkhaltige Rasen	R44	5-10	5-10	#	Veränderungen in der Artenzusammensetzung; Zunahme der Pflanzenproduktion
Heiden- und Strauchhabitate (S)					
Tundra	S1	3-5	3-5 [d]	#	Veränderungen in der Biomasse; physiologische Effekte; Veränderungen in der Artenzusammensetzung der Moospflanzen; Rückgang des Flechtenartenreichtums
Arktische, alpine und subalpine Zwergstrauchheiden	S2	5-15	**5-10** [d]	#	Rückgang der Flechten, Bryophyten und immergrünen Sträucher
Tief- bis bergländisches gemäßigtes und submediterranes *Juniperus* Buschland	S31		**5-15**	**(#)**	Verschiebung der Zusammensetzung der Vegetationsgemeinschaft;

Ökosystemtyp	**EUNIS Code**	**2011 kg N ha^{-1} a^{-1}**	**2022 kg N ha^{-1} a^{-1}**	**2022 Verlässlich-keit**	**Überschreitungsindikator**
					geringere Lebensfähigkeit der Samen
Nördliche feuchte Heide	S411				
▶ 'U' *Calluna*-dominierte feuchte Heide (Bergland)	S411	10-20	**5–15** [e]	**##**	Abnehmende Heidedominanz; Rückgang von Flechten und Moosen; erhöhte N-Auswaschung
▶ 'L' *Erica tetralix*-dominierte feuchte Heide (Tiefland)	S411	10-20	**5-15** [e]	**##**	Übergang von der Dominanz von Heidekraut zu Gräsern; Rückgang der Heidekrautbedeckung; Veränderung der Zusammensetzung der Vegetationsgemeinschaft
Trockene Heiden	S42	10-20	**5-15** [e]	##	Übergang von der Heidekraut- zur Grasdominanz; Rückgang der Flechten; Veränderungen in der Biochemie der Pflanzen; erhöhte Empfindlichkeit gegenüber abiotischem Stress
Macchia, baumförmige Hartlaubgebüsche und thermo-mediterranes Buschland	S5	20-30	**5-15**	(#)	Veränderung des Artenreichtums und der Zusammensetzung der Pflanzengemeinschaften; Nitratauswaschung; Versauerung des Bodens.
Garrigue	S6		**5-15**	**#**	Veränderungen in der Artenzusammensetzung; Rückgang der Strauchbedeckung; zunehmende Invasion von einjährigen Kräutern
Wälder und Forsten (T)					
Sommergrüne Laubwälder	T1	**10-20**	10-**15**	##	Veränderungen in Bodenprozessen; Nährstoffungleichgewicht; Veränderungen der Mykorrhiza und Bodenvegetation
Buchenwald auf nicht-sauren und sauren Böden	T17, T18	**10-20**	10-**15**	(#)	Veränderungen der Bodenvegetation und Mykorrhiza; Nährstoffungleichgewicht; Veränderungen der Bodenfauna

Ökosystemtyp	**EUNIS Code**	**2011 kg N ha^{-1} a^{-1}**	**2022 kg N ha^{-1} a^{-1}**	**2022 Verlässlich-keit**	**Überschreitungsindikator**
Mediterraner bodensaurer Buchenwald	T18		**10-15**	**(#)**	Jährliches Höhen- und Volumenwachstum der Bäume; Analogie zu Buchenwäldern der gemäßigten Zonen
Bodensaurer eichendominierter Wald	T1B	10-15	10-15	(#)	Rückgang der Mykorrhiza; Verlust von epiphytischen Flechten und Bryophyten; Veränderungen der Bodenvegetation
Eichen-Hainbuchen mesischer Laubwald	T1E	15-20	15-20	(#)	Veränderungen der Bodenvegetation
Mediterraner immergrüner Eichenwald	T21	**10-20**	10-**15**	(#)	NO_3 im Bodenwasser und Wasserläufen
Nadelwälder	T3	**5-15**	**3**-15	##	Veränderungen der Bodenprozesse; Nährstoffungleichgewicht; veränderte Zusammensetzung von Mykorrhiza und Bodenvegetation; Zunahme der Sterblichkeit bei Trockenheit
Gemäßigter Fichtenbergwald Gemäßigter Tannenbergwald	T31, T32	**10-15**	10-15	(#)	Verminderte Biomasse der Feinwurzeln; Nährstoffungleichgewicht; Rückgang der Mykorrhiza; veränderte Bodenfauna
Mediterraner Tannenbergwald	T33		**10-15**	**(#)**	Blattstöchiometrie der Bäume; Baumphysiologie; N-Verluste im Boden
Gemäßigter kontinentaler *Pinus sylvestris* Wald	T35	5-15	5-15	#	Veränderungen der Bodenvegetation und der Mykorrhiza; Nährstoffungleichgewicht; erhöhte N_2O- und NO-Emissionen
Mediterraner *Pinus sylvestris-Pinus nigra* Bergwald	T37		**5-17**	**(#)**	Flechtenchemie und Veränderungen der Lebensgemeinschaften in mediterranen Mischwäldern in den USA
Mediterraner Tiefland- bis submontaner Kiefernwald	T3A	**3-15**	**5-10**	(#)	Verringerung der Feinwurzelbiomasse; Veränderung der Flechtengemeinschaft
Dunkle Taiga	T3F	**5-10**	**3-5** [f]	##	Veränderungen der epiphytischen Flechten- und

Ökosystemtyp	EUNIS Code	2011 kg N ha^{-1} a^{-1}	2022 kg N ha^{-1} a^{-1}	2022 Verlässlich-keit	Überschreitungsindikator
					Bodenschicht-Bryophytengemeinschaften; Zunahme der freilebenden Algen; Rückgang der N-Fixierung
Pinus sylvestris helle Taiga	T3G	**5-10**	**2-5** [f]	#	Veränderungen der epiphytischen Flechten- und Bodenschicht-Bryophytengemeinschaften; Zunahme der freilebenden Algen; Rückgang der N-Fixierung

[a)] Die Untergrenze des $CL_{emp}N$-Bereichs sollte für Seen in kleinen Einzugsgebieten (mit einem hohen Verhältnis zwischen See und Einzugsgebiet) angewandt werden, da diese der atmosphärischen Deposition am stärksten ausgesetzt sind, da ein relativ hoher Anteil ihrer N-Einträge direkt auf den Seen abgelagert und nicht in den Einzugsgebieten zurückgehalten wird. In ähnlicher Weise sollte die Untergrenze des $CL_{emp}N$-Bereichs für Seen in Einzugsgebieten mit dünnen Böden, spärlicher Vegetation und/oder einem hohen Anteil an nacktem Felsen angewendet werden.

[b)] Dieser $CL_{emp}N$-Wert sollte nur auf oligotrophe Gewässer mit geringer Alkalinität und ohne nennenswerte landwirtschaftliche oder sonstige menschliche Einträge angewendet werden. Die Untergrenze des Bereichs gilt für subarktische und alpine Klarwasserseen, der mittlere Bereich für boreale Seen und die Obergrenze des Bereichs für atlantische Weichgewässer.

[c)] Dieser $CL_{emp}N$-Wert sollte nur auf Gewässer mit geringer Alkalinität und ohne nennenswerte landwirtschaftliche oder sonstige direkte menschliche Einträge angewendet werden. Die Untergrenze des Bereichs sollte auf boreale dystrophe Seen angewendet werden.

[d)] Verwendung der Obergrenze, wenn der Phosphorgehalt begrenzt ist, und der Untergrenze, wenn der Phosphorgehalt nicht begrenzt ist.

[e)] Verwendung der Obergrenze bei hoher Intensität und Verwendung der Untergrenze bei niedriger Intensität.

[f)] Hauptsächlich basierend auf die Auswirkungen der N-Deposition auf Flechten und Moose.

1 Introduction

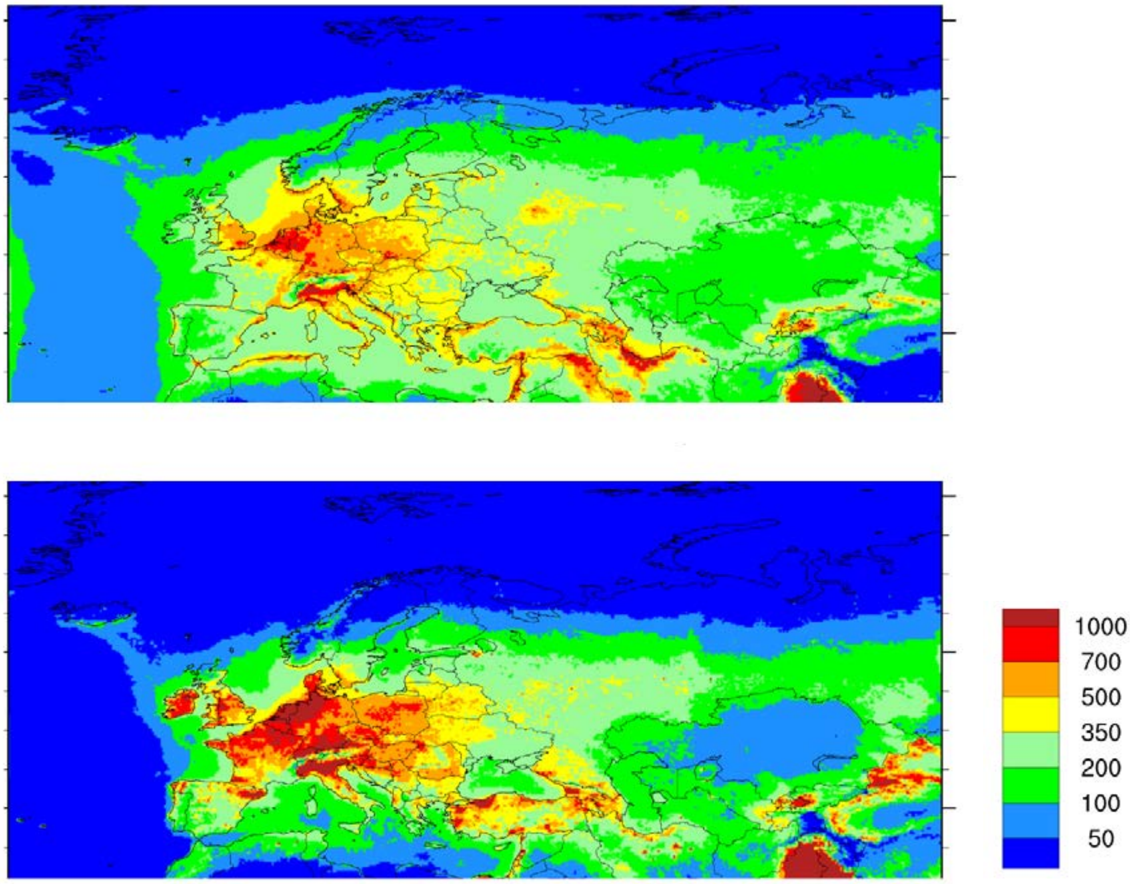
Adapted by Roland Bobbink, Christin Loran and Hilde Tomassen

Nitrogen addition experiments are of utmost importance for the setting of empirical critical loads: overview of the experimental N addition facility at Whim Bog, Scotland, UK. Photo: Roland Bobbink.

1.1 Impacts of N deposition

Emissions of ammonia (NH_3) and nitrogen oxides (NO_x) strongly increased in the second half of the 20th century. Ammonia is volatilised from intensive agricultural systems, such as dairy farming and intensive animal husbandry, whereas nitrogen oxides originate mainly from burning of fossil fuel by traffic, industry and households. Because of short- and long-range transport of these nitrogenous compounds, atmospheric nitrogen (N) deposition has clearly increased in many natural and semi-natural ecosystems across the world. Areas with high atmospheric N deposition (20-80 kg N ha^{-1} yr^{-1}) nowadays are central and western Europe, eastern United States and, since the 1990s, eastern Asia (e.g. Galloway and Cowling, 2002; Dentener et al., 2006; Fowler et al., 2020). The modelled deposition for Europe is published annually by EMEP/MSC-West (Figure 1.1).

Figure 1.1. Modelled total depositions of oxidised (upper map) and reduced (lower map) nitrogen (mg N m^{-2} yr^{-1}] across Europe in 2019. 1000 mg N m^{-2} = 10 kg N ha^{-1}.

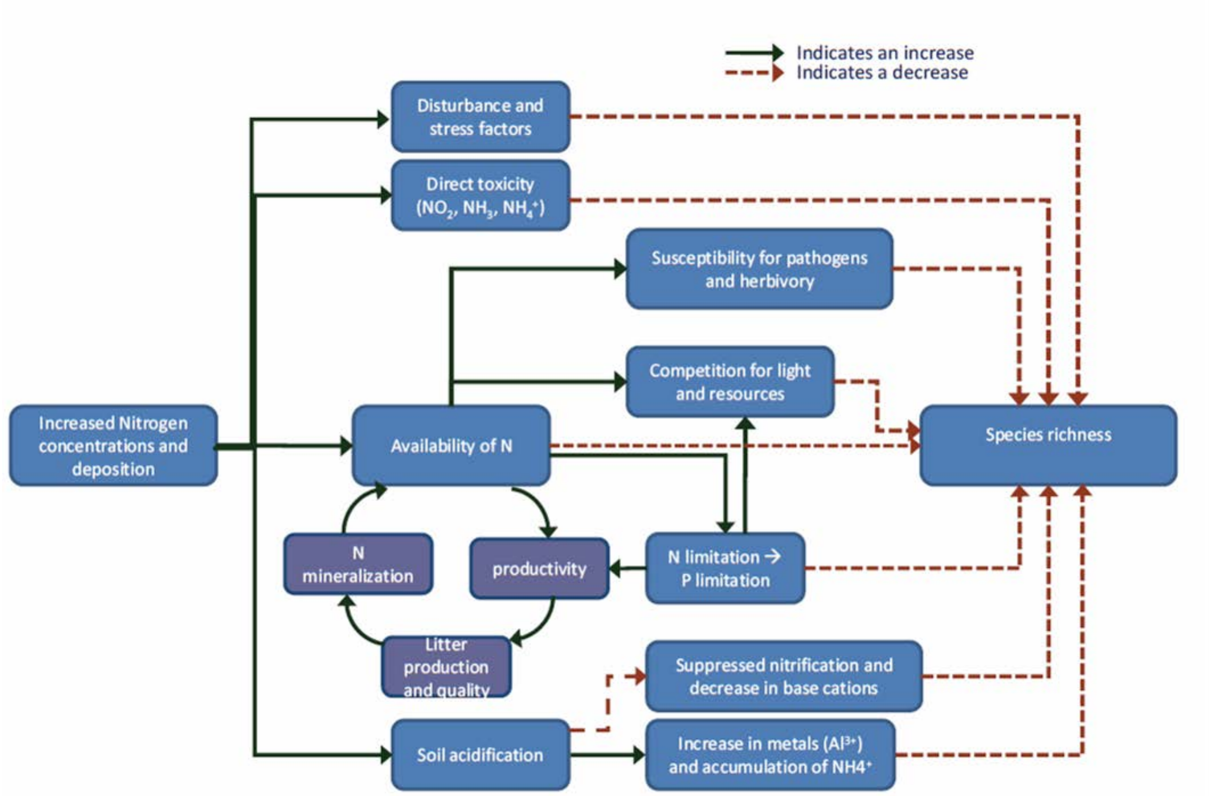


Source: EMEP Status Report, 2021

The availability of nutrients is one of the most important abiotic factors which determine plant species composition in ecosystems. N is the primary limiting nutrient for plant growth in many natural and semi-natural ecosystems, especially for oligotrophic and mesotrophic habitats. Most of the plant species in such ecosystems are adapted to nutrient-poor conditions and can only survive or compete successfully on soils with low N availability (e.g. Tamm, 1991; Aerts and Chapin, 2000). The series of events which occurs when N inputs increase in an area with originally low background deposition rates is highly complex. Many ecological processes interact and operate at different temporal and spatial scales. As a consequence, high variations in sensitivity to atmospheric N deposition have been observed between different natural and semi-natural ecosystems. Despite this diverse sequence of events, the most obvious effects of increased N deposition are significant changes in the N cycle, vegetation composition and biodiversity. For more details, see Bobbink et al. (1998, 2010), Dise et al. (2011) and Stevens et al. (2020).

Many ecological processes interact and operate at different temporal and spatial scales. Furthermore, N is the limiting nutrient for plant growth in many natural and semi-natural ecosystems, especially oligotrophic and mesotrophic habitats. The severity of the impacts of atmospheric N deposition depends on a number of factors, of which the most important are (numbers not being a ranking): 1) the duration and total amount of inputs, 2) the chemical and physical form of the airborne N input, 3) the intrinsic sensitivity of the plant and animal species present, 4) the abiotic conditions, including climate, and 5) the past and present land use or management. Acid neutralising capacity (ANC), soil nutrient availability, and soil factors that influence the nitrification potential, N immobilisation and denitrification rates, are especially important. As a consequence, different ecosystems show high variability in sensitivity to atmospheric N deposition (Bobbink et al., 2010; Pardo et al., 2011). Despite this highly diverse sequence of events, it is possible to generalise some types of impacts. A schematic overview of the potential sequence of events is given in Figure 1.2.

Figure 1.2. Scheme of the main impacts of increased N deposition on ecosystems. Stress is considered to occur when external constraints limit the rate of dry matter production of the vegetation, whereas disturbance consists of mechanisms which affect soils and plant biomass by causing its partial or total destruction.



Source: Bobbink and Hettelingh, 2011

a) **Direct toxicity of N gases and aerosols to individual species**

An important effect of nitrogenous gases, aerosols and dissolved compounds (NH_3, NO_2, NO, HNO_3 and NH_4^+) can be direct toxicity to the above-ground parts of individual plants. The impacts have been mostly studied in crops and saplings, but studies with native plant species or mixtures of species in open-top chambers (OTCs) and free-air fumigation have also demonstrated leaf injury, changes in physiology, and growth reductions at increased concentrations of just-mentioned N pollutants (e.g. Pearson and Stewart, 1993; Grupa, 2003; Sheppard et al., 2009). Direct toxicity impacts of NO_2 were observed in parts of Europe and North America in the 1980s, but are currently rare in these regions, except in cities or in the direct neighbourhood of roads with heavy traffic. However, concentrations of these nitrogen oxides in air are now increasing in large areas of Asia (primarily China and India), possibly leading again to direct foliar impacts. In addition, lichens are clearly the most sensitive group in the vegetation with respect to direct toxicity of NH_3 (e.g. Hallingbäck, 1992; Van Herk et al., 2003). This, based on data from the United Kingdom, Italy and Portugal, has led to a significant lowering of the long-term critical level of NH_3 for ecosystems in which lichens and bryophytes are important (Cape et al., 2009; Sutton et al., 2009). Furthermore, it became obvious that the exceedances of this critical level occur in many areas of North-western Europe (Sutton et al., 2009).

b) **Eutrophication**

N is the limiting nutrient for plant growth in many natural and semi-natural terrestrial ecosystems, especially under oligotrophic and mesotrophic conditions. Increased N deposition results in an increase in the availability of inorganic N in the topsoil, in the short term, except in bogs and fens. This gradually leads to an increase in plant productivity in N-limited vegetation, and thus to higher annual litter production and litter with high concentrations of N. Because of this, N mineralisation will also gradually increase, which, in turn, may increase plant productivity. This is positive feedback, because higher N mineralisation leads to higher N uptake and its subsequent effects. Local plant species diversity increases with increasing resource availability at originally very low levels of resource availability. Above a certain level of primary productivity, however, local plant species diversity declines as production increases. Observational studies across N deposition gradients, and many N addition experiments, demonstrate this effect in the long term. Competitive exclusion ('overshading') of characteristic species of oligotrophic or mesotrophic habitats occurs in the presence of relatively fast-growing nitrophilic species, with rare species at low abundances being especially at risk (Figure 1.3) (e.g. Bobbink et al., 1998; Suding et al., 2005).

Figure 1.3. A calcareous grassland (*Mesobromion erecti*) (R1A) in the Netherlands without N addition (left) and after three years of N addition (100 kg N ha^{-1} yr^{-1}) (right).

Source: R. Bobbink

The rate of N cycling in the ecosystem is clearly increased in such situations, although the response time to increased N inputs can be long in highly organic soils (with high C:N ratios), or, indeed in any soil with large potential N sinks. When N is no longer limiting in the ecosystem, plant growth becomes limited by other resources, such as phosphorus (P), potassium (K), magnesium (Mg), or water. In this situation, the productivity of the vegetation will not increase any further with continuing increases in N. However, N concentrations within the plants do tend to increase when N availability continues to increase. This may affect the palatability of the vegetation for herbivores or the sensitivity to pathogens (see below), and will influence microbial communities, too. Recently, it has been suggested that after a shift from N to P limitation or in highly P-limited situations, changes in plant species composition can gradually still occur under long-term N inputs (see Chapter 6 for examples).

c) **Acidification**
Soil acidification is characterised by a wide variety of long-term effects. It is defined as the loss of acid neutralising capacity (ANC) and may lead to a decrease in soil pH. Changes in pH are dependent on the buffering capacity of the soil (e.g. Ulrich 1983, 1991). Acidifying compounds (N and S) deposited on calcareous soils (including substrates of young moraine regions) at first will not change soil acidity. In these soils HCO_3^- and Ca^{2+} ions leach from the system, but the pH remains the same until almost all of the calcium carbonate has been depleted. In soils dominated by silicate minerals (pH 6.5-4.5), buffering is taken over by cation exchange processes of the soil adsorption complexes. In this situation, protons are exchanged with Ca^{2+} and Mg^{2+}, and these cations are leached from the soil together with anions (mostly nitrate or sulphate). Because of the restricted capacity of this buffering system, soil pH will soon start to decrease. However, in mineral soils with a large cation exchange capacity and high base saturation, this buffering may continue for several decades, even at relatively high inputs.

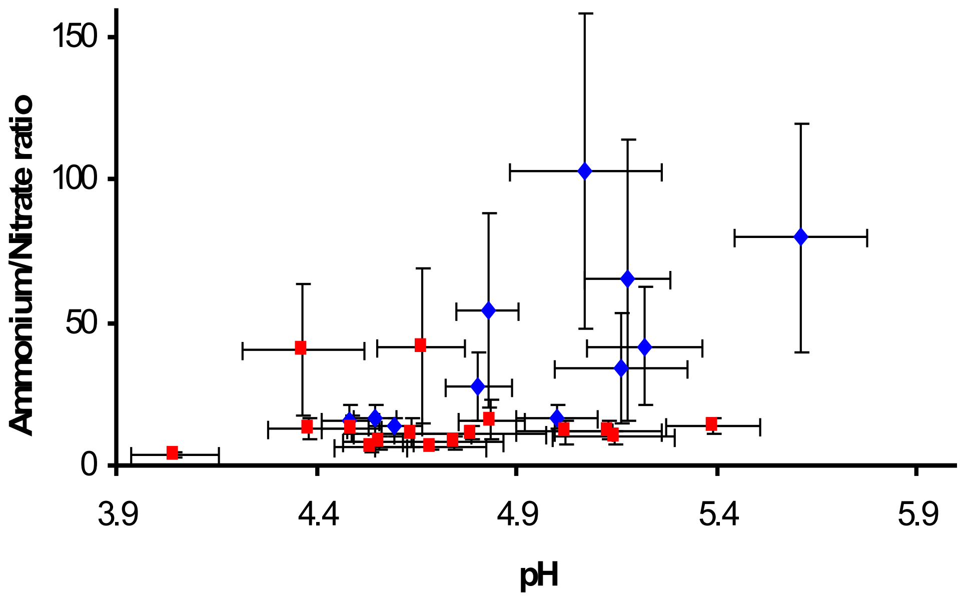
At low pH (< 5.0), hydrous oxides of several metals dissolve. This causes a strong increase in the levels of toxic Al^{3+} and other metals in the soil solution. As a result of the decrease in pH, nitrification is strongly hampered or even completely absent in most of these highly acidic soils. This may lead to accumulation of ammonium, with nitrate levels decreasing to almost zero (e.g. Roelofs et al., 1985). In addition, the decomposition rate of organic material in the soil is lower in these acidified soils, which leads to increased accumulation of litter (e.g. Van Breemen et al., 1982; Ulrich, 1983, 1991). As a result of this cascade of changes, plant growth and species composition of the vegetation can be seriously affected: acid-resistant plant species will gradually become dominant, and several species typical to intermediate and higher soil pH will disappear.

d) **Differences in effects of oxidised versus reduced N**
Emissions of ammonia (NH_3) and nitrogen oxides (NO_x) both contribute to atmospheric N deposition. Ammonia is volatilised from agricultural systems, such as dairy farming and intensive animal husbandry, whereas nitrogen oxides originate mainly from burning of fossil fuels in economic sectors including traffic (also by vehicle catalysts which may be a locally important source of N deposition), households and industry (Truscott et al., 2005). Because of this difference in sources (i.e. agriculture vs industry, households or traffic) and different rates of deposition from the atmosphere, the spatial and temporal patterns of deposition differ between reduced and oxidised compounds. Oxidised N deposition prevails in urban or industrial areas, whereas reduced N deposition clearly dominates in agricultural or rural regions. Furthermore, in most regions with a relatively high rate of N deposition, a high proportion of the deposited N originates from NH_y (e.g. Asman et al., 1998; Fowler, 2002; Sutton et al., 2008). This may cause a change in the dominant N form in the soil from nitrate to ammonium, especially in habitats with low rates of nitrification (pH < 4.5).

The response of sensitive plant species can be significantly affected by this change in N form. Species of calcareous or slightly acidic soils are able to use nitrate, or a combination of nitrate and ammonium, as their N source, whereas early studies showed that species of acidic habitats generally use ammonium (e.g. Gigon and Rorison, 1972; Kinzel, 1982), because at least some of these plants do not have nitrate reductase (Ellenberg, 1996). For several plant species reduced N appeared to be only toxic at low pH (Lucassen et al., 2003). Laboratory and field studies demonstrate that the performance of most forest understory species of deciduous forests in southern Sweden improves when not only ammonium but also nitrate can be taken up (Falkengren-Grerup, 1998; Olsson and Falkengren-Grerup, 2000).

One of the impacts of increased ammonium uptake is a reduced uptake of base cations and exchange of these cations (K^+, Ca^{2+} and Mg^{2+}) to the rhizosphere. Ultimately this can lead to severe nutritional imbalances, which are important in the decline in tree growth in areas with high ammonia/ammonium deposition (e.g. Nihlgård, 1985; Van Dijk et al., 1990; references in Bobbink et al., 2003). High concentrations of ammonium in the soil or water layer are also toxic to many sensitive plant species, disrupting cell physiology, cell acidification, accumulation of N-rich amino acids, poor root development, and finally, inhibition of shoot growth. Strong evidence exists that many endangered vascular plant species of grasslands, heathlands and soft-water lakes, and fen bryophytes, are very intolerant to increased concentrations of reduced N and to high NH_4^+:NO_3^- ratios (De Graaf et al., 1998; Paulissen et al., 2004; Kleijn et al., 2008; Van den Berg et al., 2008) (Figure 1.4).

Figure 1.4. Characterisation of growth sites of common (blue diamonds) and rare (red squares) species typical to Dutch heaths, matgrass swards and fen meadows in terms of pH and NH_4^+:NO_3^- ratio in the soil. Symbols indicate mean ± SE. In contrast to common species, almost all rare species occur only at a low NH_4^+:NO_3^- ratio.



Source: Kleijn et al., 2008

e) **Increased susceptibility to secondary stress and disturbance factors**
The sensitivity of plants to stress (defined here as external constraints, such as drought, frost, pathogens or herbivores, which limit dry-matter production rate), or disturbance factors, (mechanisms which affect plant biomass by causing its partial or complete destruction), may be significantly affected by N deposition. With increasing N deposition, susceptibility to fungal pathogens and attacks by insects also increases. This is probably due to altered concentrations of phenolic compounds (leading to lower resistance) and soluble N compounds, such as free amino acids, together with a lower vitality of individual

plants as a result of polluted deposition. Increased levels of pathogenic fungi have been found for several tree species in N addition experiments and field surveys, but for most ecosystems data are lacking and the influence of such pathogens on diversity is still unclear (e.g. Flückiger et al., 2002; Bobbink et al., 2003).

In general, herbivory is affected by the palatability of plant material, which is strongly determined by its N content. Increased organic N content in plants, caused by N deposition, can thus result in increased insect herbivory (e.g. Throop and Lerdau, 2004). Data on herbivory and N deposition are very scarce, but a link has been demonstrated in dry *Calluna* heathlands. The frequency and intensity of infestations of heather beetle (*Lochmaea suturalis*) are clearly related to atmospheric N inputs and N concentrations in the heather (e.g. Brunsting and Heil, 1985; Berdowski, 1993; Bobbink and Lamers, 2002; for details see Chapter 8). N-related changes in plant physiology, phenology, biomass allocation (root:shoot ratios) and mycorrhizal infection can also differentially influence the sensitivity of plant species to drought or frost stress, leading to reduced growth in some species and possible changes in plant interactions.

1.2 Background to, and aims of the report

Within the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention), procedures have been developed to model and map critical loads for airborne N deposition in support of effect-based European policies for the abatement of air pollution (Bull et al., 2001; Hettelingh et al., 2001, 2007). Both the steady-state mass balance method and the empirical approach are used to scientifically support European policies aiming at effective emission reductions of air pollutants (ICP M&M, 2017). For the support of these policies it is important that scientific knowledge be regularly updated with new findings. This report focuses on recent knowledge for the review and revision of empirical critical loads of N ($CL_{emp}N$).

$CL_{emp}N$ are in almost all cases based on observed changes in the structure and functioning of ecosystems, primarily in a) species abundance, composition and/or diversity ('structure'), or b) N leaching, decomposition or mineralisation rate ('functioning'). For a more complete overview of indicators, see Løkke et al. (2000). Effects have been evaluated for specific ecosystems. Statistically and biologically significant results from field addition experiments and mesocosm experiments conducted under close-to-field conditions have been used for quantifying $CL_{emp}N$. Only studies on independent N treatments with a duration of two years or more have been used. In particular data from long-term experiments in low-background areas are most useful for observing effects of N enrichment. However, since experimental studies have been conducted for a variety of reasons, their designs differ, and the methods used are carefully scrutinised to identify factors related to the experimental design or data analysis that may constrain their use. This includes evaluation of the accuracy of estimated values of background N deposition at experimental sites. In addition, the results from correlative or retrospective field studies have been used, but only as additional evidence to support conclusions from experiments, or as a basis for an 'expert judgement' rating.

$CL_{emp}N$ for natural and semi-natural ecosystems were first presented in a background document for the 1992 workshop on critical loads held under the UNECE Convention at Lökeberg (Sweden) (Bobbink et al., 1992). After detailed discussions, before and during the meeting, the proposed values were set at that meeting (Grennfelt and Thörnelöf, 1992). Additional information from the 1992-1995 period was evaluated and summarised in an updated background paper and published as Annex III (Bobbink et al., 1996) of the UNECE manual on methodologies and criteria for mapping critical levels and loads. The updated $CL_{emp}N$ were discussed and set with full consensus at the December 1995 expert meeting held under the UNECE Convention in Geneva (Switzerland). They were also used for the development of the second edition of the Air Quality Guidelines for Europe by the World Health Organization's Regional Office for Europe (WHO, 2000). Furthermore, the $CL_{emp}N$ deposition was extensively reviewed and updated in 2001-2002 (Berne workshop; Achermann and Bobbink, 2003). In that update, classification of the receptor ecosystems was brought in line with that of the European Nature Information System (EUNIS) (mostly level 3) (Davies and Moss, 2002; Hall et al., 2003; Davies et al., 2004), in addition to the incorporation of results from new N-impact studies from the 1996-2002 period (Bobbink et al., 2003). The last review and revision were in 2009/2011 (Noordwijkerhout workshop, Bobbink and Hettelingh, 2011). That revision incorporated N-impact studies from November 2002 to spring 2010. The CCE workshop in Noordwijkerhout included separate sessions of three Working Groups each addressing one or more EUNIS classes. The summary reports of Working Group 1, 2 and 3 can be found in the Appendices in Bobbink and Hettelingh (2011).

It was recognised at the CCE workshop and Task Force meetings of the International Cooperative programme on Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends in Madrid (ICP M&M, 2019) that considerable new insights into, and data on, the impacts of N deposition on natural and semi-natural vegetation have become available since the compilation of the last background document in 2011. An update of the background material based on the availability of new scientific evidence for many N-sensitive ecosystems is thus pertinent and was adopted by the Working Group on Effects at its 39th session (WGE, 2020), under the LRTAP Convention, and was included in the work plan 2020-2021. This new report will be the basis for the revision of Chapter 5.2 of the modelling and mapping manual (ICP M&M, 2017).

The aims and structure of this report are as follows:

- New relevant information from studies (2010 - summer 2021) on the impacts of N on semi-natural and natural ecosystems, with emphasis on Europe, to be added to the existing database on $CL_{emp}N$
- To update the recently revised EUNIS classes (Chytrý et al., 2020) and link, where possible, the $CL_{emp}N$ based on the EUNIS classification with Natura 2000 Annex 1 habitats
- To review and revise Bobbink and Hettelingh (2011) and provide a revised table on $CL_{emp}N$ for Europe, using the new scientific data including gradient studies (Chapters 2 to 9)
- To synthesise examples of how $CL_{emp}N$ can be used on different scales and in different European countries (Chapter 10)

Furthermore, the report ends with three appendices. The correspondence between ecosystems classified according to EUNIS and the EU Habitats according to Directive Annex 1 is provided in appendix 1. Appendices 2 and 3 present the agenda and the list of participants from the expert workshop in Berne (Switzerland, October 2021).

1.3 References

Achermann, B. and Bobbink, R. (eds.) (2003). *Empirical* critical *loads for nitrogen*. Environmental Documentation No.164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Aerts, R. and Chapin, F.S. (2000). The mineral nutrition of wild plants revisited: A re-evaluation of processes and patterns. *Advances in Ecological Research* **30**, 1-67.

Asman, W.A.H., Sutton, M.A. and Schjorring, J.K. (1998). Ammonia: emission, atmospheric transport and deposition. *New Phytologist* **139**, 27-48.

Berdowski, J.J.M. (1993). The effect of external stress and disturbance factors on *Calluna*-dominated heathland vegetation. In: Aerts, R. and Heil, G.W. (eds.), *Heathlands: patterns and processes in a changing environment*, Kluwer, Dordrecht, pp. 85-124.

Bobbink, R., Boxman, D., Fremstad, E., Heil, G., Houdijk, A. and Roelofs, J. (1992). Critical loads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna. In: Grennfelt, P. and Thörnelöf, E. (eds.), *Critical loads for nitrogen*, Nord 41, Nordic Council of Ministers, Copenhagen. pp. 111.

Bobbink, R., Hornung, M. and Roelofs, J.G M. (1996). Empirical nitrogen critical loads for natural and semi-natural ecosystems. In: *Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded*, Texte 71/96, III-1-54. Federal Environmental Agency, Berlin.

Bobbink, R., Hornung, M. and Roelofs, J.G.M. (1998). The effects of air-borne nitrogen pollutants on species diversity in natural and semi-natural European vegetation. *Journal of Ecology* **86**, 717-738.

Bobbink, R. and Lamers, L.P.M. (2002). Effects of increased nitrogen deposition. In: Bell, J.N.B. and Treshow, M. (eds.), *Air pollution and plant life* (2nd edition), John Wiley and Sons, Chichester, pp. 201-235.

Bobbink, R., Ashmore, M., Braun, S., Fluckiger, W. and Van den Wyngaert, I.J.J. (2003). *Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update.* Environmental Documentation No. 164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J-W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L., and De Vries, W. (2010). Global assessment of nitrogen deposition effects on plant terrestrial biodiversity: a synthesis. *Ecological Applications*: **20**, 30-59.

Bobbink, R. and Hettelingh, J.P. (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships*. Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM).

Brunsting, A.M.H. and Heil, G.W. (1985). The role of nutrients in the interactions between a herbivorous beetle and some competing plant species in heathlands. *Oikos* **44**, 23-26.

Bull K.R., Achermann B., Bashkin V., Chrast R., Fenech G., Forsius M., Gregor H-D., Guardans R., Haußmann T., Hayes F., Hettelingh J.-P., Johannessen T., Krzyzanowski M., Kucera V., Kvaeven B., Lorenz M., Lundin L., Mills G., Posch M., Skjelkvåle B.L. and Ulstein M.J. (2001). Coordinated effects monitoring and modelling for developing and supporting international air pollution control agreements. *Water, Air and Soil Pollution* **130**, 119-130.

Cape, J.N., Van der Eerden, L.J., Sheppard, L.J., Leith, I.D. and Sutton, M.A. (2009). Evidence for changing the critical level for ammonia, *Environmental Pollution* **157**:1033-1037

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**, 648-675.

Davies, C.E. and Moss, D. (2002). *EUNIS habitat classification, Final report.* CEH Monks Wood, UK.

Davies, C.E., Moss, D. and Hill, M.O. (2004). *EUNIS habitat classification revised 2004*. European Environment Agency, European topic centre on nature protection and biodiversity.

De Graaf, M.C.C., Bobbink, R., Roelofs, J.G.M. and Verbeek, P.J.M. (1998). Differential effects of ammonium and nitrate on three heathland species. *Plant Ecology* **135**, 185-196.

Dentener, F., Drevet, J., Lamarque, J.F., Bey, I., Eickhout, B., Fiore, A.M., Hauglustaine, D., Horowitz, L.W., Krol, M., Kulshrestha, U.C., Lawrence, M., Galy-Lacaux, C., Rast, S., Shindell, D., Stevenson, D., Van Noije, T., Atherton, C., Bell, N., Bergman, D., Butler, T., Cofala, J., Collins, B., Doherty, R., Ellingsen, K., Galloway, J., Gauss, M., Montanaro, V., Müller, J.F., Pitari, G., Rodriguez, J., Sanderson, M., Solmon, F., Strahan, S., Schultz, M., Sudo, K., Szopa, S., and Wild, O. (2006). Nitrogen and sulfur deposition on regional and global scales: A multimodel evaluation. *Global Biogeochemical Cycles* **20**, GB4003.

Dise, N.B., Ashmore, M., Belyazid, S., Bleeker, A, Bobbink, R., De Vries, W., Erisman, J.W., Van den Berg, L., Spranger, T. and Stevens, C.J. (2011). N deposition as a threat to European terrestrial Biodiversity (Chapter 20). In: Sutton M.A., Howard C.M., Erisman J.W., Billen G., Bleeker A., Grennfelt P., Van Grinsven H. and Grizzetti B. (eds.) *The European Nitrogen Assessment.* Cambridge University Press, 463-494.

Ellenberg, H. (1996). *Vegetation Mitteleuropas mit den Alpen* 5. Auflage. Ulmer, Stuttgart.

Falkengren-Grerup, U. (1998). Nitrogen response of herbs and graminoids in experiments with simulated acid soil solution. *Environmental Pollution* **102 (S)**, 93-99.

Flückiger, W., Braun, S. and Hiltbrunner, E. (2002). Effects of air pollutants on biotic stress. In: Bell, J.N.B. and Treshow, M. (eds.) *Air pollution and plant life (2nd edition)*. John Wiley and Sons, Chichester, pp. 379-406.

Fowler, D. (2002). Pollutant deposition and uptake by vegetation. In: Bell, J.N.B. and Treshow, M. (eds.) *Air Pollution and Plant Life.* Wiley, Chichester, pp. 43-67.

Fowler, D., Brimblecombe, P., Burrows, J., Heal, M.R., Grennfelt, P., Stevenson, D.S., Jowett, A., Nemitz, E., Coyle, M., Liu, X., Chang, Y., Fuller, G.W., Sutton, M.A., Klimont, Z., Unsworth M.H. and Vienoet, M. (2020). A chronology of global air quality. *Philosophical Transactions of the Royal Society A* **378**, 20190314.

Galloway, J.N. and Cowling, E.B. (2002). Reactive nitrogen and the world: 200 years of change. *Ambio* **31**, 64-71.

Gigon, A. and Rorison, I.H. (1972). The response of some ecologically distinct plant species to nitrate- and to ammonium-nitrogen. *Journal of Ecology* **60**, 93-102.

Grennfelt, P. and Thörnelöf, E. (1992). *Critical loads for nitrogen.* Nord 41, Nordic Council of Ministers, Copenhagen.

Grupa, S.V. (2003). Effects of atmospheric ammonia (NH_3) on terrestrial vegetation: a review. *Environmental Pollution* **124,** 179-221.

Hall, J, Davies, C. and Moss, D. (2003). Harmonisation of ecosystem definitions using the EUNIS habitat classification. In: Achermann, B. and Bobbink, R. (eds.) *Emperical Critical Loads for Nitrogen – Proceedings expert workshop*. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne, pp. 171-195.

Hallingback, T. (1992). The effect of air pollution on mosses in Southern Sweden. *Biological Conservation* **59**, 163-170.

Hettelingh J.-P., Posch M., De Smet P.A.M. (2001). Multi-effect critical loads used in multi-pollutant reduction agreements in Europe. *Water, Air and Soil Pollution* **130**, 1133-1138.

Hettelingh J.-P., Posch M., Slootweg J., Reinds G.J., Spranger T., Tarrason L. (2007). Critical loads and dynamic modelling to assess European areas at risk of acidification and eutrophication. *Water, Air and Soil Pollution Focus* **7**, 379-384.

ICP M&M (2019). *Report of the 35th ICP M&M Task Force Meeting on assessments of impacts of air pollution, and interactions with climate change, biodiversity and ecosystem services. International Cooperative Programme on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects, Risks and Trends (ICP M&M) under the Convention on Long-range Transboundary Air Pollution*. 2nd–4th April 2019, CIEMAT, Madrid (Spain). Available here: https://www.umweltbundesamt.de/sites/default/files/medien/4292/dokumente/35th_icp_mm_minutes.pdf

ICP M&M (2017). *Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & levels and Air Pollution Effects, Risks and Trends*, update of UBA (2004). Available here: https://www.umweltbundesamt.de/en/manual-for-modelling-mapping-critical-loads-levels?parent=68093

Kinzel, S. (1982). *Pflanzenökologie und Mineralstoffwechsel*. Ulmer, Stuttgart.

Kleijn, D., Bekker, R.M., Bobbink, R., De Graaf, M.C.C. and Roelofs, J.G.M. (2008). In search for key biogeochemical factors affecting plant species persistence in heathland and acidic grasslands: a comparison of common and rare species. *Journal of Applied Ecology* **45**, 680-687.

Løkke, H., Bak, J., Bobbink, R., Bull, K., Curtis, C., Falkengren-Grerup, U., Forsius, M., Gundersen, P., Hornung, M., Skjelkvåle, B.L., Starr, M. and Tybirk, K. (2000). *Critical Loads Copenhagen 1999, 21-25 November 1999.* Conference report prepared by members of the conference's secretariat, the scientific committee and chairmen and rapporteurs of its workshops in consultation with the UNECE secretariat, Critical loads, National Environmental Research Institute, Denmark, pp. 48.

Lucassen, E.C.H.E.T., Bobbink, R., Smolders, A.J.P., Van der Ven, P.J.M., Lamers, L.P.M. and Roelofs, J.G.M. (2003). Interactive effects of low pH and high ammonium levels responsible for the decline of *Cirsium dissectum* (L.) Hill. *Plant ecology* **165**, 45-52.

Nihlgård, B. (1985). The ammonium hypothesis - an additional explanation to the forest dieback in Europe. *Ambio* **14**, 2-8.

Olsson, M.O. and Falkengren-Grerup, U. (2000). Potential nitrification as an indicator of preferential uptake of ammonium or nitrate by plants in an oak understory. *Annals of Botany* **85**, 299-305.

Pardo, L.H., Fenn, M.E., Goodale, C.L., Geiser, L.H., Driscoll, C.T., Allen, E.B., Baron, J., Bobbink, R., Bowman, W.D., Clark, C., Emmett, B., Gilliam, F.S., Greaver, T., Hall, J., Lilleskov, E.A., Liu, L., Lynch, J.A., Nadelhoffer, K., S.S. Perakis, S.S., Robin-Abbott, M.J., Stoddard, J.L. Weathers, K.C. and Dennis, R.L. (2011). Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. *Ecological Applications*, **21**, 3049-3082.

Paulissen, M.P.C.P., Van der Ven, P.J.M., Dees, A.J. and Bobbink, R., (2004). Differential effects of nitrate and ammonium on three fen bryophyte species in relation to pollutant nitrogen input. *New Phytologist* **164**, 551-458.

Pearson, J. and Stewart, G.R. (1993). The deposition of atmospheric ammonia and its effects on plants. *New Phytologist* **125**, 283-305.

Roelofs, J.G.M., Kempers, A.J., Houdijk, L.F.M. and Jansen., J. (1985). The effect of air-borne ammonium sulphate on *Pinus nigra* var. *maritima* in the Netherlands. *Plant and Soil* **84**, 45-56.

Sheppard, L.J., Leith, I.D., Crossley, A., Van Dijk, N., Cape, J.N., Fowler, D. and Sutton, M.A. (2009). Long term cumulative exposure exacerbates the effects of atmospheric ammonia on an ombrothrophic bog: implications for critical levels. In: Sutton, M.A., Reis, S. and Baker, S.M.H. (eds.) *Atmospheric ammonia – Detecting emission changes and environmental impacts*. Springer Science + Business Media B.V.

Stevens C.J., Bell, J.N.B., Brimblecombe, P., Clark, C.M., Dise, N.B., Fowler, D., Lovett, G.M. and Wolseley, P.D. (2020). The impact of air pollution on terrestrial managed and natural habitats. *Philosophical Transactions of the Royal Society A* **378,** 20190317.

Suding, K.N., Collins, S.L., Gough, L., Clark, C., Cleland, E.E., Gross, K.L., Milchunas, D.G. and Pennings, S. (2005). Functional- and abundance-based mechanisms explain diversity loss due to N fertilization. *PNAS* **102**, 4387-4392.

Sutton, M.A., Erisman, J.W., Dentener, F. and Miller, D. (2008). Ammonia in the environment: from ancient times to present. *Environmental Pollution* **156**, 583-604.

Sutton, M.A., Reis, S. and Baker, S.M.H. (eds.) (2009). *Atmospheric ammonia – Detecting emission changes and environmental impacts.* Springer Science + Business Media B.V.

Tamm, C.O. (1991). *Nitrogen in Terrestrial Ecosystems*. Springer Verlag, Berlin.

Throop, H.L. and Lerdau, M.T. (2004). Effects of nitrogen deposition on insect herbivory: implications for community and ecosystem processes. *Ecosystems* **7**, 109-133.

Truscott, A.M., Palmer, S.C.F., McGowan, G.M., Cape, J.N., and Smart, S. (2005). Vegetation composition of roadside verges in Scotland: the effects of nitrogen deposition, disturbance and management. *Environmental Pollution* **136**, 109-118.

Ulrich, B. (1983). Soil acidity and its relation to acid deposition. In: Ulrich, B. and Pankrath, J. (eds.) *Effects of accumulation of air pollutants in ecosystems*. Reidel Publishing, Boston, pp. 127-146.

Ulrich, B. (1991). An ecosystem approach to soil acidification. In: Ulrich, B. and Summer, M.E. (eds.) *Soil acidity*. Springer, Berlin, 28-79.

Van Breemen, N., Burrough, P.A., Velthorst, E.J., Van Dobben, H.F., De Wit, T. and Ridder, T.B. (1982). Soil acidification from atmospheric ammonium sulphate in forest canopy throughfall. *Nature* **299**, 548-550.

Van den Berg, L.J.L., Peters, C.J.H., Ashmore, M.R. and Roelofs, J.G.M. (2008). Reduced nitrogen has a greater effect than oxidised nitrogen on dry heathland vegetation. *Environmental Pollution* **154**, 359-369.

Van Dijk, H.F.G., De Louw, M.H.J., Roelofs, J.G.M. and Verburgh, J.J. (1990). Impact of artificial ammonium-enriched rainwater on soils and young coniferous trees in a greenhouse. Part 2 - Effects on the trees. *Environmental Pollution* **63**, 41-60.

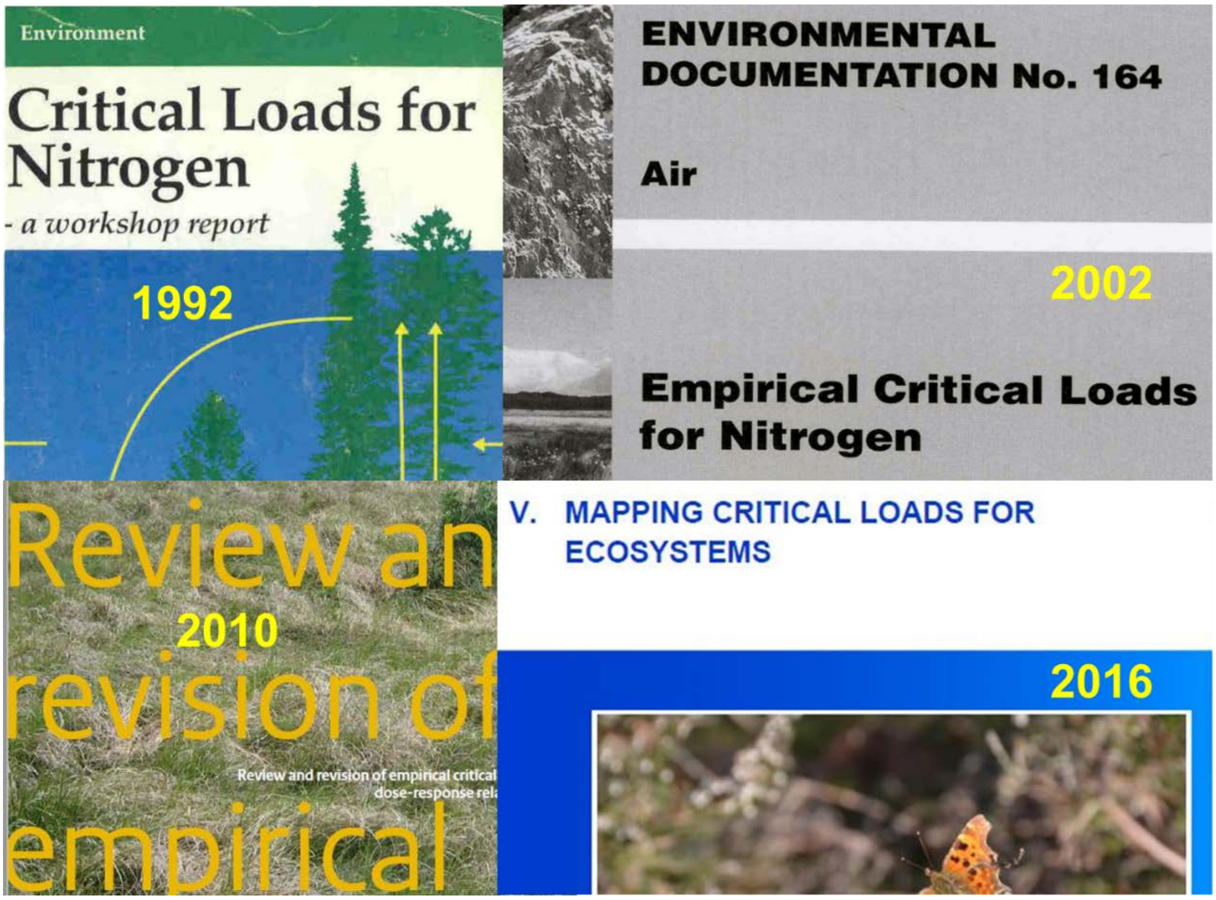
Van Herk, C.M., Mathijssen-Spiekman, E.A.M. and De Zwart, D. (2003). Long distance nitrogen air pollution effects on lichens in Europe. *Lichenologist* **35**, 347-359.

WGE (2020). *Report of the Executive Body on its thirty-ninth session*. Geneva, 9-13 December 2019. ECE/EB.AIR/144/Add.2. Economic Commission for Europe. Executive Body for the Convention on Long-range Transboundary Air Pollution. Available here: https://unece.org/info/events/event/18910

WHO (2000). *Air quality guidelines for Europe, second edition*. WHO regional publications, European series, No. 91. World Health Organisation, Regional office for Europe, Copenhagen.

2 Updating and reviewing procedures for empirical critical loads of nitrogen ($CL_{emp}N$)

Adapted by Roland Bobbink

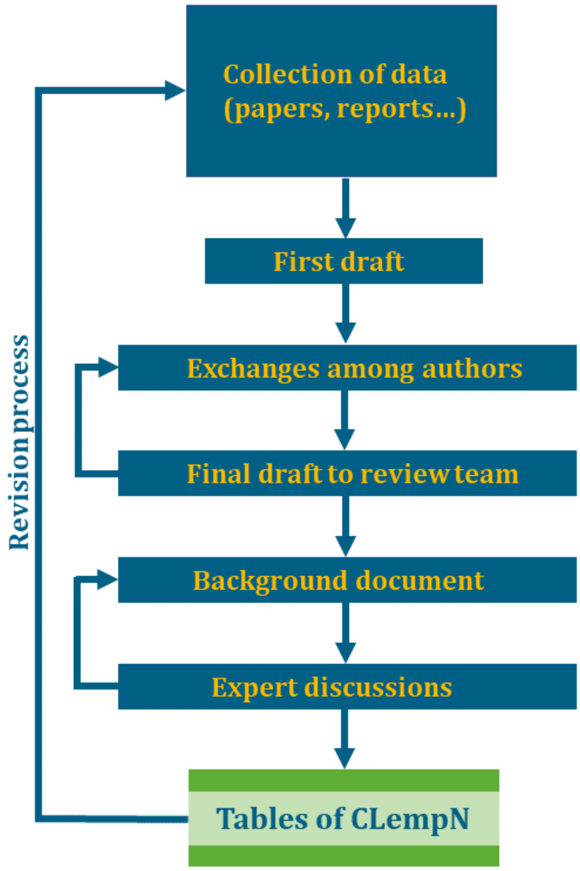


Compilation of cover pages of previous reports on empirical critical loads of nitrogen ($Cl_{emp}N$).

2.1 Updating procedure

In this updating procedure, an '***empirical approach***' has been used, similar to that of the previous background documents (Bobbink et al., 2003; Bobbink and Hettelingh, 2011), with the following phases: 1) Kick-Off meeting (June 2020), 2) data collection, 3) first drafting of the different chapters (per class according to the European Nature Information System (EUNIS)), 4) optimisation of the drafts after exchange between the contributing authors (internal review round), 5) review of the second draft by an external expert team, and 6) finalisation of the background document for the UNECE CCE expert workshop, held in October 2021 (Figure 2.1). Following the expert workshop, the background document was finalised after addition and incorporation of the comments of the workshop participants.

Figure 2.1. Schematic representation of the working procedure



a) **Data collection**

A comprehensive collection of European publications on the effects of nitrogen (N) in natural and semi-natural ecosystems has been made for the period from early 2010 to September 2021. Peer-reviewed publications, PhD theses, book chapters, nationally published papers, and 'grey' reports by institutes or organisations (if digitally available) were used. Relevant information from these studies has been collected, including location, background deposition (if available), and EUNIS classification. The correspondence between the EUNIS class and the Natura-2000 habitat type has been added in an appendix (Appendix 1), as in Bobbink and Hettelingh (2011).

In principle, only European studies have been used as the basis for the assessment of empirical critical loads of N ($CL_{emp}N$). Exceptionally, when no or very few studies were available for a particular important ecosystem, such as steppe grasslands or Mediterranean vegetation, non-European (mostly Northern American and Chinese) literature was used for an 'expert judgement' rating of ecosystem sensitivity to N deposition.

b) **Drafting of the chapters**

Following data collection, drafts of the several chapters (per EUNIS class) of the background document were produced by groups of authors, using the 2011 document (Chapters 3 to 9 in Bobbink and Hettelingh (2011)) as a starting point. When no new data were available for a specific habitat, the 2011 text was used. When new data were found to be available, the 2011 text was updated accordingly, which in places resulted in completely rewritten chapters. At the end of each chapter, a concluding table presents the CLempN.

c) **Optimisation of the chapter drafts**
All drafts of the different chapters were circulated in the author group of that chapter for discussion and review. This was coordinated by the lead author of the chapter. Comments of co-authors were discussed and incorporated into the main version of the chapter by the lead author, which resulted in the generation of several drafts. All drafts of the different chapters were then reviewed and checked for consistency by the team of lead authors. This internal review round led to a draft version of each chapter, which was then checked and integrated into the main document.

d) **External review**
The consolidated drafts of the chapters were presented to a team of international experts on the impacts of N enrichment in natural and semi-natural ecosystems. This reviewing team consisted of experts from different parts of Europe and the USA. Each chapter on a specific EUNIS class was evaluated by at least two to three experts.

e) **Finalisation of the background document**
Review comments on the second draft were incorporated into the text by the leading authors of each chapter, in close collaboration with the coordinator(s) of the revision. After a final check, the background document was sent to the participants of the UNECE CCE expert workshop on $CL_{emp}N$ (October 2021, in Berne, Switzerland). The comments and additions by participants were used to finalise the final $CL_{emp}N$ table and background document for the formal revision of the Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends (ICP Modelling & Mapping, in prep.), within the framework of the UNECE Convention on Long-range Transboundary Air Pollution.

2.2 Reviewing and setting values for $CL_{emp}N$

2.2.1 Types of empirical evidence

The type of "empirical" evidence used to set values for $CL_{emp}N$ varies between terrestrial and aquatic ecosystems. Long-term field manipulations in lakes are highly ecologically relevant but are prohibitively expensive, and are therefore also limited in terms of duration and replication. Experiments of more limited time-span in more controlled conditions in aquatic systems (mesocosms) can supply valuable additional evidence on ecosystem effects related to N addition, especially when they are done along environmental gradients. Therefore, the following text on the type of evidence largely relates to terrestrial ecosystems, including wetlands, while for aquatic ecosystems evidence has been gathered to a larger extent from gradient studies in combination with controlled experiments.

There are two major types of "empirical" evidence available to relate atmospheric N deposition to changes in the structure and/or functioning of ecosystems. The first is from long-term field addition (or manipulation) experiments, in which N deposition is artificially increased, normally by application of increased concentrations of mineral N (NH_4^+ and/or NO_3^-). If significant impacts were detected compared to the untreated controls, it was inferred with confidence that simulated N deposition would have been the cause. Experiments can provide information on how long it takes for different components of the system to respond to N addition, and can be designed to assess interactions, for example with other stresses (e.g. drought, warming), land management type or ecosystem type. Experiments can also identify thresholds for the effects of N on biodiversity. However, since most experiments examine effects of increases in deposition, it is difficult to identify thresholds from experiments in areas with a relatively long history of elevated levels of N deposition, where there may already have been significant impacts of N deposition and where those thresholds may have already been crossed in the past. Other limitations of experimental studies are that they typically assess relatively short-term responses (even the longest-running experiments seldom exceed 20-25 years) and that peculiarities of the experiment (e.g. very high concentrations of the applied pollutant compared with environmentally realistic loads) or site-specific factors might also explain part of the observed response.

A second approach – besides of the evaluation of N addition experiments - is through studies exploring changes in ecosystem structure and/or functioning over an observational N deposition gradient. Such targeted N-gradient studies may provide information on longer-term responses of increased atmospheric N deposition and serve to demonstrate that the responses observed in experiments can also be found in the real-world. They usually cover, if present, a more differentiated range of N deposition than added in the N addition experiments, particularly at the lower end of N deposition, as experiments typically deal with high N doses that are often based on future projections. Most of the studies used species richness of the vegetation – or of components of the vegetation – as a bioindicator, but sometimes (a)biotic factors including plant tissue chemistry and soil biogeochemistry were also considered. In addition, these studies avoid experimental artefacts and allow for analysis of interactions with other environmental stressors (e.g. drought). However, since gradients of N deposition may also be correlated with gradients of other potential important drivers of ecosystem structure and functioning (e.g. S deposition, rainfall, temperature or management), gradient studies need a careful and appropriate design and statistical evaluation. Thus, gradient studies have some advantages and disadvantages compared with experimental evidence. Advantages include adequate sampling of the range of climate space and variability in other factors such as soil type, elevation and management, as well as the range of N deposition present across a habitat type. The disadvantage is greater variability in ecological responses, which requires careful consideration of confounding variables, as discussed above, and larger sample sizes, needed to give sufficient statistical power to detect effects of N deposition. Confounding abiotic and biotic variables may also be considered as modulating variables when they alter the response of the ecosystem to N deposition. One special class of gradient study is the structured survey (defined here as survey not explicitly set up to detect effects of N deposition). This can be analysed post-hoc to detect N impacts (e.g. Maskell et al. 2010). As decided at the Kick-Off meeting of the 2020-2022 review and revision of the $CL_{emp}N$ and based upon the outcome of discussions within an expert group of the authors, the outcome of published N gradient studies have been incorporated in the $CL_{emp}N$ approach as important insights.

2.2.2 Setting $CL_{emp}N$

In this background document, the authors focus particularly on statistically and biologically significant outcomes of field N addition experiments and mesocosm studies for the assessment of $CL_{emp}N$. Only studies which have independent N treatments and realistic N loads and durations (below 100 kg N ha^{-1} yr^{-1}; 2 years or more, optimally > 5-10 years in low background areas) were used for the update and refinement of $CL_{emp}N$ values. Studies with higher N additions or shorter experimental periods have only been interpreted with respect to the understanding of effect mechanisms, possible N limitation or sensitivity of the studied ecosystem. The methods used in these studies have been carefully scrutinised to identify factors related to the experimental design or data analysis, which may constrain their use in assessing critical loads. This includes evaluation of the precision of the estimated values of background deposition at the experimental site, which are often based on models instead of on-site measurements. This is necessary to get insight into the total N load in both the N-treated and the control vegetation.

In general, pot or microcosm studies were not used for setting $CL_{emp}N$ values, except for bryophyte layer studies. However, the outcome of these studies, in some selected cases, was used as an indication of the N sensitivity of the most important or sensitive plant species of an ecosystem (e.g. in coastal habitats).

When available, the outcomes from dynamic ecosystem models provided additional insight into underlying mechanisms of ecosystem decline, which are difficult to incorporate or analyse in experimental studies, such as increased frequencies of pests and diseases and a greater sensitivity to environmental stressors such as frost, drought and heatwaves.

2.2.3 N gradient studies

The outcomes of N gradient studies have been used to assess the impacts of atmospheric N deposition in this 2020-2022 review and revision of the $CL_{emp}N$, combined with the results of N addition experiments. Principally, peer-reviewed publications have been analysed, together with some "grey" publications. N gradient studies show the relationships between (modelled) atmospheric annual total inorganic N deposition and ecological parameters, e.g. species richness of the vegetation (or of taxonomical or functional groups of the vegetation), species composition, plant tissue stoichiometry, soil chemistry, drought resistance and sensitivity against pathogens and pests. In almost all cases, significant relationships between the selected indicators and N deposition have been found. Several functions have been fitted through the data, such as linear, negative exponential or S-shaped curves and in several instances more rapid responses are observed at lower N loads. The relationships as presented in many N gradient studies have been originally quantified to demonstrate the potentially negative influence of N deposition on ecosystem structure or functioning, mostly not to identify N thresholds per se. Several recent studies have, however, been purposedly set up to reveal $CL_{emp}N$ values/ranges and used statistical techniques/models to identify these thresholds. These techniques may include change point models (Roth et al., 2017; Tipping et al., 2013), community change point analysis with Threshold Indicator Taxa ANalysis (TITAN) (Payne et al., 2013; Wilkins and Aherne, 2016; Wilkins et al., 2016) or simple comparison between classes of N deposition (e.g. per 5 kg N ha^{-1} yr^{-1}) (Roth et al., 2013). A comparison of the just mentioned techniques to reveal $CL_{emp}N$ values or ranges is given in the next chapter (1.3), demonstrating a good agreement between the different techniques if applied to the same data set.

To evaluate the published N gradient studies in the empirical approach, two types of studies have been identified: a) studies that did not use special techniques to quantify critical loads ("only general" regressions) and b) studies using the previously mentioned special statistical techniques. With respect to the first group of evidence (a), visual inspection of the basic figures was used to reveal a "change range" in the curves (data points). For the second group of studies (b) the presented technique has been shortly described and the outcome given as evidence with respect to the setting of the $CL_{emp}N$ of the studied ecosystem type.

Most of the N gradient studies used have been published in peer-reviewed, international scientific journals. In this case it is likely that the (modelling of) atmospheric N deposition is accurate and at a relevant scale (e.g. Braun et al., 2017) for the specific situation and thus useable for $CL_{emp}N$ evaluation, as done for N addition experiments too. If relevant, possible weaknesses in the presented N deposition values are evaluated, including suggestions with respect to the setting of $Cl_{emp}N$. In addition, it has been checked if confounding/modulating predictors (e.g. temperature, rainfall, S deposition) were included sufficiently and if appropriate statistical procedures were used. Some of the N gradient studies have so far only been available in "grey" literature reports. In that case, special attention has been paid to the outcome of the study with respect to both the method of disentangling the contributions of different predictor variables and the N deposition values used (which model etc). After this evaluation, (parts of) the results have been used to find $CL_{emp}N$ values.

2.3 A comparison of methods to estimate empirical critical loads from gradient studies

Authors: Tobias Roth, Julian Aherne, Kayla Wilkins and Roland Bobbink

2.3.1 Introduction

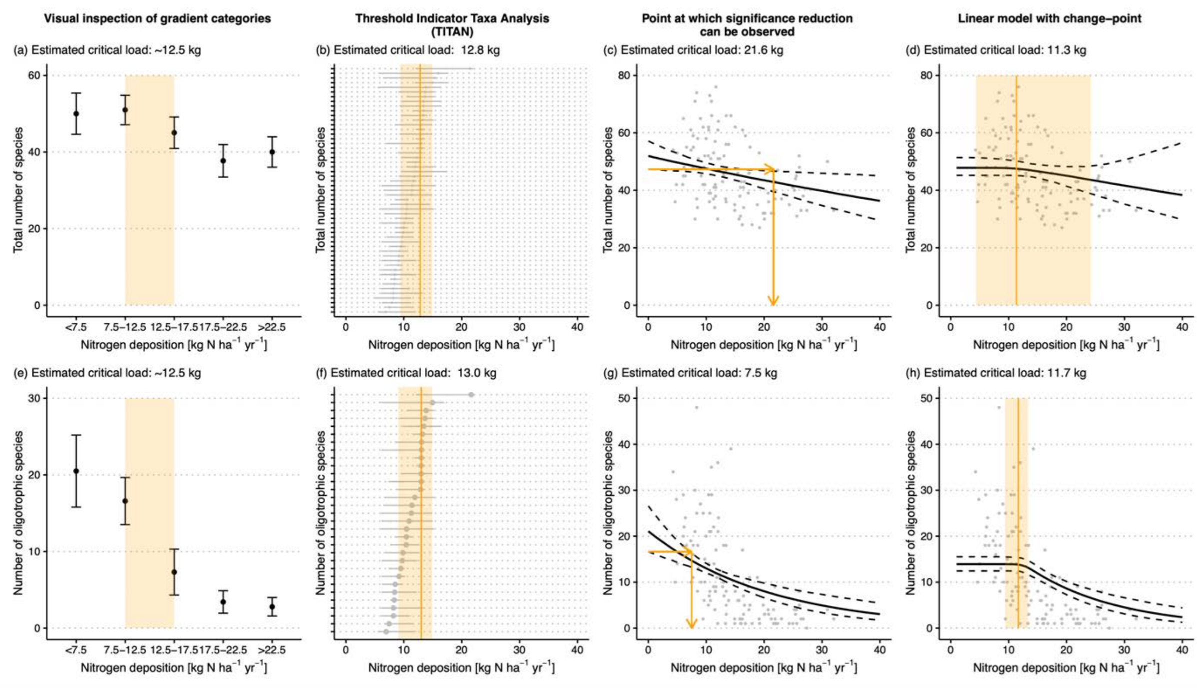
There are numerous methods to estimate an abrupt change or 'change-point' along a gradient. Several studies have used an abrupt change in plant species along a nitrogen deposition gradient as a 'quantitative estimate' of the empirical critical loads for nitrogen (e.g. Wilkins et al., 2016; Roth et al., 2017). To our knowledge, these methods have never been applied to the same data. It is thus unknown whether the different methods result in systematic differences between the estimated critical loads. Consequently, it is difficult to compare critical load estimates between studies. Here we apply four methods that were previously used in publications to estimate empirical critical loads from gradient studies to the same data sets as indicated in Chapter 2. The four methods are listed in Table 2.1. Our goals were 1) to give an overview of the differences between the methods and list their potential advantages and shortcomings, and 2) to show whether and how the results differ if they are applied to identical data sets.

2.3.2 Case studies: Mountain hay meadows in Switzerland and Atlantic oak woodlands in Ireland

We applied the four methods (Table 2.1) to mountain hay meadow data from the biodiversity monitoring in Switzerland (Weber et al., 2004) and Atlantic oak woodland data (species abundances) from the Irish National Parks and Wildlife Service (Perrin et al., 2008). For mountain hay meadows, we selected the same sites as in Roth et al. (2013) but used for each site the most recent survey conducted between 2016–2020. For the recent surveys, species cover (abundance) was recorded, whereas for the data in Roth et al. (2013) only presence / absence data were available. For these surveys, we analysed all recorded species together and the subset of species that are typically found on nutrient poor sites (i.e. oligotrophic species with N-values of one and two; Landolt et al., 2010). For Atlantic oak woodlands, we used the same sites, plant species data (surveyed between 2003–2007), and nitrogen deposition data as Wilkins and Aherne (2016). We analysed all recorded species together and the subset of species identified as positive indicators by the Irish National Parks and Wildlife Service. All analyses were conducted in R. Data sets and scripts to reproduce the analyses are available on GitHub[1].

[1] https://github.com/TobiasRoth/eCL-methods

Figure 2.2. **Comparison of the results applying the four methods (a and e: visual inspection of gradient categories; b and f: Threshold Indicator Taxa Analysis for plant species with negative change-points; c and g: point at which significant reduction can be observed; d and h: linear model with change-point) to the plant data of mountain hay meadows in Switzerland. The upper panels (a–d) give the results based on all plant species and the lower panels (e–g) give the results based on oligotrophic species only. Critical load estimates (vertical orange line) with estimate range (orange areas) are given if provided by the method.**

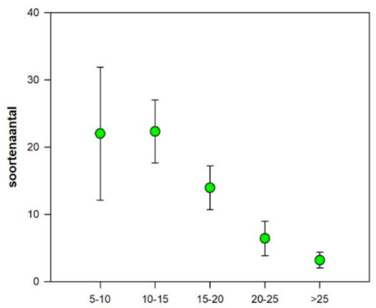
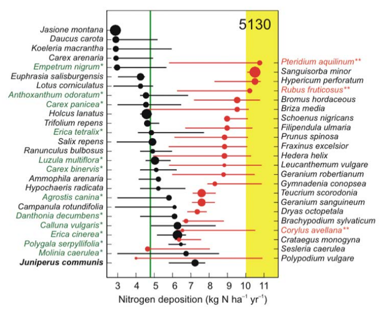
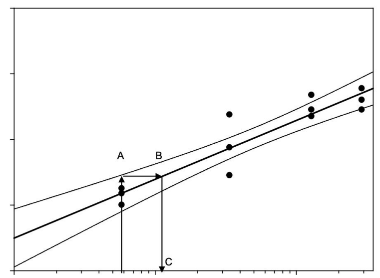
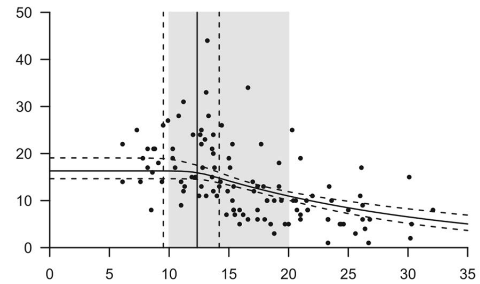


Source: https://github.com/TobiasRoth/eCL-methods

For the mountain hay meadows, the critical load estimates based on the visual inspection of gradient categories, TITAN and linear model with change-point were very similar and were stable, independent of whether the analyses were based on all recorded species or on only the oligotrophic species (Figure 2.2). The estimates were all in the range of the critical load that was established in this revision. In contrast, the method of Cape et al. (2009), based on the point at which significant reduction of species richness could be observed, showed a difference between the total and subset species data and the critical load found was outside of the established range for both (Figure 2.2). This method suggested the lowest critical load based on oligotrophic species, but the highest critical load when based on all recorded species (Figure 2.2).

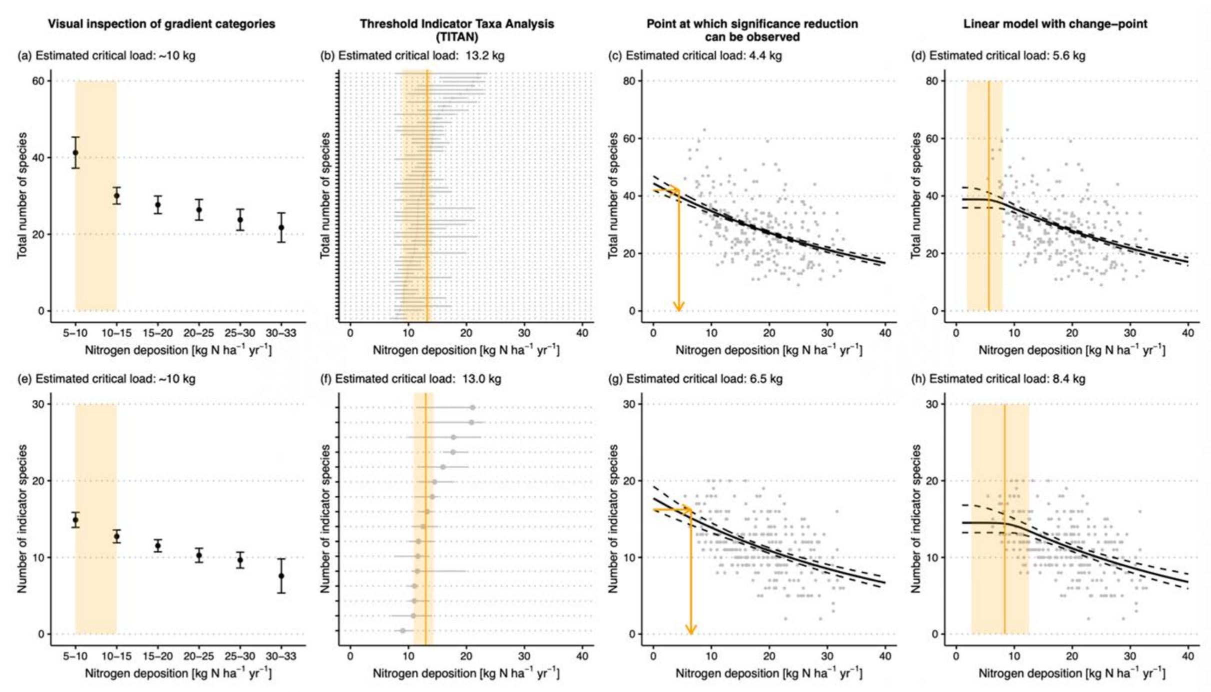
With respect to the Atlantic oak woodland, the critical load estimates of the visual inspection of gradient categories and TITAN were again very similar and within the range of the established critical load (Figure 2.3). They both showed very little difference in estimated critical loads based on the total number of species or the subset. In contrast, the method of Cape et al. (2009) and the linear model with change-points both suggested critical loads below the established critical load with lower estimates for total species richness than for the number of indicator species.

Table 2.1. Comparison of four methods to estimate empirical critical loads from nitrogen deposition gradient studies.

V.	VI. Visual inspection of gradient categories	VII. Threshold Indicator Taxa Analysis (TITAN)	VIII. Point at which significance reduction can be observed	IX. Linear model with change-point
References	Wamelink et al. (2021)	Baker and King (2010); Wilkins et al. (2016)	Cape et al. (2009)	Tipping et al. (2013); Roth et al. (2017)
Short description	The gradient (nitrogen deposition) is aggregated to equal sized classes (e.g. 5 kg N ha^{-1} yr^{-1} deposition ranges) and the average and confidence interval of the biodiversity measure is calculated for each class. The critical load is estimated 'by eye' between the classes at which the first significant decrease in the biodiversity measure occurs.	A two-step approach; 1) Individual taxa with significant change-points in species occurrence or abundance along a nitrogen deposition gradient are identified. 2) The point along the gradient at which these single species change-points accumulate (the community change-point) is identified, and interpreted as the critical load.	(Generalized) linear (mixed) model that describes the relationship between the gradient and the biodiversity measure and other variables (covariates). The nitrogen deposition at which a significant change in the biodiversity measure can be observed is considered as the critical load.	(Generalized) linear (mixed) model that describes the relationship between the gradient and the biodiversity measure using a change-point (or segmented, piecewise, broken-stick). The nitrogen deposition at which the change-point occurs is considered as the critical load.
Example figure from cited references				
Advantages	▶ Easy to apply and understand.	▶ Explicitly considers individual species' behaviour.	▶ Can account for covariables.	▶ Can account for covariables. ▶ Estimates uncertainty in critical load setting.

V.	VI. Visual inspection of gradient categories	VII. Threshold Indicator Taxa Analysis (TITAN)	VIII. Point at which significance reduction can be observed	IX. Linear model with change-point
		▶ Estimates uncertainty in change-point.		
Potential shortcomings	▶ Arbitrary classification of the continuous gradient into discrete ranges. ▶ An exact change-point is not identified (generally set to middle of range). ▶ Sufficient sample sites are needed for all categories. ▶ Limited to a summary statistic across all species (e.g. species richness, total number of individuals).	▶ For some individual species the estimated change-points seem unrealistic (e.g. eutrophic species with low change-points and vice versa). ▶ Not possible to account for covariates.	▶ Since the confidence interval decreases with the sample size, the estimated critical load will also change with sample size. ▶ Assumes that statistical significance corresponds to biological relevance. ▶ Limited to a summary statistic across all species (e.g. species richness, total number of individuals).	▶ Brute force application of the change-point model to the data; a critical load is also estimated if nitrogen deposition does not negatively affect biodiversity. ▶ Limited to a summary statistic across all species (e.g. species richness, total number of individuals).

Figure 2.3. Comparison of the results applying the four methods (a and e: visual inspection of gradient categories; b and f: Threshold Indicator Taxa Analysis for plant species with negative change-points; c and g: point at which significant reduction can be observed; d and h: linear model with change-point) to the plant data of Atlantic Oak Woodland in Ireland. The upper panels (a–d) give the results based on all plant species and the lower panels (e–g) give the results based on positive indicator species only. Critical load estimates (vertical orange line) with estimate range (orange areas) are given if provided by the method.



Source: https://github.com/TobiasRoth/eCL-methods

2.3.2 Discussion

- In general, three of the four methods (i.e. visual inspection of gradient categories, TITAN and linear model with change-point) provided critical load estimates that were similar in most cases suggesting that there is no systematic difference in the results they provide.
- Two of the methods (visual inspection of gradient categories and TITAN) appear to be less influenced by the number of species (or sample size), as they showed very little difference in estimated critical loads based on the total number of species or the species subset (oligotrophic or indicator species).
- In the two example habitats presented here, the highest and most stable critical load estimates were generally obtained from TITAN. We suggest that this may be related to the greater resolution provided by individual species abundance and frequency data compared with the summation to species richness from presence / absence data only used in the other methods.
- In some cases, the TITAN approach estimates single-species change-points that seem unrealistic (e.g. eutrophic species with low change-points and vice versa) and it was argued that this may affect the estimated community change-points. If that is the case, one would expect biased results particularly if all species (including all eutrophic species) are used in the analyses. For the mountain hay meadows and Atlantic oak woodlands, however, the

TITAN estimates were almost identical if applied to all species and if applied to the oligotrophic or indicator species subsets only. Furthermore, for mountain hay meadows the TITAN results were very similar (< 1.5 kg N ha^{-1} yr^{-1}) to the results obtained from the linear model with a change-point.

- The unrealistic single-species change-points estimated by TITAN may reflect a species with a unimodal response to nitrogen that was not captured by the observed deposition range in the case studies. Further, the unrealistic single-species response was present in the other methods, which used the species and nitrogen deposition data. However, the response of the single-species' was masked in the summation to species richness.
- Based on our results we are confident that estimates of critical loads are comparable between studies regardless of whether they were obtained using the visual inspection of gradient categories, TITAN or linear models with change-points.

2.4 Ecosystem classification

In this background document, the groups of natural and semi-natural ecosystems have been classified and ordered according to the EUNIS (European Nature Information System) habitat classification for Europe. For a general description of the updated EUNIS classification and an introduction to its use, see Davies et al. (2004) and the supporting website (https://www.eea.europa.eu/data-and-maps/data/eunis-habitat-classification). With respect to the classification in 2011, the codes for two classes changed: heathland, scrub and tundra became Class S (instead of F), whereas forest and other wooded land changed from G to T. In addition, very recently, also marine habitats changed into MA (instead of A), coastal habitats to N (formerly B), grasslands and lands dominated by forbs, mosses or lichens into R (instead of E) and wetlands to Q (instead of D) (Chytrý et al., 2020). Level 2 and 3 numerical codes changes in many cases as well, but not always. In general, the ecosystems described in this document have been classified down to level 3 of the EUNIS hierarchy, and the EUNIS code is given in the text and tables in brackets, for example, perennial calcareous grasslands and basic steppes (R1A). The Natura-2000 habitat type (e.g. H6210) have been presented in Appendix 1. Finally, as in 2011, additional attention has been paid to the classification of forest and other wooded land (T), in order to better differentiate the $CL_{emp}N$ between the wide range of European forest types. As before, studies based on pure plantation stands were not included in the chapter on forest habitats. The critical loads of N for these intensively managed systems are based upon steady-state mass balance methods, see the UNECE Mapping Manual (2015-17). Ground-living lichens and bryophytes, as before, have been incorporated in the chapters of the appropriate ecosystems, because many experimental and gradient studies incorporated these species groups. Epiphytic lichens and epiphytic bryophytes are mostly not part of the empirical approach for N critical loads, as they often were not treated or exposed in experiments, but they have been included in the critical levels for ammonia.

2.5 Revision, reliability and interpretation of $CL_{emp}N$ ranges

The indication of reliability of the $CL_{emp}N$ (Bobbink and Hettelingh, 2011) have been adapted because of the implementation of N gradient studies in this review and revision. The following guidelines have now been used:

- expert judgement (#): we only use this indication if no empirical data (experimental and/or gradient studies) were available for this type of ecosystem. For this, the $CL_{emp}N$ was based upon expert judgement and knowledge of ecosystems which were likely to be similar;

- if, for a given ecosystem, few experimental data exist (and no N gradient study) showing a significant effect at a certain range, then the $CL_{emp}N$ is considered as quite reliable #;
- if, for a given ecosystem, no experimental data exist, but an appropriate gradient study showed a significant effect at a certain range, then the $CL_{emp}N$ is considered as quite reliable #;
- if several good quality studies (both experimental and/or N gradient) revealed N deposition effects at a certain level/range, then the $CL_{emp}N$ is considered as reliable ##.

2.6 References

Baker, M.E. and King, R.S. (2010). A new method for detecting and interpreting biodiversity and ecological community thresholds: Threshold Indicator Taxa ANalysis (TITAN). *Methods in Ecology and Evolution* **1**, 25-37.

Bobbink, R., Ashmore, M., Braun, S., Fluckiger, W. and Van den Wyngaert, I.J.J. (2003). *Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update.* Environmental Documentation No. 164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Bobbink R. and Hettelingh, J.P. (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships.* Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), RIVM report 680359002/2011 (244 pp).

Braun, S., Achermann, B., De Marco, A., Pleijel, H., Karlsson, P. E., Rihm, B., Schindler C. and Paoletti, E. (2017). Epidemiological analysis of ozone and nitrogen impacts on vegetation – Critical evaluation and recommendations. *Science of The Total Environment* **603-604**, 785-792.

Cape, J.N., Van der Eerden, L.J., Sheppard, L.J., Leith, I.D. and Sutton, M.A. (2009). Evidence for changing the critical level for ammonia. *Environmental Pollution* **157**, 1033-1037.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**(4), 648-675.

Davies, C.E., Moss, D. and Hill, M.O. (2004). EUNIS habitat classification revised 2004. European Environment Agency, European topic centre on nature protection and biodiversity.

EMEP Status Report (2021). Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components. Joint MSC-W & CCC & CEIP Report 1/2021.

EUNIS Habitat classification. https://www.eea.europa.eu/data-and-maps/data/eunis-habitat-classification.

ICP Modelling & Mapping (in prep.). Manual on Modelling and Mapping Critical Loads & Levels. (https://www.umweltbundesamt.de/en/cce-publications).

Landolt, E., Bäumler, B., Erhardt, A., Hegg, O. Klötzli, F.A., Lämmler, W., Nobis, M., Rudmann-Maurer, K., Schweingruber, F.H., Theurillat, J.P., Urmi, E., Vust, M. and Wohlgemuth, T. (2010). *Flora indicativa. Ecological indicator values and biological attributes of the flora of Switzerland and the Alps*. Haupt Verlag.

Maskell, L.C., Smart, S.M., Bullock, J.M., Thompson, K. and Stevens, C.J. (2010). Nitrogen deposition causes widespread loss of species richness in British habitats. *Global Change Biology* **16**, 671-679.

Payne, R.J., Dise, N.B., Stevens, C.J., Gowing, D.J. and BEGIN Partners (2013). Impact of nitrogen deposition at the species level. *Proceedings of the National Academy of Sciences* **110**, 984-987.

Perrin, P., Martin, J., Barron, S., O'Neill, F., McNutt, K. and Delaney, A. (2008). N*ational Survey of Native Woodlands - Volume 1: Main Report*. National Parks and Wildlife Service (URL: www.npws.ie/research-projects/woodlands).

Roth, T., Kohli, L., Rihm, B. and Achermann, B. (2013). Nitrogen deposition is negatively related to species richness and species composition of vascular plants and bryophytes in Swiss mountain grassland. *Agriculture, Ecosystems and Environment* **178**, 121-126.

Roth, T., Kohli, L., Rihm, B., Meier, R. and Achermann, B. (2017). Using change-point models to estimate empirical critical loads for nitrogen in mountain ecosystems. *Environmental Pollution* **220**, 1480-1487.

Roth, T., Kohli, L., Rihm, B., Meier, R. and Achermann, B. (2017). Using change-point models to estimate empirical critical loads for nitrogen in mountain ecosystems. *Environmental Pollution* **220**, 1480-1487.

Tipping, E., Henrys, P., Maskell, L. and Smart, S. (2013). Nitrogen deposition effects on plant species diversity; threshold loads from field data. *Environmental Pollution* **179**, 218-223.

Wamelink, G.W.W., Goedhart, P.W., Roelofsen, H.D., Bobbink, R., Posch, M. and Van Dobben, H.F. (2021). *Relaties tussen de hoeveelheid stikstofdepositie en de kwaliteit van habitattypen*. Wageningen Environmental Research, Wageningen. Available from https://research.wur.nl/en/publications/b63fc771-d3e3-432f-8c08-33f26c1b2a7e (accessed December 27, 2021).

Weber, D., Hintermann, U. and Zangger, A. (2004). Scale and trends in species richness: considerations for monitoring biological diversity for political purposes. *Global Ecology and Biogeography* **13**, 97-104.

Wilkins, K. and J. Aherne (2016). Vegetation community change in Atlantic oak woodlands along a nitrogen deposition gradient. *Environmental Pollution* **216**, 115-124.

Wilkins, K., J. Aherne and A. Bleasdale (2016). Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high. *Atmospheric Environment* **146**, 324-331.

3 Effects of nitrogen deposition on marine habitats (EUNIS class MA, formerly A)

Adapted by Roland Bobbink

Upper-mid salt marsh (MA223) on a Wadden Island in the Netherlands. Photo: Bas Van de Riet.

Summary

In this chapter empirical N critical loads ($CL_{emp}N$) for Atlantic coastal salt marshes (MA223, MA224 and MA225) have been updated and revised, if necessary. Unfortunately, no experimental field studies with application of nitrogen compounds with low enough N loads have become available in the present revision period. However, the outcome of two N gradient studies of Atlantic salt marshes (MA223 and MA224) indicated that the $CL_{emp}N$ of these two salt marsh types should be lowered to 10-20 kg N ha^{-1} yr^{-1} (expert judgement). Finally, it has been concluded that long-term N addition studies with low doses of N application are highly needed in these typical intertidal communities of high conservation value. In addition, no data are presently available for ecologically important Mediterranean salt marshes (MA25).

3.1 Introduction

Marine habitats, categorised in the European Nature Information System (EUNIS) under class MA, are distinguished from other ecosystems by their direct connection to the sea. Most of these systems are either not covered with vascular plants or fully aquatic, and therefore out of the scope of this background document on the effects of atmospheric N deposition and empirical critical loads. However, coastal salt marshes around and above the high (spring) tide in tidal regions are included in marine habitats (Class MA) and therefore treated in this chapter. Since the 2011 update, hardly any new evidence has become available for this EUNIS class, thus, the content of this chapter is more or less identical to that of 2011 (Bobbink and Hettelingh, 2011), except for a few corrections and the outcome of two additional "gradient" studies for salt marshes. No N addition studies with low enough N loads have been published in the present update and revision period.

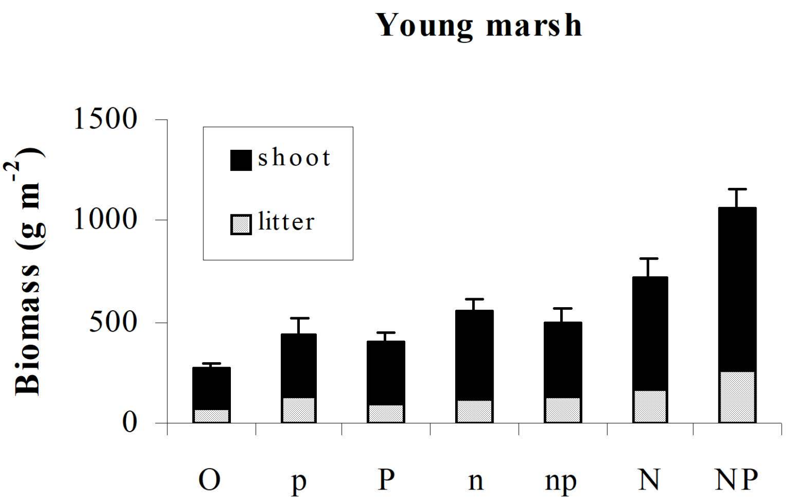
3.2 Atlantic coastal salt marshes and saline reed beds (MA22)

Salt marshes develop where fine sediments accumulate along sheltered coastlines. They are often associated with estuaries, but also frequently occur in areas where the coastline is protected by islands and sandbars. They are typically intertidal, i.e. located in areas that lie between lowest and highest tide and are periodically covered with salty water. The dominant plants are rooted macrophytes and shrubs which are adapted to the environmental stresses associated with sea water inundation (Archibold, 1995). They are characterised by an open nutrient cycle, receiving large amounts of nutrients from surface water, and exporting similarly large amounts of nutrients through surface water and denitrification (for N). This has led to the conclusion, in accordance with Morris (1991), that these systems are not vulnerable to the effects of increased atmospheric N deposition, at least not at most current deposition rates.

However, it is generally accepted that salt-marsh vegetation is primarily N limited (Mitsch and Gosselink, 2000) and N limitation has been demonstrated, for example, in European salt marshes at the Dutch island of Schiermonnikoog (Kiehl et al., 1997) and in Norfolk, in the United Kingdom (Jefferies and Perkins, 1977). During salt-marsh succession, N accumulates in organic material, and N mineralisation increases as marshes age, as shown by Olff et al. (1993) and Van Wijnen et al. (1999). This accumulation of N is considered as a major driving force behind succession, as competition for nutrients is replaced by competition for light.

Van Wijnen and Bakker (1999) added 50 kg N ha^{-1} yr^{-1} for three years to a 15-year-old salt marsh (EUNIS category MA224 - Atlantic mid-low salt marshes) and a 100-year-old salt marsh (EUNIS category MA223 - upper-mid salt marshes) in the Netherlands (background deposition 15-20 kg N ha^{-1} yr^{-1}). Biomass increased significantly after N application from the first growing season in the young salt marsh, and it continued to be higher during all three years of this treatment than in the control treatment (Figure 3.1). In the older salt marsh, however, the addition of 50 kg N ha^{-1} yr^{-1} had no significant effect on biomass, although the response to a much higher N application (250 kg N ha^{-1} yr^{-1}) showed that the vegetation was at least partly N limited (Van Wijnen and Bakker, 1999). Fertilisation increased biomass of late-successional species and decreased the floristic differences between the young and old marshes. However, these species-composition responses were measured only in the combined high N (250 kg N ha^{-1} yr^{-1}) and high P treatments, compared with the control situation. Thus, the effect of N on species composition could not be separated from the effect of adding P. However, as the effects of added P on biomass were either non-significant or quite small compared to the effects of N, there is a clear indication that increased N availability does increase the rate of succession. The successional age of these salt marshes is an important determinant of their quality as staging areas for Brent and Barnacle geese (*Branta bernicla* and *Branta leucopsis,* respectively) (Bakker, 1985). The increases in N deposition might decrease the surface area of early successional vegetation on the marsh and thereby the foraging area that is suitable for these migratory birds. Salt marsh areas that are also particularly important for migratory birds are those located in the southernmost part of Europe (e.g. Doñana National Park and Bay of Cádiz in Spain). However, there are to date no available studies that have evaluated the effects of increased N deposition on Mediterranean-type salt marshes (MA25).

Figure 3.1. Above-ground biomass of young salt marsh vegetation (MA224) in the Netherlands, after a 1-year addition of differential nutrients; lower case n = 50 kg N ha^{-1} yr^{-1}, capital N = 250 kg N ha^{-1} yr^{-1}, lower case p = 20 kg P ha^{-1} yr^{-1}, capital P = 100 kg P ha^{-1} yr^{-1}.



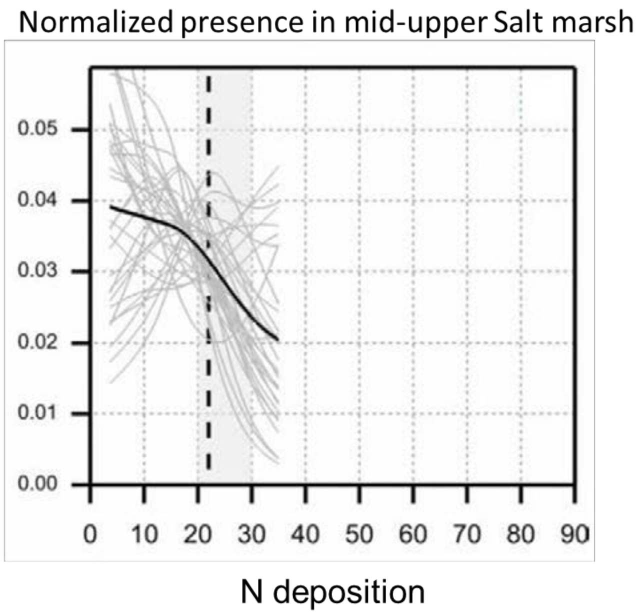
Source: adapted from Van Wijnen and Bakker, 1999

During primary succession N accumulates in organic material in the soil and is one of the main driving forces of succession. Increased N deposition will accelerate this natural process, but, because it does not affect the sediment accretion rate of salt marshes, this will result in a net loss of salt marshes of a low successional age (EUNIS categories MA225 and MA224). Information from long-term monitoring (25 years) of vegetation on one Dutch island with an estimated total N deposition of 15 to 20 kg ha^{-1} yr^{-1} showed a trend towards more eutrophic vegetation, both in grazed and ungrazed mid-successional salt marshes (Dijkema et al., 2005). This study was based primarily on mid-successional salt marshes (MA223); in early successional salt marshes the trend was less pronounced, partly because of the very low number of species. This has been considered as an indication for a $CL_{emp}N$ of 15-20 ha^{-1} yr^{-1} in the previous update and revision.

Recently, changes in species composition – with positive indicator species – along a nitrogen deposition gradient has been studied in Ireland (Aherne et al., 2020). Vegetation data (relevés) of Irish salt marshes (EUNIS code MA22x) were obtained from the Irish National Parks & Wildlife Service (Salt Marsh Monitoring Program [SMMP]). Unfortunately, no differentiation could be made in the analysis between the different salt marsh types in this data set, but the proposed habitat maps suggest that the relevés predominantly (68%) represented MA223 and MA224. The TITAN (Threshold Indicator Taxa Analysis) model has been used to detect changes in plant species distributions (e.g. plant species abundance) along an environmental gradient, such as N deposition; the location along the gradient where the greatest change occurs is called the ‘change point’ (Baker and King, 2010). Evidence for community thresholds is suggested by a convergence (or synchrony) of individual species change points along the environmental gradient. TITAN produces two change points: one represents the species that significantly decrease in abundance along the environmental gradient (z−), and the other represents the species that significantly increase along the environmental gradient (z+). This study focused on changes in positive indicator species of the salt marshes (Aherne et al., 2020). 65 plant species were present in the Irish salt marshes dataset (EUNIS code MA22x), with 11 of the 15 species with a z− change point were positive indicator species, and the z− community change point was 7.8 kg N ha^{-1} yr^{-1}. The authors suggested a new range for Atlantic salt marshes of 5-10 kg N ha^{-1} yr^{-1}, well below the previously set up $CL_{emp}N$ range (20-30 kg N ha^{-1} yr^{-1}).

Furthermore, dose-response relations for 60 Annex 1 habitat types and N deposition based on the responses of individual plant species were estimated (Wamelink et al., 2021). The deposition was linked to the plant species present in over 400,000 vegetation relevés of the European Vegetation Archive (EVA) database (http://euroveg.org/eva-database). In this study relevés from 14 countries, from Portugal to Finland, and from Ireland to Austria were used. The total N deposition (EMEP) at the site was calculated as the average deposition of the previous five years. A response curve for N deposition was estimated for each species. From this spline function, the percentiles were used to estimate the response of a habitat type to N deposition. The percentiles and the occurrence of positive indicator species of a habitat type were added together – after normalisation - and subsequently a mean response curve per habitat was estimated (Wamelink et al., 2021). This approach did not reveal a “change range” for pioneer salt marshes dominated by *Salicornia* spp. (MA225), but for mid-low salt marshes (MA224) and upper-mid salt marshes (MA223) a “change range” of 15-20 kg N ha^{-1} yr^{-1} was observed (Figure 3.2). Thus, the results of this study also indicate a lower $CL_{emp}N$, especially for the two just-mentioned salt marshes.

Figure 3.2. Normalised presence (Y-axis) of all indicator species (grey lines) and the mean of the positive indicator species (black line) for the upper-mid salt marsh (MA225) against total N deposition (kg N ha^{-1} yr^{-1}). Grey area is the $CL_{emp}N$ range in 2011, striped vertical line the $CL_{emp}N$ according to the Dutch modelling approach.



Source: Wamelink et al., 2021

Based on the previous expert judgement and the new evidence from the two "gradient studies", a new critical load range of 10 to 20 kg N ha^{-1} yr^{-1} is recommended for mid-low and upper-mid salt marsh systems (MA224 and MA223; Table 3.1). The lower limit of 10 is the mean of 5 (Aherne et al., 2020) and 15 (Wamelink et al., 2021). The upper limit represents the maximum value proposed by Wamelink et al. (2021). For pioneer salt marshes (MA225) the range of the $CL_{emp}N$ has not been revised. However, field experiments with lower N additions over a longer period of time and/or additional gradients studies to improve the reliability for these adaptations are urgently needed, particularly in southern European latitudes that harbour areas of extremely high importance for migratory birds.

Table 3.1. $CL_{emp}N$ and effects of exceedances on marine habitats (MA). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Atlantic upper-mid salt marshes	MA223	20-30	(#)	**10-20**	(#)	Increase in dominance of graminoids; decline positive indicator species
Atlantic mid-low salt marshes	MA224	20-30	(#)	**10-20**	(#)	Increase in late successional species; decline positive indicator species
Atlantic pioneer salt marshes	MA225	20-30	(#)	20-30	(#)	Increase in late successional species; increase in productivity species

3.3 References

Aherne, J., Wilkins, C. and Cathcart, H. (2020). Nitrogen-sulphur critical loads: assessment of the impacts of air pollution on habitats (2016-CCRP-MS.43). EPA research report, Wexford, Ireland.

Archibold, O.W. (1995). *Ecology of world vegetation*. Chapman & Hall, London.

Baker, M.E. and King, R.S. (2010). A new method for detecting and interpreting biodiversity and ecological community thresholds. *Methods in Ecology and Evolution* **1**, 25-37.

Bakker, J.P. (1985). The impact of grazing on plant communities, plant populations and soil conditions on salt marches. *Vegetatio* **62**, 391-398.

Bobbink, R. and Hettelingh, J.P. (2011). *Review and revision of empirical critical loads and dose-response relationships.* Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), RIVM report 680359002/2011 (244 pp).

Dijkema, K.S., Van Duin, W.E. and Van Dobben, H.F. (2005). Kweldervegetatie op Ameland: effecten van veranderingen in de maaiveldhoogte van Nieuwlandsrijd en De Hon. In: *Begeleidingscommissie Monitoring Bodemdaling Ameland, Monitoring effecten van bodemdaling op Ameland-Oost: evaluatie na 18 jaar gaswinning*.

Jefferies, R.L. and Perkins, N. (1977). The effects on the vegetation of the additions of inorganic nutrients to salt march soils at Stiffkey, Norfolk. *Journal of Ecology* **65**, 867-882.

Kiehl, K., Esselink, P. and Bakker, J.P. (1997). Nutrient limitation and plant species composition in temperate salt marches. *Oecologia* **111**, 325-330.

Mitsch, W.J. and Gosselink, J.P. (2000). *Wetlands, 3rd edition*. Wiley, New York.

Morris, J.T. (1991). Effects of nitrogen loading on wetland ecosystems with particular reference to atmospheric deposition. *Annual Review of Ecology and Systematics* **22**, 257-279.

Olff, H., Huisman, J., and Van Tooren, B.F. (1993). Species dynamics and nutrient accumulation during early primary succession in coastal sand dunes. *Journal of Ecology* **81**, 693-706.

Van Wijnen, H.J. and Bakker, J.P. (1999). Nitrogen and phosphorus limitation in a coastal barrier salt marsh: the implications for vegetation succession. *Journal of Ecology* **87**, 265-272.

Van Wijnen, H.J., Van der Wal, R. and Bakker, J.P. (1999). The impact of herbivores on nitrogen mineralization rate: consequences for salt-marsh succession. *Oecologia* **118**, 225-231.

Wamelink, G.W.W., Goedhart, P.W., Roelofsen, H.D., Bobbink, R., Posch, M., Van Dobben, H.F. and Data providers (2021). Relations between the amount of nitrogen deposition and habitat quality. Wageningen, Wageningen Environmental Research Rapport 3089; ISSN 1566-7197.

4 Effects of nitrogen deposition on coastal habitats (EUNIS class N, formerly B)

Adapted by Laurence Jones, Emiel Brouwer and Eva Remke

Coastal landscape, Terschelling (The Netherlands). Photo: Eva Remke.

Summary

In this chapter empirical N critical loads ($CL_{emp}N$) for coastal habitats have been updated and revised where necessary. Since the last $CL_{emp}N$ update in 2011, a number of experimental and gradient studies have been published for coastal dunes and sandy shores (N1). Based on new evidence, the $CL_{emp}N$ ranges were confirmed or narrowed down to a lower level. For shifting coastal dunes (N13, N14), the $CL_{emp}N$ range of 10 to 20 kg N ha^{-1} yr^{-1} was confirmed. However, it should be kept in mind that the mechanisms of N impact may differ in temperate Europe compared with the Mediterranean. Compared to the last review in 2011, it is proposed to decrease the $CL_{emp}N$ range for coastal dune grasslands (N15), coastal dune heaths (N18, N19) and moist to wet dune slacks (N1H). For these subcategories, the evidence status should be increased to reliable (dune grasslands) or quite reliable (dune heaths, dune slacks).

New in this chapter, compared to the last update by Bobbink and Hettelingh (2011), is the inclusion of the EUNIS class dune slack-pools which belong to the freshwater aquatic communities of permanent Atlantic and Baltic (N1H1) or Mediterranean and Black Sea (N1J1) dune-slack water bodies (Chytrý et al., 2020). Due to relatively little new evidence for this habitat the $CL_{emp}N$ range did not change.

Beside this good evidence for coastal dunes and sandy shores (N1), there remain major knowledge gaps for coastal shingle (N2) and rock cliffs, ledges and shores, including the supralittoral (N3), for which $CL_{emp}N$ could not be established due to a lack of evidence.

4.1 Introduction

This chapter presents an evaluation of the impacts of atmospheric nitrogen (N) deposition on coastal habitats (class N, formerly B) of the European Nature Information System (EUNIS), with respect to the setting of empirical critical loads of N ($CL_{emp}N$). Coastal habitats are defined as situated above the high spring tide limit (or above mean water level in non-tidal waters) with coastal features and characterised by their proximity to the sea. They include coastal dunes (dry grasslands, wet to moist dune slacks, dune-slack pools, scrub and wooded dunes), beaches and cliffs (Davies et al., 2004). Dune-slack pools in the EUNIS system were classified under permanent oligotrophic waters in Bobbink and Hettelingh (2011). However, due to their coastal location, this habitat is now incorporated within the coastal sand dunes chapter for the purposes of $CL_{emp}N$ evaluation.

The first subdivision within class N in EUNIS is based on underlying substrates, that is, sand, shingle or rock, but data to support proposals for $CL_{emp}N$ are only available for sand substrates (N1; coastal dune and sandy shores). The following chapter integrates information from the last review of $CL_{emp}N$ (Bobbink and Hettelingh, 2011) with information published since the last review to evaluate the total evidence base and determine whether to retain or revise the $CL_{emp}N$ values. In this background document, separate critical load values for N deposition are evaluated and reviewed for major habitats within the EUNIS category N1. There remain major knowledge gaps on the effects of atmospheric N on the other coastal habitats, including coastal shingle (N2) and rock cliffs, ledges and shores, including the supralittoral (N3).

4.2 Coastal dunes and sandy shores (N1)

Dune ecosystems in the coastal areas of Europe retain a large part of their original plant and animal life, and are thus a major reservoir of European biodiversity, especially for lowland species adapted to calcareous substrates which have been lost elsewhere due to agricultural expansion. They are found on sandy, nutrient-poor soils, and considered to be sensitive to the impacts of both eutrophication and acidification (e.g. Ellenberg, 1988a; Wellburn, 1988; De Vries et al., 1994). With respect to the setting of $CL_{emp}N$ in coastal dune and sand habitats, evidence exists only for some EUNIS categories, namely those of shifting coastal dune (N13 and N14), coastal dune grassland (grey dune) (N15), coastal dune heaths (N18 and N19), Atlantic and Baltic moist and wet dune slack (N1H), and dune slack pools (N1H1, N1J1).

4.2.1 Shifting coastal dunes (N13, N14)

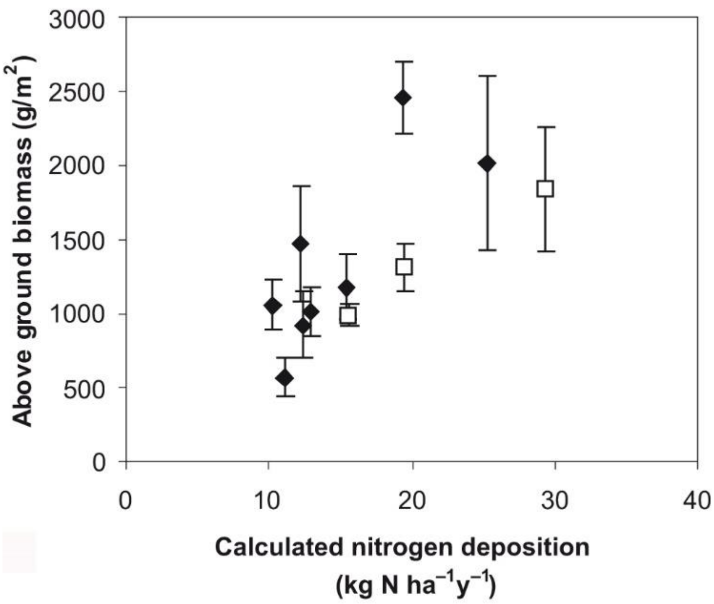
Shifting coastal dunes are coastal, mobile sand habitats of the boreal, nemoral, steppe, Mediterranean and warm-temperate humid zones of Europe. They include embryonic shifting dunes at the beach and shifting dunes along the shoreline with *Ammophila arenaria* ('white dunes'). There is only little evidence available on this EUNIS category. This includes two experimental studies: one from Iceland with rather high N addition rates but low ambient deposition (Greipsson and Davy, 1997) and evidence from a new study in Italy with low additions and also low ambient deposition (Menicagli et al., 2020). The other evidence comes from a gradient study in the UK (Jones et al., 2004). There are other, non-European studies on shifting dunes, but the studies are carried out at relatively high ambient N deposition (15 kg N ha^{-1} yr^{-1}) and with high experimental additions of three to nine times ambient N deposition (Bird and Choi, 2017). For these reasons, their results are not included in this review.

In the Iceland study (Greipsson and Davy, 1997), the effects of N addition over a two-year period were reported for coastal dunes (probably Atlantic and Baltic shifting coastal dune EUNIS N13) in a region with very low atmospheric N deposition. The number of flowering spikes and total seed weight of the tall dune grass *Leymus arenarius* strongly increased within the first year of application of either 50 or 100 kg N ha^{-1}. This study is not ideal as nitrogen fertiliser has been applied only once (in June) and in high doses, but as there are only limited studies available for this habitat type it has been included in this review.

The Italian study consisted of a one-year field experiment in Mediterranean, Macaronesian and Black Sea shifting coastal dunes (N14) on the Tuscan sandy coast (Menicagli et al., 2020). The study examined the individual and combined effects of N deposition (current and predicted in the Mediterranean for 2050), macro-plastic and biotic condition on the performance of vegetative propagules of dune plants. Two clonal grass species typical for the Mediterranean embryonic dunes were chosen: *Thinopyrum junceum* (L.) A. Love (previously known as a *Elymus farctus* or sand couch), a typical dune-building and endemic species, and *Sporobolus pumilus* (Roth) P.M. Peterson and Saarela (previously *Spartina patens*), a generalist species not involved in the dune formation process and probably introduced from North America. Background wet N deposition rate along the Tuscan coast ranged from 4.7 to 5 kg N ha^{-1} yr^{-1} (Marchetto et al., 2014). As a necessary simplification to estimate total N deposition, dry deposition has been assumed to be broadly equivalent to wet deposition (this is typical of seasonally dry Mediterranean environments), giving a background load of approximately 10 kg N ha^{-1} yr^{-1}. In experimental plots an amount of 5.2 kg N ha^{-1} yr^{-1} was added, which would give a total N input of ~15 kg N ha^{-1} yr^{-1}, which is in the range of the atmospheric N inputs predicted in the Mediterranean for 2050 (10-15 kg ha^{-1} yr^{-1}, Phoenix et al., 2006). Nitrogen additions were applied in four doses per year (liquid solution of NH_4NO_3). After one year, *T. junceum* plants had a significantly lower root biomass with added N; shoot biomass and establishment probability was slightly lower, though not significantly due to low replicate numbers. *S. pumilus* showed no changes to N addition alone, but it did so in interaction with added bio-degradable plastic where N and plastic together were found to increase root growth. This study indicates that elevated N could hinder growth of the dune-building function of the endemic species *T. junceum*.

Supporting evidence for the effects of N deposition is also available in the form of a targeted gradient study in the United Kingdom (Jones et al., 2002, 2004). Eleven sand dune sites were surveyed with a range of atmospheric N inputs from 10 to 30 kg N ha^{-1} yr^{-1}. The relationship between site parameters and N deposition was examined using linear regression. Each parameter was also checked for significant soil pH effects. Where significant relationships with pH occurred, pH was included as the first term of the regression to separate these effects from those of atmospheric N, with the assumption that these represented existing underlying gradients in sand parent material rather than effects of acidification due to atmospheric deposition in generally well-buffered systems. Above-ground biomass ($p < 0.05$) and sward height ($p < 0.10$) related positively to N inputs (Figure 4.1). Consequently, there was also a positive correlation between N deposition and the pool of N in the vegetation. The increase in biomass was largely caused by the increased height and cover of the typical grass species *Ammophila arenaria*. In general, the observed effects start to become apparent above 10 kg N ha^{-1} yr^{-1}, if there are no other modifying factors. The authors suggested that, in the long term, this increase in biomass may also lead to enhanced organic matter accumulation and thus accelerated soil development and increased succession rates. This agrees with the evidence provided by Jones et al. (2008), where accelerated dune soil development was positively correlated with both N deposition and temperature. The longer-term consequences of increased grass height and vigour in shifting dunes are likely to be a decrease in the area of bare sand which are important for many rare invertebrate and other species (Howe et al., 2010), and reduced natural dynamics which is important for a healthy dune system (Jones et al., 2021). As with all gradient studies, this survey indicates an association and not causality, but on the basis of these results it appears likely that the sites with the higher N inputs have been impacted.

Figure 4.1. Above ground biomass (g m^{-2}) in relation to N deposition in a UK survey of mobile and semi-fixed dunes. Filled diamonds represent calcareous sites, open squares represent acidic sites. Bars show ± 1 SE.



Source: Jones et al., 2004

Summary for shifting coastal dunes (N13, N14)

The 2011 $CL_{emp}N$ range was 10-20 kg ha^{-1} yr^{-1} (expert judgement). Taken together, the British gradient study and the new experimental evidence from the Mediterranean region support a negative impact on shifting coastal dunes from 10 kg N ha^{-1} yr^{-1} upwards. However, the mechanisms of N impact may differ in temperate Europe compared with the Mediterranean. In the drier Mediterranean there may be interactions with drought stress since N reduces root biomass of dune building species common in embryonic dunes, while in temperate dunes N generally led to increased total cover or biomass of the dominant grasses. Despite differences in potential mechanisms acting in the Mediterranean compared with temperate Europe, the evidence for embryonic dunes suggests retaining the $CL_{emp}N$ range for shifting coastal dunes of 10-20 kg ha^{-1} yr^{-1} but increasing the evidence status to quite reliable. Further experimental work is required to improve the evidence base for this habitat, and also to better understand the mechanisms of impact across different European climatic regions.

4.2.2 Coastal dune grassland (grey dune) (N15)

A large number of stable dune grasslands are located along the coasts of Europe, from the boreal to the Mediterranean and warm-temperate zones. They are found in fixed dunes, usually with herbs and graminoids as the dominant life form, although in certain areas in the northern and western systems, bryophytes make up a substantial component of the biomass – up to 70 %, particularly where grazed (e.g. Plassmann et al., 2009). In early *Corynephorus* stages in drier dunes, mosses and lichens may dominate, both in abundance and in species richness. They occur in dune habitats which are mostly out of reach of the water table (typically Mean Spring water Level values deeper than 85 cm) and occur on calcareous to acidic sandy soils, thus from high to low base status (e.g. Davies et al., 2004). In general, these stable dune grasslands have a high species diversity and many characteristic plant and animal species.

Synthesis of observed effects in dune grasslands

In many Dutch dry dune grasslands, tall grasses have increased since the 1970s, a period with relatively high N loading (20-30 kg N ha^{-1} yr^{-1}) and sulphur (S) deposition. The dominant tall grass species are mainly *Calamagrostis epigejos, Elymus repens* and *Elymus athericus* (Kooijman and De Haan, 1995; Remke, 2010). In more acidic or decalcified (i.e. partially acidified) dunes, *Ammophila arenaria and Carex arenaria* are usually the dominant species. Because of reduced light penetration through the tall grass canopies formed by these species, the development of several prostrate species has been reduced and management is now necessary to maintain the diversity of these systems. In the past, tall graminoids were usually not dominant on these low nutrient sandy dune soils in the Netherlands. A survey in the 1990s of dry dune grasslands along the Dutch coast revealed that non-calcareous, iron-poor dry dune ecosystems were N limited, but that in calcareous, iron-rich dunes there was co-limitation of N and phosphorus (P) (Kooijman et al., 1998; Kooijman and Besse, 2002). Kooijman et al. (1998) concluded that atmospheric N deposition may cause tall grass dominance encroachment in non-calcareous dunes, but probably only accelerates the process in calcareous dune grasslands. Yet, a strong, negative correlation between the percentage of open dunes and total N deposition, especially above 15 kg N ha^{-1} yr^{-1}, has been seen in both Dutch dune regions (Van Hinsberg and Van der Hoek, 2003). The hypothesis that the present dominance of tall grasses and increased rate of succession in the Netherlands might be a result of increased atmospheric N deposition is also supported by the fact that in many coastal areas of Britain, receiving relatively lower N deposition loads (approximately 10 kg N ha^{-1} yr^{-1}), stable dune grasslands are still rich in species (Jones et al., 2002; Field et al., 2014; Jones et al., 2018). N deposition is recognised as a major, although not the only, factor altering dune vegetation since approximately the 1960s. Other factors include reduction in traditional grazing, climate change and reductions in rabbit populations (Provoost et al., 2011). All of these factors, in combination with increased N deposition, tend to drive vegetation change in the same direction.

The principal new sources of evidence to add to the previous assessment come from Rowe et al. (2011), Hall et al. (2011), Jones et al. (2013), Field et al. (2014), Ford et al. (2016), Pakeman et al. (2016), Aherne et al. (2021) and Payne et al. (2020).

Evidence from short-term, high dose, N-manipulation experiments

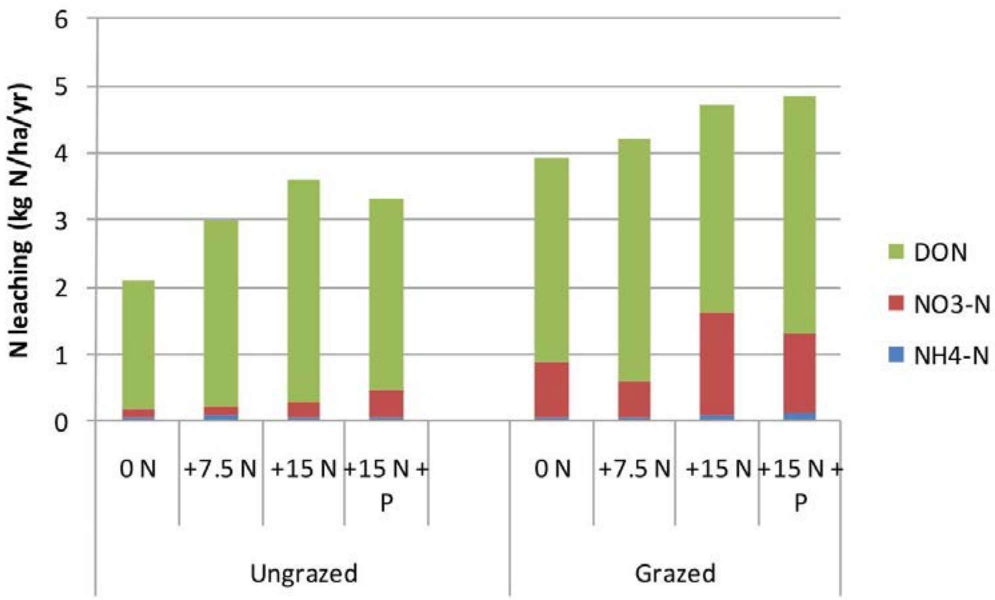
The effects of nutrients in dry dune grasslands on sandy soils (calcium carbonate 1%) were experimentally studied at Braunton Burrows (Devon, UK) by Willis (1963). Nutrients were applied during a two-year period and complete NPK fertilisation strongly stimulated the growth of grasses, such as *Festuca rubra*, *Poa pratensis* and *Agrostis stolonifera,* which significantly reduced the abundance of many small plants, such as prostrate phanerogamic species, mosses and lichens. The impacts of different combinations of N, P and potassium (K) were also investigated and N (> 100 kg N ha^{-1} yr^{-1}) proved to be more limiting for plant growth than P. Although the changes in vegetation were clearly less profound than after complete fertilisation, reduction in species numbers (especially annual species, lichens and mosses) was observed under N addition (Willis, 1963). Boorman and Fuller (1982) examined the effects of nutrient additions on species composition of rabbit grazed dune grassland in Norfolk (UK) over a five-year period. They added 80 kg N ha^{-1} yr^{-1} as $(NH_4)_2SO_4$ and $NaNO_3$ in five replicates in April, June and September of each year. The grazing prevented *F. rubra* from becoming dominant, but several species (especially annuals, mosses and lichens) declined, while two species (*Carex arenaria* and *Calystegia soldanella*) increased under all treatments containing N (80 kg N ha^{-1} yr^{-1}). In this study, no evidence was found for reduced diversity in plots that had received P and K additions. In a one-year experiment with additions of N (20, 40, 80 and 160 kg N ha^{-1} yr^{-1}; atmospheric load 15 kg N ha^{-1} yr^{-1}) or P, the above-ground biomass of a stable dune grassland at the Dutch Wadden island of Schiermonnikoog proved to be strongly N limited; plant biomass significantly increased above additions of 40 kg N ha^{-1} yr^{-1} in this 30-year-old stable dune grassland (Olff et al., 1993). In summary, these short-term experiments clearly indicate the importance of N limitation in several stable dune grasslands, but the high N doses applied prevent a reliable determination of $CL_{emp}N$. Moreover, in some cases, co-limitation with P has been observed, or rabbit grazing may have prevented the dominance of tall grasses, suggesting the importance of these two drivers as modulators of the response of these ecosystems to increased N deposition.

Evidence from longer-term, low dose, N-manipulation experiments

Two low-dose N addition experiments have been carried out in stable dune grasslands, one in the Netherlands, and one in the UK, with the specific objective to examine the effects of N deposition. In the Dutch N addition experiment, the effects of N additions and the interaction with rabbit grazing were investigated in a factorial design in two coastal stable dune grasslands, one calcareous and one partly decalcified, by Ten Harkel and Van der Meulen (1995) and Ten Harkel et al. (1998). After four years of N additions twice a year (25 kg N ha^{-1} yr^{-1} in the form of ammonium nitrate pellets; background deposition approximately 23 kg N ha^{-1} yr^{-1}) no significant changes were found in species composition, neither in the grazed nor the ungrazed situation. Exclusion of grazing by rabbits and horses, through the use of enclosures, resulted in graminoid dominance (*Festuca rubra*, *Festuca ovina* and *Poa pratensis*), especially where N additions were made, which suggested that grazing may prevent grass dominance in stable dune grasslands (Ten Harkel and Van der Meulen, 1995). Because of the high, direct leaching losses resulting from the added fertiliser pellets used in the first phase of the experiment, for the last year and a half of the experiment the treatment was changed to a fortnightly solute addition by watering of 50 kg N ha^{-1} yr^{-1} as ammonium sulphate (Ten Harkel et al., 1998). In the last year and a half, even in the no added-nitrogen controls in the ungrazed exclosures, the data suggest that there was leaching of 36% of N inputs from background deposition in the foredune plots and 13% of N inputs in the older decalcified plots, suggesting a degree of N saturation already at background deposition of 23 kg N ha^{-1} yr^{-1} (Ten Harkel et al., 1998). From this experiment in stable dune grasslands, experimental N enrichment had no effect on species composition after four years of increased N addition. This may have been related to a shift towards P limitation after the long period of high atmospheric N inputs in the Netherlands, during which time some botanical changes may already have occurred, and the relatively high rabbit grazing pressure in that period which may enhance nutrient co-limitation. However, the high N leaching from the control unfertilised vegetation is also a strong indication of N saturation of these dune grasslands, probably because of the high N deposition rates (> 20 kg N ha^{-1} yr^{-1}) over several decades, but also because the young dune soils have a thin top layer with relatively low organic matter content, lack clay particles, and therefore have fewer exchange sites with which to bind ammonium ions.

In the N addition experiment in the UK, the effects of low levels of N additions (and P) were studied on the Isle of Anglesey under relatively low background deposition (10-12 kg N ha^{-1} yr^{-1}) in a calcareous, heavily grazed fixed dune grassland (Plassmann et al., 2009). The experiment also looked at the impacts of grazing management in combination with N. Four N treatments (unwatered control, watered control, 7.5 kg N ha^{-1} yr^{-1}, 15 kg N ha^{-1} yr^{-1}, on top of the background N deposition) were nested within three grazing treatments. In a separate treatment, effects of fertilisation with both N (15 kg ha^{-1} yr^{-1}) and P (15 kg ha^{-1} yr^{-1}) on top of background N were investigated. After two years, N addition resulted in significantly greater amounts of total above-ground biomass and bryophyte biomass, under both low and high N treatment, compared to the control situation. In bryophytes, the tissue N concentration was significantly greater under high N treatment (15 kg N ha^{-1} yr^{-1}), whereas the total N pool in the bryophytes was significantly greater under both N addition treatments. No effects on vegetation composition, sward height or soil parameters occurred within the two-year research period. Furthermore, combined addition of N and P together had a greater impact on above-ground biomass, sward heights and sward structure than N addition alone. Demonstrating similarities with the Dutch experiment, further work at the site after six years of nutrient additions, on N and P mineralisation suggests the site is N and P co-limited, and that there were no longer differences in moss biomass due to elevated N, unless P was also added (Ford et al., 2016). Leaching calculations at this site show that the majority of deposited N is retained within the soil-plant system (Hall et al., 2011). A maximum of 6 % of inputs was leached at the highest N load in the fully grazed treatments (combined experimental and background load of 27 kg ha^{-1} yr^{-1}) in the form of inorganic N (or up to 18%) if dissolved organic N compounds were included in the calculations (Figure 4.2). Dynamic modelling of various scenarios of increased N deposition from a nearby poultry unit suggests that the accumulated soil N will eventually trigger changes in botanical composition over longer time scales at additions above current background levels of ~11 kg N ha^{-1} yr^{-1} (Rowe et al., 2011). These experimental and modelling results suggest that N and P co-limitation may prevent species composition changes in the short-term in dune grasslands, but that N still accumulates in the soil and plant system, and is likely to eventually cause species change at additions above ~11 kg N ha^{-1} yr^{-1}.

Figure 4.2. Leaching fluxes of inorganic and organic N with N additions (kg N ha^{-1} yr^{-1}) above a background of 10-12 kg N ha^{-1} yr^{-1} under two grazing regimes on a partially de-calcified calcareous fixed dune grassland at Newborough Warren, North Wales, UK. (DON = dissolved organic nitrogen).

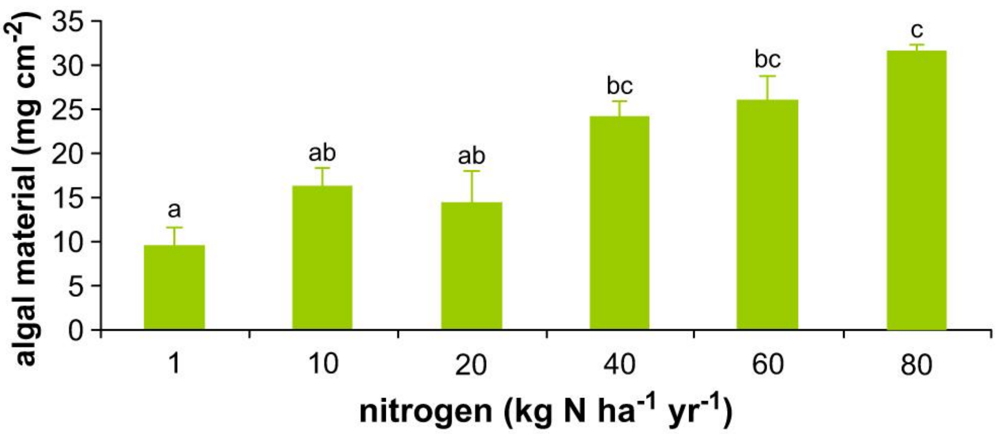


Source: Hall et al., 2011

Evidence from mesocosm studies

The effects of N loads have also been studied in a series of mesocosm studies in the Netherlands and the UK. The effects of elevated N loads in a situation of low background deposition (< 5 kg N ha^{-1} yr^{-1}) have been studied during two to three years, in recreated dry dune calcareous grassland mesocosms in a greenhouse (1 x 1 m in size) (Tomassen et al., 1999; Van den Berg et al., 2005). After a pre-treatment period of two months with clean rainwater that removed the excess of nitrate from the soil, N was added twice a week in the form of ammonium nitrate (1, 5, 10, 15, 20, 40, 60 and 80 kg N ha^{-1} yr^{-1}). The effects on soil-pore water chemistry and on two characteristic graminoid species (*Calamagrostis epigejos* and *Carex arenaria*) and two endangered herb species were monitored. Within one year of N additions, a clear difference was found in the amount of green algae (Chlorophyta) growing on the surface of the sand. The amounts of green algae increased under the treatments of between 10 and 20 kg N ha^{-1} yr^{-1}, but the difference with the two lowest treatments was especially distinct above 20 kg N ha^{-1} yr^{-1} (Figure 4.3). The strong increase in algae on the soil layer due to N deposition may have important implications as the algae prevent sand drifts that are caused by wind action. Such 'blowouts' are important for renewed vegetation succession, and biodiversity will decrease when young successional stages decline. Concentrations of nitrate in soil-pore water showed a strong seasonal fluctuation. During the first winter period an increase in nitrate was measured for the treatments ≥ 40 kg N ha^{-1} yr^{-1}. During spring, nitrate concentrations rapidly decreased. During the second and third winter, an increase in nitrate could only be observed at the highest N addition level. Ammonium concentrations remained consistently at a very low level (< 5 µmol l^{-1}) most likely due to fast nitrification.

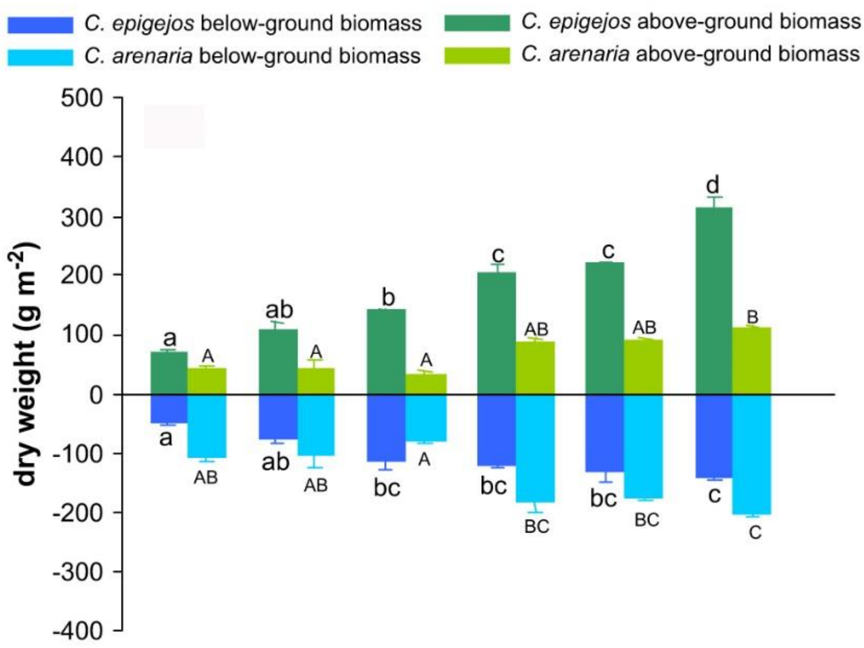
Figure 4.3. Algal material, measured as active chlorophyll concentration (mg cm^{-2}; means ± standard error; n = 4), in the top layer of the soil at different N addition rates.



Source: Van den Berg et al., 2005

Differences in plant growth were first observed after one year, and they became more obvious after two to three years. The total cover of the vegetation increased with elevated N inputs. This increase could almost completely be attributed to the growth of *Calamagrostis epigejos*. Biomass of this tall grass species increased significantly, above 20 kg N ha^{-1} yr^{-1} for shoots and above 15 kg N ha^{-1} yr^{-1} for roots (Figure 4.4). No clear effects of elevated N deposition rates on the two herbaceous species *Galium verum* and *Carlina vulgaris* were measured over the initial two years, however, in the third year, the number of individuals and dry weight of *G. verum* decreased significantly above 20 kg N ha^{-1} yr^{-1} (Van den Berg et al., 2005). After two years of treatment, the total amount of N stored in the vegetation was strongly elevated due to increased N deposition (Tomassen et al., 1999).

Figure 4.4. Above- and below-ground biomass (g m^{-2}) of *Calamagrostis epigejos* and *Carex arenaria* after two years of N application in coastal dune grassland (N15) mesocosms. From left to right: 1, 10, 20, 40, 60 and 80 kg N ha^{-1} yr^{-1}.



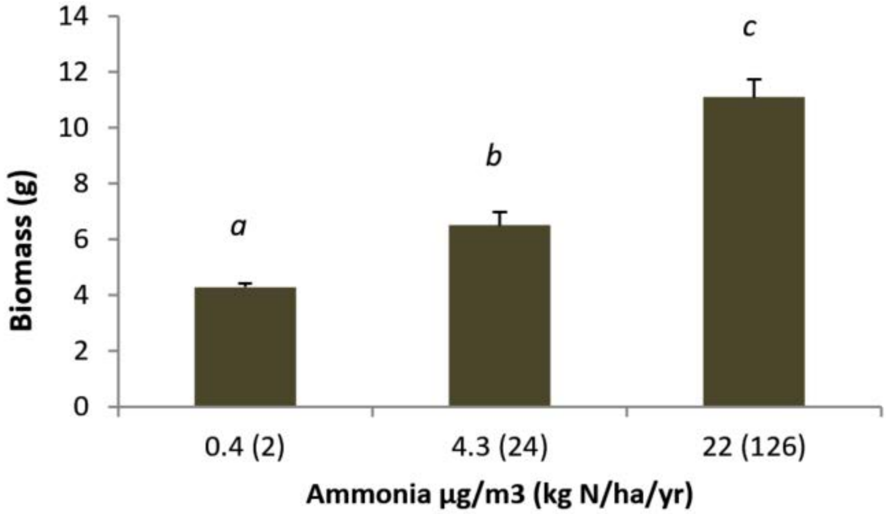
Source: Tomassen et al., 1999; Van den Berg et al., 2005

A series of mesocosm experiments were conducted in the UK (Mohd-Said, 1999; Jones et al., 2013), using a mix of seven dry dune grassland species including the following grasses and forbs: *Dactylis glomerata, Festuca rubra, Plantago lanceolata, Centaurea nigra, Achillea millefolium, Leontodon hispidus* and *Galium verum*. The species were planted in 4 litre, 20 cm diameter pots filled with sand, containing two individuals of each species in random positions, and exposed to N in three experiments: wet deposition, ammonia fumigation, and an *in-situ* exposure along a field-gradient of ammonia concentration. The simulated wet N deposition rates were an additional 2, 10, 20 and 55 kg ha^{-1} yr^{-1} on top of an estimated background of ~4 kg N ha^{-1} yr^{-1}. At additions of 10 kg N ha^{-1} yr^{-1} and over, the cover of the grass *Festuca rubra* increased, while there was no significant change in the cover of other grass or herb species (Figure 4.5). A separate experiment used the same species, exposed to different ammonia concentrations in open-top chambers, with treatments ranging from 0.4 µg m^{-3} NH_3 (equivalent to 2 kg N ha^{-1} yr^{-1}) up to 35 µg m^{-3} (200 kg N ha^{-1} yr^{-1}). The background deposition in this experiment was not measured, but it was estimated around 10-15 kg N ha^{-1} yr^{-1}. The total above-ground biomass (Figure 4.5a) showed a significant increase above an ammonia concentration of 0.4 µg m^{-3} (equivalent to N addition of 2 kg N ha^{-1} yr^{-1} on top of background of at least 10 kg N ha^{-1} yr^{-1}), and again above ammonia concentration of 4.3 µg m^{-3} (equivalent to 24 kg N ha^{-1} yr^{-1} on top of background).

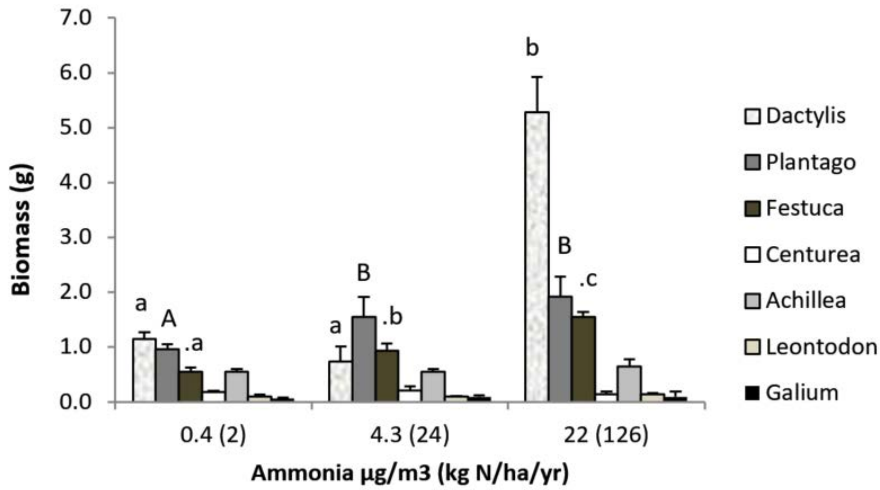
When biomass was analysed by species (Figure 4.5b), both *P. lanceolata* and *F. rubra* showed significant biomass increases above an ammonia concentration of 0.4 µg m^{-3}, and *D. glomerata* showed significant biomass increase above an ammonia concentration of 4.3 µg m^{-3}. The other species showed no significant effects of ammonia fumigation. However, all species showed significant increases in tissue N concentration with each successive treatment, i.e. showing luxury N accumulation, except *D. glomerata* which showed significant increases in tissue N at all but the lowest ammonia treatment. The third experiment involved *in situ* exposure to elevated ammonia concentrations in a field gradient study away from a poultry unit point source. Here, total biomass (Figure 4.6a), and for individual species *D. glomerata* and *P. lanceolata* (Figure 4.6b), increased between exposures of 8.3 and 11.1 kg N ha^{-1} yr^{-1}. Tissue N increased in all seven species between N exposure of 11.1 and 40 kg N ha^{-1} yr^{-1}.

Figure 4.5. Above-ground biomass showing a) total biomass of all species and b) individual dry dune grassland species after exposure to ammonia fumigation. Data from highest ammonia treatment (35 µg m^{-3}) not shown. Differing letters denote significant differences between treatments for each species ($p < 0.05$). Bars represent ± 1 s.e.

a)



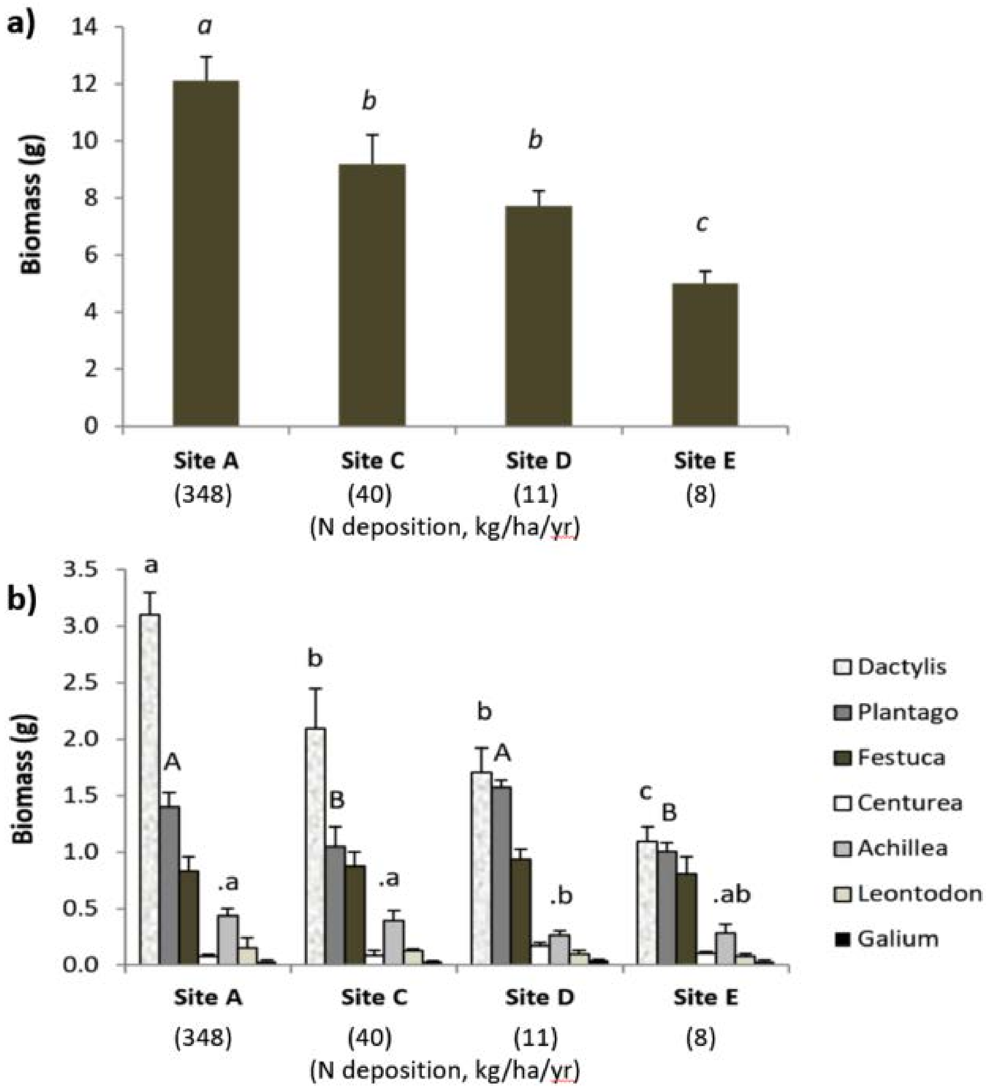
b)



Source: Jones et al., 2013

As a general conclusion, all the mesocosm experiments showed a significant increase in species cover that was often attributed to the response of a few dominant graminoid species, with the three separate UK mesocosm experiments consistently showing effects on the mainly calcareous species between total N loads of around 8 to 14 kg N ha^{-1} yr^{-1}. In all experiments, higher loads led either to dominance of graminoid species within a few years (Dutch experiment), or considerably greater biomass, primarily of graminoid species, and increased tissue N concentrations in most species with increasing N (UK experiments).

Figure 4.6. Above-ground biomass of a) all species combined, and b) seven different dry dune grassland species after exposure to a field-gradient of ammonia concentrations away from a point source. Differing letters denote significant differences between treatments for each species ($p < 0.05$). Bars represent ± 1 s.e.

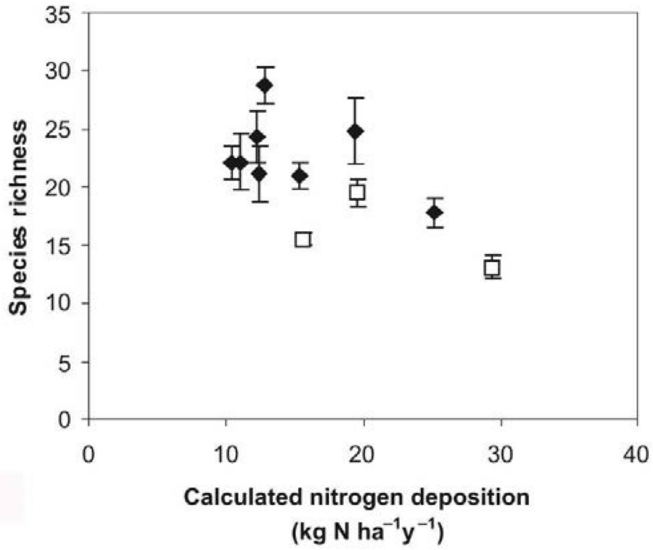


Source: Jones et al., 2013

Evidence from gradient studies

Additional field evidence on the effects of N deposition of dune grasslands is currently available from a series of gradient studies. A targeted survey in the coastal dune areas across England and Wales, in regions with much lower N deposition than in the Netherlands (Jones et al., 2004), surveyed eleven coastal dune sites with atmospheric N deposition ranging from 10 to 30 kg N ha^{-1} yr^{-1}. In these stable dune grasslands, above-ground biomass was related positively to N deposition, while species richness showed a weak negative relationship (Figure 4.7). In addition, in these grasslands, a strong positive relationship was found between dissolved organic N in groundwater and N deposition, which may be an indicator of high inorganic N inputs, or perhaps increased mineralisation in response to N deposition. Furthermore, the cover of *Carex arenaria* also related positively to N inputs. In general, the observed effects in this study started to become apparent in the range 10 to 20 kg N ha^{-1} yr^{-1}.

Figure 4.7. Species richness (2 x 2 m) of vascular plants, bryophytes and lichens in relation to N deposition in a UK survey of stable dune grasslands. Filled diamonds represent calcareous sites, open squares represent acidic sites. Bars show ± 1 s.e.



Source: Jones et al., 2004

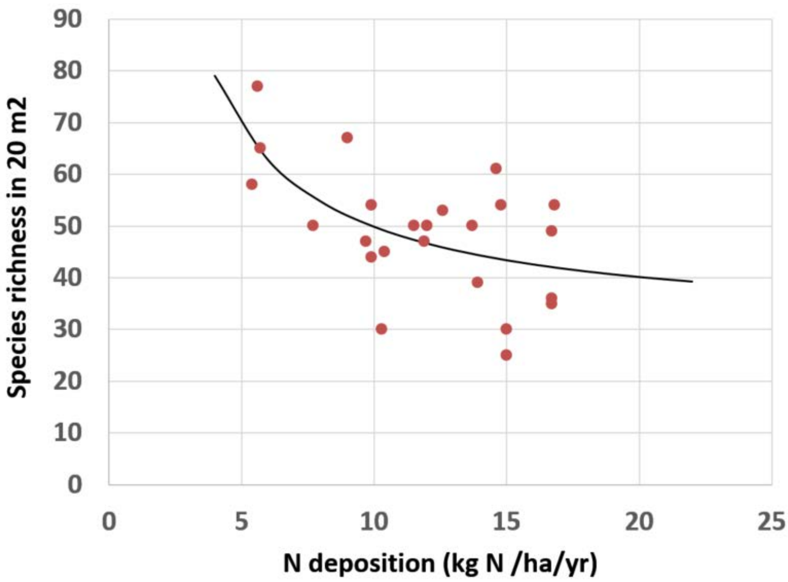
The impacts of atmospheric N deposition on dry lichen-rich dune grasslands (N151 – acidic to slightly calcareous) around the Baltic Sea was also studied in a targeted survey by Remke et al. (2009a, 2009b) and Remke (2010). Coastal dunes around the Baltic Sea are rather pristine ecosystems, to date receiving small amounts of atmospheric N. Across the 19 investigated dune sites, atmospheric wet N deposition ranged between 3 to 8 kg N ha^{-1} yr^{-1} (wet N deposition of nearby EMEP certified weather stations). The dry deposition at these cleaner sites was subsequently estimated to be at least 3 kg N ha^{-1} yr^{-1}. The N content of the dominant lichen, *Cladonia portentosa*, was demonstrated to be a suitable bio-indicator of N deposition at these low to medium N deposition levels, with tissue N concentrations increasing above 5 kg N ha^{-1} yr^{-1} wet N deposition. Comparison with EMEP deposition data showed that tissue N concentrations in *Cladonia portentosa* also reflected the deposition history of the last three to six years. Moreover, a shift from lichen-rich short grass vegetation towards species-poor vegetation dominated by *Carex arenaria* also correlated with increasing wet N deposition loads. Plant species richness, however, was not shown to correlate with these low to medium N deposition loads (Remke et al., 2009a). Accelerated soil acidification, as well as increased growth of *Carex* and accumulation of organic matter, was observed only in acidic grasslands with pH_{NaCl} of the parent material between 5.0 and 6.0. At sites with more calcareous parent material (pH_{NaCl} 6-7), these relationships with N deposition were not apparent. A trigger for grass encroachment seems to be high acidification in early successional stages to below pH_{NaCl} 4.0. Metals such as aluminium (Al) were more freely available and may inhibit more sensitive species. From the acidic sites, N mineralisation was higher at those sites with higher N deposition, which may further stimulate *Carex*. *Carex*-dominated dune grasslands are species-poor (Figure 4.8). The number of foliose lichen species, forbs and grasses was lower in regions with wet deposition above 5 kg N ha^{-1} yr^{-1}, compared with 'clean' areas (2-5 kg N ha^{-1} yr^{-1}) at the investigated acidic sites (Remke et al., 2009b). Including the estimated 3 kg N ha^{-1} yr^{-1} in dry deposition, it is likely that 5 kg N ha^{-1} yr^{-1} wet deposition will correspond with approximately 8 kg N ha^{-1} yr^{-1} total deposition.

Figure 4.8. Picture of a *Carex arenaria*-dominated stable dune grassland in the Baltic (Korshage DK).

Source: E. Remke

A further targeted gradient survey in the UK was carried out in 2009 in acidic/decalcified stable dune grasslands (Field et al., 2014; data also re-analysed in Jones et al., 2018). The gradient covered a range from 5.4 to 16.8 kg N ha^{-1} yr^{-1} total N deposition. This showed a decrease in vegetation species richness almost from the lowest N deposition points along the gradient (> 5 kg N ha^{-1} yr^{-1}), with the most rapid change in species richness of the fitted curve occurring at the cleanest sites (Figure 4.9). The number of bryophyte species was also lower above 5-10 kg N ha^{-1} yr^{-1}.

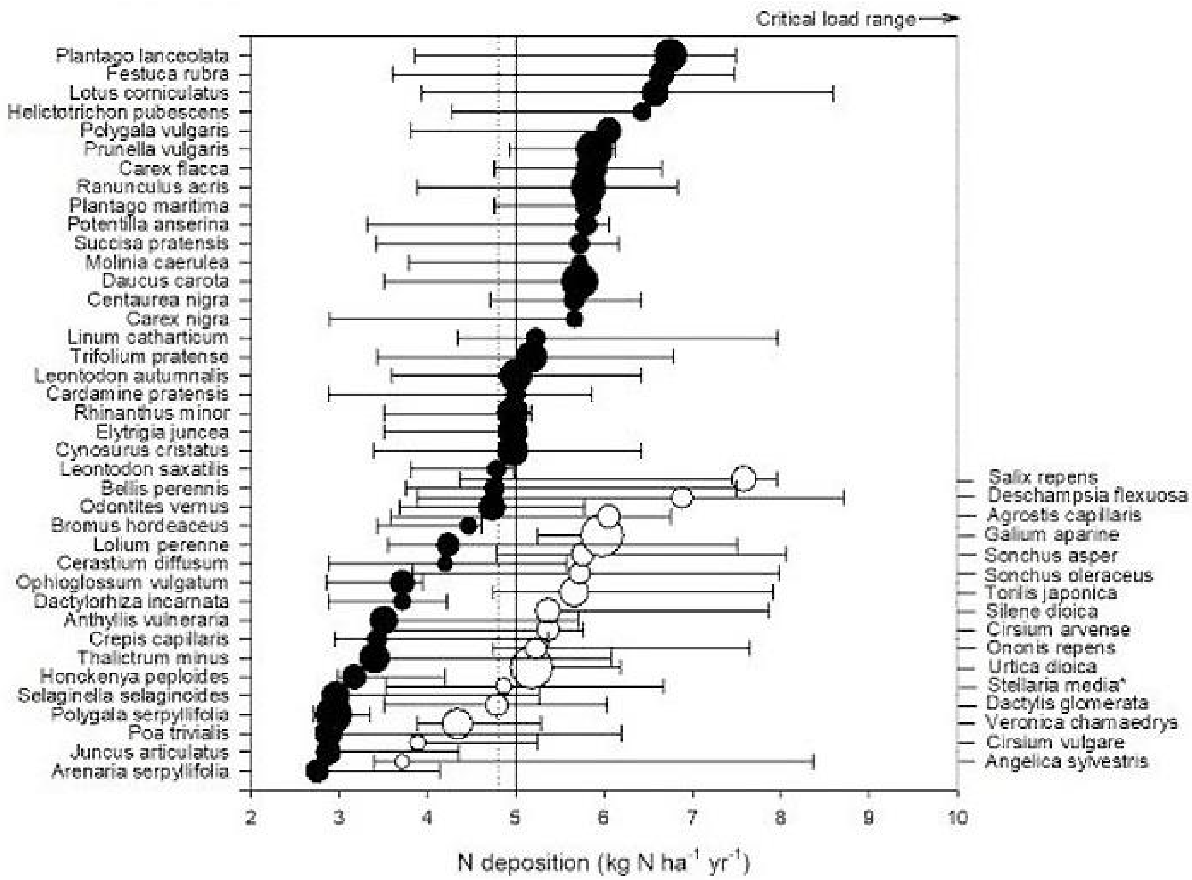
Figure 4.9 Species richness in acidic or decalcified dry dune grassland communities of UK. Line shows fitted relationship, $y = 98.351 - 20.4662 * \ln(x)$; R^2 = 33%, p = 0.003.



Data source: Field et al., 2014; Jones et al., 2018

A series of datasets including gradient surveys designed to detect impacts of N deposition, and untargeted surveys were collated by Payne et al. (2020) for analysis of N impacts. They collected 36 vegetation datasets across multiple habitats, including dunes, to conduct a TITAN (Threshold Indicator Taxa Analysis). In the final analysis, only five datasets were used as most did not pass initial screening stages, including the criterion that studies were only included when N deposition (current annual or 30-year cumulative) explained significant variation in the redundancy analysis with co-variates partialled out. Therefore, this represents a highly robust analysis of N impacts from large datasets with co-variates explicitly accounted for. The analysis used both annual deposition and 30-year cumulative deposition. Annual N deposition was calculated with the Concentration Based Estimated Deposition (CBED) model for the UK, cumulative N deposition for the past 30 years via the Fine Resolution Atmospheric Multi-Pollutant Exchange (FRAME) model. Two dune habitats, fixed dune grasslands and dune slacks (see Chapter 4.2.2 and 4.2.5), were analysed in this study. The analysis of fixed dune grasslands used data from Scottish coastal dunes (Pakeman et al., 2016), covering mostly the very clean end of the N deposition gradient, 2.7-11.8 kg N ha^{-1} yr^{-1} total N deposition, and included mostly calcareous dune habitats, but some acidic ones. In total, 39 species showed significant negative change points (i.e. a defined point above which there was a significant decrease in the abundance of that species) and 16 species showed a positive change point, analysed against annual total N deposition. The overall community sum(-z) change point was determined as 5 kg N ha^{-1} yr^{-1} (Figure 4.10).

Figure 4.10. Negative (black circles) and positive (white circles) change points for dry dune grassland species. Vertical lines show overall community sum(z-) [solid line] and sum(z+) [dotted line] change-points. Circles represent species showing high purity and reliability negative (black circles) and positive (white circles) change points in response to nitrogen deposition and bootstrap 5% and 95% quantiles.



Source: Payne et al., 2020

An entirely different dataset, this time from Ireland, also used TITAN analysis of dune habitats (Aherne et al., 2021). Their analysis was conducted across a mixture of predominantly calcareous dry dune and dune slack habitats, although the species showing significant change points were more commonly found in dry dune habitats (*Trifolium repens, Plantago lanceolata, Cerastium fontanum, Rhytidiadelphus squarrosus, Linum catharticum, Bellis perennis, Achillea millefolium*). Their estimated community change point was 6.2 kg N ha^{-1} yr^{-1}.

Lastly, additional evidence comes from a re-survey of Scottish coastal vegetation over a 35-year interval (1975-2010) (Pakeman et al., 2016). The study looked at mobile dune grasslands, fixed dune grasslands, and dune slacks, as well as other coastal vegetation types. This showed increased fertility of fixed dune grasslands, from mean Ellenberg N score of 4.003 to 4.195 over time, consistent with an increase in N deposition over that period (CBED model for N deposition). In fixed dune habitats, the greatest increases in the Ellenberg N index occurred at the more polluted end of the gradient at 4.1-5.9 kg N ha^{-1} yr^{-1} total deposition. At this higher end, the species richness declined with an average of 3.12 species per quadrat, although former S deposition cannot be excluded as a confounding factor for the change in species richness. Based on changes in overall vegetation composition and Ellenberg N indices, Pakeman et al. (2016) suggested a $CL_{emp}N$ of 4-6 kg N ha^{-1} yr^{-1} for fixed dune grasslands.

Summary for coastal dune grasslands (grey dunes) (N15)

The 2011 $CL_{emp}N$ range for coastal dune grassland (grey dune) (N15) was 8 to 15 kg N ha^{-1} yr^{-1} (quite reliable) (Bobbink and Hettelingh 2011). For this habitat, there is now strong evidence from a wide range of studies including *in situ* N additions, experimental mesocosms under a range of exposure methods, gradient studies and analysis of untargeted surveys. There is new evidence, from multiple sources, of ecological changes in both calcareous and acidic dunes at the lower end of this range. Based on this evidence we propose a reduction of the lower limit of the $CL_{emp}N$ range for coastal dune grasslands to 5 kg N ha^{-1} yr^{-1}. However, phosphorus limitation may lead to fewer botanical responses in calcareous dunes compared with acidic or decalcified dune sites. The higher end of the range is kept at 15 kg N ha^{-1} yr^{-1} as there are calcareous rich sites with P limitation where there are few botanical changes, but other receptors are still affected, such as nitrate leaching. In summary, the revised $CL_{emp}N$ range for coastal dune grasslands is 5 to 15 kg N ha^{-1} yr^{-1}, and the authors increase the evidence status to reliable.

4.2.3 Coastal dune heaths (N18 and N19)

Besides dry dune grasslands, heathland vegetation is also present in the coastal dunes in Europe (Gimingham et al., 1979; Ellenberg, 1988b). These natural coastal dune heaths are mostly dominated by the typical dwarf shrub *Empetrum nigrum*, while *Calluna vulgaris* is less common. Within EUNIS, there are now two categories of coastal heaths: Atlantic and Baltic coastal Empetrum heath (N18) and Atlantic coastal Calluna and Ulex heath (N19), which are both classified as a subcategory of coastal dune and sand habitats. There is one new study to add to the previous assessment for dune heaths, Bähring et al. (2017).

Only two N manipulation experiments have been performed in coastal heaths, one in Denmark within the Danish HEATH experiment (Riis-Nielsen, 1997; Nielsen et al., 2000), located in a coastal heath at Lodbjerg, and a new study at the island of Fehmarn, Germany (Bähring et al., 2017).

The Danish coastal heathland, dominated by *Empetrum nigrum* and *Ammophila arenaria*, is located in an area of approximately 250 to 300-year-old dunes on the coast of Jutland, with a relatively low naturally occurring N deposition (13 kg N ha^{-1} yr^{-1}) (Nielsen et al., 2000). Ammonium nitrate (0, 15, 35 and 70 kg N ha^{-1} yr^{-1}, dissolved in demineralised water) has been applied six times per year and was carried out over a period of two years. As a response to N additions, vascular plant species increased in cover, whereas lichens and bryophytes showed a very slight but non-significant decrease over those two years (Riis-Nielsen, 1997). *Hypnum cupressiforme*, a typical heathland moss, declined linearly with applications of N, whereas the cover of *Empetrum* and *Carex arenaria* increased linearly. Thus, plant productivity in this coastal heath system was obviously controlled by N. However, observed drought effects on *Empetrum* showed no interaction with N inputs (Tybirk et al., 2000). The leaching of both nitrate and ammonium was also quantified in this coastal Danish heath. In the control plots and those receiving 15 kg N ha^{-1} yr^{-1}, virtually no N leached to the subsoil. However, with higher N additions, especially 70 kg N ha^{-1} yr^{-1}, a considerable part of the N leached as nitrate from the B horizon, accompanied by aluminium, leading to soil acidification (Nielsen et al., 2000). Johansson (2000) found no effects of N additions on ericoid mycorrhizal infection of *Calluna vulgaris* in this coastal heath experiment.

At the island of Fehmarn, in the Baltic Sea, an experiment has been conducted within dry heaths on sand (EUNIS type S42; Bähring et al., 2017). Background deposition was 9 kg N ha^{-1} yr^{-1} based on modelled data of Schaap et al. (2015). Six levels of N fertilisation (0, 2.5, 5, 10, 20, and 50 kg N ha^{-1} yr^{-1}) were applied for three years in 12 doses during the growing season (May to October) as dissolved NH_4NO_3 solution. The growth responses of different plant species of different life forms (dwarf shrubs, graminoids, bryophytes, and lichens) as well as shifts in the C:N ratios of plant tissue and humus horizons were quantified. The current year's shoot increment of the dominant dwarf shrub *Calluna vulgaris* was the most sensitive indicator of N fertilisation. Shoot increment was significantly greater after additions of ≥ 5 kg N ha^{-1} yr^{-1} (on top of background, i.e. total load > 14 kg N ha^{-1} yr^{-1}) already in the first year. *Cladonia* spp. tissue C:N ratios decreased with N additions ≥ 5 kg N ha^{-1} yr^{-1} in the second year of study. After three years, an increase in the cover of graminoids and a corresponding decrease of cryptogams at N fertilisation rates of ≥ 10 kg N ha^{-1} yr^{-1} were observed.

Summary for coastal dune heaths (N18 and N19)

The $CL_{emp}N$ of coastal dune heathlands established in 2011 was 10-20 kg N ha^{-1} yr^{-1} (expert judgement). Since then, only one additional relevant study has been performed (Bähring et al., 2017). The responses observed after only a few years at total N loads of ≥ 14 kg N ha^{-1} yr^{-1} in this well-designed study suggest it would be appropriate to lower the upper end of the range of the $CL_{emp}N$ to 15, giving a $CL_{emp}N$ of 10 to 15 N ha^{-1} yr^{-1}, and to increase the evidence status to quite reliable. However, more long-term information from even cleaner environments and additional evidence would be desirable to inform this $CL_{emp}N$.

4.2.4 Moist to wet dune slacks (N1H)

Atlantic and Baltic moist and wet dune slack (EUNIS N1H) of primary or secondary origin are hot spots of plant diversity in the sandy dune regions of Europe. They are characterised by typical graminoids (sedges, rushes and grasses), together with a high diversity of forb species and bryophytes, including many rare basiphilous species. Groundwater level is usually at or above soil level in winter, whereas in the growing season the groundwater level is considerably lower in these dune slacks. A defining feature of these dune slack vegetation communities is that they dry out in most years in summer, otherwise the vegetation tends towards mire or swamp communities. Typical Mean Spring Level (as average depth from soil surface to the groundwater in March, April and May) is less than 85 cm from the ground level, but varies by community (Jones et al., 2021), and can be near the ground surface for the wetter communities. Dune slacks in nature reserves are often maintained though management, such as grazing, hay production and harvest or sod cutting (e.g. Ellenberg, 1988b; Lammerts and Grootjans, 1997; Davies et al., 2004). Because of their isolation in the landscape and their successional position, they mostly receive nutrients via atmospheric inputs, but some sites are also affected by groundwater nutrients (Rhymes et al., 2014). In addition, they are very sensitive to hydrological changes from groundwater extraction.

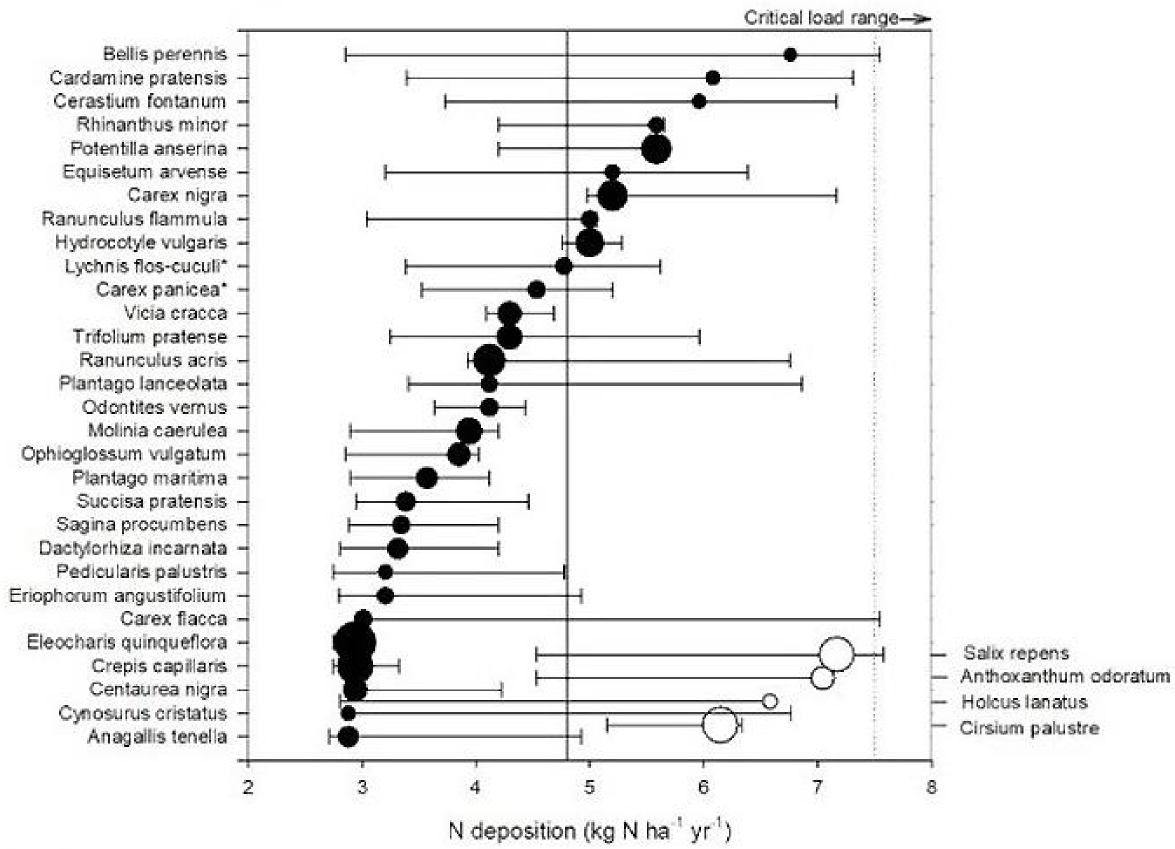
The limitation of dune slack vegetation by nutrients has been the topic of several studies (e.g. Willis, 1963; Olff et al., 1993) and reviewed by Lammerts and Grootjans (1997). Factorial fertilisation experiments have shown that in almost all studied moist to wet, primary or secondary dune slacks in the United Kingdom, the Netherlands and the United States, the above-ground biomass production is limited by N availability. Primary P limitation was found only once in a dune slack where sod cutting had been applied shortly before. Single N additions (above 100 N kg ha^{-1} yr^{-1}) have led to increased dominance of *Carex* and *Juncus* species, and of tall grasses such as *Agrostis stolonifera* and *Calamagrostis epigejos*. In some studies, typical forb species had declined in such situations (for an overview and references, see Lammerts and Grootjans, 1997). Unfortunately, none of the studies have been carried out with low N additions (< 100 N kg ha^{-1} yr^{-1}; > 1 year), and thus they are not adequate for setting a $CL_{emp}N$. The following new studies add to the previous assessment of $CL_{emp}N$ for dune slacks: Rhymes et al. (2014, 2015), Pakeman et al. (2016), and Payne et al. (2020).

A targeted field survey of eleven dune systems in the United Kingdom, with a calculated total N deposition ranging from 6.9 to 29.4 kg N ha^{-1} yr^{-1} (Jones et al., 2004), showed no significant relationship between atmospheric N deposition and either soil or bulk vegetation parameters in dune slacks. This may, in large part, be due to the relatively small sample size and the absence of wet dune slacks in two of the eleven sites. However, the cover of *Carex arenaria* and *Hypochaeris radicata* related positively to total N deposition, suggesting a response in some vegetation species of dune slacks to N enrichment at rather low loads. A second gradient study focusing on rare dune slack species at 12 sites in the United Kingdom, in a total N deposition range of 4 to 20 kg N ha^{-1} yr^{-1}, also showed no significant effects of N on species richness or soil parameters such as total N content, or available N (Jones, 2007).

However, there is experimental evidence that N may cause major shifts in the germination community of dune slacks. In a UK seedbank germination experiment, 15 kg N ha^{-1} yr^{-1} (in the form of NH_4NO_3) was applied to soils from dune slacks with a site background deposition of approximately 11 kg N ha^{-1} yr^{-1}. The emerging seedling community on N treated soil differed strongly from communities on untreated dune slack soil. Germination in response to N was generally greater in species with low Ellenberg N indicator values (Plassmann et al., 2008). This enhanced germination may deplete the seedbank of early successional species which depend on seed longevity to survive until the next sand dune mobility phase or disturbance event which exposes bare sand allowing germination.

There is new evidence since the 2011 review from gradient studies on dune slack data from Scotland, generally covering the lower part of the N deposition range, that suggest there are impacts of N on dune slack vegetation. Payne et al. (2020) analysed non-targeted survey data using TITAN change-point analysis for dune slacks in Scotland, along a gradient of 2.7-11.8 kg N ha^{-1} yr^{-1} and showed that 30 species had negative change points within this deposition range. Four species also showed positive change points (i.e. started to increase) within this range (Figure 4.11). The overall community change point for species declines was estimated at 5 kg N ha^{-1} yr^{-1}. Further analysis of the dune slack data used by Payne et al. (2020) was undertaken, since the species exhibiting change points and the order in which they responded to the N deposition gradient did not always match expectations based on their ecology. This analysis showed that there was an increase in the unweighted Ellenberg N at around 5-6 kg N ha^{-1} yr^{-1}, and a decrease in species richness at around the same point on the N deposition gradient (Figure 4.12). This is consistent with the observed community change point reported in Payne et al. (2020).

Figure 4.11. Negative (black circles) and positive (white circles) change points for dune slack species. Vertical lines show overall community sum(z-) [solid line] and sum(z+) [dotted line] change-points. Circles represent species showing high purity and reliability negative (black circles) and positive (white circles) change points in response to nitrogen deposition and bootstrap 5% and 95% quantiles.

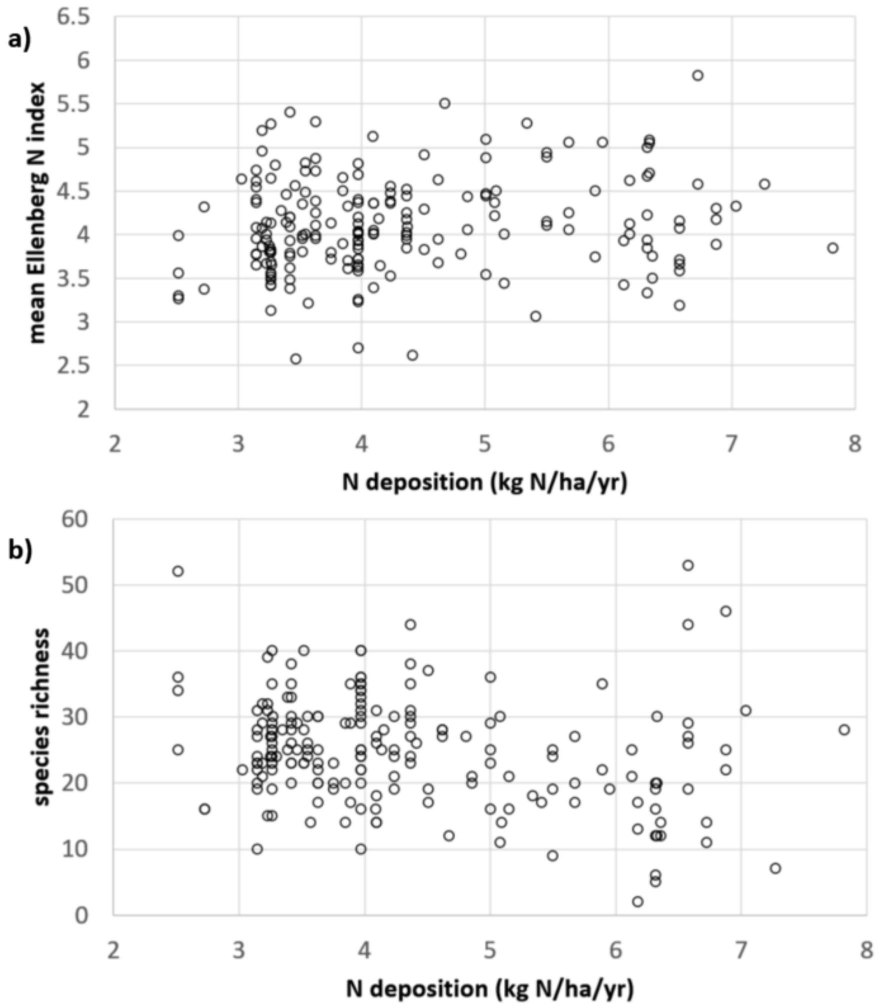


Source: Payne et al., 2020

Data looking at change in dune slack vegetation over time also provides evidence of impacts within this deposition range. The Scottish re-survey of coastal vegetation by Pakeman et al. (2016) over a 35-year interval (1975-2010) also showed a shift towards more nitrophilous vegetation in dune slacks, from a mean Ellenberg N score of 3.997 to 4.147 over time, consistent with the increase in N deposition over that period.

Separate evidence comes from a UK study (Rhymes et al., 2014, 2015) which looked at N impacts on dune slack vegetation and soils through observation of within-site gradients in groundwater contamination. While this represents a different nutrient pathway, this study provides strong additional evidence for the sensitivity of dune slacks to N. The Aberffraw dunes in Wales, UK, contain an N gradient in groundwater coming from adjacent farmland. Analysis of soils and vegetation along this gradient suggests higher available nitrate in soil where dissolved inorganic N (DIN) concentrations in groundwater were greater than 0.084 ± 0.034 mg L^{-1}, and altered vegetation composition where DIN concentrations in groundwater were greater than 0.0224 ± 0.011 mg L^{-1} (Rhymes et al., 2014), by comparison with locations towards the middle of the site where the vegetation was least impacted, and assumed only to receive N from background atmospheric deposition with no additional groundwater inputs of N (Rhymes et al., 2015). The background deposition to the site is approximately 11 kg N ha^{-1} yr^{-1}, therefore, regardless of the source and pathway of N, this study provides evidence for further impacts of N above this level. This also shows the importance of understanding all significant N inputs at any given site.

Figure 4.12. Re-analysis of dune slack data used in Payne et al. (2020), data originally from Lewis et al. (2016); Pakeman et al. (2015, 2016, 2017), showing a) mean unweighted Ellenberg N index and b) species richness, against N deposition.



Source: Payne et al., 2020; Lewis et al., 2016; Pakeman et al. (2015, 2016, 2017)

Summary for moist to wet dune slacks (N1H)

The 2011 $CL_{emp}N$ range was 10-20 kg ha^{-1} yr^{-1} (expert judgement). There is new evidence suggesting that dune slacks are sensitive to N deposition. This is based on surveys, and from monitoring of change over time, particularly from studies focused in areas of relatively low N deposition. Together, these studies suggest that the $CL_{emp}N$ range should be lowered to 5 to 15 kg N ha^{-1} yr^{-1} with the evidence status improved to quite reliable. This is reinforced by evidence of impacts from additional N inputs via groundwater in a site receiving atmospheric deposition of 11 kg N ha^{-1} yr^{-1}. Experimental studies to reinforce this new value would certainly be desirable. Due to new evidence of impacts at low N deposition loads in calcareous systems, we no longer recommend to apply a different part of the $CL_{emp}N$ range to slacks of low base status. However, acid to acidic slacks may still show greater impacts than well-buffered slacks for a given level of N deposition.

4.2.5 Dune-slack pools (freshwater aquatic communities of permanent Atlantic and Baltic (N1H1) or Mediterranean and Black Sea (N1J1) dune-slack water bodies)

These relatively small pools are found in the European coastal dune areas, both in the temperate as well as the Mediterranean zone. Dune pools can dry out totally in some years and the very shallow and small ones also every year, but they have a waterbody for more than ¾ of the year. Wet dune slacks have typically less than half year with an open water body. Dune slack pools are a typical part of young, expanding coastal dune areas. These pools with young sediments are oligotrophic to mesotrophic nutrient poor, but not extremely so. Whereas large, permanent water bodies in such dunes are often rather eutrophic, dune slack pools are characterised by clear water and a diverse submerged macrophyte vegetation (e.g. *Potamogeton* and *Chara* species, and littoral isoetids). Relative to oligotrophic inland surface waters, they show a high succession rate towards large helophytes and shrubs. A distinct influence of local groundwater from the surrounding dunes is present, transporting dissolved iron that ensures phosphorus limitation in the water layer. Contaminated groundwater may also transport nutrients to such pools. Desiccation with aeration of a large part of the sediment may frequently occur and aeration of the sediments also occurs by radial oxygen loss of the root system of isoetids like *Littorella uniflora* (Adema et al., 2005). Oxygenation of the rhizosphere stimulates denitrification of N_2 to the air via nitrate production, reducing the amount of N in the pool. In addition, the fluctuation of the water table improves phosphorus binding to iron, reducing its availability. The interactions between carbonate content, desiccation, N deposition and succession rates are yet not well studied.

Despite their well-defined geographic location, their functioning can differ in several ecologically important aspects. Water bodies can be permanent, or annual desiccation may occur. This may also vary from year to year depending on precipitation and the local hydrology. Dune sands can also vary from highly calcareous to almost without calcium carbonate and slightly acidic. Accumulation of organic matter is a dominant factor in determining the succession rates. Both desiccation as well as calcareous soils stimulate decomposition of organic material, thereby slowing down succession (Sival and Grootjans, 1996). Water eutrophication and N deposition accelerate succession rates and shorten the lifespan of dune slack pools. Eutrophication in these dune slack pools is more likely to be caused by atmospheric inputs or by high densities of waterfowl than by the inflow of enriched surface water, because of the hydrological isolation of these habitats and their location in (large) natural coastal areas but could in principle occur from contaminated groundwater.

Very few experimental data exist on the sensitivity of dune slack pools with respect to $CL_{emp}N$ setting, despite the generally well-known N limitation of dune slack wetlands (Lammerts and Grootjans, 1997; Romo et al., 2016). In Australian dune lakes, algal growth was stimulated by N additions as low as 50 µg L^{-1} ammonium nitrate, but these lakes are ancient and rather acidic (Hadwen, 2002). The impact of atmospheric N deposition was quantified in dune-pool mesocosms (approximately 2-m diameters) during a two-year experiment with different liquid N loads (1, 20, 40 and 120 kg N ha^{-1} yr^{-1} as $(NH_4)_2SO_4$ in an unheated glasshouse (Brouwer et al., 1996). No acidification of the water was found in those two years, but total biomass of water plants and helophytes increased strongly at over 20 kg N ha^{-1} yr^{-1}. Nitrogen additions clearly accelerated the rate of succession in these dune slack mesocosms, leading to more helophytes and less open water. This phenomenon has also been observed in many dune slack pools in the Netherlands receiving relatively high atmospheric N loads (15-20 kg N ha^{-1} yr^{-1}). A recent problem in temperate dune pools is invasion by an exotic plant species, *Crassula helmsii* (Dean, 2015). Experiments have shown that the colonisation of phosphorus poor soils is strongly enhanced after three months of adding N at a rate of 15 kg N ha^{-1} y^{-1}(Brouwer et al., 2017).

Summary for dune-slack pools (N1H1, N1J1)

The 2011 $CL_{emp}N$ range was 10-20 kg ha^{-1} yr^{-1} (expert judgement) rated as 'expert judgement'. There is relatively little new evidence for this habitat and it does not change the previous assessment. Therefore, we retain the existing $CL_{emp}N$ range.

In this case we also recommend that long-term field experiments are held, with realistic doses of N added to these waters, especially in regions with low naturally occurring N deposition. Mediterranean dune pools differ from pools in the Atlantic/Baltic zone by higher temperatures, longer desiccation periods, on average more alkaline conditions and less accumulation of organic matter (Romo et al., 2016). This highly dynamic situation stimulates N losses through cycles of nitrification and denitrification and prevents accumulation of nitrogen in organic matter. Therefore, Mediterranean pools might be less sensitive to increased N deposition but could be more affected by N pollutants from surrounding agricultural areas.

4.2.6 Coastal forests

Coastal forests on sand occur as natural Atlantic dunes woodland or other natural assemblages, in addition to plantation woodland. For N impacts on these woodlands, please refer to the forest chapter (EUNIS class T).

4.3 Overall summary of $CL_{emp}N$ for coastal dunes and sandy shores (N1)

The $CL_{emp}N$ for coastal habitats (N) are summarised in Table 4.1.

Table 4.1. $CL_{emp}N$ and effects of exceedances on coastal habitats (N). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Shifting coastal dunes	N13, N14	10-20	(#)	10-20	**#**	Biomass increase; increased N leaching; reduced root biomass
Coastal dune grasslands (grey dunes)	N15	8-15	#	**5**-15	**##**	Increased biomass and cover of graminoids and mesophilic forbs; decrease in oligotrophic species including lichens; increased tissue N; increased N leaching; soil acidification
Coastal dune heaths	N18, N19	10-20	(#)	10-**15**	**#**	Increased plant production; increased N leaching; accelerated succession; typical lichen C:N decrease; increased yearly increment *Calluna*
Moist and wet dune slacks	N1H	10-20	(#)	**5-15**	**#**	Increased cover of graminoids and mesophilic forbs; decrease in oligotrophic species, increased Ellenberg N

Ecosystem type	EUNIS code	2011 kg N $ha^{-1} yr^{-1}$	2011 reliability	2022 kg N $ha^{-1} yr^{-1}$	2022 reliability	Indication of exceedance
Dune-slack pools (freshwater aquatic communities of permanent Atlantic and Baltic or Mediterranean and Black Sea dune-slack water bodies)	N1H1, N1J1	10-20	(#)	10-20	(#)	Increased biomass and increased rate of succession

4.4 References

Adema, E.B., Van de Koppel, H., Meijer, A.J. and Grootjans, A.P. (2005). Enhanced nitrogen loss may explain alternative stable states in dune slack succession. *Oikos* **109**, 374-386.

Aherne, J., Wilkins, K. and Cathcart, H. (2021). *Nitrogen-sulphur critical loads: assessment of the impacts of air pollution on habitats*. EPA Research Report (2016-CCRP-MS.43).

Bähring, A., Fichtner, A., Ibe, K., Schütze, G., Temperton, V. M., von Oheimb, G. and Härdtle, W. (2017). Ecosystem functions as indicators for heathland responses to nitrogen fertilisation. *Ecological Indicators* **72**, 185-193.

Bird, E. J. and Choi, Y. D. (2017). Response of native plants to elevated soil nitrogen in the sand dunes of Lake Michigan, USA. *Biological Conservation* **212**, 398-405

Bobbink, R. and Hettelingh, J.P. (2011). *Review and revision of empirical critical loads and dose-response relationships: Proceedings of an expert workshop*, Noordwijkerhout, 23-25 June 2010. Rijksinstituut voor Volksgezondheid en Milieu RIVM.

Boorman, L.A. and Fuller, R.M. (1982). Effects of added nutrients on dune swards grazed by rabbits. *Journal of Ecology* **70**, 345-355.

Brouwer, E., Bobbink, R., Roelofs, J.G.M. and Verheggen, G.M. (1996). *Effectgerichte maatregelen tegen verzuring en eutrofiëring van oppervlaktewateren*. University of Nijmegen, Nijmegen, 25-50 (in Dutch).

Brouwer, E., Denys, L., Lucassen, E.C.H.E.T., Buks, M. and Onkelinx, T. (2017). Competitive strength of Australian swamp stonecrop (*Crassula helmsii*) invading moorland pools. *Aquatic invasions* **12**, 321-331.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**(4), 648-675.

Davies, C.E., Moss, D. and Hill, M.O. (2004). *EUNIS habitat classification revised 2004*. European Environment Agency, European topic centre on nature protection and biodiversity.

Dean, C.E. (2015). *The Ecology, Impacts and control of Crassula helmsii.* Thesis Bournemouth University & National Trust & Royal Society for the Protection of Birds.

De Vries, W., Klijn, J.A. and Kros, J. (1994). Simulation of the long-term impact of atmospheric deposition on dune ecosystems in the Netherlands. *Journal of Applied Ecology* **31**, 59-73.

Ellenberg, H. jr. (1988a). Floristic changes due to nitrogen deposition in central Europe. In: Nilsson, J. and Grennfelt, P. (eds.). *Critical loads for sulphur and nitrogen, Report from a workshop held at Skokloster, Sweden 19-24 March, 1988*. Miljörapport/Nord 15 Nordic Council of Ministers, Kopenhagen.

Ellenberg, H. (1988b). *Vegetation Ecology of Central Europe*. Cambridge University Press, Cambridge.

Field C., Dise N., Payne, R., Britton, A., Emmett, B., Helliwell R., Hughes S., Jones L., Leake J., Leith I., Phoenix G., Power S., Sheppard L., Southon G., Stevens C. and Caporn S.J.M. (2014). Nitrogen drives plant community change across semi-natural habitats. *Ecosystems* **17**, 864-877.

Ford, H., Roberts, A. and Jones, L. (2016). Nitrogen and phosphorus co-limitation and grazing moderate nitrogen impacts on plant growth and nutrient cycling in sand dune grassland. *Science of the Total Environment* **542**, 203-209.

Gimingham, C.H., Chapman, S.B. and Webb, N.R. (1979). European heathlands. In: Specht, R.L. (ed.). *Ecosystems of the world, 9A*. Elsevier, Amsterdam, 365-386.

Greipsson, S. and Davy, A.J. (1997). Responses of *Leymus arenarius* to nutrients: improvement of seed production and seedling establishment for land reclamation. *Journal of Applied Ecology* **34**, 1165-1176.

Hall, J., Emmett, B., Garbutt, A., Jones, L., Rowe, E., Sheppard, L., Vanguelova, E., Pitman, R., Britton, A., Hester, A. and Ashmore, M. (2011). *UK Status Report July 2011: Update to empirical critical loads of nitrogen*. Report to Defra under contract AQ801 Critical Loads and Dynamic Modelling.

Hadwen, W.L. (2002). *Effects of Nutrient Additions on Dune Lakes on Fraser Island, Australia*. Thesis Centre for Catchment and In-Stream Research, Griffith University.

Howe, M.A., Knight, G.T. and Clee, C. (2010). The importance of coastal sand dunes for terrestrial invertebrates in Wales and the UK, with particular reference to aculeate Hymenoptera (bees, wasps & ants). *Journal of Coastal Conservation* **14**, 91-102.

Johansson, M. (2000). The influence of ammonium nitrate on the root growth and ericoid mycorrhizal colonization of *Calluna vulgaris* (L.) Hull from a Danish heathland. *Oecologia* **123**, 418-424.

Jones, M.L.M., Hayes, F., Brittain, S.A., Haria, S., Williams, P.D., Ashenden, T.W., Norris, D.A. and Reynolds, B. (2002). *Changing nutrient budget of sand dunes: consequences for the nature conservation interest and dune management*. CCW contract No. FC 73-01-347. Field survey. Centre for Ecology and Hydrology, Bangor, 1-31.

Jones, M.L.M., Wallace, H.L., Norris, D., Brittain, S.A., Haria, S., Jones, R.E., Rhind, P.M., Reynolds, B.R. and Emmett, B.A. (2004). Changes in vegetation and soil characteristics in coastal sand dunes along a gradient of atmospheric nitrogen deposition *Plant Biology* **6**, 598-605.

Jones, M.L.M. (2007). Model based risk assessment of the vulnerability of rare coastal species to N deposition. Task 16, pp. 191-198. In: Emmett, B., Ashmore, M., Britton, A., Broadmeadow, M., Bullock, J., Cape, N., Caporn, S. J. M., Carroll, J. A., Cooper, J. R., Cresser, M. S., Crossley, A., d'Hooghe, P., De Lange, I., Edmondson, J., Evans, C. D., Field, C., Fowler, D., Grant, H., Green, E., Griffiths, B., Haworth, B., Helliwell, R., Hicks, K., Hinton, C., Holding, H., Hughes, S., James, M., Jones, A., Jones, M., Jones, M. L. M., Leake, J., Leith, I., Maskell, L., McNamara, N., Moy, I., Oakley, S., Ostle, N., Pilkington, M., Power, S., Prendergast, M., Ray, N., Reynolds, B., Rowe, E., Roy, D., Scott, A., Sheppard, L., Smart, S., Sowerby, A., Sutton, M., Terry, A., Tipping, E., Van den Berg, L., Van Dijk, N., Van Zetten, E., Vanguelova, E., Williams, B., Williams, D. and Williams, W. *Terrestrial Umbrella: effects of eutrophication and acidification on terrestrial ecosystems. Final report.* NERC/Centre for Ecology & Hydrology, 288pp. (CEH Project Number: C02613, Defra Contract No. CPEA 18).

Jones, M.L.M, Sowerby, A., Williams, D.L. and Jones, R.E. (2008). Factors controlling soil development in sand dunes: evidence from a coastal dune soil chronosequence. *Plant and Soil* **307**, 219-234.

Jones L., Nizam M.S., Reynolds B., Bareham S. and Oxley E.R.B. (2013). Upwind impacts of ammonia from an intensive poultry unit. *Environmental Pollution* **180**, 221-228.

Jones, L., Milne, A., Hall, J., Mills, G., Provins, A. and Christie, M., (2018). Valuing improvements in biodiversity due to controls on atmospheric nitrogen pollution. *Ecological Economics* **152**, 358-366.

Jones, L., Rooney, P., Rhymes. J. and Dynamic Dunescapes partners (2021). *The Sand Dune Managers Handbook*. Version 1, June 2021. Produced for the Dynamic Dunescapes (DuneLIFE) project: LIFE17 NAT/UK/000570; HG-16-0864361

Kooijman, A.M. and De Haan, M.W.A. (1995). Grazing as a measure against grass-encroachment in Dutch dry dune grasslands: effects on vegetation and soil. *Journal of Coastal Conservation* **1**, 127-134.

Kooijman, A.M., Dopheide, J.C.R., Sevink, J., Takken, I. and Verstraten, J.M. (1998). Nutrient limitations and their implications on the effects of atmospheric deposition in coastal dunes; lime-poor and lime-rich sites in the Netherlands. *Journal of Ecology* **86**, 511-526.

Kooijmann, A.M., and Besse, M. (2002). The higher availability of N and P in lime-poor than in lime-rich coastal dunes in the Netherlands. *Journal of Ecology* **90**, 394-403.

Lammerts, E.J. and Grootjans, A.P. (1997). Nutrient deficiency in dune slack pioneer vegetation: a review. *Journal of Coastal Conservation* **3**, 87-94.

Lewis, R.J., Marrs, R.H., Pakeman, R.J., Milligan, G. and Lennon, J.J. (2016). Climate drives temporal replacement and nested-resultant richness patterns of Scottish coastal vegetation. *Ecography* **39**, 754-762.

Marchetto, A., Arisci, S., Tartari, G.A., Balestrini, R. and Tait, D. (2014). Current state and temporal evolution of the chemical composition of atmospheric depositions in forest areas of the CONECOFOR network. *Forest@ - Journal of Silviculture and Forest Ecology* **11**, 72-85.

Menicagli, V., Balestri, E., Vallerini, F., Castelli, A. and Lardicci, C. (2020). Combined effect of plastic litter and increased atmospheric nitrogen deposition on vegetative propagules of dune plants: A further threat to coastal ecosystems. *Environmental Pollution* **266**, 115281.

Mohd-Said M.N. (1999). *Effects of anthropogenic nitrogen inputs on dune grassland*. PhD thesis, University of Wales, Bangor.

Nielsen, K.E., Hansen, B., Ladekarl, U.L. and Nornberg, P. (2000). Effects of N-deposition on ion trapping by B-horizons of Danish heathlands. *Plant and Soil* **223**, 265-276.

Olff, H., Huisman, J., and Van Tooren, B.F. (1993). Species dynamics and nutrient accumulation during early primary succession in coastal sand dunes. *Journal of Ecology* **81**, 693-706.

Pakeman, R.J., Alexander, J., Beaton, J., Brooker, R., Cummins, R., Eastwood, A., Fielding, D., Fisher, J., Gore, S., Hewison, R., Hooper, R., Lennon, J., Mitchell, R., Moore, E., Nolan, A., Orford, K., Pemberton, C., Riach, D., Sim, D., Stockan, J., Trinder, C. and Lewis, R. (2015). Species composition of coastal dune vegetation in Scotland has proved resistant to climate change over a third of a century. *Global Change Biology* **21**, 3738-3747.

Pakeman, R.J., Alexander, J., Brooker, R., Cummins, R., Fielding, D., Gore, S., Hewison, R., Mitchell, R., Moore, E., Orford, K., Pemberton, C., Trinder, C. and Lewis, R. (2016). Long-term impacts of nitrogen deposition on coastal plant communities. *Environmental Pollution* **212**, 337-347.

Pakeman, R.J., Hewison, R.L. and Lewis, R.J. (2017). Drivers of species richness and compositional change in Scottish coastal vegetation. *Applied Vegetation Science* **20**, 183-193.

Payne, R.J., Campbell, C., Stevens, C.J., Pakeman, R.J., Ross, L.C., Britton, A.J., Mitchell, R.J., Jones, L., Field, C., Caporn, S.J., Carroll, J., Edmondson, J.L., Carnell, E.J., Tomlinson, S., Dore, A., Dragosits, U. and Dise, N. (2020) Disparities between plant community responses to nitrogen deposition and critical loads in UK semi-natural habitats. *Atmospheric Environment* **239**, 117478.

Phoenix, G.K., Hicks, W.K., Cinderby, S., Kuylenstierna, J.C., Stock, W.D., Dentener, F.J., Giller, K.E., Austin, A.T., Lefroy, R.D., Gimeno, B.S. and Ashmore, M.R. (2006). Atmospheric nitrogen deposition in world biodiversity hotspots: the need for a greater global perspective in assessing N deposition impacts. *Global Change Biology* **12**, 470-476.

Plassmann, K., Brown, N., Jones, M.L.M. and Edwards-Jones, G. (2008). Can atmospheric input of nitrogen affect seed bank dynamics in habitats of conservation interest? The case of dune slacks. *Applied Vegetation Science* **11**, 413-420.

Plassmann, K., Edward-Jones, G. and Jones, M.L.M. (2009). The effects of low levels of nitrogen deposition and grazing on dune grassland. *Science of the total environment* **407**, 1391-1404.

Provoost, S., Jones, M.L.M. and Edmondson, S.E. (2011). Changes in landscape and vegetation of coastal dunes in northwest Europe: a review. *Journal of Coastal Conservation* **15**, 207-226.

Remke, E., Brouwer, E., Kooijman, A., Blindow, I., Esselink, H. and Roelofs, J.G.M. (2009a). Even low to medium nitrogen deposition impacts vegetation of dry, coastal dunes around the Baltic Sea. *Environmental Pollution* **157**, 792-800.

Remke, E., Brouwer, E., Kooijman, A., Blindow, I. and Roelofs, J.G.M. (2009b). Low atmospheric nitrogen loads lead to grass encroachment in Coastal Dunes, but only on acid soils. *Ecosystems* **12**, 1173-1188.

Remke, E. (2010). *Impact of atmospheric nitrogen deposition on lichen-rich, coastal dune grasslands.* PhD thesis, Radboud University Nijmegen.

Romo, S., Soria, J., Olmo, C., Flor, J., Cavlo, S., Ortells, R. and Armengol, X. (2016). Nutrients and carbon in some Mediterranean dune ponds. *Hydrobiologia* **782**, 97-109.

Rhymes J., Wallace H., Fenner N. and Jones L. (2014). Evidence for sensitivity of dune wetlands to groundwater nutrients. *Science of the Total Environment* **490**, 106-113.

Rhymes J., Jones L., Lapworth, D.J., White, D., Fenner, N., McDonald, J.E. and Perkins, T.L. (2015). Using chemical, microbial and fluorescence techniques to understand contaminant sources and pathways to wetlands in a conservation site. *Science of the Total Environment* **511**, 703-711.

Riis-Nielsen, T. (1997). *Effects of nitrogen on the stability and dynamics of Danish heathland vegetation*. PhD thesis, University of Copenhagen.

Rowe, E.C., Jones, M.L.M., Henrys, P.A., Smart, S.M., Tipping, E., Mills, R.T. and Evans, C.D. (2011). *Predicting effects of N pollutant load on plant species based on a dynamic soil eutrophication indicator*. Final report on Nitrogen Effects on Dune Species (NEDS) project.

Schaap, M., Kruit, R.W., Hendriks, C., Kranenburg, R., Segers, A., Builtjes, P., Banzhaf, S. and Scheuschner, T., (2015). *Atmospheric deposition to German natural and semi-natural ecosystems during 2009*. Intermediate Report UBA FE-Nr.371263, 240-241.

Sival, F.P. and Grootjans, A.P. (1996). Dynamics of seasonal bicarbonate supply in a dune slack: effects on organic matter, nitrogen pool and vegetation succession. *Vegetatio* **126**, 39-50

Ten Harkel, M.J. and Van der Meulen, F. (1995). Impact of grazing and atmospheric nitrogen deposition on the vegetation of dry coastal dune grasslands. *Journal of Vegetation Science* **7**, 445-452.

Ten Harkel, M.J., Van Boxel, J.H. and Verstraten, J.M. (1998). Water and solute fluxes in dry coastal dune grasslands. The effects of grazing and increased nitrogen deposition. *Plant and Soil* **202**, 1-13.

Tomassen, H., Bobbink, R., Peters, R., Van der Ven, P., and Roelofs, J. (1999). *Kritische stikstofdepositie in heischrale graslanden, droge duingraslanden en hoogvenen: op weg naar meer zekerheid*. Eindrapport in het kader van het Stikstof Onderzoek Programma (STOP), 1997-1999. Nijmegen & Utrecht, Katholieke Universiteit Nijmegen en Universiteit Utrecht, 1-46 (in Dutch).

Tybirk, K., Nilsson, M.C., Michelson, A., Kristensen, H.L., Shevtsova, A., Strandberg, M.T., Johansson, M., Nielsen, K.E., Riis-Nielsen, T., Strandberg, B. and Johnsen, I. (2000). Nordic *Empetrum* dominated ecosystems: Function and susceptibility to environmental changes. *Ambio* **29**, 90-97.

Van den Berg, L., Bobbink, R. and Roelofs, J.G.M. (2005). Effects of nitrogen enrichment in coastal dune grassland: a mesocosm study. *Environmental Pollution* **138**, 77-85.

Van Hinsberg, A. and Van der Hoek, D.C.J. (2003). Oproep: meer onderzoek naar oorzaken van verstruiking. *De Levende Natuur* **104**, 58-59 (in Dutch).

Wellburn, A.R. (1988). *Air pollution and acid rain - The biological impact*. Longman Scientific and Technical, Harlow.

Willis, A.J. (1963). Braunton Burrows: the effects on the vegetation of the addition of mineral nutrients to the dune soils. *Journal of Ecology* **51**, 353-374.

5 Effects of nitrogen deposition on inland surface water habitats (EUNIS class C)

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Summary

In this chapter empirical N critical loads ($CL_{emp}N$) for standing inland surface water habitats (C1) have been reviewed and revised. The assessment is given for the permanent oligotrophic lakes, ponds and pools (C1.1) and for the permanent dystrophic lakes, ponds and pools (C1.4). New, compared to the last report (Bobbink and Hettelingh, 2011), is the differentiation of the permanent oligotrophic class (C1.1) into the following three subcategories: alpine and sub-Arctic clear-water lakes (C1.1), boreal clear-water lakes (C1.1) and Atlantic soft-water bodies (C1.1/C1.2). New $CL_{emp}N$ ranges are proposed for the first two subcategories, respectively 2-4 and 3-6 N ha^{-1} yr^{-1}. For the permanent dystrophic class (C1.4) the lower end of the $CL_{emp}N$ range has been increased from three to five (5 to10 kg N ha^{-1} yr^{-1}). New in this chapter, compared to the last update by Bobbink and Hettelingh (2011), is the exclusion of the EUNIS class dune slack-pools, which has been moved to Chapter 4 in this report (EUNIS class N). It belongs to the freshwater aquatic communities of permanent Atlantic and Baltic (N1H1) or Mediterranean and Black Sea (N1J1) dune-slack water bodies (Chytrý et al., 2020).

5.1 Introduction

In this chapter, the effects of atmospheric N deposition on freshwater ecosystems (inland surface water habitats; class C of the European Nature Information System (EUNIS)) are evaluated. Inland surface water habitats are non-coastal, open, fresh or brackish water bodies (e.g. lakes and pools, rivers, streams, ditches and springs), including their littoral zones. This class includes constructed inland freshwater, brackish or saline water bodies (e.g. canals and ponds) which support a semi-natural community of both plants and animals, and seasonal water bodies which may dry out for part of the year (temporary or intermittent rivers and lakes and their littoral zones). Freshwater littoral zones include those parts of banks or shores that are sufficiently frequently inundated to prevent the formation of closed terrestrial vegetation. Permanent snow and ice are excluded from this EUNIS class.

The main subcategories of EUNIS class C are: surface standing waters (C1), surface running waters (C2), and the littoral zone of inland surface water bodies (C3) (Davies et al., 2004). This chapter summarises field and experimental evidence to establish critical loads for atmospheric N deposition with respect to eutrophication or effects of adverse ammonium. This chapter only discusses surface standing waters (C1) because of limited data availability for class C2 and C3. As usual in earlier empirical critical load reports, rivers (C2) were not assessed because most rivers with potentially available data received high loads of nitrogen from agricultural sources. Only surface waters with no significant agricultural or other human impacts besides atmospheric deposition are considered in this chapter. Atmospheric nitrogen inputs also has acidifying effects on surface waters (Stoddard et al., 1994; Marchetto et al., 1994; Wright et al., 2001). Critical loads for acidification in relation to atmospheric N deposition have been established using mass- and charge balance models (Forsius et al., 2021).

Surface standing waters vary according to nutrient status, depth, sediment type, alkalinity and colour (Moe et al., 2008; Arts, 2002) and these characteristics are also used in the classification of ecological status of surface waters for the EU Water Framework Directive (European Communities, 2003). These characteristics determine their suitability as habitat for aquatic organisms, most importantly fish, phytoplankton (free-floating algae), water plants (macrophytes), organisms growing on solid substrates (epilithon), zooplankton and macro-invertebrates. Nitrogen leaching from semi-natural catchments, which remains challenging to predict (Dise et al., 2009), is related to both N deposition and catchment characteristics, including habitat management and land use within the catchment (Dise and Wright, 1995; Aber et al., 1998; Oulehle et al., 2013). Lake nitrate is often found to be positively related to the catchment ratio (Helliwell et al., 2001; Helliwell et al., 2007).

Nutrient status, in particular phosphorus (P) concentrations, is likely to be an important control of aquatic biological responses to changes in N inputs (Bergström, 2010). Terrestrial and aquatic food webs have similarities and differences related to nutrient limitations (Elser et al., 2000). There are structural contrasts between terrestrial and aquatic food webs that regulate trophic pathways, apparently related to nutritional quality of autotrophs (Shurin et al., 2006). The effects of N deposition on biology in freshwater habitats will depend on catchment N retention capacity, and on the sensitivity of habitats and organisms to changed N availability. The assessment is given for three subcategories of C1, i.e. Atlantic soft-waters, clear-water boreal and alpine lakes, and dystrophic boreal waters. No changes were made in empirical critical loads for Atlantic soft-waters whereas new ranges are now supplied for clear-waters and dystrophic surface waters.

5.2 Surface standing waters (C1)

The main division of permanent standing waters containing fresh water (i.e. non-saline) is based on the nutrient status of the waters, from oligotrophic (C1.1) to mesotrophic (C1.2) and eutrophic lakes, ponds and pools (C1.3). In addition, dystrophic water bodies (C1.4) which are rich in humic substances and often with a brown colour, are also included in C1. Unfortunately, the important gradient in alkalinity (from very soft to hard water), which is at least partly independent of the nutrient status of the water (e.g. Moss, 1988; Brouwer et al., 2002), is not separately treated in the EUNIS habitat hierarchy, making it difficult to classify these systems under EUNIS. As in the previous update (Bobbink and Hettelingh, 2011), the impacts of N deposition are first discussed for the elements of category C1.1 Atlantic soft-water lakes. The effects of N deposition in pristine boreal and alpine oligotrophic water bodies across Europe and North America have received considerable attention in the last 15 years, leading to new evidence, experimental and from gradient studies, to set $CL_{emp}N$ for this important group of category C1.1.

5.2.1 Atlantic soft-water lakes (part of permanent oligotrophic lakes, pools and ponds (C1.1) and some elements of permanent mesotrophic water bodies (C1.2))

In the lowlands of western Europe, many shallow soft waters are found on sandy sediments that are poor in, or almost devoid of, calcium carbonate. These waters are poorly buffered and their concentrations of calcium are very low. Also, they are shallow, fully mixed water bodies with fluctuating water levels that are mainly fed by rain water or water from acidic catchment soils. Thus, they are oligotrophic. In the EUNIS classification scheme, these waters are classified as either permanent oligotrophic (C1.1) or permanent mesotrophic (C1.2) lakes, pools and ponds. These ecosystems are characterised by plant communities from the phytosociological alliance Littorelletea (Schoof-Van Pelt, 1973; Wittig, 1982; Arts, 1990; 2002). Stands of these communities are characterised by the presence of rare and endangered isoetids (with the growth form of *Isoetes*), such as *Littorella uniflora*, *Lobelia dortmanna*, *Isoetes lacustris*, *Isoetes echinospora*, *Echinodorus* species, *Luronium natans* and other soft-water macrophytes. Nowadays, these soft-waters have become very rare in western Europe (Arts, 2002) and are almost all located within nature reserves. This decline is illustrated by the fact that *Littorella uniflora*, which was recorded at more than 230 sites in the Netherlands in the early 1950s, was found at about only 40 locations by the end of the 1980s. In addition, these soft waters have also seen a marked decline in their amphibian species (Leuven et al., 1986). It has been suggested that systems in western Europe (e.g. The Netherlands, northern Germany, Belgium) have been affected more than in other European countries (e.g. Poland) because they are smaller and shallower than elsewhere (Arts, 2002).

The effects of atmospheric N pollutants on these hydrologically isolated soft-water systems – with (hardly) any catchment – have been studied intensively in the Netherlands, in both field surveys and experimental studies. Field observations in approximately 70 soft-water systems that supported well-developed isoetid vegetation in the 1950s showed that the waters in which characteristic macrophytes were still abundant in the early 1980s were those that were poorly buffered (alkalinity 50-500 meq l^{-1}), circumneutral (pH 5-6) and very low in N (Roelofs, 1983; Arts et al., 1990). The 53 soft-water sites from which these plant species had disappeared fell into two groups. In 12, eutrophication resulting from the inflow of enriched water seemed to be the main cause of the decline. In this group of non-acidified waters, plants such as *Lemna minor* had become dominant and high concentrations of phosphate and ammonium were measured in the sediment of these waters. In some of the larger water bodies in this group, no macrophytes were found due to dense plankton blooms. In the second group of lakes and pools (41 out of 53), the isoetid species were found to have been replaced by dense stands of *Juncus bulbosus* or aquatic mosses, such as *Sphagnum cuspidatum* and *Drepanocladus fluitans*. This indicates, clearly, that acidification has occurred in these soft-waters over recent decades, probably as a result of increases in atmospheric N deposition. Also, this field study found that N levels in the water were higher in ecosystems from which the natural vegetation had disappeared, compared to those in which isoetid stands were still present (Roelofs, 1983). This strongly suggests a detrimental effect of atmospheric N deposition on these lakes.

A number of ecophysiological studies have revealed that 1) low inorganic carbon availability due to intermediate levels of alkalinity, and 2) low N concentrations in the water are important to the success of endangered isoetid macrophytes communities. Furthermore, most of the typical soft-water plants had a relatively low potential growth rate. Increased acidity and higher concentrations of ammonium in the water clearly stimulated the development of *Juncus bulbosus* and submerged mosses, such as *Sphagnum* and *Drepanoclad*us species (Roelofs et al., 1984; Den Hartog, 1986) (Figure 5.1). Laboratory experiments have also shown that the form of N involved (ammonium or nitrate) differentially influences the growth of aquatic plant species. For example, almost all of the characteristic soft-water isoetids developed more effectively when nitrate was added compared to ammonium, whereas the growth of *Juncus bulbosus* and aquatic mosses (*Sphagnum* and *Drepanoclad*us) were stimulated by higher levels of ammonium (Schuurkes et al., 1986). The importance of ammonium for the growth of these aquatic mosses is also reported by Glime (1992).

Figure 5.1. Mass growth of *Juncus bulbosus* in a soft-water in an area with high deposition of reduced N.

Source: E. Lucassen

The effects of atmospheric deposition on macrophyte communities were studied over a two-year period in soft-water mesocosms contained in an unheated greenhouse; the mesocosms were treated with different types of artificial rain (Schuurkes et al., 1987). Acidification due to sulphuric acid, without N inputs, did not result in increased growth of *Juncus bulbosus,* and a diverse isoetid vegetation remained. However, when N concentrations of 19 kg N ha^{-1} yr^{-1} or higher were applied in the form of ammonium sulphate, changes in floristic composition were observed compared to the control treatments (< 2 kg N ha^{-1} yr^{-1}). These changes were similar to those seen under field conditions, i.e. a dramatic increase in dominance of *Juncus bulbosus*, submerged aquatic mosses and *Agrostis canina* (Schuurkes et al., 1987). This strongly suggests that the observed changes in the field may have occurred due to ammonium sulphate deposition, which led to both eutrophication and acidification. The increased levels of ammonium in the system also stimulated the growth of plants such as *Juncus bulbosus*, and any surplus ammonium was nitrified in these soft-water systems (pH > 4.0). During this nitrification process, H^+ ions are produced that increase the acidity of the system. The results from this study demonstrate that large changes occurred within two years of treatment with ≥ 19 kg N ha^{-1} yr^{-1}. In addition, the strongest decline in macrophyte species composition in Dutch soft-water communities was found to occur in areas with atmospheric N loads of approximately 10 to 13 kg N ha^{-1} yr^{-1} (Arts, 1990). In addition, Brouwer et al. (1997) showed that, after ten years of treatment with clean rainwater, there was only a partial recovery within the soft-water mesocosms that had been treated with ammonium sulphate, and that *Juncus* and *Molinia* were still the dominant species, suggesting a strong legacy effects of the N deposition impacts.

The previously established $CL_{emp}N$ for shallow soft-water bodies (most in C1.1**,** but some elements in C1.2) was based on experimental evidence and was set at 3 to 10 kg N ha^{-1} yr^{-1} and was considered as reliable (Bobbink et al., 1996, 2003; Bobbink and Hettelingh, 2011). As no new evidence from experimental studies or targeted gradient studies has been published since then on the impacts of N deposition in (shallow) soft-water lakes, the $CL_{emp}N$ range for these systems remains unchanged. This has been incorporated into the more general $CL_{emp}N$ range for oligotrophic lakes, ponds and pools (C1.1) of 2 to 10 kg N ha^{-1} yr^{-1} (reliable), with a recommendation that the upper part of the range (5 to10 kg N ha^{-1} yr^{-1}) is applied in practice (see also Chapter 5.2.2).

5.2.2 Oligotrophic boreal and alpine lakes (part of C1.1 and C1.4)

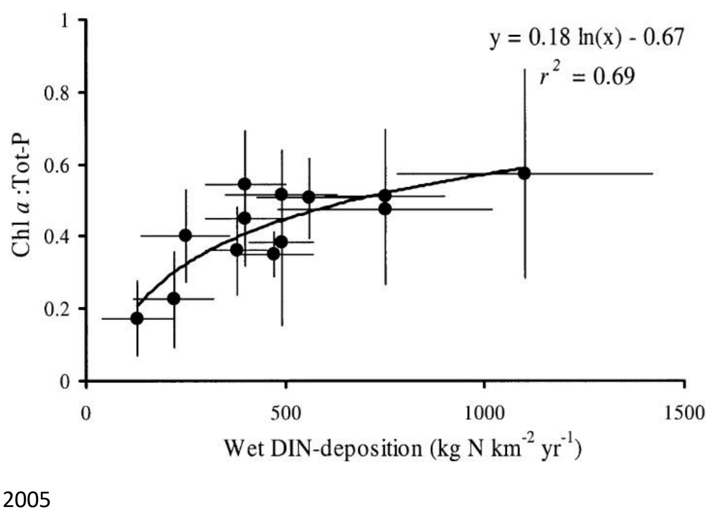
There is ample evidence that an increase in acidic and acidifying compounds in atmospheric deposition has resulted in the acidification of lakes and streams in geologically sensitive regions of Scandinavia, western Europe, Canada and the United States (e.g. Hultberg, 1988; Muniz, 1991). This acidification is characterised by a decrease in pH, alkalinity and acid neutralising capacity (ANC), and by increases in concentrations of sulphate, aluminium, and sometimes nitrate and ammonium. Since the 1970s, various research approaches (field surveys, laboratory studies, whole-lake experiments) have shown that surface water acidification can have dramatic consequences for plant and animal species (macrofauna, fish), and for the functioning of these aquatic ecosystems (Havas and Rosseland, 1995). However, due to the strong reduction in sulphur deposition over the last decades, a (partial) chemical recovery from acidification in these very sensitive waters has been observed in both North America and Europe (e.g. Stoddard et al., 1999; Skjelkvale et al., 2005; Van Kleef et al., 2010; Monteith et al., 2014; Garmo et al., 2014). Currently, critical loads for acidification of surface waters take into account acidifying effects of S and N deposition (Forsius et al., 2021).

Before the 1990s, effects of eutrophication by atmospheric N deposition were hardly viewed as relevant with regards to surface waters, except for some permanent oligotrophic or mesotrophic water bodies (see Chapter 5.2.1). This is because primary production in almost all surface waters was thought to be limited by phosphorus (e.g. Moss, 1988). However, in the past decades it has become obvious that this paradigm does not hold for all freshwater systems, especially in pristine areas of alpine, sub-Arctic, Arctic or boreal regions and that N limitation or co-limitation is widespread (e.g. Saros et al., 2005; Bergström and Jansson, 2006; Wolfe et al., 2006; Sterner, 2008; Elser et al., 2007; De Wit and Lindholm, 2010). This view appears to be widely accepted now, as evidenced in the review from Howarth et al. (2021), on N and P limitation in aquatic ecosystems. The authors conclude that for freshwaters located in catchments with no local N and P sources, and in low N deposition areas, N limitation or co-limitation is common and that lakes can be driven towards P-limitation when N deposition is high and/or catchment N retention is low (Howarth et al., 2021). An update of the literature review in De Wit and Lindholm (2010) also concludes that evidence for the limiting role of N in alpine and boreal lakes has increased (Thrane et al., 2021).

Bergström et al. (2005, 2008) conducted lake sampling and in situ nutrient enrichment enclosure experiments with N and P in oligotrophic boreal lakes (C1.1) along a gradient of increasing atmospheric N deposition (0.9 to 8.0 kg N ha^{-1} yr^{-1} wet deposition) in Sweden. Regional and seasonal patterns of nutrient limitation of phytoplankton were clearly related to the levels of N deposition and catchment N inputs that these lakes received. In areas of low N deposition in northern Sweden (< 3 kg ha^{-1} yr^{-1} wet deposition), N limitation of phytoplankton growth was evident in the summer season due to high N retention in the catchment areas and very low dissolved inorganic N (DIN) inputs during the early summer. Higher N deposition in the south (> 3 kg ha^{-1} yr^{-1} wet deposition) was accompanied by high DIN concentrations in the lakes during the early summer and subsequent P limitation of phytoplankton. However, in these lakes P limitation did not persist over the summer and, as a result of a declining DIN pool, co-limitation by N and P subsequently occurred, followed by N limitation only. Generally, in the summer, the studied oligotrophic Swedish lakes were N limited rather than P limited. The authors concluded that N limitation is probably a natural state of boreal and sub-Arctic oligotrophic lakes, but that P limitation of varying intensities and duration had been induced by elevated atmospheric N deposition (> 3-4 kg ha^{-1} yr^{-1} wet deposition).

In a lake survey that included over 4000 oligotrophic lakes in Europe and North America, Bergström and Jansson (2006) showed that phytoplankton biomass per unit P increased with increasing wet N deposition. The range in wet N deposition was < 1 to 14 kg N ha^{-1} yr^{-1}, and the largest increase in algal biomass per unit P occurred at wet N deposition < 5 kg N ha^{-1} yr^{-1}. Lake sampling, bioassays and physiological assays in four acid-sensitive lakes in the United States (5 to 9 kg N ha^{-1} yr^{-1}) demonstrated that N enrichment gave growth responses in phytoplankton that were similar to or larger than those of P enrichment, and that N deposition was large enough to satisfy daily algal demand for N (Axler et al., 1994). These results support findings from Sweden (Bergström et al., 2005; Figure 5.2) and indicate that N deposition has contributed to higher algal productivity in oligotrophic lakes, including dystrophic ones.

Figure 5.2. The relationship between mean ratios of chlorophyll a and total phosphorus (Chl a: tot P, g to g) and mean wet inorganic nitrogen deposition (wet DIN deposition) in unproductive lakes in different Swedish regions for the period 1995-2001.



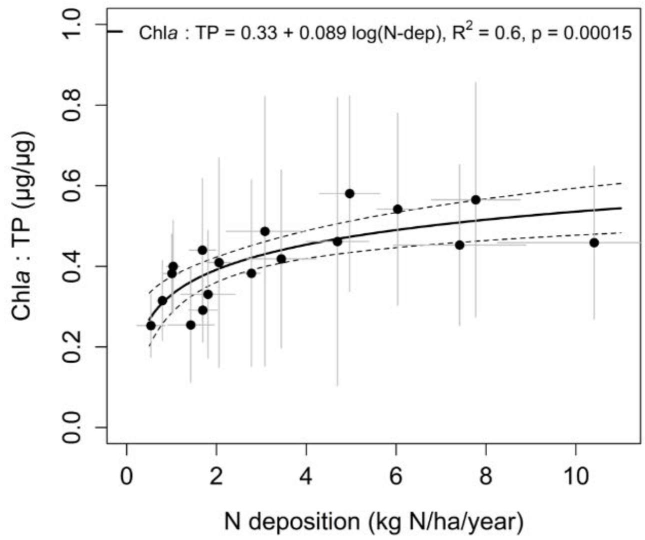
Source: Bergström et al., 2005

The impact of atmospheric N deposition on element stoichiometry was investigated in a survey of oligotrophic lakes in Norway, Sweden and the United States (Colorado), in low and high N-deposition areas, including dystrophic waters, but the dystrophic waters were not addressed separately (Elser et al., 2009). The survey showed that atmospheric wet N deposition increased the stoichiometric N:P ratio in these lakes, indicative of a shift in ecological nutrient limitation. In a subset of lakes, bioassays indicated that phytoplankton growth was generally found to be N limited under low N deposition (approximately 4.5 kg N ha^{-1} yr^{-1} in Norway and approximately 2 kg N ha^{-1} yr^{-1} in Sweden). However, in lakes with high N deposition (approximately 8.5 kg N ha^{-1} yr^{-1} in Norway and approximately 6 kg N ha^{-1} yr^{-1} in Sweden), the growth of phytoplankton was consistently limited by P. Elser et al. (2009) concluded that even relatively low levels of N deposition affected nutrient limitation of phytoplankton growth in oligotrophic lakes in Scandinavia (and the United States). The authors hypothesised that in the long term, in regions with still increasing atmospheric N loads, future functioning of the food web, even in lakes far away from direct human disturbance may be seriously disrupted. Whole-lake manipulations designed to examine effects of experimentally added N on lake food webs (Deininger et al., 2017a, 2017c) have not found strong cascading impacts of N at higher trophic levels.

The referred studies on nutrient limitation in lakes from surveys in Europe and North America are based on data collection from the 1990s and early 2000s. Deposition of N in Europe has declined since 1990 (Engardt et al., 2017; Torseth et al., 2012) while concentrations of DIN have either remained constant or have declined, probably driven by trends in deposition and climate (Kaste et al., 2020; Lucas et al., 2016; Rogora et al., 2012). Surface waters in catchments that receive high N deposition have a higher risk for elevated inorganic nitrogen concentrations (Wright et al., 2001), but this risk is mediated by catchment properties such as vegetation cover or lack thereof, which is found in a dataset of water chemistry, catchment characteristics, climate and deposition from North America and Europe (Austnes et al., 2022).

A data analysis from 2006-2018 from > 300 Nordic lakes in natural catchments (range in wet N deposition from < 2 to > 12 kg N ha^{-1} yr^{-1}) shows that chlorophyll *a* to total phosphorus (Chl*a*:TP) ratios increase with N deposition where total N deposition is below 2 to 4 kg N ha^{-1} yr^{-1} (Thrane et al., 2021; Figure 5.3), suggesting N limitation, while no further responses are found above deposition of 3 to 5 kg N ha^{-1} yr^{-1}. Thus, the threshold range where the lakes turn from N limitation to P or co-limitation is in the same range as indicated by Bergström and Jansson (2006) and Bergström et al. (2005).

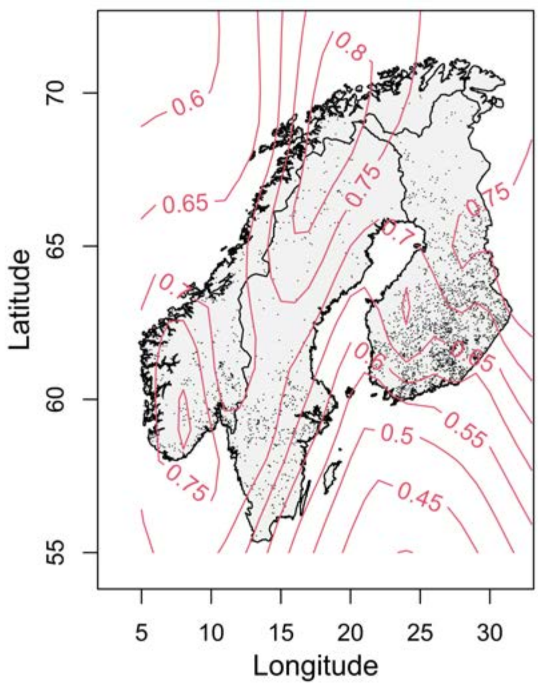
Figure 5.3. The relationship between median values of chlorophyll *a* per unit total phosphorus (chl*a* : TP) and median values of total N deposition for lakes in 18 regions in Norway, Sweden and Finland. Grey lines show median ± one standard deviation for chl*a* : TP (vertical lines) and N deposition (horizontal lines) within each region



Source: Thrane et al., 2021

It should be noted that Thrane et al. (2021) use the sum of dry and wet deposition, while many other studies, including Bergström and Jansson (2006), use wet N deposition. Thrane et al. (2021) calculate that median wet N deposition in their dataset is 68% (25% and 75% percentiles: 63% and 73%, respectively) of total N deposition (EMEP data, for 2006-2018) (Figure 5.4).

Figure 5.4. Mean fraction of wet N deposition to total N deposition in the Nordic countries for 2006-2018.



Source: Thrane et al., 2021

Although this chapter on surface waters does not include an assessment of running waters, it is worthwhile to note that Thrane et al. (2021) also present seasonal element stoichiometry (DIN to TP) from natural rivers in Norway. They find that DIN:TP in river from catchments receiving less than 2.2 kg N ha^{-1} yr^{-1} (total deposition) are most frequently in the range indicative for N limitation established by Bergström (2010) for lakes. River catchments receiving more than 4 ha^{-1} yr^{-1} had DIN:TP ratios that usually indicated P-limitation. Thus, the critical threshold for the shift from N limitation to P limitation for these natural rivers is similar to what the analysis of ratios of chlorophyll-a to total phosphorus from lakes indicates, i.e. 2 to 4 kg N ha^{-1} yr^{-1}. Myrstener et al. (2018) found that biofilm communities in oligotrophic Arctic streams display N limitation, based on nutrient bioassays. Others have found N and co-limitation of biofilm in polluted rivers (Reisinger et al., 2016), suggesting that rivers may display somewhat similar responses to increases in N availability as standing waters.

In recent years, additional focus has been drawn to the role of dissolved organic matter (DOM) as a possible factor that can affect productivity in contrasting ways, i.e. by limiting light availability and by transporting organic forms of N and P (Karlsson et al., 2009; Seekell et al., 2015). Northern lakes have become more DOM-rich in the past decades, related to reduced acid deposition (Monteith et al., 2007) and to increased precipitation (De Wit et al., 2016). DOM in boreal and alpine lakes varies with land cover (forests and peatlands) and climate (Larsen et al., 2011). Results from a whole lake experiment (Deininger et al., 2017b) suggested that effects of enhanced N are weakened along a positive DOM gradient, indicating that algal productivity in clear-water lakes is more sensitive to increased N availability than darker lakes. This implies that clear-water lakes are especially sensitive to N deposition because low DOM and low N retention capacity are causally linked through low pools of soil organic matter in their catchments, a consequence of little vegetation and shallow soils (Evans et al., 2006).

Combined effects of DOM and nutrient availability on food web dynamics suggested that DOM affected food quality for secondary producers (Deininger et al., 2017b). A meta-analysis of field studies and experimental data from a global lake dataset focused on effects of nutrients and DOM on primary and secondary lake productivity supported that N was a limiting nutrient but could not draw definite conclusions on interactions between DOM and N availability for lake productivity (Faithfull et al., 2011). However, the nutritional value of pelagic lake food webs appears to be negatively related with nutrient availability (DIN to total P ratios) and lake temperature (Lau et al., 2021).

Studies of nutrient limitation in epilithic communities of oligotrophic Swedish lakes along a north–south gradient concluded that increased atmospheric N deposition (10-12 kg N ha^{-1} yr^{-1}) intensified P limitation of epilithic algae and invertebrate grazers, compared to those in low deposition areas (2-6 kg N ha^{-1} yr^{-1}), although more studies would be needed to generalise these findings (Liess et al., 2009). In-situ nutrient-diffusion strata placed in alpine lakes in French Alps receiving N deposition of 7 kg N ha^{-1} yr^{-1}, with and without grazing, showed that phyto-benthic biomass was higher in N-enriched substrates, and green filamentous algae were favoured over diatoms and cyanobacteria in N-enriched substrates (Lepori and Robin, 2014). Myrstener et al. (2018) presented nutrient-diffusion strata in Arctic streams in northern Sweden, receiving less than 1 kg N ha^{-1} yr^{-1} to test for various limitations on microbial communities in biofilms and found strong responses to N availability clearly signaling N limitation, but also found that such responses were mitigated by temperature and light availability.

The $CL_{emp}N$ for Atlantic soft-water lakes (see Chapter 5.2.1) has been based on experiments. For boreal, sub-Arctic and alpine lakes, the $CL_{emp}N$ was based on survey and gradient studies supported by experimental evidence from bioassays and whole-lake experiments. Emerging evidence points at factors that mitigate responses to nitrogen – such as DOM (regulating light) and temperature. Still, many studies published after 2010 have further substantiated that N limitation is common in boreal, sub-Arctic and alpine lakes. Studies from North America and Europe come to similar conclusions. This is in particular illustrated by the empirical critical loads developed in the US shortly after the $CL_{emp}N$ developed for the LTRAP Convention (Bobbink and Hettelingh, 2011) presented in Baron et al. (2011). Baron et al. (2011) differentiate between the high-elevation western lakes with sparse vegetation and north-eastern lakes in forest-covered regions, considering catchment differences in capacity to retain atmospherically deposited N. Their results are summarised as follows: The nutrient enrichment $CL_{emp}N$ for western lakes ranged from 1.0 to 3.0 kg N ha^{-1} yr^{-1}, reflecting the nearly non-existent watershed vegetation in complex, snowmelt-dominated terrain. The nutrient enrichment $CL_{emp}N$ for north-eastern lakes ranged from 3.5 to 6.0 kg N ha^{-1} yr^{-1}.

In the 2011 report (Bobbink and Hettelingh 2011), a single $CL_{emp}N$ range for C1.1 (permanent oligotrophic water bodies) of 3 to 10 kg ha^{-1} yr^{-1} (reliable) was proposed. The range should only be applied to oligotrophic waters with low alkalinity with no significant agricultural or other direct human inputs. Furthermore, the lower end of the range would apply to boreal, sub-Arctic and alpine lakes, whereas the upper end of the range applies to Atlantic soft-water lakes. It was also concluded in 2011 that dystrophic lakes (C1.4) may be sensitive to N deposition. This is now further substantiated and it can be stated with more confidence that dystrophic lakes are indeed sensitive to N deposition, but less so than clear-water lakes, which is due to the fact of a) the effect of DOM on light availability, that limits the ecological response to increases in nitrogen, b) the catchment properties that lead to high DOM also lead to higher retention of atmospheric nitrogen deposition.

The evidence that has accumulated since 2010 allows for a better quantification of the $CL_{emp}N$ for C1.1 and C1.4. Clear-water sub-Arctic and alpine lakes (C1.1) in catchments with little vegetation and limited soil cover have a slightly lower threshold for responses to N deposition than boreal lakes (C1.1) from forested regions. Based on the evidence referred to above, we propose a $CL_{emp}N$ range of 2-6 kg ha^{-1} yr^{-1}, where Alpine and sub-Arctic clear-water lakes have a $CL_{emp}N$ range of 2-4 kg N ha^{-1} yr^{-1} (reliable) while boreal (non-dystrophic) lakes have a $CL_{emp}N$ range of 3-6 kg N ha^{-1} yr^{-1}(reliable). Dystrophic, humic lakes (C1.4) in catchments with forests, wetlands and well-developed forest soils are less sensitive to N deposition because more N is retained in the catchment, while high DOM limits the eutrophying effects because of light limitation. Hence, their $CL_{emp}N$ range is proposed to be 5-10 kg N ha^{-1} yr^{-1}(expert judgement).

5.3 Overall summary of $CL_{emp}N$ for inland surface waters (C)

Table 5.1 provides an overview of the $CL_{emp}N$ for inland surface waters (C). The $CL_{emp}N$ should only be applied to waters with low alkalinity and no significant agricultural or other direct human inputs.

Table 5.1. $CL_{emp}N$ and effects of exceedances on surface standing water habitats (C1)[a]. ## reliable, # quite reliable, and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Permanent oligotrophic lakes, ponds and pools	C1.1	3-10	##	**2**-10 [b]	##	Increased algal productivity and a shift in nutrient limitation of phytoplankton

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
(including soft-water lakes)						from N to P; shifts in macrophyte community
▶ Alpine and sub-Arctic clear-water lakes	C1.1			**2-4**	##	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
▶ Boreal clear-water lakes	C1.1			**3-6**	##	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
▶ Atlantic soft-water bodies	C1.1, elements C1.2	3-10	##	**5**-10	##	Change in species composition of macrophyte communities
Permanent dystrophic lakes, ponds and pools	C1.4	3-10	(#)	**5**-10 [c]	(#)	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P

a) The lower part of the $CL_{emp}N$ range should be applied for lakes in small catchments (with high lake to catchment ratios), because these are most exposed to atmospheric deposition, given that a relatively high fraction of their N inputs is deposited directly on the lakes and is not retained in the catchments. Similarly, the lower part of the range should be applied for lakes in catchments with thin soils, sparse vegetation and/or with a high proportion of bare rock.

b) This $CL_{emp}N$ should only be applied to oligotrophic waters with low alkalinity and with no significant agricultural or other human inputs. Apply the lower end of the range to clear-water sub-Arctic and alpine lakes, the middle range to boreal lakes and the higher end of the range to Atlantic soft waters.

c) This $CL_{emp}N$ should only be applied to waters with low alkalinity and with no significant agricultural or other direct human inputs. Apply the lower end of the range to boreal dystrophic lakes.

5.4 References

Aber, J., McDowell, W., Nadelhoffer, K., Magill, A., Berntson, G., Kamakea, M., McNulty, S., Currie, W., Rustad, L. and Fernandez, I. (1998). Nitrogen saturation in temperate forest ecosystems – Hypotheses revisited. *Bioscience* **48,** 921-934.

Arts, G.H.P. (1990). *Deterioration of atlantic soft-water systems and their flora, a historical account*. PhD thesis, University of Nijmegen, Nijmegen.

Arts, G.H.P., Van der Velde, G., Roelofs, J.G.M. and Van Swaay, C.A.M. (1990). Successional changes in the soft-water macrophyte vegetation of (sub)atlantic, sandy, lowland regions during this century. *Freshwater Biology* **24**, 287-294.

Arts, G.H.P. (2002). Deterioration of Atlantic soft water macrophyte communities by acidification, eutrophication and alkalinisation. *Aquatic Botany* **73**, 373-393.

Austnes, K., Hjermann, D.Ø., Sample, J., Wright, R.F., Kaste, Ø. and De Wit, H.A. (2022). *Nitrogen in surface waters: time trends and geographical patterns explained by deposition levels and catchment characteristics.* NIVA report 7728-/2022; ICP Waters report 149/2022. Norwegian Institute for Water Research.

Axler, R.P., C. Rose, and C.A. Tikkanen (1994). Phytoplankton nutrient deficiency as related to atmospheric nitrogen deposition in northern Minnesota acid-sensitive lakes. *Canadian Journal of Fisheries and Aquatic Sciences* **51,** 1281-1296.

Baron J.S., Driscoll, C.T., Stoddard, J.L and E.E. Richer (2011). Empirical critical loads of atmospheric nitrogen deposition for nutrient enrichment and acidification of sensitive US lakes. *Bioscience* **61**, 602-613.

Bergström, A-K., Blomqvist, P. and Jannson, M. (2005). Effects of atmospheric nitrogen deposition on nutrient limitation and phytoplankton biomass in unproductive Swedish lakes. *Limnology and Oceanography* **50**, 987-994.

Bergström, A-K. and Jannson, M. (2006). Atmospheric nitrogen deposition has caused nitrogen enrichment and eutrophication of lakes in the northern hemisphere. *Global Change Biology* **12**, 635-643.

Bergström, A-K., Jonsson, A., and Jansson, M. (2008). Phytoplankton responses to nitrogen and phosphorus enrichment in unproductive Swedish lakes along a gradient of atmospheric nitrogen deposition. *Aquatic Biology* **4**, 55-64.

Bergström, A-K. (2010). The use of TN:TP and DIN:TP ratios as indicators for phytoplankton nutrient limitation in oligotrophic lakes affected by N deposition. *Aquatic Sciences* **72**(3), 277-281.

Bobbink, R., Ashmore, M., Braun, S., Fluckiger, W. and Van den Wyngaert, I.J.J. (2003). *Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update.* Environmental Documentation No. 164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Bobbink, R., Hornung, M., and Roelofs, J.G M. (1996). Empirical nitrogen critical loads for natural and semi-natural ecosystems. Texte 71 – 96, III-1. Federal Environmental Agency, Berlin.

Bobbink, R. and J.P. Hettelingh (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships.* Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), Bilthoven, The Netherlands.

Brouwer, E., Bobbink, R., Meeuwsen, F. and Roelofs, J.G.M. (1997). Recovery from acidification in aquatic mesocosms after reducing ammonium and sulphate deposition. *Aquatic Botany* **56**, 119-130.

Brouwer, E., Bobbink, R. and Roelofs, J.G.M. (2002). Restoration of aquatic macrophyte vegetation in acidified and eutrophied softwater lakes: an overview. *Aquatic Botany* **73**, 405-431.

Davies, C.E., Moss, D. and Hill, M.O. (2004). EUNIS habitat classification revised 2004. European Environment Agency, European Topic Centre on Nature Protection and Biodiversity.

De Wit, H.A. and Lindholm, M. (2010). *Nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters - a review*. In: NIVA report 6007/2010; ICP Waters report 101/2010. Norwegian Institute for Water Research, Oslo. p 39

De Wit, H.A., Valinia S., Weyhenmeyer G.A., Futter, M.N., Kortelainen, P., Austnes, K., Hessen, D.O., Räike, Laudon, H. and Vuorenmaa, J. (2016). Current Browning of Surface Waters Will Be Further Promoted by Wetter Climate. *Environ. Sci. Technol. Lett.* **3**(12), 430-435.

Deininger, A., Faithfull, C.L. and Bergström, A.K. (2017a). Nitrogen effects on the pelagic food web are modified by dissolved organic carbon. *Oecologia* **184**, 901-916.

Deininger, A., Faithfull, C.L. and Bergström, A.K. (2017b). Phytoplankton response to whole lake inorganic N fertilization along a gradient in dissolved organic carbon. *Ecology* **98**, 982-994.

Deininger, A., Faithfull, C.L., Karlsson, J. Klaus, M. and Bergström, A.K. (2017c). Pelagic food web response to whole lake N fertilization. *Limnology and Oceanography* **62**, 1498-1511.

Den Hartog, C. (1986). The effects of acid and ammonium deposition on aquatic vegetations in the Netherlands. In: *Proceedings 1st. Internat. Symposium on water milfoil (Myriophyllum spicatum) and related Haloragaceae species*. Vancouver, Canada, 51-58.

Dise, N.B. and R.F. Wright (1995). Nitrogen leaching from European forests in relation to nitrogen deposition. *Forest Ecology and Management* **71,**153-161.

Dise, N.B., Rothwell, J.J., Gauci, V., Van der Salm, C. and De Vries, W. (2009). Predicting dissolved inorganic nitrogen leaching in European forests using two independent databases. *Science of the Total Environment*, **407**, 1798-1808.

Engardt, M., Simpson, D., Schwikowski, M. and Granat, L. (2017). Deposition of sulphur and nitrogen in Europe 1900-2050. Model calculations and comparison to historical observations. *Tellus Ser. B-Chemical and Physical Meteorology,* **69**, 20.

Elser, J.J., Fagan, W.F., Denno, R.F., Dobberfuhl, D.R., Folarin, A., Huberty, A., Interlandi, S., Kilham, S.S., McCauley, E., Schulz, K.L., Siemann, E.H. and Sterner, R.W. (2000). Nutritional constraints in terrestrial and freshwater food webs. *Nature*, **408**, 578-580.

Elser, J.J., Bracken, M.E.S., Cleland, E.E., Gruner, D.S., Harpole, W.S., Hillebrand, H., Ngai, J.T., Seabloom, E.W., Shurin, J.B. and Smith, J.E. (2007). Global analysis of nitrogen and phosphorus limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecology Letters* **10**, 1135-1142.

Elser, J.J., Andersen, T., Baron, J.S., Bergström, A.K., Jansson, M., Kyle, M., Nydick, K.R., Steger, L. and Hessen, D.O. (2009). Shifts in lake N:P stoichiometry and nutrient limitation driven by atmospheric nitrogen deposition. *Science* **326,** 835.

European Communites (2003). Common Implementation Strategy for the Water Framework Directive (2000/60/EC). Guidance Document No. 10. Rivers and Lakes – Typology, Reference Conditions and Classification Systems. Produced by Working Group 2.3 – REFCOND. Available here: https://circabc.europa.eu/sd/a/dce34c8d-6e3d-469a-a6f3-b733b829b691/Guidance%20No%2010%20-%20references%20conditions%20inland%20waters%20-%20REFCOND%20(WG%202.3).pdf

Evans, C.D., Reynolds, B., Jenkins, A., Helliwell, R.C, Curtis, C.J., Goodale, C.L., Ferrier, R.C., Emmett, B.A., Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Norris, D., Davies, J. and Coull, M.C. (2006). Evidence that soil carbon pool determines susceptibility of semi-natural ecosystems to elevated nitrogen leaching. *Ecosystems* **9**, 453-462.

Faithfull, C.L., Wenzel, A., Vrede, T. and Bergstrom, A.K. Testing the light: nutrient hypothesis in an oligotrophic boreal lake. *Ecosphere* **2**, 1-15.

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., De Wit, H.A., Dirnbock, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Kram, P., Lindroos, A.J., Lofgren, S., Pecka, T., Ronnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S. and Vana, M. (2021). Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe. *Science of the Total Environment* **753**, 12.

Garmo, O.A., Skjelkvale, B.L., De Wit, H.A., Colombo, L., Curtis, C., Folster, J., Hoffmann, A., Hruska, J., Hogasen, T., Jeffries, D.S., Keller, W.B., Kram, P., Majer, V., Monteith, D.T., Paterson, A.M., Rogora, M., Rzychon, D., Steingruber, S., Stoddard, J.L., Vuorenmaa, J. and Worsztynowicz, A. (2014). Trends in Surface Water Chemistry in Acidified Areas in Europe and North America from 1990 to 2008. *Water Air and Soil Pollution* **225**, 1880.

Glime, J.M. (1992). Effects of pollutants on aquatic species. In: Bates, J.W. and Farmer, A.M. (eds.) *Bryophytes and lichens in a changing environment.* Clarendon Press, Oxford, 333-361.

Havas, M., and Rosseland, B.O. (1995). Response of zooplankton, benthos, and fish to acidification: An overview. *Water Air and Soil Pollution* **85,** 51-62.

Helliwell, R. C., M. C. Coull, J. J. L. Davies, C. D. Evans, D. Norris, R. C. Ferrier, A. Jenkins and B. Reynolds (2007). The role of catchment characteristics in determining surface water nitrogen in four upland regions in the UK. *Hydrology and Earth System Sciences* **11**,356-371.

Helliwell, R.C., Ferrier, R.C. and Kernan, M.R. (2001). Interaction of nitrogen deposition and land use on soil and water quality in Scotland: issues of spatial variability and scale. *Science of the Total Environment* **265**, 51-63.

Howarth, R.W., Chan, F., Swaney, D.P., Marino, R.M. and Hayn, M. (2021). Role of external inputs of nutrients to aquatic ecosystems in determining prevalence of nitrogen vs. phosphorus limitation of net primary productivity. *Biogeochemistry* **154**, 293-306.

Hultberg, H. (1988). Critical loads for sulphur to lakes and streams. In: Nilsson, J. and Grennfelt, P. (eds.). *Critical loads for sulphur and nitrogen*. Nordic Council of Ministers, Kopenhagen, 185-200.

Karlsson, J., Bystrom, P., Ask, J., Persson, L. and Jansson, M. (2009). Light limitation of nutrient-poor lake ecosystems. *Nature,* **460**, 506-509.

Kaste, Ø., Austnes, K. and De Wit, H.A. (2020). Streamwater responses to reduced nitrogen deposition at four small upland catchments in Norway. *Ambio*, **49**, 1759-1770.

Larsen, S., Andersen, T. and Hessen, D.O. (2011). Predicting organic carbon in lakes from climate drivers and catchment properties. *Global Biogeochemical Cycles* **25**, GB3007.

Lau, D.C.P., Jonsson, A., Isles, P.D.F., Creed, I.F. and Bergström, A.K. (2021). Lowered nutritional quality of plankton caused by global environmental changes. *Global Change Biology*, **13**.

Lepori, F. and Robin, J. (2014). Nitrogen limitation of the phytobenthos in Alpine lakes: results from nutrient-diffusing substrata. *Freshwater Biology* **59**, 1633-1645.

Leuven, R.S.E.W., Den Hartog, C., Christiaans, M.C.C. and Heyligers, W.H.C. (1986). Effects of water acidification on the distribution pattern and the reproductive success of amphibians. *Experientia* **42**, 495-503.

Liess, A., Drakare, S. and Kahlert, M. (2009). Atmospheric nitrogen-deposition may intensify phosphorus limitation of shallow epilithic periphyton in unproductive lakes. *Freshwater Biology* **54**, 1759-1773

Lucas, R.W., Sponseller, R.A., Gundale, M.J., Stendahl, J., Fridman, J., Högberg, P. and Laudon, H. (2016). Long-term declines in stream and river inorganic nitrogen (N) export correspond to forest change. *Ecological Applications* **26**, 545-556.

Marchetto, A., Mosello, R., Psenner, R., Barbieri, A., Bendetta, G., Tait, D. and Tartari, G.A. (1994). Evaluation of the level of acidification and the critical loads for Alpine lakes. *Ambio* **23**, 150-154.

Moe, S.J., Dudley, B. and Ptacnik, R. (2008). REBECCA databases: experiences from compilation and analyses of monitoring data from 5,000 lakes in 20 European countries. *Aquatic Ecology* **42,**183-201.

Monteith, D.T., Stoddard, J.L., Evans, C.D., De Wit, H.A., Forsius, M., Høgåsen, T., Wilander, A., Skjelkvåle, B.L., Jeffries, D.S., Vuorenmaa, J., Keller, B., Kopácek, J. and Vesely, J. (2007). Dissolved organic carbon trends resulting from changes in atmospheric deposition chemistry. *Nature* **450**, 537-540.

Monteith, D.T., Evans, C.D., Henrys, P.A., Simpson, G.L and Malcolm, I.A. (2014). Trends in the hydrochemistry of acid-sensitive surface waters in the UK 1988-2008. *Ecological Indicators* **37**, 287-303.

Myrstener, M., Rocher-Ros, G., Burrows, R.M., Bergström, A.K., Giesler, R. and Sponseller, R.A. (2018). Persistent nitrogen limitation of stream biofilm communities along climate gradients in the Arctic. *Global Change Biology* **24**, 3680-3691.

Moss, B. (1988). *Ecology of Fresh Waters Man and Medium*. Blackwell, Oxford.

Muniz, I.P. (1991). Freshwater acidification: its effects on species and communities of freshwater microbes, plants and animals. *Proceedings of the Royal Society of Edinburgh* **97B**, 227-254.

Oulehle, F., Chuman, T., Majer, V. and Hruska, J. (2013). Chemical recovery of acidified Bohemian lakes between 1984 and 2012: the role of acid deposition and bark beetle induced forest disturbance. *Biogeochemistry* **116**, 83-101.

Reisinger, A.J., Tank, J.L. and Dee, M.M. (2016). Regional and seasonal variation in nutrient limitation of river biofilms. *Freshwater Science* **35**, 474-489.

Roelofs, J.G.M. (1983). Impact of acidification and eutrophication on macrophyte communities in soft waters in the Netherlands 1. Field observations. *Aquatic Botany* **17**, 139-155.

Roelofs, J.G.M., Schuurkes, J.A.A.R. and Smits, A.J.M. (1984). Impact of acidification and eutrophication on macrophyte communities in soft waters. II Experimental studies. *Aquatic Botany* **18**, 389-411.

Rogora, M., Arisci, S. and Marchetto, A. (2012). The role of nitrogen deposition in the recent nitrate decline in lakes and rivers in Northern Italy. *Science of the Total Environment* **417,** 214-223.

Saros, J.E., Michel, T.J., Interlandi, S.J. and Wolfe, A.P. (2005). Resource requirements of *Asterionella Formosa* and *Fragilaria crotonsis* in oligotrophic alpine lakes: implications for recent phytoplankton community reorganisations. *Canadian Journal of Fisheries and Aquatic Sciences* **62,** 1681-1689.

Schoof-van Pelt, M.M. (1973). Littorelletea, a study of the vegetation of some amphiphytic communities of western Europe. PhD thesis. Catholic University of Nijmegen.

Schuurkes, J.A.A.R., Kok, C.J. and Den Hartog, C. (1986). Ammonium and nitrate uptake by aquatic plants from poorly buffered and acidified waters. *Aquatic Botany* **24**, 131-146.

Schuurkes, J.A.A.R., Elbers, M.A., Gudden, J.J.F. and Roelofs, J.G.M. (1987). Effects of simulated ammonium sulphate and sulphuric acid rain on acidification, water quality and flora of small-scale soft water systems. *Aquatic Botany* **28**, 199-225.

Seekell, D.A., Lapierre, J.F., Ask, J., Bergström, A.K., Deininger, A., Rodriguez, P. and Karlsson, J. (2015). The influence of dissolved organic carbon on primary production in northern lakes. *Limnology and Oceanography* **60**, 1276-1285.

Shurin, J.B., Gruner, D.S. and Hillebrand, H. (2006). All wet or dried up? Real differences between aquatic and terrestrial food webs. *Proceedings of the Royal Society B-Biological Sciences* **273**, 1-9.

Skjelkvale, B.L., Stoddard, J.L., Jeffries, D.S., Torseth, K., Hogasen, T., Bowman, J., Mannio, J., Monteith, D.T., Mosello, R., Rogora, M., Rzychon, D., Vesely, J., Wieting, J., Wilander, A. And Worsztynowicz, A. (2005). Regional scale evidence for improvements in surface water chemistry 1990-2001. *Environmental Pollution* **137**, 165-176.

Sterner, R.W. (2008). On the phosphorus limitation paradigm for lakes. *International Review of Hydrobiology* **93,** 433-445

Stoddard, J.L. (1994). Long-term changes in watershed retention of nitrogen-its causes and aquatic consequences, p. 223-284, In: Baker, L.A. (ed.) *Environmental Chemistry of Lakes and Reservoirs*, Vol. 237.

Stoddard, J.L., Jeffries, D.S., Lukewille, A., Clair, T.A., Dillon, P.J., Driscoll, C.T., Forsius, M., Johannessen, M., Kahl, J.S., Kellog, J.H., Kemp, A., Mannio, J., Monteith, D.T., Murdoch, P.S., Patrick., S., Rebsdorf, A., Skjelkvale, B.L., Staiton, M.P., Traaen, T., Van Dam, H., Webster, K.E., Wieting, J. and Wilander, A. (1999). Regional trends in aquatic recovery from acidification in North America and Europe. *Nature* **401**, 575-578.

Thrane, J.E. De Wit, H.A. and Austnes, K. (2021) *Eutrophying effects of nitrogen in boreal surface waters in natural catchments.* In: NIVA report 7680-2021; ICP Waters report 146/2021. Norwegian Institute for Water Research, Oslo.

Torseth, K., Aas, W., Breivik, K., Fjaeraa, A.M., Fiebig, M., Hjellbrekke, A.G., Myhre, C.L., Solberg S. and Yttri, K. E. (2012). Introduction to the European Monitoring and Evaluation Programme (EMEP) and observed atmospheric composition change during 1972-2009. *Atmospheric Chemistry and Physics* **12**, 5447-5481.

Van Kleef, H.H., Brouwer, E., Leuven, R.S.E.W., Van Dam, H., De Vries-Brock A., Van der Velde, G. and Esselink, H. (2010) Effects of reduced nitrogen and sulphur deposition on the water chemistry of moorland pools. *Environmental Pollution* **158**, 2679-2685.

Wittig, R. (1982). Verbreitung der Littorelletea-Arten in der Westfälischen Bucht. *Decheniana (Bonn)* **135**, 14-21.

Wolfe, A.P., Cooke, C.A. and Hobbs, W.O. (2006). Are current rates of atmospheric Nitrogen deposition influencing lakes in the eastern Canadian Arctic? *Arctic, Antarctic and Alpine Research* **38,** 465-476.

Wright, R.F., Alewell C., Cullen, J.M., Evans, C.D., Marchetto, A., Moldan, F., Prechtel, A. and Rogora, M. (2001). Trends in nitrogen deposition and leaching in acid-sensitive streams in Europe. *Hydrology and Earth System Sciences* **5,** 299-310.

6 Effects of nitrogen deposition on mire, bog and fen habitats (EUNIS class Q, formerly D)

Adapted by Chris Field, Julian Aherne, Roland Bobbink and Hilde Tomassen

Sphagnum moss species in the Forest of Alyth Mires, Scotland, UK. Photo: Chris Field.

Summary

Mire, bog and fens (EUNIS class Q) are among the most sensitive habitats to nitrogen deposition, and this is reflected in their low critical load ranges. A limited number of new experimental addition studies have been published since the last update; in contrast, a large number of gradient studies have been published. For raised and blanket bogs (Q1), recent experimental research and gradient studies confirm the previous recommended empirical N critical load ($CL_{emp}N$) of 5-10 kg ha^{-1} yr^{-1}. However, several studies indicate that effects already occur at the lower end of this range and additional work is needed to understand these responses further. In addition, recent experimental studies show that reduced nitrogen is probably more harmful than oxidised nitrogen. For poor fens (Q22) based on limited new evidence we recommend a reduction to the low end of the critical load and a revised $CL_{emp}N$ range of 5 to 15 kg N ha^{-1} yr^{-1}. In rich fens (Q41-Q44) a recent gradient study provides strong indications that the upper limit of 30 kg N ha^{-1} yr^{-1} is too high, so we propose a new range of 15 to 25 kg N ha^{-1} yr^{-1}. This range corresponds with the $CL_{emp}N$ for arctic-alpine rich fens (Q45). Based on expert judgement, we were able to set a new $CL_{emp}N$ range for palsa and polygon mires (Q3) of 3 to 10 kg N ha^{-1} yr^{-1}.

6.1 Introduction

Class Q of the European Nature Information System (EUNIS) includes a wide range of wetland systems that have their water table at or above soil or sediment level for at least half of the year, dominated by either herbaceous or ericoid vegetation (Davies and Moss, 2002; Davies et al., 2004). EUNIS Q encompasses the broad habitat classifications: Raised and blanket bogs (Q1), Valley mires, poor fens and transition mires (Q2), Palsa and polygon mires (Q3), Base-rich fens and calcareous spring mires (Q4) and Helophyte beds (Q5). Nutrient budgets in these wetland ecosystems are characterised by inputs and outputs of nutrients via groundwater and surface water and are tightly linked with local hydrology. The extent to which these systems receive and lose nutrients with in- and out-flowing water largely determines their sensitivity to excess nitrogen (N) from atmospheric deposition. Little to very little effects from N deposition are to be expected in several open wetland systems, such as reed marshes and sedge beds (EUNIS category Q5, e.g. Morris, 1991). A larger impact of atmospheric N deposition is expected in systems with a closed N cycle. This, of course, is especially clear in the case of ombrotrophic bogs (EUNIS Q1), which receive the majority of their nutrients from the atmosphere. In addition, attention should be paid to the existing hydrological state of the studied wetlands, as decreasing water tables may enhance decomposition and thus nutrient availability and allow encroachment by shrubs, whereas in areas of higher water table, hydrology may constrain responses to N.

The following chapter builds on the last review of $CL_{emp}N$ in 2011 (Bobbink and Hettelingh, 2011) and adds a synthesis of information published since this review to determine retention or revision of the $CL_{emp}N$ values. Most research undertaken to date has been on raised and blanket bogs (Q1), which is the focus of much of this chapter. However, we also cover valley mires, poor-fens and transition mires (Q2), base-rich fens and calcareous spring mires (Q4), and for the first time, we are able to provide a $CL_{emp}N$ for palsa and polygon mires (Q3).

6.2 Raised and blanket bogs (Q1)

Introduction

Ombrotrophic (raised) and blanket bogs are nutrient-poor habitats that receive all their nutrients from the atmosphere, and therefore are particularly sensitive to airborne N deposition. These bogs are systems of acidic, wet areas and are very common in the boreal and temperate regions of Europe. Due to a high-water table and often anaerobic conditions, decomposition rates are low, favouring the development of peat. Typical plant species of bogs include peat mosses (*Sphagnum*), sedges (*Carex*, *Eriophorum*) and ericaceous plants (*Andromeda*, *Calluna* and *Erica*). Within the EUNIS system, these communities have been classified under Q1 (raised and blanket bogs) for which the criterion is that precipitation is their continuous or primary water supply. EUNIS category Q1 is subdivided into raised bogs (Q11) and blanket bogs (Q12). Raised bogs are highly oligotrophic, strongly acidic peatlands with a raised centre from which water drains towards the edges. Blanket bogs are formed on flat or gently sloping grounds with poor surface drainage, in oceanic climates with high levels of precipitation (north-western Europe) (Davies et al., 2004). For the purpose of critical load definition, there is no basis on which to differentiate between raised and blanket bogs. Both bog types support the same Ericoid vegetation, and both are acidic and oligotrophic which suits the formation of carpets of *Sphagnum* spp. and hummocks of *Eriophorum vaginatum*.

The current $CL_{emp}N$ for ombrotrophic and blanket bogs is between 5-10 kg ha^{-1} yr^{-1} based on evidence gathered up to 2011. Since the last update of the $CL_{emp}N$ for ombrotrophic and blanket bogs (Bobbink and Hettelingh, 2011), a significant number of publications on the effects of N on bogs have appeared. The results from these studies confirm the importance of *Sphagnum* mosses for the immobilisation of N; when *Sphagnum* becomes saturated, N availability increases for vascular plants, leading to increased vascular plant biomass (Berendse et al., 2001; Bobbink and Hettelingh, 2011) and shifts in species composition. Experiments with realistic additions of N that are within the range of the $CL_{emp}N$ (5-10 kg ha^{-1} yr^{-1}) and that may be used to validate the existing critical load, are still limited. However, evidence is emerging from experimental N addition studies from Scotland (addition levels from 8-56 kg N ha^{-1} yr^{-1}) and Canada (addition levels from 5-25 kg N ha^{-1} yr^{-1} in a low-N area (bulk [wet] deposition < 2 kg N ha^{-1} yr^{-1})), as well as gradient studies from Canada, Ireland, Norway and the UK that support experimental responses.

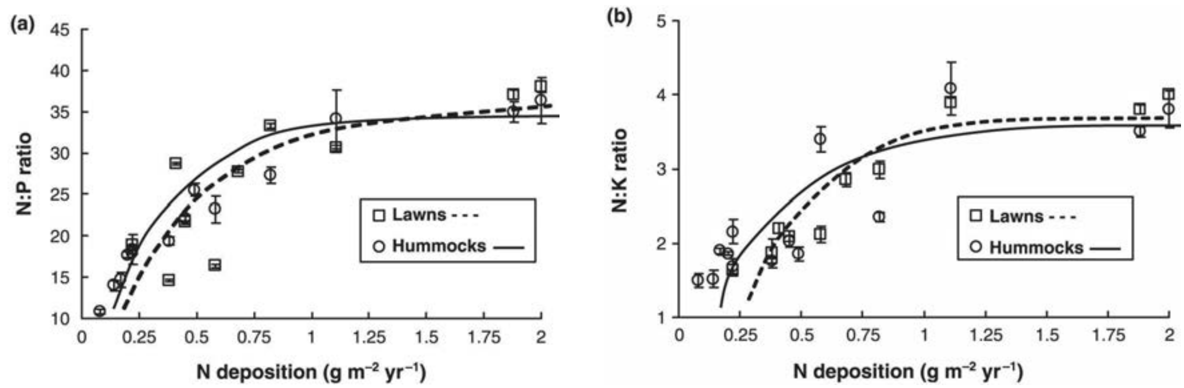
Physiological responses in bryophytes and lichens

This section on physiological responses drawers heavily on work captured during the previous review of $CL_{emp}N$ in 2011, however, recent work from Canada and northern Europe is incorporated alongside the earlier body of evidence. Several studies on British bogs have shown that increased supplies of N are rapidly absorbed and utilised by bog mosses (*Sphagnum* species), reflecting the significance of N deposition as a nutrient and its scarcity in unpolluted regions (e.g. Press et al., 1988). High N loadings, however, are supra-optimal for the growth of many characteristic *Sphagnum* species, as demonstrated by restricted development in growth experiments and transplantation studies between clean and polluted locations. In areas with high N loads such as the Pennines, the growth of *Sphagnum* in general was lower than in unpolluted areas (Lee and Studholme, 1992). After transplantation of *Sphagnum* from an 'unpolluted' site to a bog in the southern Pennines, a rapid increase in N content from ca 12 to 20 mg g^{-1} dry weight was observed (Press et al., 1988). Furthermore, a large increase in N-containing amino acids (arginine) in the shoots of these bog mosses was found after application of N, indicating nutritional imbalance in this species.

At Whim Moss, Scotland, *Sphagnum papillosum* tissue N content increased in response to N additions of 8 kg N ha^{-1} yr^{-1} on top of ambient N deposition of around 8-11 kg N ha^{-1} yr^{-1} (Millet et al., 2012). The same experiment also revealed tissue stoichiometry responses in the lichen *Cladonia portentosa*, albeit to higher levels of N addition (+56 kg N ha^{-1} yr^{-1}; Hogan et al., 2010) and changes in proteins responsible for physiological processes such as respiration and photosynthesis (Munzi et al., 2017; 2020), highlighting a potential underlying response mechanism. The work highlighted that elevated N levels can also shift nutrient limitation to phosphorus (P) availability (Hogan et al., 2010) as found in earlier studies (e.g. Aerts et al., 1992; Gunnarsson and Rydin, 2000).

Bragazza et al. (2004) determined nutrient limitation of ombrotrophic *Sphagnum* plants across a natural gradient of bulk atmospheric N deposition ranging from 1 to 20 kg ha^{-1} yr^{-1} in Europe. Nutrient ratios increased steeply at low atmospheric input, but above a threshold of ~10 kg ha^{-1} yr^{-1} the N:P and N:K ratios tended to saturate (Figure 6.1) and *Sphagnum* growth changed from being N limited to K and P co-limited.

Figure 6.1. Mean values (± 1 SE) of (a) N: P and (b) N: K ratios in hummock and lawn *Sphagnum* plants across a natural gradient of bulk atmospheric N deposition ranging from 1 to 20 kg ha^{-1} yr^{-1} in Europe (1 g m^{-2} yr^{-1} is equal to 10 kg ha^{-1} yr^{-1}). Dashed and continuous lines represent the theoretical patterns based on regression model calculations.



Source: Bragazza et al., 2004

Phuyal et al. (2008) studied the effects of increased atmospheric N deposition on phosphatase activity in *Sphagnum capillifolium* tissue in the lowland ombrotrophic bog at Whim Moss (Scotland) over a four-year period. Phosphatase activity (an enzyme that catalyses the cleavage of inorganic P from organic P compounds) of *Sphagnum capillifolium* was found to be significantly increased by addition of 56 kg N^{-1} ha^{-1} yr^{-1} in both ammonium (NH_4Cl) and nitrate ($NaNO_3$) treatments, ambient N deposition was around 8 kg ha^{-1} yr^{-1}. Phosphatase activity was found to be positively related with tissue N and negatively related to tissue P concentrations. This implies that, when N starts to accumulate in *Sphagnum* tissue, the relative availability of P may be (temporarily) increased by increasing phosphatase activity, leading to increased growth and thereby dilution of tissue N concentration.

Although these and other studies strongly indicate the detrimental effects of high N deposition rates on the development of lichens and bog-forming *Sphagnum* species, there is also evidence of growth stimulation in response to small increments in N deposition. Field experiments by Aerts et al. (1992) at a site with low atmospheric deposition (0.4 kg N ha^{-1} yr^{-1}) in northern Sweden showed that *Sphagnum balticum* increased its growth four-fold within three years of N addition (20 and 40 kg N ha^{-1} yr^{-1}), whilst no effect was found on *Sphagnum magellanicum*[2] at southern Sweden sites with higher atmospheric deposition (7-9 kg N ha^{-1} yr^{-1}). Responses at very low atmospheric deposition of N have also been observed in lichens including the epiphyte *Evernia mesomorpha* which, in a Canadian study, showed increases in chlorophyll fluorescence as N deposition increased above a background of 1.3 kg N ha^{-1} yr^{-1} (Vitt et al., 2020).

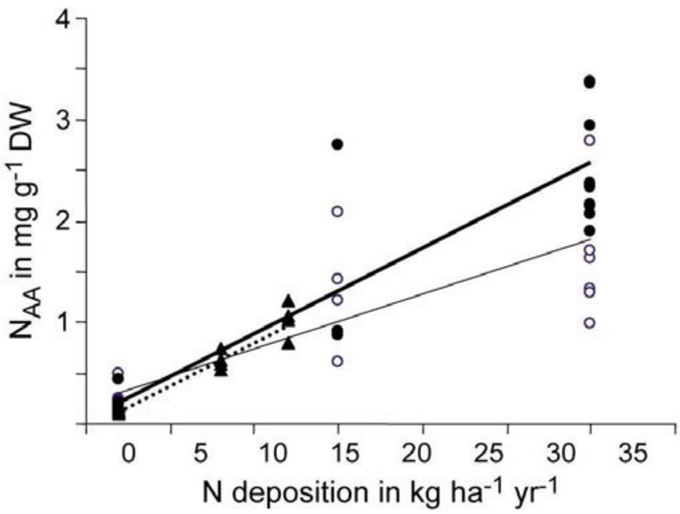
A three-year field manipulation experiment used a range of N addition rates (0, 10, 30, 50, 100 kg N ha^{-1} yr^{-1}) at two Swedish mires with differing existing atmospheric loads (Gunnarsson and Rydin, 2000; Nordin and Gunnarsson, 2000). N:P ratios and experimental results confirmed that *Sphagnum* growth at the site in central Sweden, with very low ambient deposition (~3-4 kg N background), was N limited, while the southern site (~7-11 kg N background) was P limited (Gunnarsson and Rydin, 2000). After two years, the experimental addition of N increased free amino acid concentrations in *Sphagnum* capitula, whereas it decreased *Sphagnum* extension growth at deposition rates of 30 kg N ha^{-1} yr^{-1} and over. However, at low N deposition rates

2 *Sphagnum magellanicum* has recently been split into three species: *S. magellanicum*, *S. divinum* and *S. medium* (Hassel et al., 2018), however, it is not possible to determine which of these specific species in this review of critical loads are referred to hence we used the collective name '*S. magellanicum*'.

(lower than 7-11 kg N ha^{-1} yr^{-1}), no correlation was observed between *Sphagnum* total amino acid N concentrations and growth rates (Nordin and Gunnarsson, 2000). After three years of treatment, biomass production and length increment decreased, while capitulum dry mass increased, with increasing N addition. This experiment included a treatment of 10 kg N ha^{-1} yr^{-1}, but the biomass and length responses showed a steady decline from the control treatment, especially in areas dominated by *Sphagnum rubellum*. Thus, even if the control and 10 kg N ha^{-1} yr^{-1} treatments are not statistically distinguished, these data suggest a threshold for effects on such a bog community exists below 10 kg N ha^{-1} yr^{-1}.

Accumulation of free amino acids in *Sphagnum* capitula was found in several other studies. Tomassen et al., (2003) treated transplanted turfs from an ombrotrophic floating bog with a range of N deposition rates in the laboratory over three years. Tissue N concentrations in *Sphagnum fallax* showed a linear response to the experimental N addition. Excess N was accumulated as N-rich free amino acids, starting already at very low N addition rates (> 2.5 kg ha^{-1} yr^{-1}), indicating N saturation. Wiedermann et al., (2009b) also concluded that N accumulation in the form of free amino acids (N_{AA}) is a sensitive indicator signalling N saturation and future shifts in vegetation composition, although no clear relationship between this indicator and a shift in plant diversity could be drawn (Figure 6.2).

Figure 6.2. The relationship between soluble amino acid N tissue content (mg g^{-1} DW) and N deposition (kg N ha^{-1} yr^{-1}) for *S. balticum* samples from a gradient study (filled triangles, dashed line), and from a field experimentin 1997 (open circles, thin line) and 2004 (filled circles, solid line).



Source: Wiedermann et al., 2009b

The linear increase in total N and amino acid N concentrations in lichen tissue paralleling increased N deposition (Figure 6.2) raises the question whether or not *Sphagnum* species are capable of adjusting to high N supply through N uptake regulation. Wiedermann et al. (2009a) exposed *S. balticum* and *S. fuscum* from three peatland sites differing in N deposition (2, 8 and 12 kg N ha^{-1} yr^{-1}) to different N forms labelled with 15N (15NH_4+, 15NO_3- and the amino acids [15N] alanine and [15N] glutamic acid). All the forms of applied N were taken up by both *Sphagnum* species. Uptake rates were highest for NH_4+, followed by alanine and glutamic acid, with only very small amounts of NO_3- taken up. At the site with the highest background deposition (12 kg N ha^{-1} yr^{-1}) N uptake was lower compared to the two other sites (2 and 8 kg N ha^{-1} yr^{-1}). The potential of *Sphagnum* to adjust to high N exposure through N uptake regulation will not prevent tissue N accumulation but is likely to delay the toxic effects of high tissue NH_4+ concentrations.

Increased pathogen infections in *Sphagnum* have also been linked to N. Following three consecutive years of N fertilisation in field experiments in Ireland and the Netherlands, Limpens et al., (2003b) observed increased fungal infections, caused by the parasite *Lyophyllum palustre* (a basidiomycete fungus). Total background N deposition in these experiments ranged from 15 (Irish site) to 37 (Dutch sites) kg N ha^{-1} yr^{-1}, with additions of 40 kg N ha^{-1} yr^{-1} in N treatments. In a subsequent greenhouse experiment, they inoculated *Sphagnum* to verify that the necrosis found had indeed been caused by *Lyophyllum palustre* and was related to tissue N concentration. This experiment confirmed that *Lyophyllum palustre* was responsible for the necrosis or defoliation of *Sphagnum* and disease severity was related to the N concentration in the capitula (Limpens et al., 2003b).

In a north–south transplant experiment, covering a latitudinal N deposition gradient across northern Europe ranging from 2.8 kg ha^{-1} yr^{-1} in the north to 14.9 kg ha^{-1} yr^{-1} in the south, Granath et al. (2009a) measured photosynthetic responses to increasing N deposition in *Sphagnum balticum* and *Sphagnum fuscum*. The maximum photosynthetic rate increased southwards and was mainly explained by tissue N concentrations. For *Sphagnum fuscum* photosynthetic rate continued to increase up to a deposition level of 14.9 kg N ha^{-1} yr^{-1}, whereas for *Sphagnum balticum* this seemed to level out at 11.4 kg N ha^{-1} yr^{-1}. The results suggest that *Sphagnum* species may be able to adapt or physiologically adjust to higher N depositions, and that *Sphagnum balticum* might be more sensitive to N deposition than *Sphagnum fuscum.* The maximum photosynthetic rate was not (*Sphagnum balticum*) or only weakly (*Sphagnum fuscum*) correlated with biomass production, indicating that production is largely governed by factors other than photosynthetic capacity (Granath et al., 2009a).

However, in a 12-year field fertilisation experiment in Sweden. The maximum photosynthetic rate in *Sphagnum balticum* did not differ between the control (2 kg ha^{-1} yr^{-1}) and the high N treatment (30 kg ha^{-1} yr^{-1}) but was higher for the middle N treatment (15 kg ha^{-1} yr^{-1}) (Granath et al., 2009b). The optimum tissue N concentration for photosynthetic rate in *S. balticum* was found to be ≈ 13 mg N g^{-1}. Granath et al., (2009b) suggest that negative effects on *Sphagnum* productivity under high N deposition are not related to negative effects on the photosynthetic apparatus. However, differences in optimum N concentration levels between the various *Sphagnum* species may affect their competitive ability under different N deposition regimes. In a subsequent greenhouse experiment, increases in tissue N concentrations were observed when a range of N between 0 and 5.6 kg N ha^{-1} yr^{-1} were added to plants collected from a background N deposition area of around 2 kg N ha^{-1} yr^{-1} (Granath et al., 2012). *Sphagnum balticum* showed the same unimodal response as the field experiment, whilst productivity of *S. fallax* and *S. fuscum* declined with the authors suggesting P limitation.

For the assessment of $CL_{emp}N$, these pot or microcosm studies, generally, are not accepted, except for bryophyte layer studies (which are relevant for bogs). We have focused particularly on statistically and biologically significant outcomes of field addition experiments and mesocosm studies. Wiedermann et al. (2009b) examined whether small-scale field experiments could predict the direction and magnitude of ecosystem responses to increased N supply. In order to do so, they compared data from a ten-year field experiment [involving deposition of 2 (ambient), 15 and 30 kg N ha^{-1} yr^{-1}] with field data from sites representing a gradient of increasing N deposition (2, 8 and 12 N ha^{-1} yr^{-1}). They found a highly significant correlation between the two data sets which was attributed to the key function of *Sphagnum* species that monopolise N availability and control the water balance, creating an environment hostile to vascular plants (Wiedermann et al., 2009b). These results support the use of data from small-scale experiments on the effects of N deposition on vegetation dominated by bryophytes.

Effects on bryophytes and lichen species composition

This section on species composition summarises earlier work included during the previous review of $CL_{emp}N$ in 2011 and brings in more recent experiment work from Canada and Scotland. Nitrogen has differential effects on the growth of different *Sphagnum* species that causes changes in species composition across the N deposition gradient; this has been demonstrated in several experimental studies. For example, in an early controlled-environment experiment, Risager (1998) examined the growth responses of *Sphagnum fallax* at low N addition rates (0, 5, 10 and 20 kg N ha^{-1} yr^{-1}). Growth was significantly stimulated by the application of N (especially with 5 and 10 kg NH_4-N ha^{-1} yr^{-1}), but because of the growth dilution effect, tissue N concentration did not change. Risager (1998) also investigated the responses of *Sphagnum magellanicum* after addition of N in a comparable study involving similar levels of N addition, *Sphagnum magellanicum* showed no increase in height, but addition of N decreased the production density of capitula. In contrast to *Sphagnum fallax*, the tissue N concentration of *Sphagnum magellanicum* increased with increasing additions of N. In both cases, uptake of NO_3- was considerably lower than of NH_4+. Risager (1998) concluded that the form of N is also important (see later in this chapter) in species response, and that increased N availability may cause shifts in species composition in favour of *Sphagnum fallax* at higher N deposition levels. This expansion of *Sphagnum fallax* under high N deposition rates was studied in a field experiment at sites with low and high background deposition of N (Limpens et al*.,* 2003c). At the low N deposition site *Sphagnum fallax* area expanded when extra N (40 kg ha^{-1} yr^{-1}) was applied. At the high N deposition sites such expansion was limited by P. The authors concluded that *Sphagnum fallax* will gradually colonise an increasing number of new habitats in areas with low, albeit increasing, N deposition, but may only grow to dominate when P supply is adequate (Limpens et al., 2003c).

Clear effects of N eutrophication have also been observed in Dutch ombrotrophic bogs. The composition of the moss layer in small remnants of formerly large bog areas has changed markedly as N loads increased to between 20 and 40 kg N ha^{-1} yr^{-1}, especially in the form of ammonium/ammonia; the most characteristic *Sphagnum* species were replaced by more nitrophilous moss species (Greven, 1992). These dramatic changes in species composition were also observed in British bogs (e.g. Lee and Studholme, 1992); many characteristic *Sphagnum* species were eradicated from affected ombrotrophic bog areas, such as those in the South Pennines in England, where atmospheric N deposition increased to over 30 kg N ha^{-1} yr^{-1} following many decades of elevated sulphur deposition.

More recent experimental work at the low end of the N range (bulk [wet] deposition < 2 kg N ha^{-1} yr^{-1}) in Canada observed responses in different species to N additions of 5, 10, 15 and 20 kg N ha^{-1} yr^{-1} over five years. Initially, *Sphagnum fuscum* growth responded positively to N, then declined. It also decreased in abundance whereas *S. magellanicum* increased. No effects were observed on *S. angustifolium*. The authors recommended a $CL_{emp}N$ of 3 kg N ha^{-1} yr^{-1} (Wieder et al., 2019).

Bog micro-habitat may also be important in determining species responses. A two-year German N addition experiment showed such effects on species composition in an established *Sphagnum* community (Lütke Twenhöven, 1992) in the field. In bog hollows, *Sphagnum fallax* was significantly promoted by the addition of both nitrate and ammonium (10 kg N ha^{-1} yr^{-1} with an estimated ambient atmospheric deposition of 5 kg N ha^{-1} yr^{-1}), but less so on the bog lawns. This resulted in *Sphagnum fallax* outcompeting *Sphagnum magellanicum* in the hollows and, when water supply was sufficient, also on the lawns. However, on the hummocks in the bog, nitrate and, to a lesser extent, ammonium reduced the growth of both species. Results from Gunnarsson and Rydin (2000) also suggest that lawn communities are less vulnerable to increased N deposition than hummock communities. Because of the differences in vegetation structure, the rate of supply of N to a hummock community dominated by dwarf shrubs is about 40% greater than to a lawn community (Bobbink et al., 1992; Malmer and Wallén, 1999).

Sphagnum capillifolium is often associated with good quality bogs and its vitality has been observed to suffer at 16 kg N ha^{-1} yr^{-1} (background was 8 kg N ha^{-1} yr^{-1}) at Whim Moss, Scotland (differential responses of N form were observed - see section Effect of nitrogen form) (Sheppard et al., 2011). Declines in lichens were also observed at 16 kg N ha^{-1} yr^{-1} (Phoenix et al., 2012), and the pleurocarpous bryophyte *Pleurozium schreberi* declined from 16 kg N ha^{-1} yr^{-1} and was eradicated by 64 kg N ha^{-1} yr^{-1} (Sheppard et al., 2014) whilst in contrast, *Hypnum jutlandicum* increased at 16, 32 and 64 N ha^{-1} yr^{-1} to oxidised N addition.

The importance of competition between moss species is also indicated by the earlier study of Mitchell et al. (2002) using higher rates of N deposition. This experiment examined the effect of an addition of 30 kg N ha^{-1} yr^{-1} in a cutover bog in the Jura Mountains of Switzerland, where ambient deposition was estimated to be around 15 kg N ha^{-1} yr^{-1}. The normal pattern of succession in the restoration of these sites is that keystone species such as the moss *Polytrichum strictum* create favourable micro- environments for the establishment of *Sphagnum fallax* and hence of typical bog vegetation. However, three years of study showed that cover and density of *Polytrichum* strictum was strongly increased while cover and density of *Sphagnum fallax* decreased; *Polytrichum* was observed to overgrow the *Sphagnum* which might prevent the typical regeneration process in central European bogs. Thus, in the study period, biomass production of *P. strictum* increased by almost 75% in response to N addition, whereas production of *Sphagnum fallax* decreased by close to 50% (Mitchell et al., 2002).

Effects on vascular plants and community composition

This section synthesises information on N effects on vascular plant growth and species composition, it builds on work included in the previous reviews of critical loads by incorporating evidence from N addition experiments in Canada and Scotland. A Danish survey of national ombrotrophic bogs showed a decline in the original bog vegetation together with an increase in more N-demanding grass species (such as *Molinia caerulea* and *Deschampsia flexuosa*) and trees (such as *Betula pubescens*) in areas with wet ammonium (NH_4^+) deposition loads of more than 10 to 15 kg N ha^{-1} yr^{-1} (Aaby, 1994), together with increased NH_3 concentrations, rapidly deposited to ombrotrophic mires. For a bog in southern Sweden, in a region with an ambient N deposition of 7 to 9 kg ha^{-1} yr^{-1}, Gunnarsson et al. (2002) showed that the total number of species (vascular plants + mosses) per plot did not change much between 1954 and 1997. However, there were large changes in species composition that were indicative particularly of a drier mire surface and an increased availability of N. The increased growth of trees may also have triggered further changes in plant cover.

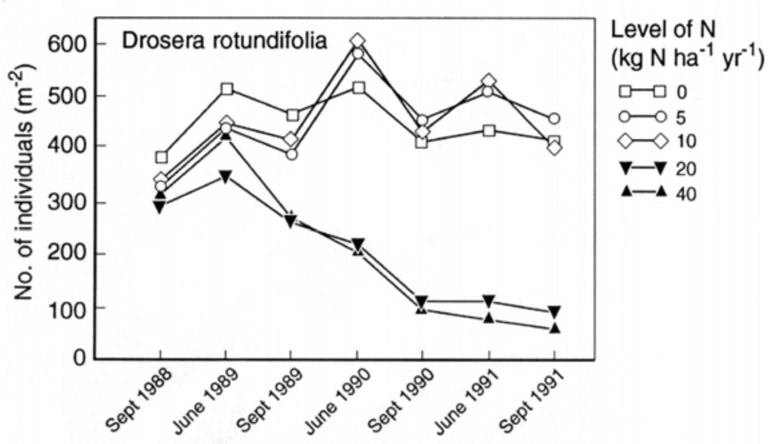
Responses from other gradient surveys are discussed later in this chapter, however, such observations highlight that increased N deposition may influence the competitive relationships between mosses and vascular plants in nutrient-deficient vegetation such as bogs. For mineral nutrients, interactions between vascular plants and mosses are partly unbalanced as *Sphagnum* mosses rely only on atmospheric supply while vascular plants also rely on mineralisation (Malmer et al., 1994). Thus, in a field experiment in southern Sweden (ambient N deposition rate 7-9 kg ha^{-1} yr^{-1}) a supply of both N and P (20 and 4 kg ha^{-1} yr^{-1}, respectively) only affected the growth of the mosses if N and P were applied on the surface, and only affected the vascular plants if applied below the moss surface.

To determine the N available to quantitatively important boreal bog plants, Nordbakken et al. (2003) measured the $\delta^{15}N$ signature after a three-year N addition experiment in a Norwegian bog with an estimated total N background deposition of 7.9 kg N ha^{-1} yr^{-1}. An addition of only 5 kg N ha^{-1} yr^{-1} was sufficient to significantly increase the N concentration in *Sphagnum* mosses, liverworts and shallow rooted vascular plants. An addition of 40 kg N ha^{-1} yr^{-1}, however, was not sufficient to increase the N concentration in deep rooted plants even after three years of N addition. Over the course of three years, the *Sphagnum* layer was able to completely immobilise the relatively high N load of 40 kg N ha^{-1} yr^{-1} and is therefore, likely to have significant effects on the N availability to vascular plants. However, an earlier synthesis of *Sphagnum* tissue N concentrations across a number of studies covering a gradient of N deposition, suggested saturation and failure of the 'natural nitrogen filter' at above 20 kg N ha^{-1} yr^{-1} (Lamers et al., 2000). This highlights the importance of long-term experiments in reproducing real- world effects and that experiments of short durations at higher N loads may not produce effects observed in the real world over a longer period at lower N. N accumulation and saturation are therefore important.

These responses were also demonstrated experimentally by Heijmans et al. (2001), who studied the effect of added N deposition (50 kg N ha^{-1} yr^{-1}) at an ambient atmospheric deposition of the same magnitude, on peat monoliths taken from a mire in the northern Netherlands. The N:P ratio in the mosses indicated P limitation (24:1), which corresponded with the observed lack in growth response to N addition. The mosses were still able to capture a large part of the deposited N (Heijmans et al., 2002a) and three years after the start of the experiment, all species showed increased N concentrations. The mosses showed decreased height increment, but no changes in dry matter production, indicating an effect on moss morphology (Heijmans et al., 2001). The fertilised mesocosms showed a significantly higher N uptake by deep-rooting vascular plants (based on 15N enrichment; Heijmans et al., 2002a), but only the growth, in terms of cover area, of *Vaccinium oxycoccus* had increased significantly. There was a negative relationship between litter (also increased by N) and vascular plant cover, on the one hand, and *Sphagnum* species, on the other hand (Heijmans et al., 2001), suggesting that *Sphagnum* growth might also be limited by increased shading.

Sphagnum mosses are not the only plants that encounter negative effects from N deposition due to changes in competitive interactions with vascular plants. The effects of the supply of extra N on the population ecology of *Drosera rotundifolia* were studied in a four-year fertilisation experiment in Swedish ombrotrophic bogs, using a range of deposition rates (Redbo-Torstensson, 1994). It was demonstrated that experimental applications of 20 kg N ha^{-1} yr^{-1} (as NH_4NO_3 above an ambient deposition of 5 kg N ha^{-1} yr^{-1}) significantly reduced the survivorship of the plants after one year, and also negatively affected flowering after two years (Figure 6.3). The N driven decrease in the population density of the characteristic carnivorous bog species *Drosera* was associated with increased density of tall species such as *Eriophorum* and *Andromeda*, which resulted in increased competition for light and further reduction in *Drosera*.

Figure 6.3. Numbers of individuals of *Drosera rotundifolia* in an ombrotrophic raised bog (D1) near Stockholm (Sweden) during three years of N additions.



Source: Redbo-Torstensson, 1994

Limpens et al. (2004) conducted a three-year N fertilisation experiment (+40 kg ha^{-1} yr^{-1}) at six sites; one with moderate N deposition and five with high N deposition. Adding N increased the concentration of inorganic N in the rhizosphere at the site with moderate deposition and at two of the sites with high deposition. The addition of N depressed *Sphagnum* height increment but shading by vascular plants was of minor importance in explaining the negative effects of N on *Sphagnum*. P alleviates the negative impact that N has on *Sphagnum* by enhancing its capability to assimilate the deposited N. P availability is therefore a major factor determining the impact of deposition on *Sphagnum* production and, thus, on carbon sequestration in bogs (Limpens et al., 2004).

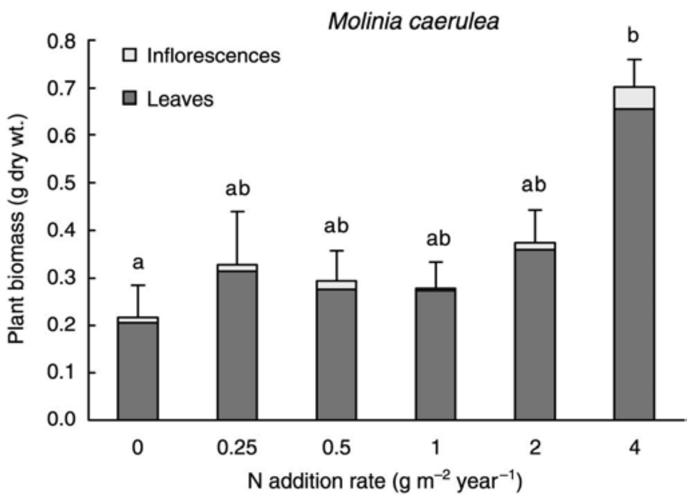
Gerdol et al. (2007) studied the effect of N addition over a period of four years in an Italian bog. *Sphagnum* production was depressed by high levels (30 kg ha^{-1} yr^{-1}) of N addition, but not at an intermediate level of 10 kg ha^{-1} yr^{-1}. Vascular plant cover increased at the expense of *Sphagnum* mosses, but this was probably triggered by an exceptional heat wave in one of the summer periods (year 2003; Gerdol et al., 2008). A proportionally greater accumulation of vascular plant litter, together with an increased potential decay of *Sphagnum* litter, may result in decreased carbon fixation. Gunnarsson et al. (2008) also suggested that one of the main causes of the low carbon input rates to the peat layer was the high level of N deposition, which increased decomposition and changed the vegetation from peat-forming *Sphagnum*-dominance to dominance by dwarf shrubs and graminoids.

There is information about the effects of N addition on species composition at high levels of deposition. In a greenhouse competition experiment between two graminoid species, *Eriophorum vaginatum* and *Rhynchospora alba*, and two ericoid species, *Calluna vulgaris* and *Vaccinium oxycoccos*, Kool and Heijmans (2009) found that at a high N supply (50 kg ha^{-1} yr^{-1}), ericoid species responded more strongly than graminoids. This suggests that under increased N availability, bogs could potentially turn into dwarf-shrub-dominated ecosystems than into grasslands. However, the balance between the functional groups is likely to be also mediated by hydrology, with *Calluna* favouring drier sites and sedges, wetter areas - in another greenhouse study, Heijmans et al. (2002b) found that *Rhynchospora alba* became the dominant vascular plant species at a high N deposition of 50 kg ha^{-1} yr^{-1}.

Experimental work at Whim Moss, Scotland, showed increases of *Eriophorum vaginatum* at high levels of N (64 kg N ha^{-1} yr^{-1}) at the expense of *Calluna vulgaris*. In the early stages of the experiment, other shrubs including *Erica tetralix*, *Vaccinium myrtillus* and *Empetrum nigrum* all responded positively to N as a result of the *Calluna* canopy opening (Phoenix et al., 2012), although after seven years the positive increase in *E. tetralix* had subsided and other ericoids had declined (Sheppard et al., 2014). The nitrophilous fern *Dryopteris dilatata* became invasive at a high rate of 64 kg N ha^{-1} yr^{-1} (Sheppard et al., 2011). N form effects were observed and are discussed in section Effect of nitrogen form. The potential negative effects of N to *Erica tetralix* were also observed in Danish wet heath where in these drier conditions, *Calluna* did well (Strandberg et al., 2012). Acidification due to N was determined as the cause.

In Dutch and Danish bogs, the first signs of the negative effect of high atmospheric N deposition of around 40 kg were invasions of the graminoid species *Molinia caerulea* and tree species such as *Betula pubescens* and *Pinus* species (e.g. Aaby, 1994; Tomassen et al., 2002). In order to confirm the hypothesis that high atmospheric N loads had triggered the observed vegetation changes, Tomassen et al. (2003) studied the effects of N on *Molinia caerulea* and *Betula pubescens* in a three-year N addition laboratory experiment. *Betula pubescens* and *Molinia caerulea* plants were introduced into transplanted turfs from an ombrotrophic floating bog in the Netherlands, and were treated with various N deposition rates. After three years, above-ground biomass of *Molinia caerulea* plants was significantly higher in the turfs that received 40 kg N ha^{-1} yr^{-1} (Figure 6.4). *Betula pubescens* was unable to increase its above-ground biomass, probably due to P limitation.

Figure 6.4. Individual above-ground biomass of *Molinia caerulea* after three years at different rates of experimental N addition (means +1 SE; n = 4). Different letters indicate significant differences (P < 0.05) between N treatments (one-way ANOVA). Discussion in text has converted to kg ha^{-1} by multiplying g m^{-2} by 10.



Source: Tomassen et al., 2003

In addition, Tomassen et al. (2004) studied the effects of N on *Molinia caerulea* and *Betula pubescens* in a three-year N addition experiment in an Irish raised bog. The water table at the experimental site had been drastically reduced by peat cutting activities in the past. Tomassen et al. (2004) concluded that the invasion of *Molinia* and *Betula* in bogs is likely to be less affected by desiccation than by increased N availability. *Molinia* is known to be well adapted to P-limiting conditions (Ellenberg, 1988; Kirkham, 2001), which may explain its success in regions with increased N deposition.

Limpens et al. (2003a) examined the effects of N deposition on the competition between shrubs and mosses and the establishment and growth of the invasive *Betula pubescens* and *Molinia caerulea* in intact bog vegetation removed from a site subject to 40 kg N ha^{-1} yr^{-1}. *Molinia* biomass was positively related to the inorganic N concentration in the interstitial water which was positively affected by N additions. N deposition increased the N availability to vascular plants in the rhizosphere, thus encouraging vascular plant growth. Water-table level and availability of P were found to be important in explaining species-specific responses to N deposition (Limpens et al., 2003a).

Shifts in species composition have also been observed much lower down the N range (bulk deposition < 2 kg N ha^{-1} yr^{-1}) at a bog N addition experiment in Alberta, Canada, where increased N above background produced an increase in shrub cover and vascular plants in general (Wieder et al. 2019). The authors recommended a $CL_{emp}N$ of 3 kg N ha^{-1} yr^{-1} and commented that responses above background N had no clear threshold but were linear across the N deposition range, however, it is important to note that, depending on the relative contribution of dry N deposition, bulk deposition may underestimate total N deposition, and this would mean their critical load should be slightly higher.

Whilst peatlands in Alberta receive relatively low N deposition compared to most of Europe, bogs in northern and western Europe in Finland, Norway, Sweden, Ireland, and areas of the UK are at these low levels of N.

Effect of nitrogen form

Recently, several studies have been focussed on the effects of different forms of N on bog vegetation. In most fertilisation experiments N is added as NH_4NO_3, reflecting the current ratio of reduced and oxidised N in precipitation (e.g. Boxman et al., 2008), but a number of experiments have studied the effects of ammonium (NH_4Cl) and nitrate ($NaNO_3$), separately. As discussed earlier, the uptake of nitrate by *Sphagnum* mosses is much lower than the uptake of ammonium (Risager, 1998; Phuyal et al., 2008; Wiedermann et al., 2009a), something that could lead to different effects on vegetation.

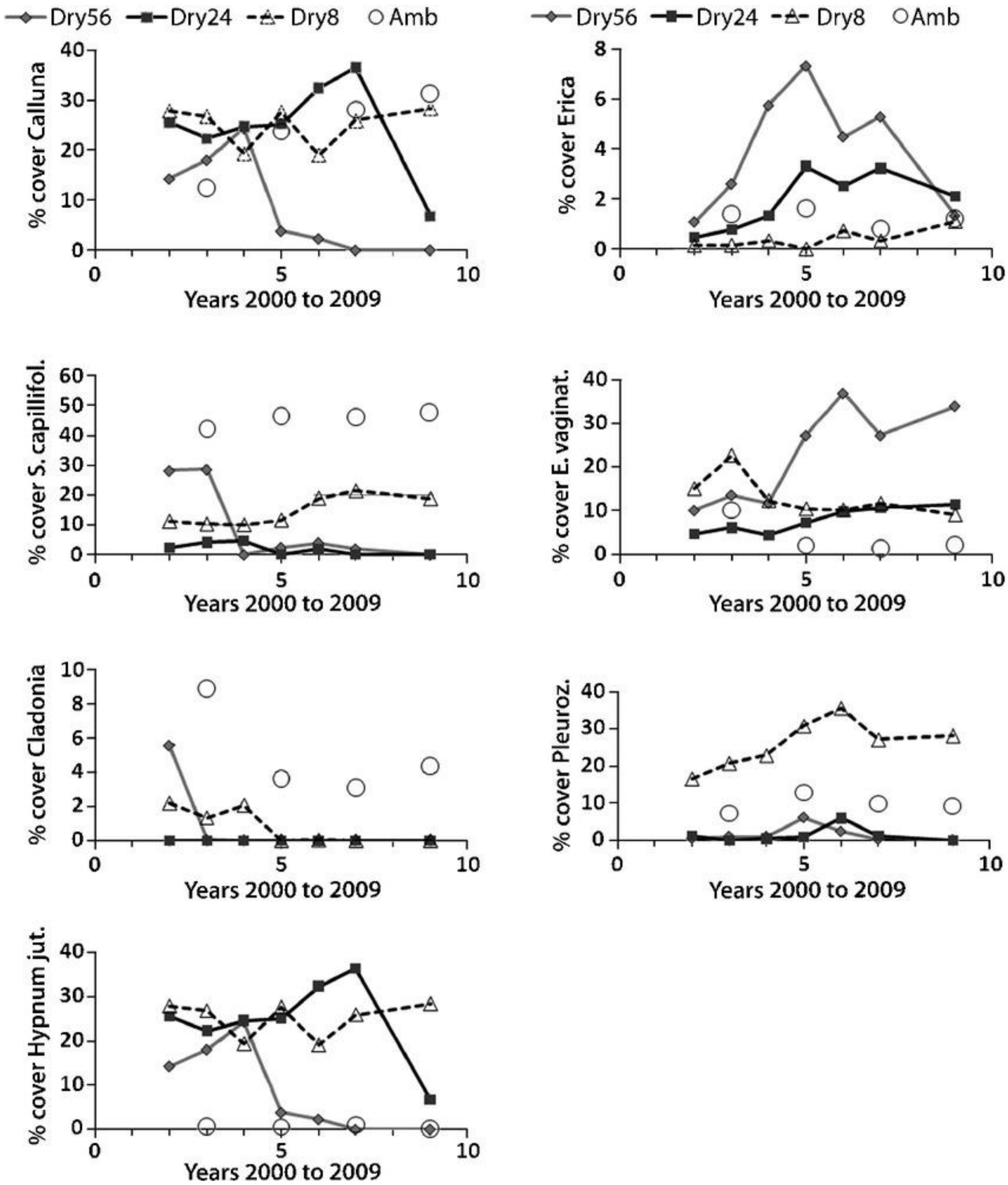
In an early controlled-environment experiment, Risager (1998) examined the growth responses of *Sphagnum fallax* to different forms of N (NO_3^-, NH_4^+ or NH_4NO_3) at low addition rates (0, 5, 10 and 20 kg N ha^{-1} yr^{-1}). This growth was significantly stimulated by the application of reduced N only, especially with 5 and 10 kg NH_4-N ha^{-1} yr^{-1}.

Differential evidence of N uptake is also evident outside of manipulation experiments. Bragazza et al. (2005) determined the $\delta^{15}N$ isotopic signatures of *Sphagnum* plants collected from sites with an ambient N deposition ranging from 2 to 20 kg N ha^{-1} yr^{-1}. The $\delta^{15}N$ isotopic signatures were found to be more related to the ratio of reduced to oxidised N forms in atmospheric deposition than to the total amount of atmospheric N deposition, indicating that $\delta^{15}N$ signatures can be used as an integrated measure of $\delta^{15}N$ signature of atmospheric precipitation. Bogs located in areas dominated by NH_3 emissions had *Sphagnum* plants with a more negative $\delta^{15}N$ signature, compared to areas dominated by NO_x emissions (Bragazza et al., 2005).

At Whim Moss, Scotland, a large-scale, automated N addition field experiment with different forms of N (ammonium and nitrate) at 8 (background), 16 and 64 kg N ha^{-1} yr^{-1} was started in 2002 (Sheppard et al., 2004). Within one or two years of increased N input, branching and the height of photosynthetically active material of *Sphagnum capillifolium* were reduced. The effect was more pronounced when the water table was low, but responses did not differ between oxidised and reduced forms of N addition (Carfrae et al., 2007). The experiment also contains a gaseous ammonia (NH_3) release to mimic an intensive farm where rapid responses in vegetation were observed following the start of the experiment including the total loss of *Calluna vulgaris*, *Sphagnum capillifolium* and *Cladonia portentosa* at a high 56 kg N ha^{-1} yr^{-1} dry NH_3 deposition (calculated from NH_3 concentration data) on top of ambient deposition of 8-11 kg N ha^{-1} yr^{-1} (Figure 6.5, Sheppard et al., 2011). Visible injury was observed in *Calluna vulgaris* at 17 kg N ha^{-1}

yr^{-1} and *Sphagnum capillifolium* at 22 kg N ha^{-1} yr^{-1} in the dry NH_3 treatment (not shown), with *Sphagnum capillifolium* vitality reducing in the wet plots (N applied in solution as either $NaNO_3$ or NH_4Cl) at 16 kg N ha^{-1} yr^{-1} (Figure 6.6, Sheppard et al., 2011). The N load at which the responses were observed declined over time. Wet N did not have a detrimental effect on *Calluna* cover (Sheppard et al., 2013).

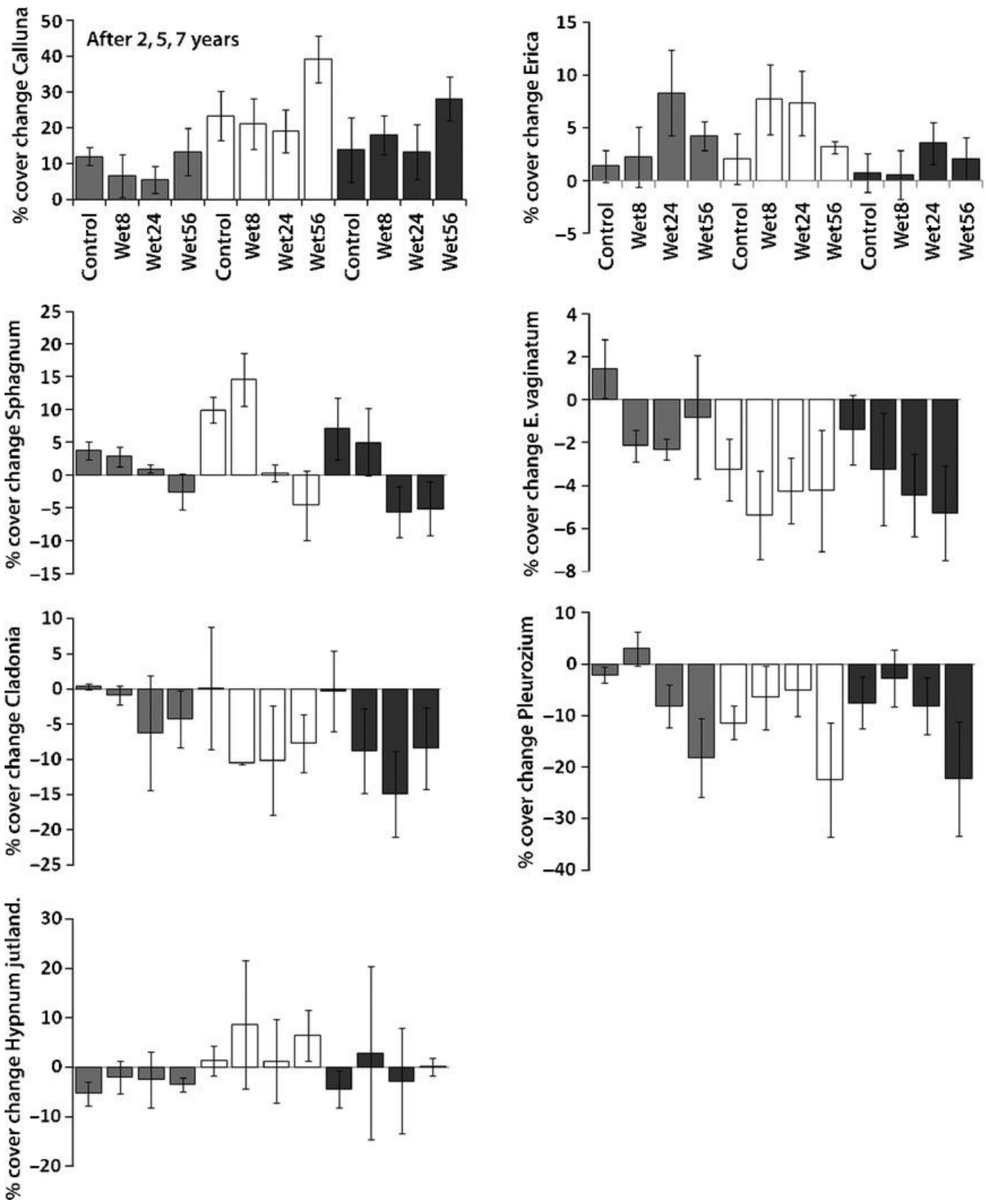
Figure 6.5. Chronological percent cover of *Calluna*, *Erica tetralix*, *Sphagnum capillifolium*, *Eriophorum vaginatum*, *Cladonia portentosa*, *Pleurozium schreberi* and *Hypnum jutlandicum* in permanent quadrats receiving 8, 24 and 56 kg N ha^{-1} yr^{-1} dry N deposition and ambient (assessed from Year 1, 2003) from the start (2002) through to 2009.



Source: Sheppard et al., 2011

In pleurocarpous bryophytes, *Pleurozium schreberi* was eradicated at high doses of 56 kg N ha^{-1} yr^{-1} in the wet-reduced N plots, whereas *Hypnum jutlandicum* increased with oxidised N at +8 and +56 kg N ha^{-1} yr^{-1} (Figure 6.6, Sheppard et al., 2011). The lichen *Cladonia portentosa* was also strongly responsive to dry NH_3, declining rapidly close to the ammonia source (~56 kg N ha^{-1} yr^{-1}) in the first year of the experiment and disappearing entirely up to 48 m away from the source (~16 kg N ha^{-1} yr^{-1}) after four years, with damage at lower N levels emerging (Sheppard et al., 2011). *Cladonia* also decreased in the wet-N plots but much more slowly (Figure 6.6). Responses in lichen proteomics as discussed earlier also exhibit differential responses to N form, although the direction of change is inconsistent (Munzi et al., 2017; 2020).

Figure 6.6. Changes (difference from start) in *Calluna*, *Erica tetralix*, *Sphagnum capillifolium*, *Eriophorum vaginatum*, *Cladonia portentosa*, *Pleurozium schreberi* and *Hypnum jutlandicum* in permanent quadrats in control and treatments receiving 8, 24 and 56 kg N ha^{-1} yr^{-1} wet N deposition, pre-treatment and repeated after 2 (grey), 5 (open) and 7 (black) years, ± SEM.



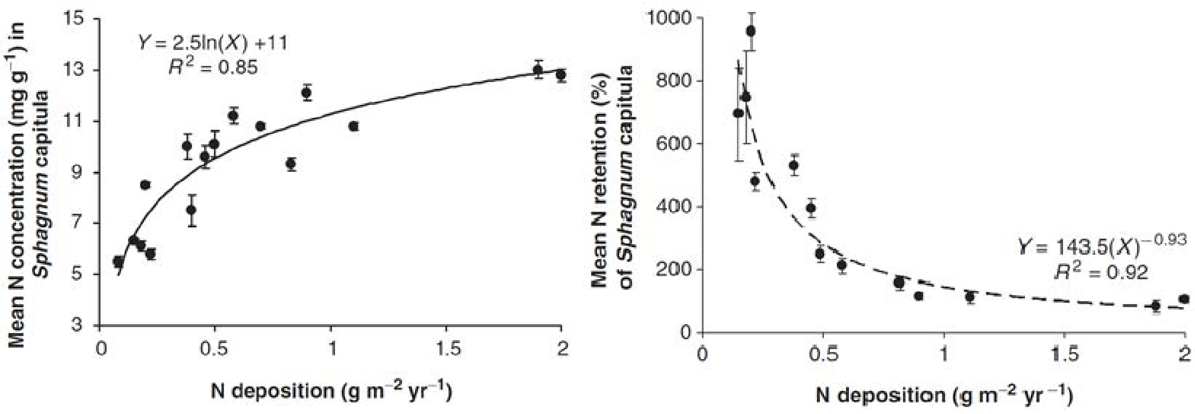
Source: Sheppard et al., 2011

Consistently, the responses to reduced N, especially gaseous, appear more acute than responses to oxidised N, particularly in non-vascular plants (Sheppard et al., 2014). The authors recommend the lowest value of the $CL_{emp}N$ range where vegetation communities include sensitive lower plants and where deposition is dominated by reduced N (Sheppard et al., 2014). However, the authors highlight that cumulative effects of N are important too, with oxidised N appearing to drive a greater response at low N and reduced N at higher N loads. The accumulation of the ammonium cation in soil in contrast to the mobility of the anion nitrate was suggested as the driving mechanism behind this response (Sheppard et al., 2014). Response of soil processes and leaching to N forms are incorporated into the following section.

Effects on *Sphagnum*, peat and peat water chemistry as evidence of nitrogen saturation

Along a natural gradient of bulk atmospheric N deposition varying from 2 to 20 kg N ha^{-1} yr^{-1}, Bragazza and Limpens (2004) found that concentrations of dissolved inorganic N (DIN) and dissolved organic N (DON) in pore water increased with N deposition. The increase in concentrations of DIN was related to the reduced capacity of the moss layer to trap atmospheric N, which in turn was a result of the moss layer's N saturation. The increased concentrations of DON appeared to be the result of increased leaching of organic compounds by *Sphagnum* (Bragazza and Limpens, 2004). Survey work demonstrated increasing tissue N concentrations as N deposition increased which saturated at low N loads (Bragazza et al., 2005), with the percentage of N retained falling rapidly from around 2 kg N ha^{-1} yr^{-1} (Figure 6.7). This in turn led to elevated inorganic N in pore waters which increased exponentially at N deposition loads lower than the top of the current $CL_{emp}N$ range of 5-10 kg N ha^{-1} (Bragazza et al., 2005).

Figure 6.7 a) Trend of mean (± SE) nitrogen (N) concentration in *Sphagnum* capitula along the gradient of atmospheric N and b) Mean (± SE) N retention by *Sphagnum* capitula along the gradient in atmospheric N deposition. To convert g m^{-2} to kg ha^{-1} multiply by 10. N retention was calculated as the ratio between capitulum N concentration per square metre and atmospheric N deposition.

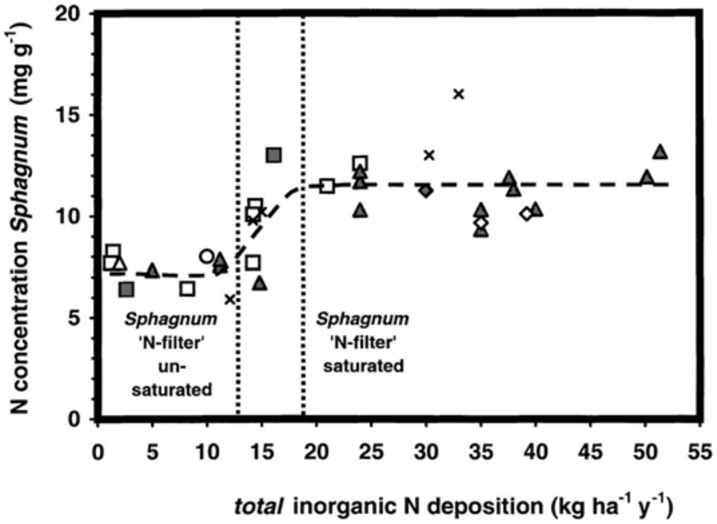


Source: Bragazza et al., 2005

Lamers et al. (2000) had previously used field data from Ireland and the Netherlands to produce an assessment of the capacity of moss layers to filter the amount of N deposition reaching the rhizosphere, thereby decreasing the growth of competitive graminoids and other species (Figure 6.8). The estimates were based on field data on *Sphagnum* species from sites covering a range of N- deposition values. At a deposition level of below 10 kg N ha^{-1} yr^{-1}, Lamers et al. (2000) suggest that growth stimulation may absorb additional N inputs, while between 10 and 20 kg ha^{-1} yr^{-1} an increase in N content of *Sphagnum* species, primarily from accumulation of amino acids, might assimilate further increase in N deposition.

Above 20 kg ha^{-1} yr^{-1}, the 'natural nitrogen filter' of *Sphagnum* is suggested to fail completely, as was observed in regions with high deposition levels (Lamers et al., 2000). It is important to note that Lamers et al. (2000) estimated the total deposition to be double the measured bulk deposition, however, at the remote sites with low deposition levels, where dry deposition is likely to be very low, their method would significantly overestimate the actual deposition inputs; according to this figure, this would thus reduce the threshold N deposition rate for response as illustrated by Braggaza et al. (2005).

Figure 6.8. The N concentration (mg g dry weight^{-1}) in raised bog (D1) *Sphagnum* species (apical parts) in Europe and the United States, in relation to total atmospheric N inputs (estimated at twice the wet deposition). Data taken from literature (◆ Ferguson et al., 1984; □ Malmer, 1988; ■ Aerts et al., 1992; ⬨ Lütke Twenhöven, 1992; ○ Van der Molen, 1992; × Pitcairn et al., 1995; Δ Johnson and Maly, 1998); ▲ collected by the authors in 1998 in Europe and the United States).



Data source: Lamers et al., 2000

The significance of the N saturation of the *Sphagnum* layer, and increased availability of N in peat and in peat waters, has also been investigated in several experimental studies. Tomassen et al. (2003) found that peat water ammonium concentrations had significantly increased after three years of N addition (40 kg N ha^{-1} yr^{-1}). Ammonium concentrations increased to 25 µmol l^{-1} (a common level in Dutch ombrotrophic bogs); in all other N-treatment experiments, levels were between 5 and 10 µmol l^{-1} due to the very high N retention in the peat mosses. Limpens et al. (2003a) found that lowering of the N input from 40 to 0 kg ha^{-1} yr^{-1} decreased both interstitial water and *Sphagnum* N concentrations, while doubling the N input to 80 kg ha^{-1} yr^{-1}, increased N concentrations.

A study at Whim Moss, Scotland, analysed responses in the upper profile of peat following five years of experimental N additions, revealed rapid increases in soil solution N in response to N addition above a background N of 8 kg N ha^{-1} yr^{-1} (Field et al., 2013). The authors suggested that bog vegetation was not efficient at buffering N deposition and commented that N retention in the peat appeared to be very poor in comparison to organic heath soils with the site leaching N at high C:N ratios (Field et al., 2013). However, an experiment on boreal peatland at very low background N (~1.6 kg N ha^{-1} yr^{-1}) in Canada did not find increases in pore water N in response to added N over the period of the study (Wieder et al., 2019). The authors suggested that additional N was being taken up by shrubs which increased in growth at the expense of *Sphagnum*. At Whim Moss, a positive growth response in shrubs (*Erica tetralix*) was observed in the early years of the experiment but subsequently declined (Sheppard et al., 2014). Hydrology can also play a role in the balance between shrubs and peat mosses, limiting the response of the former in wet situations.

Effects on carbon and nutrient cycling and biological processes

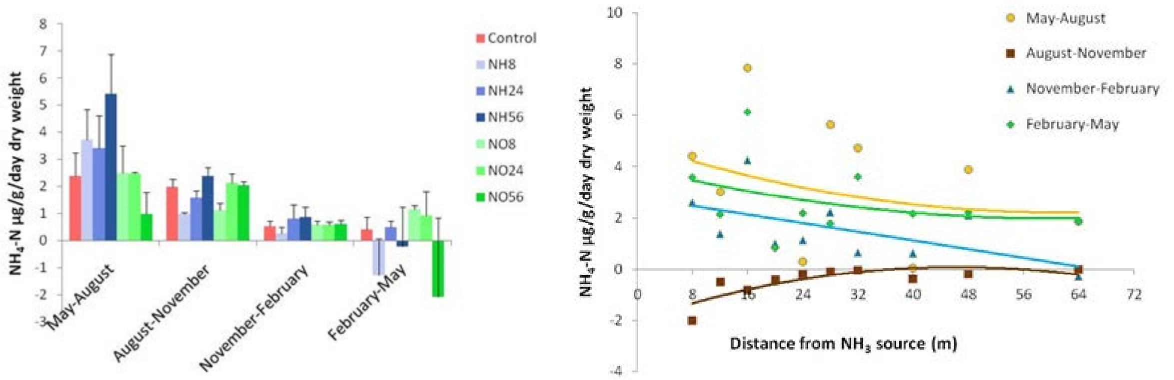
Concern over rising levels of atmospheric CO_2 has directed increasing attention towards bogs and peatlands due to their large storage capacity of carbon (Joosten, 2009; Nichols and Peteet, 2019), with the sequestration of carbon in peat determined by the ratio between primary production and decomposition of plant material, mainly bryophytes (Risager, 1998; Mitsch and Gosselink, 2000).

Increased carbon as well as N availability may increase primary production under pristine (nutrient- poor) conditions, whilst increased N will reduce the C:N ratio of litter and thus hypothetically increase peat decomposition rates. However, peat mosses (*Sphagnum* species), similar to bryophytes in other systems (e.g. grasslands, forests), have been proved to be sensitive to increasing N availability, and to react with decreased growth to high doses of N. Furthermore, the C stores within peat are themselves threatened by climate change (e.g. by increased drying of the peat layer and thus increased mineralisation).

This section of the chapter considers the effect of increasing N addition on carbon and nutrient cycling and biological processes. It builds on previous work through the addition of recent experimental work from Canada, Scotland and Sweden, and a European gradient study. Early work, based on studies on the northern and southern Swedish sites discussed previously in this chapter, concluded that a high atmospheric N supply may affect the carbon balance of ombrotrophic bogs (Aerts et al., 1992), because productivity under these circumstances is P limited, rather than N limited, but decomposition is probably increased by the high N loads. This hypothesis was supported by results from short-term experiments by Williams et al. (1999) and Williams and Silcock (2000) where N was added for three years on Moidach More, Scotland. The addition of 30 kg N ha^{-1} yr^{-1} resulted in a decrease in peat C:N ratios (Williams et al., 1999), which could increase rates of first-stage organic matter decomposition and N mineralisation (Aerts et al., 1992). Furthermore, additions of N to cores of *Sphagnum capillifolium* and *Sphagnum recurvum*, although taken up mainly by the moss, had significant effects on C and N values of the microbial biomass in the underlying peat (Williams and Silcock, 2000).

Nitrogen stimulates mineralisation across many habitats (e.g. Curtis et al., 2004, Pilkington et al., 2005; Emmet et al., 2007) including peatlands where N driven increases in mineralisation were observed at the Whim Moss experiment, Scotland (Field et al., 2010, 2013). In this study, N increased mineralisation but only in warmer periods when the water table was lower. Due to high site heterogeneity, the data were only significant in the reduced-N plots (Figure 6.9a, Field et al., 2010, 2013). The authors highlighted strong responses along the gaseous NH_3 transect over a concentration range equivalent to 8 to 56 kg N ha^{-1} yr^{-1}, with elevated mineralisation closer to the NH_3 source in all but one season (Figure 6.9b; Field, 2010) and observed that the site appeared to leach at high C:N ratios, with N being poorly retained in the soil (not shown). The authors suggested that other factors such as hydrology or P or potassium (K) availability may constrain responses.

Figure 6.9. Summer 2006 to Spring 2007 seasonal net daily mineralisation rates from the top 10 cm surface peat from Whim a) Wet N plots (reduced N addition plots in blue, oxidised N addition in green, control plots in pink) and b) dry NH_3 release transect. Ambient deposition was circa 8-11 kg N ha^{-1} yr^{-1}.



Source: Field et al., 2010

An early study of *Sphagnum* peat in Swedish ombrotrophic bogs along a gradient of N deposition (Hogg et al., 1994) also indicated that the decomposition rate of *Sphagnum* peat was more influenced by the P content of the material, than by N. This finding that P content is significant is consistent with results from a further study on the same two Swedish sites by Aerts et al. (2001), involving four years of fertilisation with 5 or 10 kg N ha^{-1} yr^{-1}. These treatments were found to have no significant effects on potential decay rates at either site, measured by taking litter formed in the first three years of the experiment and monitoring time courses of CO_2 evolution in the laboratory. There was a significant relationship between the potential rate of peat decay and nutrient concentrations in litter, but the effects of the N and P content in the litter were comparable, and these relationships were primarily governed by differences between sites rather than treatments. Moreover, Tomassen et al. (2002) determined the decay rate of peat, which had been treated with six levels of N deposition for three years (see previous section), by measuring both CH_4 and CO_2 evolution. Despite significant differences in C:N ratios, carbon mineralisation rates were not at all affected by N treatments. Tomassen et al. (2004) also found no effect of N addition on the C mineralisation, despite significant differences in C:N ratios.

However, decomposition rates of recently formed peat litter collected in nine European countries under a natural gradient of atmospheric N from ca 2 to 20 kg N ha^{-1} yr^{-1}, increased with increasing N deposition rates, resulting in higher carbon dioxide emissions and dissolved organic carbon (DON) release (Bragazza et al., 2006). Increasing N concentrations in *Sphagnum* litter, as a result of increased exogenous N availability, was found to be accompanied by a decreasing concentration of polyphenols (Bragazza and Freeman, 2007). The lower content of decay-inhibiting polyphenols could accelerate litter peat decomposition. Other studies have observed that an increase in vascular plant cover in response to N can lead to increased peat decomposition (Breeuwer et al., 2008).

In contrast, Saarnio et al. (2003) found that, over a two-year period, additions of 30 kg N ha^{-1} yr^{-1} had very little effect on the C gas exchange in lawns of boreal oligotrophic mires. To verify long-term changes in C balance, however, experiments over longer periods of time would be needed. Kivimäki et al. (2013) observed increased ecosystem respiration at Whim Moss, Scotland, but only between the control and a high +56 kg N ha^{-1} yr^{-1} treatment. Currey et al. (2010) observed enhanced mineralisation of labile carbon at Whim Moss, but either a decrease or no change in the mineralisation of complex carbon with an increasing effect of N. Effects were strongest at the highest N of +56 kg ha^{-1} yr^{-1} but no thresholds were identified. Eriksson et al. (2009) studied the effects of long-term N addition (12 years) on the production of methane. Long-term deposition of N increased methane production, which may be attributed to a shift in plant community composition. The percentage of cover of the sedge *Eriophorum vaginatum* and the dwarf shrubs *Andromeda polifolia* and *Vaccinium oxycoccos* increased in response to experimental N deposition, with a concomitant reduction in the cover of *Sphagnum* species (Wiedermann et al., 2007); *Eriophorum* species in particular have been linked to elevated methane emissions due to the presence of aerenchyma. These findings differ from other studies, which found no effects of long-term deposition of N in the field on methane production in peat samples, despite similar changes in vegetation cover (Nykänen et al., 2002; Keller et al., 2005). These studies, however, lasted for five to six years, which may not be enough to induce changes in community structure of the methanogenic population or its substrate supply (Eriksson et al., 2009).

Lund et al. (2009) conducted a short-term N fertilisation experiment (+40 kg ha^{-1} yr^{-1}) in two Swedish bogs subjected to low (2 kg ha^{-1} yr^{-1}) and high (15 kg ha^{-1} yr^{-1}) background N deposition. At the low background deposition site, after two years, both gross primary production and ecosystem respiration were already significantly increased by N addition. At the site with high N background deposition, primary production was limited by P. N addition had no effect on CH_4 exchange, but elevated N_2O emissions were detected in N-fertilised plots. This corresponds with the results from Glatzel et al. (2008), who examined the effects of atmospheric N deposition on greenhouse gas release in a restoring peat bog in north-western Germany. They found that N fertilisation did not increase decomposition of surface peat, but under high N deposition it would be important to avoid frequent water table fluctuations which may increase N_2O release. Lund et al. (2009) concluded that, in the long term, increased nutrient availability will cause changes in plant composition, which will further act to regulate peatland greenhouse gas exchange. Higher N_2O emission were associated with elevated N at Whim Moss but only in the +56 kg ha^{-1} yr^{-1} wet-oxidised N and dry-reduced N plots (Sheppard et al., 2013); in the wet-reduced N plots, a lower pH was suggested to inhibit denitrification. The authors here also highlighted the poor N-retention of the system, linking it to P or K limitation; growth responses in *Sphagnum capillifolium* when P/K was added were previously observed at the site (Carfrae et al., 2007). The potential of increase methane release was suggested following an 18-year fertilisation and warming experiment in a Swedish oligotrophic peatland. Marti et al. (2019) studied the response of the methanogen community to an N addition of 30 kg ha^{-1} yr^{-1} (above ambient deposition of around 2 kg N ha^{-1} yr^{-1}) and simultaneous warming, they found that N amplified the effects of warming with an increase in the abundance of methanogens.

Whilst much of this work has occurred at N deposition loads at or above the upper end of the existing $CL_{emp}N$ range, experimental work in Canada at a low background N < 2 kg ha^{-1} yr^{-1}, found that biological nitrogen fixation (BNF) was progressively inhibited at N deposition > 3 kg (Wieder et al., 2019). The study also observed stimulation of cellulose decomposition by N but no response in mineralisation rates. Similar N mediated declines in BNF were observed in a comparative study of 4 bogs across Europe spanning a N range of 2-27 kg ha^{-1} yr^{-1}; BNF at the second lowest N site at 6 kg ha^{-1} yr^{-1} was lower than BNF at 2 kg ha^{-1} yr^{-1} (Saiz et al. 2021). Whilst the number of sites in this study were low, replication was high (39 at each over two years) and a simultaneous experiment which added +15 kg N ha^{-1} yr^{-1} found similar responses in BNF, demonstrating the link with N (Saiz et al. 2021). Hinting at an underlying mechanism and demonstrating a link with ecosystem functioning, a survey of 25 European bogs over a 7 to 30 kg N range found linear declines in the diversity and community structure of ericoid mycorrhizal fungi (EMF) in response to increasing N deposition (Van Geel et al., 2020). The authors also linked soil P with a reduction in EMF richness.

In summary, these results do indicate that effects of elevated N deposition on *Sphagnum* and shrub growth, litter chemistry, and on the microbial community microbial biomass, are likely to affect the decomposition processes and ecosystem functioning, but that this process is regulated by more than just the C:N ratio of the peat with hydrology, climate and the availability of other nutrients being key. However, further evidence is still necessary to properly evaluate the long-term effects of increased N supply on the decomposition of *Sphagnum* peat, rates of nutrient cycling and the stability of carbon stored within peat.

Observed responses in vegetation composition from real-world gradient surveys

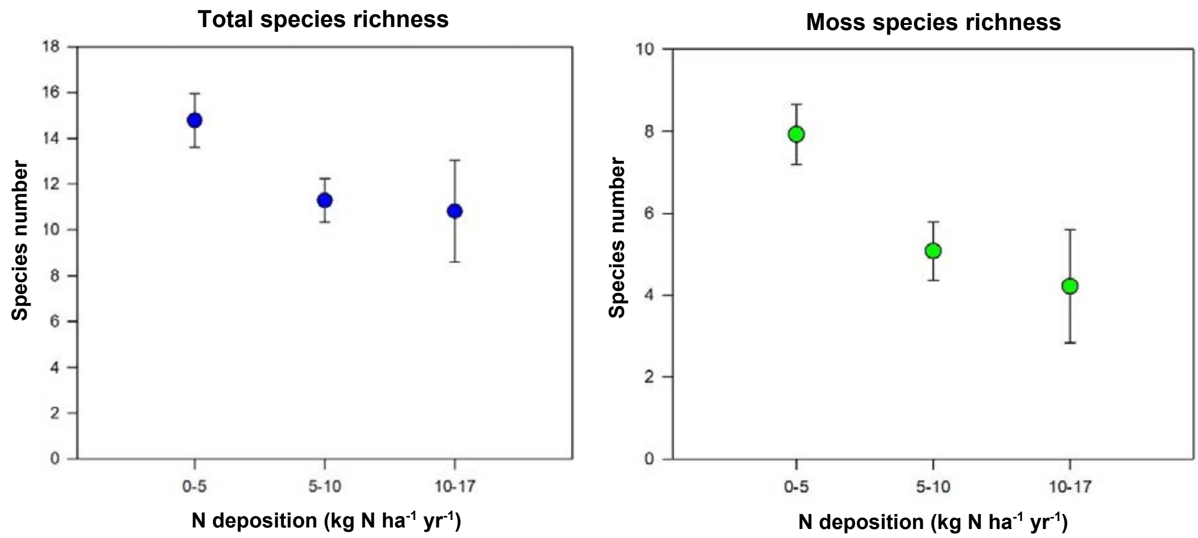
Since Bobbink and Hettelingh (2011), published gradient studies have provided strong, field-scale evidence of links between N deposition (in both oxidised and reduced forms) and widespread changes in the composition and diversity of plant species in bogs (Q1). Many of these studies have used vegetation data from national-scale surveys to assess changes in species richness and plant community composition along a N deposition gradient (Stevens et al., 2011; Tipping et al., 2013; Wilkins et al., 2016; Aherne et al., 2021). A number of these studies have attempted to identify an N deposition threshold where change occurs (Tipping et al., 2013; Wilkins et al., 2016; Aherne et al., 2021).

Further, these gradient studies typically include regions with low N deposition (e.g. 5 kg N ha^{-1} yr^{-1}) allowing for the assessment of impacts at lower N inputs compared with experimental additions.

A number of broad-scale gradient studies have assessed the impacts of N deposition on bogs in the UK (Stevens et al., 2011; Tipping et al., 2013; Field et al., 2014; Van den Berg et al., 2016; using total N deposition, CBED model at 5 × 5 km resolution). Field et al. (2014) assessed species richness and plant community composition along gradients of climate and pollution for five habitats including bogs (n = 29) with N deposition ranging from 5.9 to 30.9 kg N ha^{-1} yr^{-1}. They found linear reductions species richness and changed species composition associated with higher N deposition along the N gradient studied. Furthermore, N deposition was the dominant correlate to species richness of lichens (as a negative relationship). Species richness declined by about 20% of maximum species richness from the lowest to the highest N deposition bog sites; 80% of maximum species richness occurred at 7.1 kg N ha^{-1} yr^{-1}. Similarly, Van den Berg et al. (2016) assessed species richness of vascular plants as a measure of biodiversity in bogs and mires (n = 1136 quadrats) from the 2007 UK Countryside Survey; total N deposition ranged from 5.1 to 54.2 kg N ha^{-1} yr^{-1}. Their results provide clear evidence that N deposition affects species richness, after factoring out correlated explanatory variables (climate and sulphur deposition). The observed decline in species richness was accompanied by an increased grass:forb ratio. Approximately 80% of species richness occurred at 11 kg N ha^{-1} yr^{-1}. Stevens et al. (2012) analysed the probability of presence of individual lichen taxa (n = 6) in bogs at a given level of N deposition together with driver data for climate, change in sulphur deposition, and land-use using generalised additive models. Two taxa showed a significant relationship with N deposition, with *Cladonia portentosa* showing a steep reduction in the chance of occurrence from 10 to 25 kg N ha^{-1} yr^{-1}. Tipping et al. (2013) used a broken stick median regression to estimate the thresholds above which N deposition definitely had an effect on plant species richness in bogs (n = 203) from the 1998 UK Countryside Survey under an N deposition gradient of 5.3 to 40 kg N ha^{-1} yr^{-1}. The average relative loss of species in bogs was 1.7% per kg N ha^{-1} yr^{-1}. The threshold N deposition for declines in species richness was estimated to be 14.3 kg N ha^{-1} yr^{-1} with a 95% confidence interval range of 13.2 to 15.9 kg N ha^{-1} yr^{-1}, suggesting a conservative threshold of 13.2 kg N ha^{-1} yr^{-1}.

Outside of the UK, Jokerud (2012) evaluated the possible impact of N deposition on ombrotrophic mire vegetation (species richness and composition) in Western Norway. Linear multiple regression with backward selection was used to assess which environmental gradient best explained species richness along a gradient between the northern and southern study areas (20 sites with N deposition ranging from 2.7 to 17.5 kg N ha^{-1} yr^{-1}; observation-based total N deposition at 50 × 50 km resolution). Latitude showed a strong correlation with total species richness, vascular plant species richness and bryophyte species richness. Nitrogen deposition was the only variable that explained the significant decrease in total species richness and bryophyte species richness southwards. However, vascular plant species richness showed only a weak relationship with nitrogen. There was a significant decrease in average total species richness between the two deposition ranges 0-5 kg N ha^{-1} yr^{-1} and 5-10 kg N ha^{-1} yr^{-1} (Figure 6.10). However, the greatest change occurred after a threshold deposition of 5.8 kg N ha^{-1} yr^{-1}.

Figure 6.10. Total species richness of the vegetation (left) and moss species richness (right) in bogs in Norway over a gradient in nitrogen deposition from 2.6 to 17.5 kg. Mean values (0-5 kg: n = 5; 5-10 kg: n = 7 and 10-17 kg: n = 8) ± 95% confidence intervals are shown.



Source: Wamelink et al., 2021; Jokerud, 2012

Wilkins et al. (2016), with updates in Aherne et al. (2021), identified vegetation community change points (thresholds) for raised and upland blanket bogs in Ireland using species abundance data from relevé plots (389 and 247, respectively) spanning a deposition gradient of 4 to 17 kg N ha^{-1} yr^{-1} (observation-based total N deposition at 5 × 5 km resolution). For raised bogs, species data were limited to 15 indicator species, compared with all species data for blanket bogs (i.e. 115 plant species). The community change point for declining species in raised bogs was estimated at 6.7 kg N ha^{-1} yr^{-1} with 4 species decreasing in abundance (*Trichophorum germanicum*, *Sphagnum cuspidatum*, *Sphagnum denticulatum*, *Sphagnum capillifolium s. rubellum*). For blanket bogs the threshold for declining species was 6.2 kg N ha^{-1} yr^{-1} with 15 species decreasing in abundance (8 identified as positive indicator species; *Trichophorum germanicum*, *Sphagnum tenellum*, *Racomitrium lanuginosum*, *Pleurozia purpurea*, *Sphagnum denticulatum*, *Breutelia chrysocoma*, *Rhynchospora alba*, *Schoenus nigricans*).

The community change points or thresholds for bogs in Norway and Ireland align with the lower end of the existing $CL_{emp}N$ range (5-10 kg N ha^{-1} yr^{-1}), whereas those for the UK centre on approximately 10 kg N ha^{-1} yr^{-1}, the upper end of the existing range for bogs (Q1).

Summary of Q1

Even though there has been only a limited number of long-term N manipulation experiments in bog ecosystems, a clear picture has emerged of the potential impact of elevated N deposition on bog habitats. In Canadian and Swedish work, responses to N deposition as low as 2 kg N ha^{-1} yr^{-1} have been observed in a number of bog species (in terms of survivorship, flowering, and density).

Bryophyte species, in particular *Sphagnum* species, appear to be susceptible to the rise in anthropogenic N pollution, showing a decline in favour of grass and other competitive species, changes in competition between *Sphagnum* species, and changes in physiological and biochemical characteristics. Experimental work in Canada and a gradient survey of European bogs observed an inhibition of biological nitrogen fixation (BNF) and a consequent disruption of N cycling with onward impacts of vegetation communities has been observed at above 3 kg N ha^{-1} yr^{-1}. There appears to be a limited capacity for retention of N in the moss layer, above which N availability in the rhizosphere increases, offering a tool for assessment of factors which may modify the critical loads for these systems. Studies of N cycling and retention are consistent with a long-term response threshold of around 10 kg N ha^{-1} yr^{-1} but with an indication that N retention is less effective from around 2 kg N ha^{-1} yr^{-1}. It is likely that there is a time component linked to N saturation and the accumulation of N in a system when it is added at loads above which biological processes can accommodate. There are also likely to be more severe effects when N is added over a shorter time period or a site is subject to high concentrations of N.

The $CL_{emp}N$ recommended by Bobbink and Hettelingh (2011), 5-10 N kg ha^{-1} yr^{-1}, was based on a considerable body of field and experimental evidence and, hence, was judged to be reliable. Additional studies since then have generally provided results that further support this rating, although experimental work at very low background N as highlighted above has now suggested that responses are observed at even lower N deposition. Gradient surveys in the UK (Field et al., 2014), Ireland (Wilkins et al., 2016; Aherne et al., 2021) and Norway (Jokerud, 2012) have all observed changes in species richness or composition at the low end of the $CL_{emp}N$ range. Whilst we propose that the $CL_{emp}N$ range for bog ecosystems (Q1) encompassing lowland raised bogs (Q11) and blanket bogs (Q12) remains at 5 to 10 kg ha^{-1} yr^{-1} (classification 'reliable'), we highlight the emerging body of evidence of responses at very low N loads and recommend further experimental study to understand the long-term effects on ecosystem functioning of these loads.

Much of Europe, North America, Central and Western Asia has received N deposition well above the critical load for many decades and, as a result, we have witnessed a shifted baseline across many of our peatlands compared to northern latitude systems with low deposition histories. However, many of the studies in this chapter highlight the lack of an N threshold and the progressive nature of N responses as deposition increases from a low background, this suggests that any reduction in N deposition is worthwhile and will, over time, lead to a recovery of ecosystem functioning.

6.3 Valley mires, poor fens and transition mires (Q2)

The last review of Q2 habitats established a $CL_{emp}N$ of 10-15 kg N ha^{-1} yr^{-1} based on the evidence available at that time. In this review, we summarise this earlier evidence and incorporate results from experimental work at very low background N levels in Canada and a gradient study in The Netherlands. Valley mires, poor fens and transition mires are weakly to strongly acidic peatlands, flushes and vegetated rafts formed in situations where they receive water from the surrounding landscape or are intermediate between land and water (Davies et al., 2004). All systems have permanently waterlogged soils, with groundwater just below or at the soil surface.

This water supply is rather poor in base cations, leading to an acidic system with a pH slightly above bog systems, where peat mosses, but also small sedges and some brown moss species, dominate the vegetation. The distinction between valley mires, poor fens and transition mires is made on the basis of water level and water origin and may have some implication for the level of their critical loads. However, the low number of studies does not allow a further distinction, and the limited information that is available to date comes mainly from poor-fen systems (Q22). When compared to poor fens, based on the generalisation from Morris (1991) on the link between N sensitivity and hydrology (see introduction Chapter 6.1), valley mires are expected to be slightly less sensitive, and quaking bogs and transition mires to be more sensitive to excess N.

The significance of competition for light to the N response in *Sphagnum* was demonstrated by the study of Hogg et al. (1995) in a small valley mire near York in the United Kingdom. The growth of the mosses *Sphagnum palustre* and *Sphagnum fimbriatum* was reduced by 50% after additions of 12 kg N ha^{-1} yr^{-1} over two years; a rate which was probably comparable to ambient deposition. Where *Sphagnum* was growing poorly and the dominant grass species *Molinia caerulea* was abundant, adding N had no effect, but cutting *Molina caerulea* in the summer was beneficial to *Sphagnum*, re- invigorating its growth.

In an experiment in central France, Francez and Loiseau (1999) studied the fate of N in poor fens in Côte de Braveix by tracking 5 kg N ha^{-1} yr^{-1} labelled with 15N (background deposition 10 kg N ha^{-1} yr^{-1}). All of this N, added between June and August, remained in the system until October. Most of this N (55-65%) had accumulated in the *Sphagnum* layer. The upper peat layer of up to 10 cm accumulated about 15 to 30% of the added N, and from all deeper peat layers less than 5% of the added N was retrieved (Francez and Loiseau, 1999). These results indicate that *Sphagnum* functions as a N filter in poor fens as it does in ombrotrophic bogs (Lamers et al., 2000; Malmer and Wallén, 2005). Microcosm studies with *Sphagnum magellanicum*, a species characteristic of poor fens in Scandinavia, have shown significant negative effects of N additions (30 kg N ha^{-1} yr^{-1}) on concentrations of nutrients such as P, K and Ca, in mosses after a period of three months (Jauhiainen et al., 1998b).

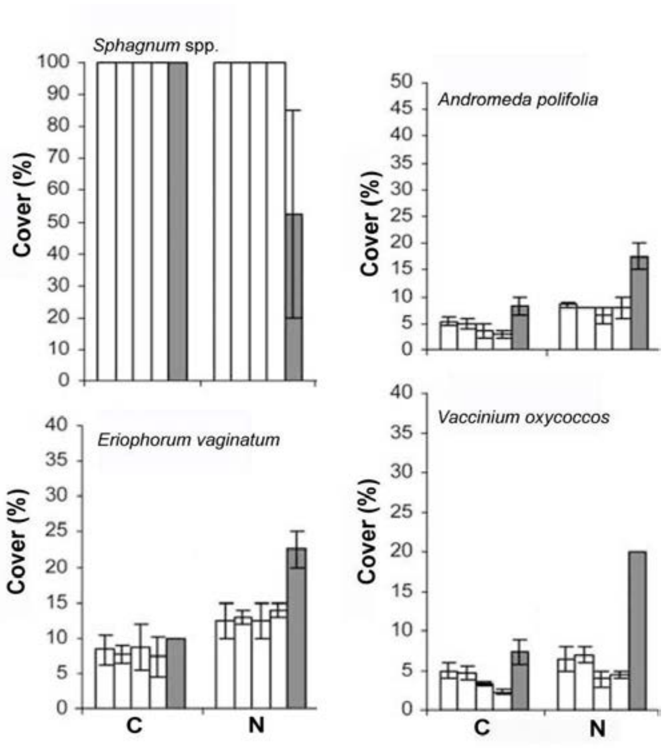
Malmer et al. (2003) studied the competition between vascular plants and peat mosses in a fertilisation experiment, and with respect to competition for light in a removal experiment in poor fens during two growing seasons. N was added in the form of ammonium nitrate at a level of 20 kg N ha^{-1} yr^{-1} (4-5 times the ambient supply rate) and was added both on and below the moss surface. Adding N confirmed the hypothesis that mosses rely on atmospheric supply, while *Narthecium ossifragum* depends on mineralisation in the peat (Malmer et al., 2003). N addition significantly increased shoot length of *Narthecium* and length increment of *Sphagnum* (but not its biomass). The negative relationship between the growth of *Narthecium* and of *Sphagnum* demonstrated a symmetric competition for light, the intensity of which increased with an increasing availability of N (Malmer et al., 2003).

In 1995, a long-term field experiment (addition of 15 and 30 kg N ha^{-1} yr^{-1}) was started in northern Sweden on a poor fen, at an ambient deposition of 2 kg N ha^{-1} yr^{-1} (Granberg et al., 2001). After three growing seasons, Granberg et al. (2001) reported results on the possible effects of climate change on CH_4 emission. They expected (and confirmed) sedges to be an important factor in CH_4 release into the atmosphere. Similar to effects found in ombrotrophic bogs (see Chapter 6.2), sedge cover significantly increased with additions of increasing amounts of N (Granberg et al., 2001). Unfortunately, the regression analysis used did not permit a distinction between effects at 15 and 30 kg N ha^{-1} yr^{-1}. Using sedge cover as a covariate, the effect of N addition on CH_4 emission changed over time from non- significant in the first year (1995) to a significant negative effect during the last year (1997). This cumulative effect was probably linked to the significant accumulation of total N in the upper 5 (15 kg N ha^{-1} yr^{-1} treatment) or 10 (30 kg N ha^{-1} yr^{-1} treatment) cm of the soil (Granberg et al., 2001).

In the same field fertilisation experiment, Gunnarsson et al. (2004) focused on the growth and production of *Sphagnum balticum* and interspecific competition between *S. balticum* and either *Sphagnum lindbergii* or transplanted *Sphagnum papillosum*. Production and length increment of *Sphagnum balticum* in nutrient-poor lawn communities was significantly reduced after four years of N addition. The area covered by *Sphagnum lindbergii* was increased on the N-treated plots, which may reflect its greater tolerance to high N influx in relation to *Sphagnum balticum* (Gunnarsson et al., 2004). The hummock-forming *Sphagnum papillosum* was found to increase at the expense of *Sphagnum balticum* under climatic regimes with a more negative water balance, probably because of the low tolerance of *Sphagnum balticum* to drier conditions. Reduced growth of peat mosses may have positive effects on vascular plants, as numerous studies on bogs have shown (see Chapter 6.2).

Gunnarsson et al. (2004) concluded that increased N deposition may transform mires that are dominated by *Sphagnum* into vascular-plant-dominated mires.

Figure 6.11. Time series for the years 1995, 1996, 1997 and 1998 (white bars) and 2003 (grey bars). Data represent cover (mean ± SE) of *Sphagnum species, Eriophorum vaginatum, Andromeda polifolia and Vaccinium oxycoccos* for two treatments (C = control and N = +30 kg N ha^{-1} yr^{-1}).

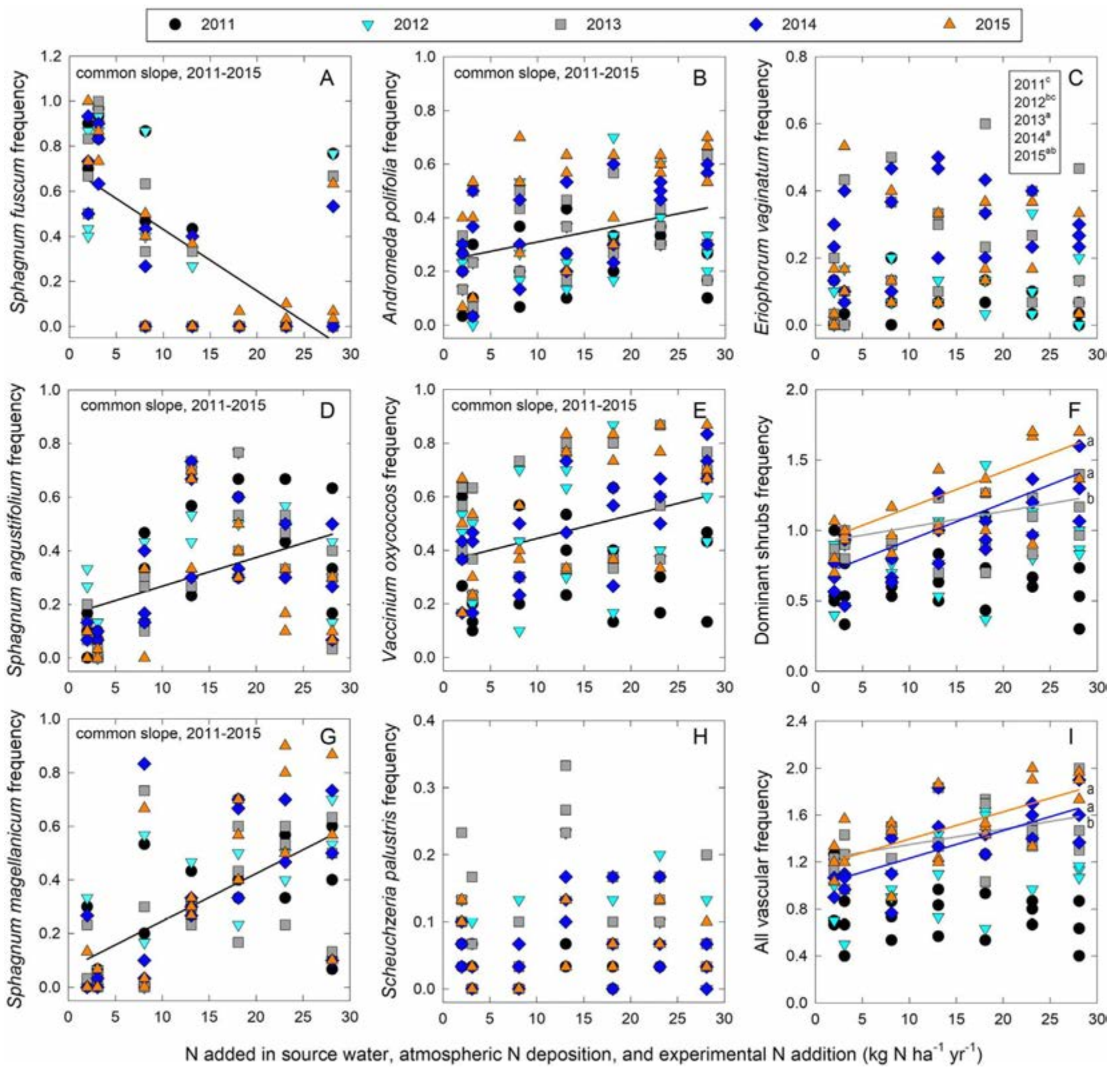


Source: Wiedermann et al., 2007

The experiment described above was continued for four more years, but unfortunately only the results from the highest treatment dose of 30 kg N ha^{-1} yr^{-1} were provided. Wiedermann et al. (2007) found that the vegetation responses were negligible for the first four years, but after eight years of continuous N addition, the closed *Sphagnum* carpet had been drastically reduced from 100% down to 41% (Figure 6.11). The total vascular plant cover (*Eriophorum vaginatum*, *Andromeda polifolia* and *Vaccinium oxycoccus*) increased from 24% to an average of 70% (Figure

6.10). Wiedermann et al. (2007) stress that both bryophytes and vascular plants in boreal mires receiving background levels of N of only about 2 kg ha^{-1} yr^{-1} exhibit a time lag of more than five years before responding to N, emphasising the need for long-term experiments. However, it should be observed that climate was also observed to be higher in 2003 and could therefore play an interactive role in the response.

Figure 6.12. Frequency of occurrence from point frame measurements for the three most frequently occurring *Sphagnum* species (*Sphagnum fuscum, Sphagnum angustifolium, Sphagnum magellanicum)*, the two most frequently occurring shrub species (*Andromeda polifolia, Vaccinium oxycoccos*), the most frequently occurring graminoid species (*Eriophorum vaginatum*), the most frequently occurring forb species (*Scheuchzeria palustris*), dominant shrubs combined (*Andromeda polifolia, Vaccinium oxycoccos, Chamaedaphne calyculata, Kalmia polifolia*), and all vascular plant species combined (*Scheuchzeria palustris, Eriophorum vaginatum, Andromeda polifolia, Vaccinium oxycoccos, Chamaedaphne calyculata, Kalmia polifolia, Drosera rotundifolia, Rubus chamaemorus, Maianthemum trifolia, Carex aquatilis, Carex limosa, Picea mariana* below 1 m tall) as a function of N addition. When there were differences between years, but no significant effect of N addition, years with the same letter superscript do not differ significantly (ANCOVA, a posteriori Tukey's Honestly Significant Difference test). Slopes with the same lower-case letter to the right of regression lines plotted for individual years do not differ significantly (ANCOVA).

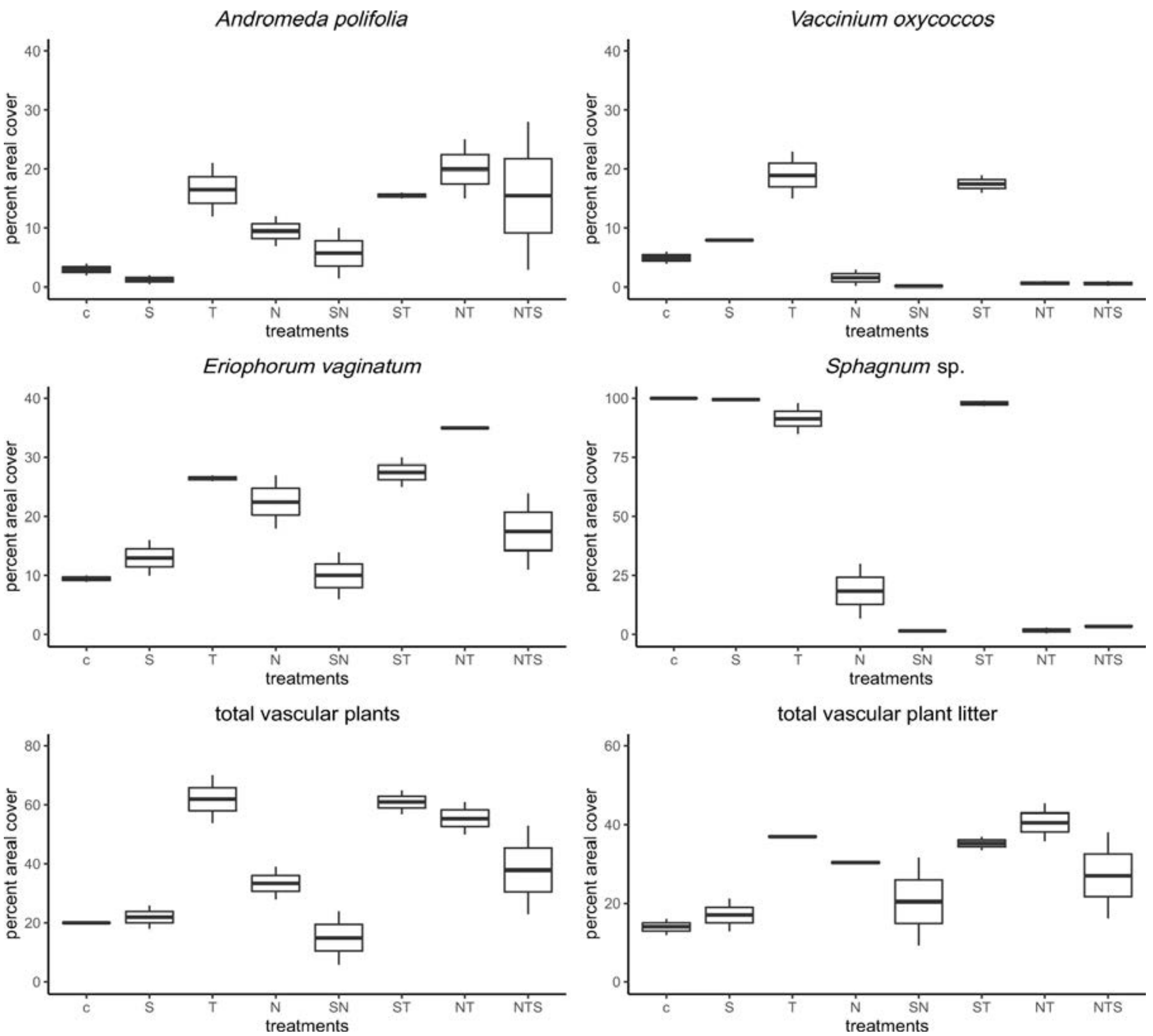


Source: Wieder et al., 2020

Experimental work completed since the last review of $CL_{emp}N$ has taken place on a poor fen in Alberta, Canada, at very low background N (bulk deposition < 2 kg N ha^{-1} yr^{-1}), where Wieder et al., (2020) found N-driven reductions in biological nitrogen fixation (BNF), decreased abundance of *Sphagnum fuscum* and increases in *S. angustifolium*, and increases in vascular plants in general (Figure 6.12). A stimulation in decomposition of cellulose was also observed but not of more recalcitrant *Sphagnum* or vascular plant litter. No response thresholds were observed, with changes suggested as progressive from above background N. A $CL_{emp}N$ of 3 kg N ha^{-1} yr^{-1} was proposed by the authors (Wieder et al., 2020) based on the observation that, above this level, N cycling and species composition was disrupted.

A recent gradient study at higher N loads in the Netherlands, spanning a range of 17-24 kg N ha^{-1} yr^{-1}, observed decreased base saturation at higher N in poor fens and an observation that Dutch fens are more acidic than fens in regions with lower N deposition (Van Diggelen et al., 2018).

Figure 6.13. Cover estimates (% areal cover) of vascular plants and *Sphagnum* vegetation as well as vascular plant litter at the long-term fertilisation experiment at Degerö Stormyr in July 2016; y-axes: cover estimate; x- axes: experimental treatments: c = control, S = sulfur (20 kg ha^{-1} yr^{-1}), T = temperature (+3.6 °C), N = nitrogen (30 kg ha^{-1} yr^{-1}), SN = sulfur + nitrogen, ST = sulfur + temperature, NT = nitrogen + temperature, NTS = nitrogen + temperature + sulfur; total number of plots = 16.



Source: Wiedermann and Nilsson, 2020

Interactions with climate change and warming have also been observed in fens. A long-term (21 year) experiment in a *Sphagnum* dominated poor-fen peatland in Degerö Stormy, Sweden, applied 30 kg N ha^{-1} yr^{-1} on top of a low background N ~2 kg N ha^{-1} yr^{-1} in conjunction with, and separately from, a warming effect created by open topped chambers (Wiedermann and Nilsson, 2020). The authors found that *Sphagnum* cover fell sharply when N was added (from ~80% to ~20%) and further still in conjunction with elevated temperature (Figure 6.13). *Vaccinium oxycoccus* reduced a little with N alone (but not significantly so) but declined further with elevated temperature and N (note that with temperature alone, the shrub increased in cover). *Andromeda polifolia* decreased with N alone but elevated temperature reversed this. As with Q1, *Eriphorum vaginatum* increased with N but more so when temperature was also elevated. In general, the overall cover of vascular plants was increased by both temperature and N (Figure 6.13; Wiedermann and Nilsson, 2020).

Other work on a transition alpine mire consisting of hummocks and lawns added 0, 10, and 30 kg N for eight years (background N ~8 kg) also observed temperature and N effects, although in response to a short-term natural heatwave event (Brancaleoni and Gerdol, 2014). In hummocks *Sphagnum* mosses recovered better from heat wave under N addition, however, in lawns vascular plants expanded faster under high N, suggesting that persistent climate change could reduce the carbon sink. Species richness also fell in lawns (Brancaleoni and Gerdol, 2014). The differential response between hummocks and lawns highlights the susceptibility of some bog species to climate change.

Summary of Q2

The $CL_{emp}N$ previously recommended for poor fens (Q22) by Bobbink and Hettelingh (2011) was 10-15 kg N ha^{-1} yr^{-1}, with the low end of the range emphasised for oceanic valley mires (Q21) where limited studies at low N were available. These critical loads represented a reduction on the earlier critical load based on the evidence of the failure of the *Sphagnum* N filter function observed in bogs. Recent studies in Q2 are still limited, however, emerging work at the low end of N deposition has found responses at very low N loads (e.g. Wieder et al., 2020). Based on this new but limited evidence we would recommend a reduction of the low end of the $CL_{emp}N$ and a new range of 5 to 15 kg N ha^{-1} yr^{-1} (classification 'reliable') and encourage further research into low N responses in these habitats to determine if the higher end of the range is appropriate.

6.4 Palsa and polygon mires (Q3)

Palsa and polygon mires are patterned mire complexes of the arctic, subarctic and northern boreal zones. The formation and maintenance of these types of mires are dependent on the action of frost or ice. Palsa mires (Q31) are formed by elevated frozen mounds or ridges (palsas) 0.5 to 8 m high and up to 50 m in diameter, interspersed with wet hollows of similar area (Davies et al., 2004). Polygon mires (Q32) are complex mires of the arctic and subarctic patterned by surface microrelief of large, 10-30 m in diameter, low-centre or high-centre polygons formed by the juxtaposition of dry, 0.3-0.5 m high ridges (Davies et al., 2004). The distinction between palsa and polygon mires is made on the perpetual presence of ice, which is only the case for palsa mires. Intact palsa mounds show a patterning of weakly minerotrophic vegetation with different assemblages of mosses, herbs and sub-shrubs on their tops and sides (Schaminee et al., 2019**)**. In polygon mires, cover of non-sphagnaceous mosses and lichens outweigh *Sphagnum* species and together with dwarf shrubs occur on the ridges (Schaminee et al., 2019). Wet hollows are occupied by grasses, sedges and mosses including *Sphagnum* species. According to Schaminee et al. (2019) palsa and polygon mires cannot be distinguished on floristic criteria. Since both types of mires are limited to the arctic, subarctic and northern boreal zones, we assume that the N sensitivity for both types of mires will be comparable.

For palsa and polygon mires no $CL_{emp}N$ was set by Bobbink and Hettelingh (2011), due to the lack of empirical data. While there are no experimental studies available from Europe, there is a study from polygon mires in the Canadian arctic (Bylot Island; Marchand-Roy, 2009). The aim of this fertilisation experiment was to simulate the nutrient input by the annual presence of the Great Snow Goose. Nitrogen was added as NH_4NO_3 at a dose of 10, 30 and 50 kg N ha^{-1} yr^{-1} for five years (ambient yearly nitrogen deposition is not given). Nitrogen was added in one yearly dose in late June or early July, after the snow melted and the polygon was no longer flooded. At an N fertilisation rate above 10 kg N ha^{-1} yr^{-1} a significant effect on the growth of graminoid plants was observed (Marchand-Roy, 2009). Compared to the control, the aboveground biomass of graminoid plants increased by 24% at the lowest dose of 10 kg N ha^{-1} yr^{-1}, 41% at the intermediate dose of 30 kg N ha^{-1} yr^{-1} and 103% at the highest dose of 50 kg N ha^{-1} yr^{-1}. This significant effect was not observed after two years of fertilisation, due the N filtering effect of bryophytes (Pouliot et al., 2009). Nitrogen input had no effect on the primary productivity of bryophytes. Tissue N concentrations of both graminoid plants and bryophytes were increased significantly by N addition. In addition, the decomposition of organic matter was significantly promoted by N.

Summary of Q3

The experiment in the Canadian arctic showed significant effects of 10 kg N ha^{-1} yr^{-1} on the production of graminoids, tissue N concentrations and decomposition rate within a short experimental period of five years. However, ambient nitrogen deposition is very low in the region (< 1-2 kg N ha^{-1} yr^{-1}), indicating the $CL_{emp}N$ is likely to be below 10 kg N ha^{-1} yr^{-1}. Since (*Sphagnum*) mosses function as an N filter in polygon mires, like ombrotrophic bogs, we think that the $CL_{emp}N$ for polygon mires is similar to that of raised and blanket bogs (Q1). However, as palsa and polygon mires only occur in pristine areas, we believe that the low end of the range is should be reduced. We therefore propose, based on expert judgement, a $CL_{emp}N$ range for palsa and polygon mires (Q3) of 3 to 10 kg N ha^{-1} yr^{-1}.

6.5 Base-rich fens and calcareous spring mires (Q4)

Similar to poor fens, base-rich fens and calcareous spring mires have developed on permanently waterlogged soils, but in these systems there is a base-rich, nutrient-poor and often calcareous water supply buffering the system from high levels of acidity. They are largely occupied by calcicolous small sedges and brown moss communities (Davies et al., 2004). Although rich fens are the habitat of a range of specialised and rare species, very few field experiments have been conducted with enrichments of ecologically relevant doses of N to determine the effects of increased N deposition. Early work set the $CL_{emp}N$ range for mesotrophic fen ecosystems at between 15 and 35 kg N ha^{-1} yr^{-1} (Bobbink et al., 2003). This was based mainly on nutrient budget studies on rich fens (Q41-Q44) in the Netherlands and on several field experiments, but these were all with (very) high N additions (> 100 kg N ha^{-1} yr^{-1}) (e.g. Beltman et al., 1996; Boeye et al., 1997; Wassen et al., 1998). This was then revised down in the last review of $CL_{emp}N$ to 15-30 kg N ha^{-1} yr^{-1} for rich fens (Q41-Q44) (Bobbink and Hettelingh, 2011) and 15-25 kg N ha^{-1} yr^{-1} for arctic-alpine rich fens (Q45). These reductions were based on experiments in Ireland that observed severe negative effects on the bryophyte layer within five years after additions of 35 kg N ha^{-1} yr^{-1} under low ambient N depositions (6-8 kg N ha^{-1} yr^{-1}). This previous work is briefly summarised below.

Koerselman and Verhoeven (1992) assumed that the input of N needed to be counterbalanced by the output of N through usual management (mowing). Increased N input, compared to N removal, results in a considerable increase in tall graminoids and a subsequent decrease in diversity of the subordinate plant species (Vermeer, 1986; Verhoeven and Schmitz, 1991).

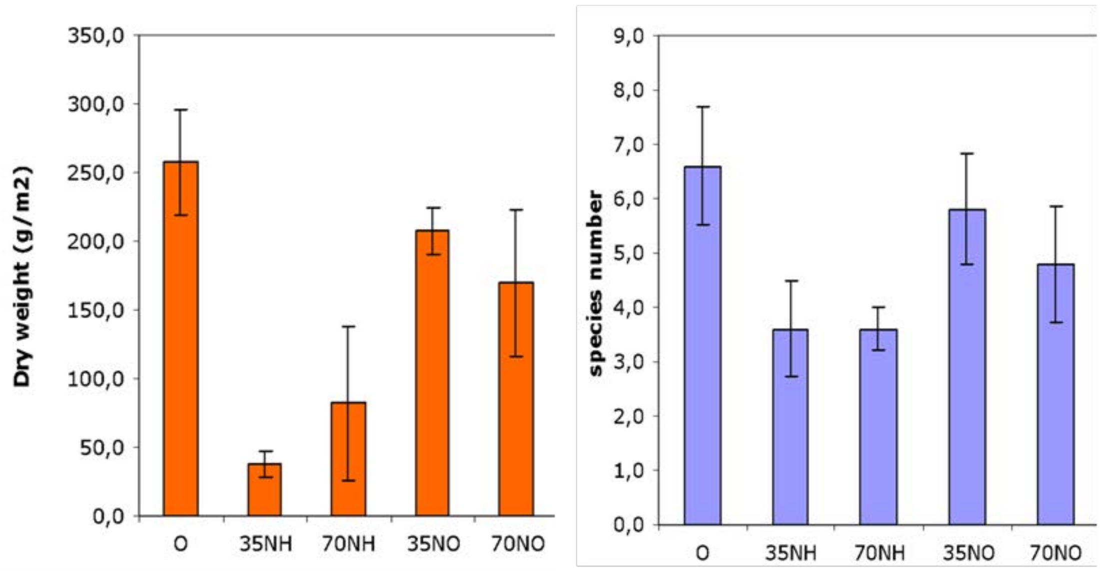
Although in some fens strong P limitation inhibits changes in diversity with increased N, it is expected that such a situation leads to increased losses of inorganic N to the surface or groundwater, thus leading to a similar critical load as those for P or N limited situations (Bobbink et al., 1996). Five locations in Belgium, Ireland and Poland were used in factorial fertilisation experiments with both N and P. Two sites (in the north- eastern part of Belgium and in Poland) gave clear evidence of N as the most important growth-limiting factor, while growth at the other three sites was strongly limited by P (Beltman et al., 1996; Boeye et al., 1997; Wassen et al., 1998).

Microcosm experiments with bryophytes (*Sphagnum wanstorfii*) and vascular plants (*Carex rostrata*) collected from rich fens at higher latitudes have indicated sensitivity to additions of ammonium nitrate of 30 kg N ha^{-1} yr^{-1} and higher. Within three months, a decrease in moss growth could be measured, as well as an increase in N and free amino acid concentrations in both mosses and vascular plants, both of which are clear indications of N enrichment effects (Jauhiainen et al., 1998a; Saarinen, 1998). Although, by themselves, the conditions in these experiments were too artificial to justify a decrease of the present critical load, they do indicate that moss species in particular, which are an important and prominent part of biodiversity in rich fens, may be very sensitive to increased N deposition.

Paulissen (2004) studied the effects of different forms of pollutant N in a rich fen in central Ireland (ambient load 6-10 kg N ha^{-1} yr^{-1}). N was added in the form of ammonium or nitrate, at a level of 50 kg N ha^{-1} yr^{-1}. After two years, no effects of N treatment could be found in the vascular plant cover, nor in the dominant bryophyte species (*Scorpidium revolvens* and *Sphagnum contortum*). Tissue N:P ratio indicated that the bryophyte layer was P limited (Paulissen, 2004). The surface phosphatase (an enzyme that allows plants to take up organically bound P under conditions of increased P limitation) activity of the typical brown moss *Scorpidium revolvens* was significantly stimulated by nitrate addition, whereas ammonium addition did the same in the invasive *Sphagnum contortum*. After four years, ammonium significantly reduced the biomass of *Scorpidium revolvens* although *Sphagnum contortum* biomass was not affected by N addition (Paulissen et al., 2016). This implies that, in the longer term, *Sphagnum* will profit from high ammonium deposition, while it will negatively affect the typical brown mosses. Paulissen (2004) concluded that, in the short term, the vegetation of rich fens would not be very sensitive to increased N deposition. In the longer term (> 2 years), however, growth of brown mosses, such as *Scorpidium revolvens*, in P-limited fens could become negatively affected by ammonium, although not (yet) by nitrate. In contrast, *Sphagnum contortum* was found not to be sensitive to increased ammonium availability. The negative effect of ammonium on brown mosses was confirmed in additional experiments (Paulissen et al., 2004; 2005).

The effects of different loads and forms of N on rich fen vegetation were studied in the same fen in central Ireland (ambient load 6-10 kg N ha^{-1} yr^{-1}), for five years (Dorland et al., 2008; Verhoeven et al., 2011 – this later study as noted as 'in-press' in the last review). N was added either as ammonium or nitrate in the following doses: 35 and 70 kg N ha^{-1} yr^{-1}. Peat water was characterised by a high pH, very low concentrations of nitrate, ammonium and phosphate, and high base-cation concentrations (especially Calcium; Verhoeven et al., 2011). Ammonium additions at rates of 35 and 70 kg N ha^{-1} yr^{-1} significantly increased tissue N concentrations in the bryophyte *Calliergonella cuspidata* and reduced the number of bryophyte species and bryophyte biomass production (Figure 6.14; Dorland et al.,2008; Verhoeven et al., 2011). In contrast, vascular plant species were not affected by N addition, and their biomass was even increased as a result of ammonium addition (Dorland et al., 2008). Vascular plants benefited from the opening up of the thick bryophyte layer, and from the reduced N uptake by bryophytes.

Figure 6.14. Dry biomass (left) and species number (right) of bryophytes after five years of N addition in a rich fen in central Ireland (Scragh Bog). Treatments: O = control treatment; NH = ammonium addition; NO = nitrate addition. Number before code = annual load of element in kg ha^{-1} yr^{-1}.



Source: Verhoeven et al., 2011

There are limited new publications to consider since the last review of $CL_{emp}N$. A survey of alkaline fens (Q41) over a low N deposition range (4.2 to 10.0 kg N ha^{-1} yr^{-1}) in Ireland used TITAN change point analysis on vegetation data collected from 32 relevé plots (Wilkins et al., 2016; Aherne et al., 2021). The authors identified a change point threshold at 5.5 kg N ha^{-1} yr^{-1} with decreases in the abundance of 5 species including 2 positive indicator species. The threshold in this study is substantially below the existing $CL_{emp}N$ range of 15-30 kg N ha^{-1} yr^{-1}, however, the study was limited by the small number of plots spanning a narrow deposition gradient. In contrast, Van den Berg et al. (2016) assessed the effects of total N deposition on species richness across eight habitats. They found clear evidence that N deposition affects species richness in all habitats except base-rich mires (n = 274 over a deposition gradient of 5.1 to 54.2 kg N ha^{-1} yr^{-1}), after factoring out correlated explanatory variables (climate and sulphur deposition).

Summary of Q4

It is difficult to make recommendations to drastically change the existing $CL_{emp}N$ based upon the limited new evidence. We have strong indications that the upper limit of 30 kg N ha^{-1} yr^{-1} for rich fens is too high. Therefore, we propose a $CL_{emp}N$ range of 15-25 kg N ha^{-1} yr^{-1} for rich fens (Q41-Q44), similar to arctic-alpine rich fens (Q45). In addition, we have increased the reliability for rich fens from 'expert judgement' to 'quite reliable'. Long-term fertilisation experiments with ecologically relevant additions of N and observational studies in temperate regions, northern countries and arctic-alpine fens would increase the reliability of these figures. For artic-alpine rich fens (Q45; $CL_{emp}N$ based on expert judgement), it is likely that the critical load should be lower with further research particularly recommended in this habitat.

6.6 Overall summary for mire, bog and fen habitats (EUNIS class Q)

An overview of the $CL_{emp}N$ ranges for mire, bog and fen habitats is presented in Table 6.1.

Table 6.1. $CL_{emp}N$ and effects of exceedances on different mire, bog and fen habitats (Q). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Raised and blanket bogs	Q1	5-10	##	5-10	##	Increase in vascular plants, decrease in bryophytes; altered growth and species composition of bryophytes; increased N in peat and peat water
Valley mires, poor fens and transition mires	Q2	10-15 [a]	#	**5**-15	**##**	Increase in sedges and vascular plants; negative effects on bryophytes
Palsa and polygon mires	Q3			**3-10**	**(#)**	Increase in graminoids; tissue N concentrations and decomposition rate
Rich fens	Q41-Q44	15-30	(#)	15-**25**	**#**	Increase in tall vascular plants (especially graminoids); decrease in bryophytes
Arctic-alpine rich fens	Q45	15-25	(#)	15-25	(#)	Increase in vascular plants; decrease in bryophytes

a) The $CL_{emp}N$ previously recommended for poor fens (Q22) by Bobbink and Hettelingh (2011) was 10-15 kg N ha^{-1} yr^{-1}, with the low end of the range emphasised for oceanic valley mires (Q21) where limited studies at low N were available.

6.7 Recommendations and knowledge gaps

Since the last update a limited number of experiments with realistic additions of N that are within or below the range of the critical load have been published. We recommend establishing field experiments in areas with low deposition histories to understand responses at low N. We also recommend experimental work in habitat types where the reliability of the critical load is based on expert judgement or is only quite reliable (e.g. Q3, Q41–Q44). Especially for arctic-alpine montane fens (Q45), fertilisation experiments are urgently needed to increase the reliability of its critical load (still based on expert judgement). For poor fens (Q2) we encourage further research into low N responses to determine if the higher end of the range (15 kg N ha^{-1} yr^{-1}) is appropriate. In this ecosystem, ground water N is also important but the interactive effects of this and atmospheric deposition are poorly understood.

Long-term fertilisation experiments (> 10 years) are needed to determine the cumulative effects of nitrogen. In addition, reduced nitrogen appears to be more harmful than oxidised nitrogen, but the number of studies on the form of nitrogen are very limited (only for Q1). Nitrogen research is often limited to the effects on the vegetation, leaving the fauna (e.g. birds and butterflies) underexposed.

6.8 References

Aaby, B. (1994). Monitoring Danish raised bogs. In: A. Grünig (ed.), *Mires and Man. Mire conservation in a densely populated country – the Swiss experience*. Kosmos, Birmensdorf, 284-300.

Aerts, R., Wallén, B. and Malmer, N. (1992). Growth-limiting nutrients in *Sphagnum*-dominated bogs subject to low and high atmospheric nitrogen supply. *Journal of Ecology* **80**, 131-140.

Aerts, R., Wallén, B., Malmer, N. and De Caluwe, H. (2001). Nutritional constraints on *Sphagnum*-growth and potential decay in northern peatlands. *Journal of Ecology* **89**, 292-299.

Aherne, J., Wilkins, K. and Cathcart, H. (2021). *Nitrogen-sulphur critical loads: assessment of the impacts of air pollution on habitats* (2016-CCRP-MS.43). EPA Research Report, Trent University, Ireland.

Beltman, B., Kooijman, A.M., Rouwenhorst, G. and Van Kerkhoven, M. (1996). Nutrient availability and plant growth limitation in blanket mires in Ireland. *Proceedings of the Royal Irish Academy* **96B**, 77-87.

Berendse, F., Van Breemen, N., Rydin, H., Buttler, A., Heijmans, M., Hoosbeek, M. R., Lee, J. A., Mitchell, E., Saarinen, T., Vasander, H. and Wallén, B. (2001). Raised atmospheric CO_2 levels and increased N deposition cause shifts in plant species composition and production in *Sphagnum* bogs. *Global Change Biology* **7**, 591-598.

Bobbink, R., Heil, G.W. and Raessen, M.B.A.G. (1992). Atmospheric deposition and canopy exchange processes in heathland ecosystems. *Environmental Pollution* **75**, 29-37.

Bobbink, R., Hornung, M. and Roelofs, J.G.M. (1996). Empirical nitrogen critical loads for natural and semi-natural ecosystems. In: *Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded,* UN ECE Convention on long-range transboundary air pollution, Federal Environmental Agency, Berlin.

Bobbink, R., Ashmore, M., Braun, S., Flückiger, W. and Van den Wyngaert, I.J.J. (2003). Empirical nitrogen loads for natural and semi-natural ecosystems: 2002 update. In: *Empirical critical loads for nitrogen*, Expert Workshop Convention on long-range transboundary air pollution UNECE), Swiss Agency for the Environment, Forests and Landscape SAEFL, Berne.

Bobbink, R. and J.P. Hettelingh (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships.* Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), Bilthoven, The Netherlands.

Boeye, D., Verhagen, B., Van Haesebroeck, V. and Verheyen, R.F. (1997). Nutrient limitation in species-rich lowland fens. *Journal of Vegetation Science* **8**, 415-424.

Boxman, A.W., Peters, R.C.J.H. and Roelofs, J.G.M. (2008). Long term changes in atmospheric N and S throughfall deposition and effects on soil solution chemistry in a Scots pine forest in the Netherlands. *Environmental Pollution* **156**, 1252-1259.

Bragazza, L. and Limpens, J. (2004). Dissolved organic nitrogen dominates in European bogs under increasing atmospheric N deposition. *Global Biogeochemical Cycles* **18**, GB4018.

Bragazza, L., Tahvanainen,T., Kutnar, L., Rydin, H., Limpens, J., Hàjek, M., Grosvernier, P., Hansen, I., Iacumin, P. and Gerdol, R. (2004). Nutritional constraints in ombrotrophic *Sphagnum* subject to increasing levels of atmospheric nitrogen deposition in Europe. *New Phytologist* **163**, 609-616.

Bragazza, L., Limpens, J., Gerdol, R., Grosvernier, P., Hajèk, M., Hajkova, P., Lacumin, P., Kutnar, L., Rydin, H. and Tahvanainen, T. (2005) Nitrogen content and $\delta^{15}N$ signature of ombrotrophic *Sphagnum* plants in Europe: to what extent is the increasing atmospheric N deposition altering the N-Status of nutrient- poor mires? *Global Change Biology* **11**, 106-114.

Bragazza, L., Freeman, C., Jones, T., Rydin, H., Limpens, J., Ellis, T., Fenner, N., Gerdol, R., Hájek, M., Hájek, T., Iacumin, P., Kutnar, L., Tahvanainen, T. and Toberman, H. (2006). Atmospheric nitrogen deposition promotes carbon loss from European bogs. *PNAS* **103**, 19386-19389.

Bragazza, L. and Freeman, C. (2007). High nitrogen availability reduces polyphenol content in *Sphagnum* peat. *Science of the Total Environment* **377**, 439-443.

Brancaleoni, L. and Gerdol, R. (2014). Habitat-dependent interactive effects of a heatwave and experimental fertilization on the vegetation of an alpine mire. *Journal of Vegetation Science* **25**, 427-438.

Breeuwer, A.J.G., Heijmans, M.M.P.D., Robroek, B.J.M., Limpens, J. and Berendse, F. (2008). The effect of increased temperature and nitrogen deposition on decomposition in bogs *Oikos* **117**, 1258 - 1268.

Carfrae, J.A., Sheppard, L.J., Raven, J.A., Leith, I.D. and Crossley, A. (2007). Potassium and phosphorus additions modify the response of *Sphagnum capillifolium* growing on a Scottish ombrotrophic bog to enhanced nitrogen deposition. *Applied Geochemistry* **22**, 1111-1121.

Clymo, R.S. (1970). The growth of *Sphagnum*: methods of measurements. *Journal of Ecology* **58**, 13-49.

Currey, P.M., Johnson, D., Sheppard, L.J., Leith, I.D., Toberman, H., Van der Wal, R., Dawson, L.A. and Artz, R.R.E. (2010) Turnover of labile and recalcitrant soil carbon differ in response to nitrate and ammonium deposition in an ombrotrophic peatland. *Global Change Biology* **6**, 2307-2321.

Curtis C.J., Emmett B.A., Reynolds B. and Shilland J. (2004). Nitrate Leaching from Moorland Soils: Can Soil C:N Ratios Indicate N Saturation?. In: Wieder R.K., Novák M. and Vile M.A. (eds.) *Biogeochemical Investigations of Terrestrial, Freshwater, and Wetland Ecosystems across the Globe*. Springer, Dordrecht.

Davies, C.E. and Moss, D. (2002). *EUNIS habitat classification, Final report.* CEH Monks Wood, UK. Davies, C.E., Moss, D. and Hill, M.O. (2004). EUNIS habitat classification revised 2004. Winfrith Technology Centre, Dorian Ecological Information Ltd. and Monks Wood, UK.

Davies, C.E., Moss, D. and Hill, M.O. (2004). *EUNIS habitat classification revised 2004*. Report to: European environment agency-European topic centre on nature protection and biodiversity, pp.127-143.

Dorland, E., Bobbink, R. and Robat, S. (2008). *Impacts of changing ratios of reduced and oxidized nitrogen deposition: case studies in acid grasslands and fen ecosystems*. Proceedings 6th European Conference on Ecological Restoration, Ghent, Belgium.

Ellenberg, H. (1988). *Vegetation Ecology of Central Europe*. Cambridge University Press, Cambridge.

Eriksson, T., Öquist, M.G. and Nilsson, M.B. (2009). Production and oxidation of methane in a boreal mire after a decade of increased temperature and nitrogen and sulfur deposition. *Global Change Biology* **16**, 2130-2144.

Ferguson, P., Robinson, R.N., Press, M.C. and Lee, J.A. (1984). Element concentrations in five *Sphagnum* species in relation to atmospheric nitrogen pollution. *Journal of Bryology* **13**, 107-114.

Field, C. (2010). *The effect of atmospheric nitrogen deposition on carbon sequestration in semi-natural ericaceous dominated ecosystems*, The Manchester Metropolitan University.

Field, C., Sheppard, L.J., Caporn, S. and Dise, N. (2013). The ability of contrasting ericaceous ecosystems to buffer nitrogen leaching. *Mires & Peat* **11**, 1-11.

Field, C.D., Dise, N.B., Payne, R.J., Britton, A.J., Emmett, B.A., Helliwell, R.C., Hughes, S., Jones, L., Lees, S., Leake, J.R., Leith, I.D., Phoenix, G.K., Power, S.A., Sheppard, L.J., Southon, G.E., Stevens, C.J. and Caporn, S.J.M. (2014). The role of nitrogen deposition in widespread plant community change across semi-natural habitats. *Ecosystems* **17**, 864-877.

Francez, A.J. and Loiseau, P. (1999). The fate of mineral nitrogen in a fen with S*phagnum fallax* Klinggr. And *Carex rostrata* Stokes (Massif-central, France). *Canadian Journal of Botany-Revue Canadienne de Botanique* **77**, 1136-1143.

Gerdol, R., Petraglia, A., Bragazza, L., Iacumin, P. and Brancaleoni, L. (2007). Nitrogen deposition interacts with climate in affecting production and decomposition rate in *Sphagnum* mosses. *Global Change Biology* **13**, 1810-1821.

Gerdol, R., Bragazza, L. and Brancaleoni, L. (2008). Heatwave 2003: high summer temperature, rather than experimental fertilization, affects vegetation and CO_2 exchange in an alpine bog. *New Phytologist* **179**, 142-154.

Glatzel, S., Forbrich, I., Krüger, Lemke, S. and Gerold, G. (2008). Small scale controls of greenhouse gas release under elevated N deposition rates in a restoring peat bog in NW Germany. *Biogeosciences* **5**, 925-935.

Granath, G., Strengbom, J., Breeuwer, A., Heijmans, M.M.P.D., Berendse, F. and Rydin, H. (2009a). Photosynthetic performance in *Sphagnum* transplanted along a latitudinal nitrogen deposition gradient. *Oecologia* **159**, 705-715.

Granath, G., Wiedermann, M.M. and Strengbom, J. (2009b). Physiological responses to nitrogen and sulphur addition and raised temperature in *Sphagnum balticum*. *Oecologia* **161**, 481-490.

Granath, G., Strengbom, J. and Rydin, H. (2012). Direct physiological effects of nitrogen on Sphagnum: a greenhouse experiment. *Functional Ecology* **26**, 353-364.

Granberg, G., Sundh, I., Svensson, B.H. and Nilsson, M. (2001). Effects of temperature, and nitrogen and sulfur deposition, on methane emission from a boreal mire. *Ecology* **82**, 1982-1998.

Greven, H.C. (1992). Changes in the moss flora of the Netherlands. *Biological Conservation* **59**, 133-137.

Gunnarsson, U. and Rydin, H. (2000). Nitrogen fertilization reduces *Sphagnum* production in bog communities. *New Phytologist* **147**, 527-537.

Gunnarsson, U., Malmer, N. and Rydin, H. (2002). Dynamics or constancy in *Sphagnum* dominated mire ecosystems: a 40-year study. *Ecography* **25**, 685-704.

Gunnarsson, U., Granberg, G. And Nilsson, M. (2004). Growth, production and interspecific competition in *Sphagnum*: effects of temperature, nitrogen and sulphur treatments on a boreal mire. *New Phytologist* **163**, 349-359.

Gunnarsson, U., Boresjö Brongo, L., Rydin, H. and Ohlson, M. (2008). Near-zero recent carbon accumulation in a bog with high nitrogen deposition in SW Sweden. *Global Change Biology* **14**, 2152-2165.

Hassel, K., Kyrkjeeide, M.O., Yousefi, N., Prestø, T., Stenøien, H.K. Shaw, A.J. and Flatberg, K.I. (2018). *Sphagnum divinum* (sp. nov.) and *S. medium* Limpr. and their relationship to *S. magellanicum* Brid. *Journal of Bryology* **40**, 197-222.

Heijmans, M.M.P.D., Berendse, F., Arp, W.J., Masselink, A.K., Klees, H., De Visser, W. and Van Breemen, N. (2001). Effects of elevated carbon dioxide and increased nitrogen deposition on bog vegetation in the Netherlands. *Journal of Ecology* **89**, 268-279.

Heijmans, M.M.P.D., Klees, H. and Berendse, F. (2002a). Competition between *Sphagnum magellanicum* and *Eriophorum angustifolium* as affected by raised CO_2 and increased N deposition. *Oikos* **97**, 415-425.

Heijmans, M.M.P.D., Klees, H., De Visser, W. and Berendse, F. (2002b). Effects of increased nitrogen deposition on the distribution of ^{15}N-labeled nitrogen between *Sphagnum* and vascular plants. *Ecosystems* **5**, 500-508.

Hogan, E.J., Minnullina, G., Sheppard, L.J., Leith, I.D. and Crittenden, P.D. (2010). Response of phosphomonoesterase activity in the lichen *Cladonia portentosa* to nitrogen and phosphorus enrichment in a field manipulation experiment. *New Phytologist* **186**, 926-933.

Hogg, E.H., Malmer, N. and Wallen, B. (1994). Microsite and regional variation in the potential decay of *Sphagnum magellanicum* in south Swedish raised bogs. *Ecography* **17**, 50-59.

Hogg, P., Squires, P. and Fitter, A.H. (1995). Acidification, nitrogen deposition and rapid vegetational change in a small valley mire in Yorkshire. *Biological Conservation* **71**, 143-153.

Jauhiainen, J., Silvola, J. and Vasander, H. (1998a). The effects of increased nitrogen deposition and CO_2 on *Sphagnum angustifolium* and *Sphagnum warnstorfii*. *Annales Botanicae Fennici* **35**, 247-256.

Jauhiainen, J., Vasander, H. and Silvola, J. (1998b). Nutrient concentration in *Sphagna* at increased N-deposition rates and raised atmospheric CO_2 concentrations. *Plant Ecology* **138**, 149-160.

Johnson, K.W. and Maly, C.C. (1998). Greenhouse studies of *Sphagnum papillosum* for commercial harvest and peatland restoration in Minnesota. *Proceedings of the 1998 International Peat Symposium*, 49-55.

Jokerud, M. (2012). *Impact of nitrogen deposition on species richness and species composition of ombrotrophic mires in Western Norway*. MSc thesis, University of Bergen, Norway.

Joosten, H. (2009). *The Global Peatland CO_2 Picture. Peatland status and drainage related emissions in all countries of the world*. Wetlands International, Ede, the Netherlands.

Keller, J.K., Bridgham, S.D., Chapin, C.T. and Iversen, C.M. (2005). Limited effects of six years of fertilization on carbon mineralization dynamics in a Minnesota fen. *Soil Biology and Biochemistry* **37**, 1197-1204.

Kirkham, F.W. (2001), Nitrogen uptake and nutrient limitation in six hill moorland species in relation to atmospheric nitrogen deposition in England and Wales. *Journal of Ecology* **89**, 1041-1053.

Kivimäki, S. K., Sheppard, L.J., Leith, I.D. and Grace, J. (2013). Long-term enhanced nitrogen deposition increases ecosystem respiration and carbon loss from a *Sphagnum* bog in the Scottish Borders. *Environmental and Experimental Botany* **90**, 53-61.

Koerselman, W. and Verhoeven, J.T.A. (1992). Nutrient dynamics in mires of various trophic status: nutrient inputs and outputs and the internal nutrient cycle. In: Verhoeven, J.T.A. (ed.). *Fens and bogs in the Netherlands; Vegetation, history, nutrient dynamics and conservation*. Kluwer, Dordrecht, 397-432.

Kool, A. and Heijmans, M.M.P.D. (2009). Dwarf shrubs are stronger competitors than graminoid species at high nutrient supply in peat bogs. *Plant Ecology* **204**, 125-134.

Lamers, L.P.M., Bobbink, R. and Roelofs, J.G.M. (2000). Natural nitrogen filter fails in polluted raised bogs. *Global Change Biology* **6**, 583-586.

Lee, J.A. and Studholme C.J. (1992). Responses of *Sphagnum* species to polluted environments. In: Bates, J.W. and Farmer A.M. (eds.). *Bryophytes and lichens in a changing environment.* Clarendon Press, Oxford, 314-322.

Limpens, J., Berendse, F. and Klees, H. (2003a). N deposition affects N availability in interstitial water, growth of *Sphagnum* and invasion of vascular plants in bog vegetation. *New Phytologist* **157**, 339-347.

Limpens, J., Raymakers, J.T.A.G., Baar, J., Berendse, F. and Zijlstra, J.D. (2003b). The interaction between epiphytic algae, a parasitic fungus and *Sphagnum* as affected by N and P. *Oikos* **103**, 59-68.

Limpens, J., Tomassen, H.B.M. and Berendse, F. (2003c). Expansion of *Sphagnum fallax* in bogs: striking the balance between N and P availability. *Journal of Bryology* **25**, 83-90.

Limpens, J., Berendse, F. and Klees, H. (2004). How phosphorus availability affects the impact of nitrogen deposition on *Sphagnum* and vascular plants in bogs. *Ecosystems* **7**, 793-804.

Lund, M., Christensen, T.R., Mastepanov, M. Lindroth, A. and Ström, L. (2009). Effects of N and P fertilization on the greenhouse gas exchange in two northern peatlands with contrasting N deposition rates. *Biogeosciences* **6**, 2135-2144.

Lütke Twenhöven, F. (1992). Competition between two *Sphagnum* species under different deposition levels. *Journal of Bryology* **17**, 71-80.

Malmer, N. (1988). Patterns in the growth and accumulation of inorganic constituents in the *Sphagnum* cover on ombrotrophic bogs in Scandinavia. *Oikos* **53**, 105-120.

Malmer, N., Svensson, B. and Wallén, B. (1994). Interactions between *Sphagnum* mosses and field layer vascular plants in the development of peat-forming systems. *Folia Geobotanica & Phytotaxonomica* **29**, 483-496.

Malmer, N. and Wallén B. (1999). The dynamics of peat accumulation on bogs: mass balance of hummocks and hollows and its variation throughout a millennium. *Ecography* **22**, 736-750.

Malmer, N., Albinsson, C., Svensson, B., and Wallén, B. (2003). Interferences between *Sphagnum* and vascular plants: effects on plant community structure and peat formation. *Oikos* **100**, 469-482

Malmer, N. and Wallén, B. (2004). Input rates, decay losses and accumulation rates of carbon in bogs during the last millennium: internal processes and environmental changes. *The Holocene* **14**, 111-117.

Malmer, N. and Wallén, B. (2005). Nitrogen and phosphorus in mire plants: variation during 50 years in relation to supply rate and vegetation type. *Oikos* **109**, 539-554.

Marchand-Roy, M. (2009). *L'effet fertilisant de la grande oie des neiges: cinq ans de suivi de l'azote et du phosphore dans les polygones de tourbe de l'île bylot au Nunavut*. PhD thesis Université Laval, Quebec, Canada.

Martí, M., Nilsson, M.B., Danielsson, Å., Lindgren, P-E. and Svensson, B.H. (2019). Strong long-term interactive effects of warming and enhanced nitrogen and sulphur deposition on the abundance of active methanogens in a boreal oligotrophic mire. *Mires and Peat* **24**, 1-14.

Millett, J., Leith, I.D., Sheppard, L.J. and Newton, J. (2012). Response of *Sphagnum papillosum* and *Drosera rotundifolia* to reduced and oxidized wet nitrogen deposition. *Folia Geobotanica* **47**, 179-191.

Mitchell, E.A.D., Buttler, A., Grosvernier, P., Rydin, H., Siegenthaler, A. and Gobat, J.M. (2002). Contrasted effects of increased N and CO_2 supply on two keystone species in peatland restoration and implications for global change. *Journal of Ecology* **90**, 529-533.

Mitsch, W.J. and Gosselink, J.G. (2000). *Wetlands* (3rd ed.). Wiley, Chichester.

Morris, J.T. (1991). Effects of nitrogen loading on wetland ecosystems with particular reference to atmospheric deposition. *Annual Review of Ecology and Systematics* **22**, 257-279.

Munzi S., Sheppard, L.J., Leith, I.D., Cruz, C., Branquinho, C., Bini, L., Gagliardi, A., Cai, G. and Parrotta, L. (2017). The cost of surviving nitrogen excess: energy and protein demand in the lichen *Cladonia portentosa* as revealed by proteomic analysis. *Planta* **245**, 819-833.

Munzi, S., Cruz, C., Branquinho, C., Cai, G., Faleri, C., Parrotta, L., Bini, L., Gagliardi, A., Leith, I.D. and Sheppard, L.J. (2020). More tolerant than expected: Taking into account the ability of *Cladonia portentosa* to cope with increased nitrogen availability in environmental policy. *Ecological Indicators* **119**, 106817.

Nichols, J.E. and Peteet, D.M. (2019) Rapid expansion of northern peatlands and doubled estimate of carbon storage. *Nature Geoscience* **12**, 917-921.

Nordbakken, J.F., Ohlsen, M. and Högberg, P. (2003). Boreal bog plants: nitrogen sources and uptake of recently deposited nitrogen. *Environmental Pollution* **126**, 191-200.

Nordin, A. and Gunnarsson, U. (2000). Amino acid accumulation and growth of *Sphagnum* under different levels of N deposition. *Ecoscience* **7**, 474-480.

Nykänen, H., Vasander, H., Huttunen, J.T. and Martikainen, P.J. (2002). Effect of experimental nitrogen load on methane and nitrous oxide fluxes on ombrotrophic boreal peatland. *Plant and Soil* **242**, 147-155.

Paulissen, M.P.C.P. (2004). *Effects of nitrogen enrichment on bryophytes in fens*. PhD thesis, University of Utrecht.

Paulissen, M.P.C.P., Van der Ven, P.J.M., Dees, A.J. and Bobbink, R. (2004). Differential effects of nitrate and ammonium on three fen bryophyte species in relation to pollution nitrogen input. *New Phytologist* **164**, 451-458.

Paulissen, M.P.C.P., Espasa Besalú, L., De Bruijn, H., Van der Ven, P.J.M. and Bobbink, R. (2005). Contrasting effects of ammonium enrichment on fen bryophytes. *Journal of Bryology* **27**, 109-117.

Paulissen, M.P.C.P., Bobbink, R., Robat, S.A. and Verhoeven J.T.A. (2016). Effects of reduced and oxidised nitrogen on rich-fen mosses: a 4-year field experiment. *Water Air Soil Pollution* **227**: 18.

Phoenix, G. K., Emmett, B.A., Britton, A.J., Caporn, S.J.M., Dise, N.B., Helliwell, R., Jones, L., Leake, J.R., Leith, I.D., Sheppard, L.J., Sowerby, A., Pilkington, M.G., Rowe, E.C., Ashmore, M.R. and Power, S.A. (2012). Impacts of atmospheric nitrogen deposition: responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments. *Global Change Biology* **18**, 1197-1215.

Phuyal, M., Artz, R.R.E., Sheppard, L., Leith, I.D. and Johnson, D. (2008). Long-term nitrogen deposition increases phosphorus limitation of bryophytes in an ombrotrophic bog. *Plant Ecology* **196**, 111-121.

Pilkington, M., Caporn, S. J. M., Carroll, J. A., Cresswell, N., Lee, J., Ashenden, T. W., Brittain, S. A., Reynolds, B. and Emmett, B. A. (2005). Effects of increased deposition of atmospheric nitrogen on an upland moor: leaching of N species and soil solution chemistry. *Environmental Pollution* **135**, 29-40.

Pitcairn, C.E.R., Fowler, D. And Grace, J. (1995). Deposition of fixed atmospheric nitrogen and foliar nitrogen content of bryophytes and *Calluna vulgaris* (L.) Hull. *Environmental Pollution* **88**, 193-205

Pouliot, R., L. Rochefort and G. Gauthier (2009). Moss carpets constrain the fertilizing effects of herbivores on graminoids plants in arctic polygon fens. *Botany* **87**: 1209-1222.

Press, M.C., Woodin, S.J. and Lee, J.A. (1988). The potential importance of increased atmospheric nitrogen supply to the growth of ombrotrophic *Sphagnum* species. *New Phytologist* **103**, 45-55.

Redbo-Torstensson, P. (1994). The demographic consequences of nitrogen fertilization of a population of sundew, *Drosera rotundifolia*. *Acta Botanica Neerlandica* **43**, 175-188.

Risager, M. (1998). *Impacts of nitrogen on* Sphagnum *dominated bogs with emphasis on critical load assessment.* PhD thesis. University of Copenhagen.

Saarinen, T. (1998) Internal C N balance and biomass partitioning of *Carex rostrata* grown at 3 levels of nitrogen supply. *Canadian Journal of Botany* **76**, 762-768.

Saarnio, S., Järviö, S., Saarinen, T., Vasander, H. and Silvola, J. (2003). Minor changes in vegetation and carbon balance in a boreal mire under a raised CO_2 or NH_4NO_3 supply. *Ecosystems* 6, 46-60.

Saiz, E., Sgouridis, F., Drijfhout, F.P., Peichl, M., Nilsson, M.B. and Ullah, S. (2021). Chronic atmospheric reactive nitrogen deposition suppresses biological nitrogen fixation in peatlands. *Environmental Science & technology* **55**, 1310-1318.

Schaminée J.H.J., M. Chytrý, M. Hájek, S.M. Hennekens, J.A.M. Janssen, M. Jiroušek, I. Knollová, C. Marcenò, T. Peterka, J.S. Rodwell, L. Tichý and all data providers (2019). *Updated crosswalks of the revised EUNIS Habitat Classification with the European Vegetation Classification and the European ed List Habitats for EUNIS coastal habitats and wetlands Formal query routines and indicator species*. Report Wageningen Environmental Research, Wageningen, the Netherlands.

Sheppard, L.J., Crossley, A., Leith, I.D., Hargreaves, K.J., Carfrae, J.A., Van Dijk, N., Cape, J.N., Sleep, D., Fowler, D. and Raven, J.A. (2004). An automated wet deposition system to compare the effects of reduced versus oxidized N on ombrotrophic bog species: practical considerations. *Water Air Soil Pollution: Focus* **4**, 197-205.

Sheppard, L. J., Leith, I.D., Mizunuma, T., Cape, J.N., Crossley, A., Leeson, S., Sutton, M.A., Van Dijk, N. and Fowler, D. (2011). Dry deposition of ammonia gas drives species change faster than wet deposition of ammonium ions: evidence from a long-term field manipulation. *Global Change Biology* **17**, 3589- 3607.

Sheppard L.J., Leith, I.D., Leeson, S.R., Van Dijk, N., Field, C. and Levy, P. (2013). Fate of N in a peatland, Whim bog: immobilisation in the vegetation and peat, leakage into pore water and losses as N_2O depend on the form of N. *Biogeosciences* **10**, 149-160.

Sheppard, L. J., Leith, I.D, Mizunuma, T., Leeson, S., Kivimaki, S., Cape, J.N., Van Dijk, N. Leaver, D., Sutton, M.A. and Fowler, D. (2014). Inertia in an ombrotrophic bog ecosystem in response to 9 years' realistic perturbation by wet deposition of nitrogen, separated by form. *Global Change Biology* **20**, 566-580.

Stevens C.J., Manning, P., Van den Berg, L.J.L., De Graaf, M.C.C., Wamelink, G.W.W., Boxman, A.W., Bleeker, A. Vergeer, P., Arroniz-Crespo, M. Limpens, J. Lamers, L.P.M., Bobbink, R. and Dorland, E. (2011). Ecosystem responses to reduced and oxidised nitrogen inputs in European terrestrial habitats. *Environmental Pollution* **159**, 665-676.

Stevens, C.J., Smart, S.M., Henrys, P.A., Maskell, L.C., Crowe, A., Simkin, J., Cheffings, C.M., Whitfield, C., Gowing, D.J.G., Rowe, E.C., Dore, A.J. and Emmett, B.A. (2012). *Ecological Indicators* **20**, 196-203.

Strandberg, M., Damgaard, C., Degn, H.J., Bak, J. and Nielsen, K.E. (2012). Evidence for acidification-driven ecosystem collapse of Danish *Erica tetralix* wet heathland. *AMBIO* **41**, 393-401.

Tipping, E., Henrys, P.A., Maskell, L.C. and Smart, S.M. (2013). Nitrogen deposition effects on plant species diversity; threshold loads from field data. *Environmental Pollution* **179**, 218-223.

Tomassen, H.B.M., Smolders, A.J.P., Limpens, J., Van Duinen, G., Van der Schaaf, S., Roelofs, J., Berendse, F., Esselink, H. and Van Wirdum, G. (2002). *Onderzoek ten behoeve van herstel en beheer van Nederlandse hoogvenen*. Leerstoelgroep Aquatische Ecologie en Milieubiologie. University of Nijmegen, Nijmegen (in Dutch).

Tomassen, H.B.M., Smolders, A.J.P., Lamers, L.P.M. and Roelofs, J.G.M. (2003). Stimulated growth of *Betula pubescens* and *Molinia caerulea* on ombrotrophic bogs: role of high levels of atmospheric nitrogen deposition. *Journal of Ecology* **91**, 357-370.

Tomassen, H.B.M., Smolders, A.J.P., Limpens, J., Lamers, L.P.M. and Roelofs, J.G.M. (2004). Expansion of invasive species on ombrotrophic bogs: desiccation or high N deposition levels? *Journal of Applied Ecology* **41**, 139-150.

Van Breemen, N. (1995). How *Sphagnum* bogs down other plants. *Trends in Ecology and Evolution* **10**, 270- 275.

Van den Berg, L.J.L., Jones, L., Sheppard, L.J., Smart, S.M, Bobbink, R., Dise, N.B. and Ashmore, M.R. (2016). Evidence for differential effects of reduced and oxidised nitrogen deposition on vegetation independent of nitrogen load. *Environmental Pollution* **208**, 890-897.

Van der Molen, P.C. (1992). *Hummock-hollow complexes on Irish raised bogs*. PhD thesis, University of Amsterdam, Amsterdam.

Van Diggelen, J.M.H., Van Dijk, G., Cusell, C., Van Belle, J., Kooijman, A., Van den Broek, T., Bobbink, R., Mettrop, I.S., Lamers, L.P.M. and Smolders, A.J.P. (2018). *Onderzoek naar de effecten van stikstof in overgangs- en trilvenen. Ten behoeve van het behoud en herstel van habitattype H7140 (Natura 2000)*. Report 2018/OBN220-LZ, VBNE, Driebergen, the Netherlands (summary in English).

Van Geel, M., Jacquemyn, H., Peeters, G., Van Acker, K., Honnay, O. and Ceulemans, T. (2020). Diversity and community structure of ericoid mycorrhizal fungi in European bogs and heathlands across a gradient of nitrogen deposition. *New Phytologist* **228**, 1640-1651.

Verhoeven, J.T.A. and Schmitz, M.B. (1991). Control of plant growth by nitrogen and phosphorus in mesotrophic fens. *Biogeochemistry* **12**, 135-148.

Verhoeven, J.T.A., Beltman, B., Dorland, E., Robat, S.A. and Bobbink, R. (2011). Differential effects of ammonium and nitrate deposition on fen phanerogams and bryophytes. *Applied Vegetation Science* **14**, 149-157.

Vermeer, J.G. (1986). The effects of nutrients on shoot biomass and species composition of wetland and hayfield communities. *Acta Oecol./Oecol. Plant.* **7**, 31-41.

Vitt, D. H., House, M., Kitchen, S. and Wieder, R.K. (2020). A protocol for monitoring plant responses to changing nitrogen deposition regimes in Alberta bogs. *Environmental Monitoring and Assessment* **192**, 1-25.

Wamelink, G.W.W., Goedhart, P.W., Roelofsen, H.D., Bobbink, R., Posch, M. and Van Dobben, H.F. (2021). *Relaties tussen de hoeveelheid stikstofdepositie en de kwaliteit van habitattypen*. Wageningen Environmental Research.

Wassen, M.J., Van der Vliet, R.E. and Verhoeven, J.T.A. (1998). Nutrient limitation in the Biebrza fens and floodplain (Poland). *Acta Botanica Neerlandica* **47**, 241-253.

Wieder, R.K., Vitt, D.H., Vile, M.A., Graham, J.A., Hartsock, J.A., Fillingim, H., House, M. Quinn, J.C., Scott, K.D. and Petix, M. (2019). Experimental nitrogen addition alters structure and function of a boreal bog: critical load and thresholds revealed. *Ecological Monographs* **89**, e01371.

Wieder, R. K., Vitt, D.H., Vile, M.A., Graham, J.A., Hartsock, J.A., Popma, J.P.A., Fillingim, H., House, M. Quinn, J.C., Scott, K.D., Petix, M. and McMillen, K.J. (2020). Experimental nitrogen addition alters structure and function of a boreal poor fen: Implications for critical loads. *Science of the Total Environment* **733**, 138619.

Wiedermann. M.M., Nordin, A., Gunnarsson, U., Nilsson, M.B., and Ericson, L. (2007). Global change shifts vegetation and plant-parasite interactions in a boreal mire. *Ecology* **88**, 454-464.

Wiedermann. M.M., Gunnarsson, U., Ericson, L. and Nordin, A. (2009a). Ecophysiological adjustment of two Sphagnum species in response to anthropogenic nitrogen deposition. *New Phytologist* **181**, 208-217.

Wiedermann. M.M., Gunnarsson, U., Nilsson, M.B., Nordin, A. and Ericson, L. (2009b). Can small-scale experiments predict ecosystem responses? An example from peatlands. *Oikos* **118**, 449-456.

Wiedermann, M. M. and M. B. Nilsson (2020). Peatland vegetation patterns in a long-term global change experiment find no reflection in belowground extracellular enzyme activities. *Wetlands* **40**, 2321- 2335.

Wilkins K., Aherne, J. and Bleasdale, A. (2016). Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high. *Atmospheric Environment* **146**, 324-331.

Williams, B.L., Buttler, A., Grosvernier, P., Francez, A.J., Gilbert, D., Ilomets, M., Jauhiainen, J., Matthey, Y., Silcock, D.J. and Vasander, H. (1999). The fate of NH_4NO_3 added to *Sphagnum magellanicum* carpets at five European mire sites. *Biogeochemistry* **45**, 73-93.

Williams, B.L. and Silcock, D.J. (2000). Impact of NH_4NO_3 on microbial biomass C and N and extractable DOM in raised bog peat beneath *Sphagnum capillifolium* and *S. recurvum*. *Biogeochemistry* **49**, 259-276.

Woodin, S.J. (1986). *Ecophysiological effects of atmospheric nitrogen deposition on ombrotrophic* Sphagnum *species*. PhD thesis, University of Manchester.

7 Effects of nitrogen inputs in grasslands and lands dominated by forbs, mosses and lichens (EUNIS class R, formerly E)

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Annual xeric grassland constituting the understory of a Dehesa, an open evergreen oak woodland, in Tres Cantos (central Spain). Dehesas and montados are traditional agroforestry systems characteristics of the Mediterranean area. Photo: CIEMAT.

Summary

In this chapter empirical N critical loads ($CL_{emp}N$) for grasslands and lands dominated by forbs, mosses and lichens have been updated and revised, if necessary. A number of experimental N addition experiments and gradient studies have become available in the present revision period. Based on these studies the $CL_{emp}N$ of four grassland types should be lowered. Calcareous grassland communities (R1A and R1B) are given a $CL_{emp}N$ of 10-20 kg N ha^{-1} yr^{-1}, Mediterranean grassland communities (R1D, R1E and R1F) are given a $CL_{emp}N$ of 5-15 kg N ha^{-1} yr^{-1}, acidic grassland communities (R1M) are given a $CL_{emp}N$ of 6-10 kg N ha^{-1} yr^{-1}, and inland dune grassland communities (R1P and R1Q) are given a $CL_{emp}N$ of 5-15 kg N ha^{-1} yr^{-1}. Although there is good evidence to support the $CL_{emp}N$ in some habitats long-term N addition studies with low doses of N application and gradient studies are needed in Mediterranean grassland (R1D, R1E and R1F), inland dune grasslands (R1P and R1Q), moist or wet mesotrophic to eutrophic hay (R35) and low and medium altitude hay meadows (R22) where $CL_{emp}N$ are based on expert judgement.

7.1 Introduction

A large number of grassland ecosystems (class R of the European Nature Information System (EUNIS), formerly E) are found across Europe: from very dry to wet habitats, acidic to alkaline conditions, inland saline soils, those adapted to high concentrations of heavy metals and very different climatic regimes (e.g. Ellenberg, 1988; Davies et al., 2004, Dengler et al., 2020). Only a small proportion of these grasslands are of natural origin (e.g. dry steppe grasslands, alpine grasslands), while some of these habitats are strongly intensified, some are covered by semi-natural vegetation (Dengler et al., 2020). Traditional agricultural use or management is thus an important ecological factor influencing the structure and function of these grassland systems. These grasslands have long been an important part of the European landscape and contain many rare and endangered plant and animal species; a number of them have been set aside as nature reserves in several European countries (e.g. Ellenberg, 1988; Dengler et al., 2020). Semi-natural grasslands of conservation importance are generally nutrient-poor, because of the agricultural use involving low levels of manure addition and the removal of plant material by grazing or hay making. The vegetation is often characterised by many species of low stature due to the nutrient-poor nature of soils (Ellenberg, 1988). However, some semi-natural meadow communities of high nature conservation value, particularly those on deep alluvial soils in river flood plains subjected to periodic inundation or inputs of manure, can be moderately fertile with soil macro-nutrients levels at the higher end of the spectrum covering species-rich grasslands. These are likely to have a higher proportion of relatively fast-growing species than, for example, oligotrophic acidic or calcareous grasslands. To maintain high species diversity, it is recommended that artificial fertilisers and slurry should be avoided. Given the sensitivity to artificial fertilisers, it is thus likely that several of these species-rich grasslands, especially those of oligotrophic or mesotrophic soils, will also be sensitive to increased atmospheric nitrogen (N) inputs. Moreover, several of the most species-rich grasslands are found under weakly buffered or almost neutral soil conditions, which make them sensitive to soil acidification and very sensitive to the negative impacts of ammonium accumulation in the case of high deposition of reduced N (see Chapter 1).

Grasslands and lands dominated by forbs, mosses or lichens (EUNIS class R), which are dry or only seasonally wet (i.e. with the water table at or above ground level for less than half of the year) usually have a vegetation cover greater than 30%. The dominant part of the vegetation is grasses, sedges and other non-woody plants, including moss-, lichen- and fern- and sedge-dominated communities. An important level of division is based on soil water availability (dry (R1), mesic (R2) and wet (R3) grasslands). Most of the studies on the effects of N in grassland habitats have been carried out for ecosystems which are classified as dry grasslands (R1). Furthermore, the impacts of N inputs have only been studied in parts of the other major EUNIS categories (R2-R7). Some information exists on mesic grasslands (R2) and wet grasslands (R3), the classification of which particularly relates to present land use or management. Evidence also exists on the impacts of N deposition in alpine and subalpine grasslands (R4).

7.2 Dry grasslands (R1)

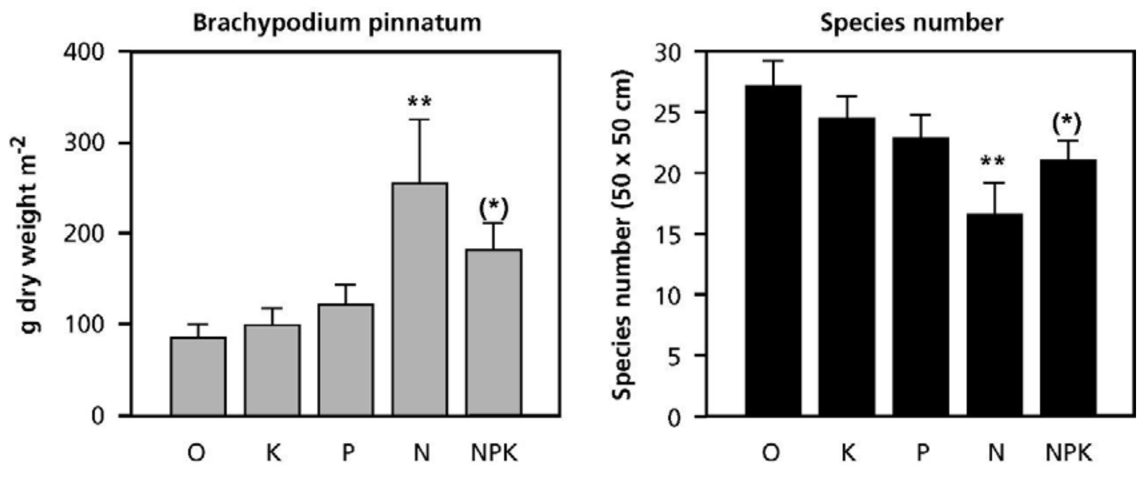
7.2.1 Semi-dry perennial calcareous grassland (meadow steppe, R1A)

Calcareous grasslands (EUNIS category R1A) are communities developed on soils derived from limestone as parent materials, which are widespread in the hilly and mountainous regions of western and central Europe. Subsoils consist of different kinds of limestone with high contents of calcium carbonate (> 90%), covered by shallow rendzina soils, low in plant-available phosphorus (P) and N (pH of the top soil: 7-8 with a calcium carbonate content of approximately

10%). Plant productivity is low and calcareous grasslands are among the most species-rich plant communities in Europe, including a large number of rare and endangered species (Dengler et al., 2020). These grasslands decreased strongly in area during the second half of the last century due to urbanisation and conversion to agriculture (e.g. Wolkinger and Plank, 1981; Ratcliffe, 1984). To maintain the characteristic calcareous vegetation, a specific management is needed in most situations to prevent its natural succession towards woodland (Wells, 1974; Dierschke, 1985).

In the late 1970s and early 1980s, a gradual increase in one grass species (*Brachypodium pinnatum)* had been observed in Dutch calcareous grasslands, although management of these grasslands (hay making in autumn) had not been changed since the mid-1950s. Since then, it has been hypothesised that the increase in atmospheric deposition of N (from 10-15 kg N ha^{-1} yr^{-1} in the 1950s to 25-35 kg N ha^{-1} yr^{-1} in the 1980s) caused this drastic change in vegetation composition (Bobbink and Willems, 1987). These effects of N enrichment were investigated in two field experiments in the Netherlands (Bobbink et al., 1988; Bobbink, 1991). Application of ammonium nitrate (50-100 kg N ha^{-1} yr^{-1}, in addition to an ambient load of 30-35 kg N ha^{-1} yr^{-1}) over a three-year period resulted in a drastic increase in the grass *B. pinnatum,* and in a strong reduction in species diversity (including several Dutch Red List species) (Figure 7.1). *B. pinnatum* is very efficient at both acquiring N from the soil and re-allocating it from senescing shoots to its well-developed rhizome system. It benefits from the extra N invested into the below-ground rhizomes by enhanced growth in the next spring. In this way *B. pinnatum* strongly monopolised the N storage (>75%) in both the above-ground and below-ground plant compartments in response to increasing N availability (Bobbink et al., 1988; 1989; De Kroon and Bobbink, 1997). Besides this decrease in phanerogamic plant species due to increased N deposition and the spread of *Brachypodium pinnatum*, many characteristic lichens and mosses have disappeared from Dutch calcareous grasslands since the 1960s (During and Willems, 1986). This has partly been caused by the (indirect) effects of extra N inputs, as shown in experiments by Van Tooren et al. (1990).

Figure 7.1. Above-ground biomass of *Brachypodium pinnatum* (g m^{-2}) and vascular plant species number (per 50 x 50 cm) on Dutch perennial calcareous grassland (R1A) after three years of N application in the form of ammonium nitrate (100 kg N ha^{-1} yr^{-1}). O = control.



Source: Bobbink, 1991

From the 1950s to the mid-1980s, almost all of the calcareous grasslands in the Netherlands were mowed and the hay was removed. By the removal of the hay, between 17 and 22 kg N ha^{-1} yr^{-1} was taken away from the system under the usual land management (Bobbink, 1991).

Legume species (*Fabaceae*) also occur in these grasslands, and provide an additional N input associated with the N-fixing bacteria in their root nodules (approximately 5 kg N ha^{-1} yr^{-1}). The N mass balance of Dutch calcareous grasslands was summarised in Bobbink et al. (1992), and a critical load of N was estimated using a steady-state mass balance model (e.g. De Vries et al., 1994). Assuming a long-term immobilisation rate for N of between 0 and 6 kg N ha^{-1} yr^{-1}, the critical load of N could be derived by adding the N fluxes due to net uptake, denitrification and leaching, corrected for the N input from fixation. In this way, the authors determined 15 to 25 kg N ha^{-1} yr^{-1} as the critical load for N (Bobbink et al., 1992). This range is in close accordance with the results found by Neitzke (1998, 2001) for calcareous grasslands in the eastern Eifel in south-west Germany. In a gradient of nutrient enrichment extending along a transect away from an agricultural field, N mineralisation was found to explain the variation in species composition and species degradation, from the nutrient enriched border zone to the intact central calcareous grassland. Comparing the soil N mineralisation rates of the undisturbed calcareous grasslands and the plots with significantly altered species composition, *B. pinnatum* increased in cover, and species diversity decreased when N mineralisation increased from 6 to 10 kg N ha^{-1} yr^{-1} in the unaffected parts to between 35 and 55 kg N ha^{-1} yr^{-1} in the areas adjacent to the agricultural fields (Neitzke, 1998, 2001).

Following a survey of data from a number of conservation sites in southern England, Pitcairn et al. (1991) concluded that *B. pinnatum* had expanded in the United Kingdom (UK) during the last century. They considered that much of the early spread could be attributed to a decreased grazing pressure, but that more recent increases in the grass, in some cases, had occurred despite grazing or mowing, and may have been related to increased N inputs. A retrospective study of a heavily grazed calcareous grassland at Parsonage Downs (UK), however, showed no substantial change in species composition between 1970 and 1990, a period for which N deposition is thought to have increased from probably below 10 kg N ha^{-1} yr^{-1} to 15 to 20 kg N ha^{-1} yr^{-1} (Wells et al., 1993). *B. pinnatum* was present in the sward but did not expand as in the Dutch grasslands.

In studies on calcareous grasslands in England, additions of N hardly stimulated a dominance of grasses (Smith et al., 1971; Jeffrey and Pigott, 1973). However, with applications of 50 to 100 kg N ha^{-1} yr^{-1} and further additions of P, a dominance of the grasses *Festuca rubra*, *Festuca ovina* or *Agrostis stolonifera* was observed. However, *B. pinnatum* or *Bromus erectus*, the most frequent species on continental calcareous grasslands, were absent from these British sites, so the data are not comparable in that respect. Van den Berg et al. (2011) conducted a survey of 46 plots on calcareous grassland in nature reserves in the UK. The plots were first surveyed between 1990 and 1993 and then re-surveyed between 2005 and 2009. The survey found a linear increase in the grass:herb ratio that was apparent from the lowest levels of deposition. The results also showed a decline in Shannon diversity and evenness as well as a decline in characteristic calcareous grassland species and rare species. The change in both Shannon diversity and evenness became a negative one around 20-25 kg N ha^{-1} yr^{-1}. Structural equation modelling indicated that direct effects of N deposition were the dominant mechanism for change in Shannon diversity. Changes in the abundance of individual species were observed even in the lowest N deposition range of 0-15 kg N ha^{-1} yr^{-1}. Despite these changes in species composition there was no decline in species richness associated with N deposition nor a change in average Ellenberg N or the number of eutrophic or oligotrophic species.

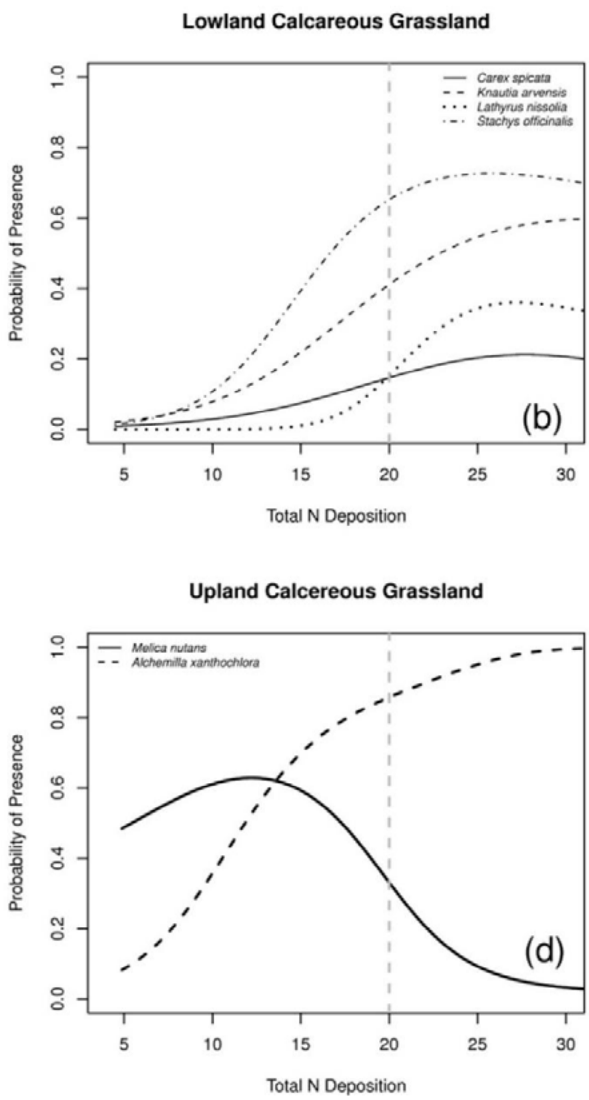
Diekmann et al. (2014) also found changes in species composition and richness by analysing published data from 1061 plots in *Bromion erecti* grasslands in north-west Germany dating from 1936 onwards plus 125 plots sampled in 2008. Whilst declines in species richness were not observed, there was a decline in specialist and rare species and an increase in generalist and taller species. Detrended correspondence analysis (DCA) showed that Ellenberg N score was aligned with the primary axis of variation. *B. pinnatum* did not increase in relation to N deposition.

A series of studies from the UK have used data from the UK Countryside Survey to examine relationships between species richness and composition and N deposition. The UK Countryside Survey is a national survey of randomly located 1 x 1 km squares across the UK. Within these squares are various quadrat sizes including 2 x 2 m plots where vegetation is recorded. Surveys were conducted in 1998 and 2007. There were 94 calcareous grassland plots used in the analysis by Maskell et al. (2010), and 92 in the analysis by Tipping et al. (2013) from 1998, and 869 in Van den Berg et al. (2016) from 2007 where a different definition of calcareous grasslands was used. Maskell et al. (2010) found no relationship between species richness and N deposition for calcareous grassland. Using cover-weighted values from trait databases they noticed significant increases in canopy height, grass:forb ratio, Ellenberg N and leaf N concentration. Also using data from 2007, Tipping et al. (2013) applied non-parametric quantile regression with a breakpoint aiming at estimating the breakpoint where there was a clear effect of N deposition on species richness. They estimated 23.6 kg N ha^{-1} yr^{-1} as the breakpoint. Van den Berg et al. (2016) undertook further analysis once new data were collected in 2007. They found that total N deposition was positively related to species richness. The larger number of plots enabled them to examine relationships with NH_x and NO_y separately. NH_x:NO_y ratio was significantly negatively related to richness. Grass:forb ratio significantly increased as N deposition increased. There was a large variation between communities classified as calcareous grasslands in Van den Berg et al. (2016) and the authors found that breaking the category down to examine the most common grassland groups showed no significant relationships with N deposition. The authors suggested variation in local management may be responsible for the lack of observable impact.

A four-year N addition experiment was performed in a calcareous grassland (R1A) in Voeren, Belgium (Jacquemyn et al., 2003). In this study, the effects of N additions at three levels (30, 60 and 90 kg N ha^{-1} yr^{-1}), in the form of ammonium nitrate (added once a year), were examined in a factorial experiment with two management treatments (high density cattle grazing and mowing with removal of the hay). The background deposition at the site was approximately 20 kg N ha^{-1} yr^{-1} (modelled EMEP data). Species richness decreased significantly, from around 25 to 29 vascular plant species per m^2 in the control area, to between 18 and 20 species after additions of 30 kg N ha^{-1} yr^{-1} and between 15 and 18 species after the two highest N additions. Much of the reduction in species richness could be attributed to decreased light availability, resulting from increased above-ground productivity of tall grasses and forb species. In contrast to the results in Dutch calcareous grasslands, *B. pinnatum* was not present on this study site, although other tall grasses and forbs became very abundant.

Henrys et al. (2011) undertook a spatial analysis using data collected from two surveys on species presence across the UK. The Vascular Plant Database records vascular plant species at 10 km resolution for the period 1987-1999. The second data set was the BSBI Local Change Database which records wild vascular plants in 811 2 x 2 km tetrads set within a regular grid of hectads (10 x 10 km) across the UK. For both datasets, species were assigned to a habitat, habitat generalists were excluded, and survey squares were designated as upland or lowland. Generalised additive models (GAMs) were employed to look at relationships between species occurrence or Ellenberg scores averaged per habitat, and N deposition, whilst accounting for the effect of other environmental variables (intensity of land-use in each grid square; minimum January temperature; maximum July temperature; total annual rainfall; and change in sulphur deposition). For the Vascular Plant Database 40 species in lowland calcareous grasslands were analysed by the GAM models, 17 of these species showed a significant relationship in their presence with N deposition, 9 of these declined from 10 kg N ha^{-1} yr^{-1} or less. Four species showed positive relationships, all increasing their presence at around 25 kg N ha^{-1} yr^{-1}. In upland calcareous grasslands, there were only sufficient data to analyse 7 species, 2 of these showed significant relationships with N, one increased steadily from lowest levels of N deposition the other declined rapidly after 15 kg N ha^{-1} yr^{-1} (Figure 7.2). In the BSBI Local Change dataset 17 species were analysed in lowland calcareous grasslands, 8 of these showed a significant relationship with N deposition, some of these were complex relationships but a number showed steep declines from the lowest levels of deposition. In upland grasslands, there were three species which showed significant relationships with N deposition and all showed a hump-shaped relationship. Only one species showed a decrease in both datasets (*Ononis repens*- a typical Mesobromion species), while other species tended to have a significant relationship in one dataset but non-significant in the other. In the Vascular Plant database, Ellenberg N increased with increasing N deposition for lowland and upland calcareous grasslands, whereas in the BSBI dataset these increases were not always linear, but tended to increase from lowest levels of N deposition in lowland calcareous grasslands.

Figure 7.2. Change in probability of presence for species showing statistically significant relationships to N deposition in calcareous grasslands in the Vascular Plant database against increasing total inorganic N deposition (kg N ha^{-1} yr^{-1}).



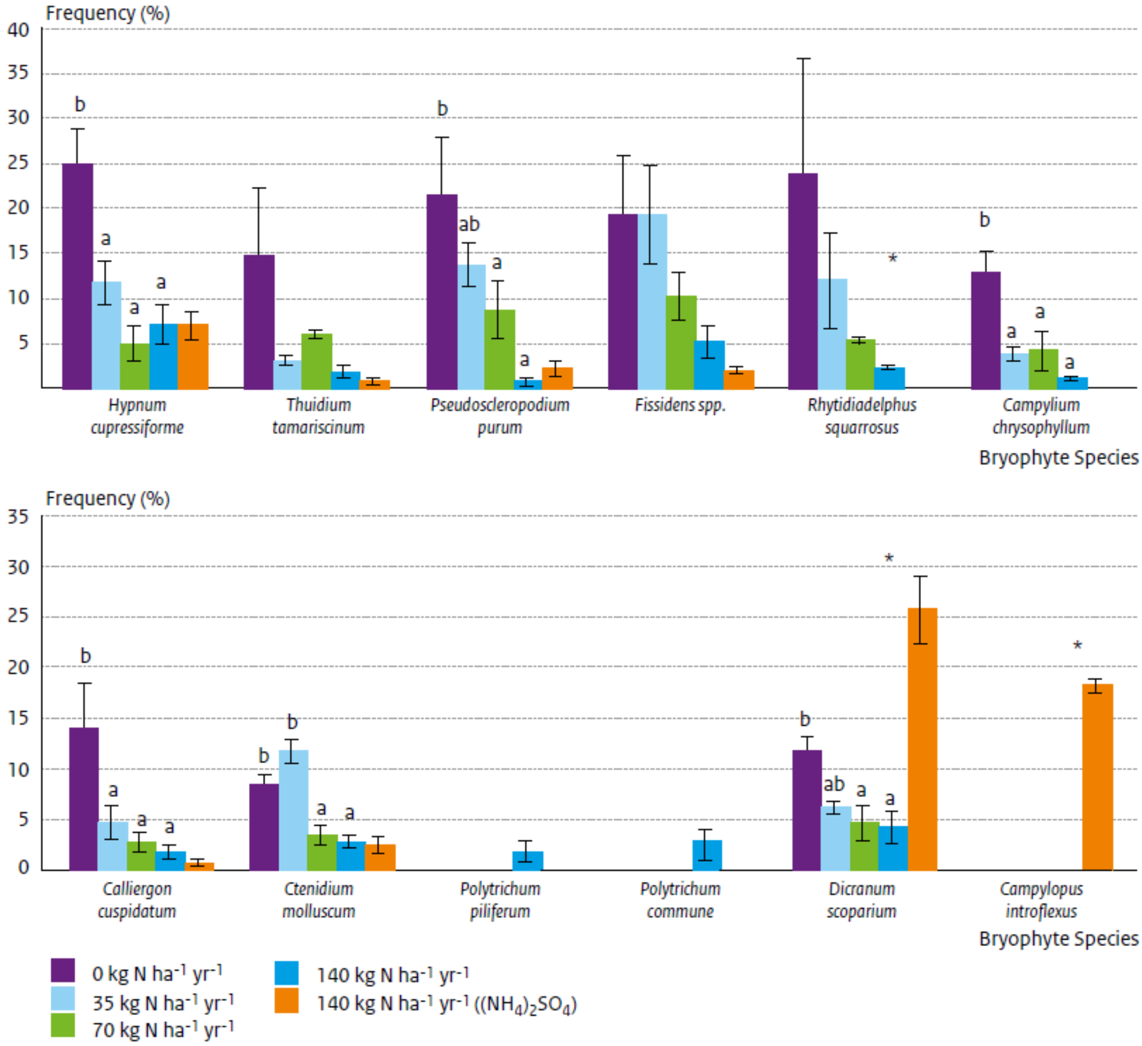
Source: Henrys et al., 2011

Stevens et al. (2012) took a very similar approach using spatial analysis with data from the British Lichen Society database over a 50-year period. Data were analysed at a 10 km resolution and as in Henrys et al. (2011), generalised additive models were applied to look at occurrence in relation to N deposition accounting for other environmental drivers. To be included in the analysis, species had to be terricolous (i.e. growing on the soil or ground), largely associated with one habitat, not too scarce or regionally distributed and accurately represented in the database. Sixteen lichen species were analysed of which three showed a significant relationship with N deposition. *Cladonia foliacea* declined significantly with increasing N deposition above 10 kg N ha^{-1} yr^{-1}.

Long-term effects of ammonium nitrate additions (35, 70 and 140 kg N ha^{-1} yr^{-1}) were studied between 1990 and 2008, in a calcareous grassland area (Wardlow Hay Cop) with a shallow soil (< 10 cm) on limestone bedrock in Derbyshire, in the UK (Morecroft et al., 1994; Carroll et al., 1997; Lee and Caporn, 1999; Carroll et al., 2003; Phoenix et al., 2003; 2004; Haworth et al., 2007; Horswill et al., 2008, O'Sullivan, 2011). The highest N addition was also applied in the form of ammonium sulphate. Within the first four years of N addition, no significant change in species composition of vascular plants was observed. From the sixth year onwards, however, there was a marked and significant dose-related decline in vascular plant cover with increased N addition (Carroll et al., 1997; Lee and Caporn, 1999; Carroll et al., 2003). The species that were negatively affected by N included a range of subordinate species (e.g. *Thymus*) typical of calcareous grasslands. In contrast, *Hypochaeris radicata* increased in the N-enriched vegetation (Carroll et al., 1997; Lee and Caporn, 1999). Overall, vegetation responses of vascular plants were slow, and significant changes in individual species cover were found mainly at the higher levels of N addition, although the trend could be witnessed from the lowest treatment upwards. Where growth was concerned, no significant increases in response to N were seen at any stage in this experiment, suggesting the strong P limitation of plant production on this calcareous grassland area (Carroll et al., 2003).

Significant changes in bryophyte species composition were also observed in this UK experiment after 12 years of N additions (Haworth et al., 2007) (Figure 7.3). The lowest NH_4NO_3 additions (35 kg N ha^{-1} yr^{-1}) produced significant declines in frequency of *Hypnum cupressiforme*, *Campylium chrysophyllum*, and *Calliergon cuspidatum*. Significant reductions in frequency at higher NH_4NO_3 application rates were recorded for *Pseudoscleropodium purum*, *Ctenidium molluscum*, and *Dicranum scoparium*. The highest NH_4NO_3 and $(NH_4)_2SO_4$ additions provided acidic conditions for the establishment of two typical calcifuges – *Polytrichum* species and *Campylopus introflexus*, respectively. Substrate-surface (0-2.5 cm) pH measurements showed a dose-related reduction in pH with increasing NH_4NO_3 addition rates, with a difference of 1.6 pH units between the control and highest deposition rate, and a further significant fall in pH of more than 1 pH unit, between the NH_4NO_3 and $(NH_4)_2SO_4$ treatments. *Dicranum scoparium* was strongly promoted by the $(NH_4)_2SO_4$ treatment, maybe also due to the strongly decreased pH. There is a clear indication that in this experiment increased N inputs led to soil acidification, especially in the top centimetres of the soil, as was also seen for the full soil layer after ten years of N addition (Horswill et al., 2008). They also found significant losses of soil base cations and increases in aluminium (Al) and manganese (Mn) following the highest N treatments (140 kg N ha^{-1} yr^{-1}; especially with $(NH_4)_2SO_4$); clear signs of soil acidification, which might lead to declines in sensitive calcicole species.

Figure 7.3. Frequency of bryophyte species at Wardlow Hay Cop after 12 years of treatment with different N loads and the impact of sulphate rather than nitrate at 140 kg N a^{-1} yr^{-1}. Significance between N forms is denoted by *



Source: adapted by Bobbink and Hettelingh (2011) based on Haworth et al., 2007

In contrast to the slow vegetation responses, N concentrations in shoots, nitrate reductase activities, and soil N mineralisation and nitrification rates significantly increased during the early years of increased inputs of N (≥ 70 kg N ha^{-1} yr^{-1}) (Morecroft et al., 1994; Carroll et al., 1997). After six years of treatments, N mineralisation rates in summer were significantly higher following all N treatments, but in autumn or winter they were only significant after the two highest N additions. Nitrification was also clearly higher in summer on all N-treated plots, but in autumn and winter, nitrification rates following the 35 kg N ha^{-1} yr^{-1} addition were not significantly different from those of the controls. Soil microbial activity was not significantly affected by six years of ammonium nitrate additions, but root-surface phosphomonoesterase activity increased significantly following the addition of ammonium sulphate (Johnson et al., 1998). In the eighth year of N additions, significant increases in root-surface phosphomonoesterase activities were found for three plant species (a forb, a grass and a sedge) after additions of both 35 and 140 kg N ha^{-1} yr^{-1} in the form of ammonium nitrate (Phoenix et al., 2004). This showed an increased demand for P induced by N enrichment.

Nitrogen cycling and accumulation in calcareous grasslands can be significantly reduced by two major processes: 1) leaching from the soil, and 2) removal through management regimes. N losses by denitrification in dry calcareous grasslands are low (< 1 kg N ha^{-1} yr^{-1}; e.g. Mosier et al., 1981). The N budget and fluxes of this calcareous grassland site have been reported by Phoenix et al. (2003) following six years of N additions. Their major finding was that even sustained, very high inputs of N did not lead to large losses of N. In general, 80 to 90% of the additional N was retained in the system; even 65% remained in the plots with the high 140 N ha^{-1} yr^{-1} additions, a treatment that represented a seven-fold increase of the ambient N deposition. The major fluxes of N loss from this grassland were from biomass removal (simulated grazing) and leaching of organic N, constituting 90% of leached N under ambient conditions. Leaching of nitrate contributed significantly to the output flux of N under the highest N treatment only. Even in this P-limited grassland, a (very) high fraction of the extra N was accumulated (immobilised) in the soil system. After 12 years of N addition O'Sullivan et al. (2011) reported no significant differences in soil ammonium N concentrations between treatments and controls but increases in concentrations of oxidised N.

Unkovich et al. (1998) also found N limitation in their field study in Wytham (UK). They noticed more than a doubling in plant production after weekly additions of 11.5 kg N ha^{-1} for 6 weeks (total N addition of almost 70 kg N ha^{-1}), independently of N form (ammonium or nitrate) and no response to P or any other nutrient. Adding the N in the form of ammonium sulphate or ammonium nitrate increased foliar N concentrations significantly, compared with the controls and also with plots where N was added in the form of potassium nitrate. Plants took up more than 40% of the added N; the remaining additions (almost 60%) were immobilised in the soil (Unkovich et al., 1998).

In a gradient study of 120 plots across 21 calcareous and 19 acidic grasslands in eight countries in Europe with a N deposition range of 4-31 kg N ha^{-1} yr^{-1}, Ceulemans et al. (2014) performed a molecular analysis to identify the impacts of N deposition on mycorrhizal fungi. Richness of arbuscular mycorrhizal operational taxonomic units (OTUs) was negatively related to N deposition in calcareous grasslands, declining steadily from the lowest levels of N deposition. Fungal community composition was significantly affected by N deposition. There were also signs of soil acidification related to N deposition. TITAN (threshold indicator taxa analysis, Baker and King, 2010) was conducted on a combined acidic and calcareous grassland dataset to identify a breakpoint in the data. This analysis revealed a threshold of 7.7 kg N ha^{-1} yr^{-1}; however, this is difficult to relate to $CL_{emp}N$ for a specific type of habitat because calcareous and acidic habitats were combined.

Less information is available for calcareous grasslands in the Mediterranean areas of Europe. A three-year N addition experiment performed in central Italy (Bonamoni et al., 2006) was considered in the 2010 update, but it was included in the 'Mediterranean xeric grassland' (Chapter 7.2.2). In this experiment, the effects of N enrichment, cutting and litter removal were investigated in a species-poor dry Mediterranean grassland. The site represented an abandoned area dominated by *Brachypodium rupestre*, 15 to 20 years after it had last been in agricultural use (rotating crops with legumes), and no management had been carried out since. In the experiment, N had been added once a year, in the form of commercial urea fertiliser, at a rate of 35 kg N ha^{-1} yr^{-1}. Estimated background deposition was less than 15 kg N ha^{-1} yr^{-1}. As for most grasslands without appropriate management, cutting of the vegetation highly reduced above-ground biomass, compared with the uncut controls. However, N additions significantly increased total above-ground biomass in each of three management treatments (uncut, litter removal and cutting), with the cutting treatment leading to the smallest increase in the vegetation. In this experiment, species diversity was quantified using the Shannon index (H), with diversity increasing particularly after cutting of the vegetation, mostly of annual and biennial species, and to a lesser extent after litter removal. Species diversity was lower following all N addition treatments, but this effect was only significant in combination with litter removal. Furthermore, it was evident that the species diversity was highly negatively correlated ($R^2 = 0.85$) with the biomass percentage of *B. rupestre*, a finding similar to that for *B. pinnnatum* in Dutch calcareous grasslands (Bobbink and Willems, 1987).

A similar experiment was conducted in an area nearby but in a species-rich mowed grassland (Bonamoni et al., 2009). In this case it was a calcareous grassland ascribed to the association *Brizo mediae–Brometum erecti*, which is rich in rare and endemic species. The experiment was performed in both the mowed grassland and in an adjacent abandoned area. The mowed site was co-dominated by the grasses *Bromus erectus*, *Briza media*, *Anthoxanthum odoratum* and *Festuca circummediterranea* and the forbs *Rhinanthus personatus*, *Tragopogon pratensis*, *Onobrychis viciifolia* and *Trifolium pratense*, while the abandoned treatment was dominated by the perennial grasses *B. rupestre* and *Dactylis glomerata*. In this experiment a similar methodology to Bonamoni et al. (2006) was followed, investigating the interactive effects of three years of N enrichment, cutting and litter removal. Nitrogen was added once a year, in the form of commercial urea fertiliser, at a rate of 35 kg N ha^{-1} yr^{-1} and estimated background deposition was less than 15 kg N ha^{-1} yr^{-1}. As found in the previous experiment, cutting significantly increased species diversity in the abandoned area expanding the cover of almost all annual and biennial species and several perennial forbs, and reducing the dominance of perennial grasses. On the other hand, in the mowed area, interrupting cutting did not reduce species diversity, although some rare species reduced their abundance while the perennial grasses started to increase their cover. Litter removal in this case had no significant effect on increasing species diversity, contrasting with the results found in the previous study. Nitrogen enrichment over three growing seasons did not affect species diversity in the abandoned and mowed areas and caused a limited increase of plant biomass, suggesting that these communities are only partially limited by nitrogen.

Summary for semi-dry perennial calcareous grassland (meadow steppe, R1A)

Considering the findings from gradient studies and spatial analyses, which revealed changes in species composition beginning from the lowest levels of deposition, it is necessary to reduce the $CL_{emp}N$ from 15 to 25 kg N ha^{-1} yr^{-1} down to 10 to 20 kg N ha^{-1} yr^{-1} (reliable). Whilst there was some evidence of impacts below 10 kg N ha^{-1} yr^{-1}, the number of sites below this level was relatively low since many of the gradient studies were from the UK with very few areas with low deposition. Increased N availability is probably of major importance in a number of European calcareous grasslands. In N-limited calcareous grasslands, increased availability of N is indicated by increased growth of some 'tall' grasses, especially of species which have a slightly higher potential growth rate and more efficient N utilisation. Nitrogen retention by the system is very high with hardly any leaching, and N mineralisation rates may also be increased because of N inputs. However, under P-limited conditions, vegetation responses are slow and loss of species is associated with changing soil conditions (acidification and decreased base saturation). Nitrogen mineralisation and nitrification are increased under P limitation, and in N-saturated systems with shallow soils this would result in somewhat higher leaching losses, although a large proportion of the N inputs would still be retained in the system. Most data from calcareous grasslands are from studies conducted in temperate, sub-Atlantic regions, and from sites with relatively high atmospheric N deposition. There is a need for experimental studies in continental regions, and for experiments with low N doses at sites with low ambient atmospheric deposition (< 10 kg N ha^{-1} yr^{-1}).

7.2.2 Mediterranean closely grazed dry grassland (R1D) or Mediterranean tall perennial dry grassland (R1E) or Mediterranean annual-rich dry grassland (R1F)

EUNIS categories R1D, R1E and R1F include xeric, thermophilic and mostly open Mediterranean perennial grasslands rich in therophytes growing on usually eutrophic, but also oligotrophic soils. The diversity of plants, but also of invertebrate and vertebrate species, is usually high. The conservation of Mediterranean xeric grasslands has been favoured by traditional management and contributes to prominent cultural landscapes. Grazing is essential for the long-term preservation of these communities and a reduction in grazing will result in scrub encroachment. Mineral fertilisation with P has been traditionally used to improve pasture quality since it increases spontaneous legume species and therefore forage protein content. There was limited information available on the effects of N enrichment on these grasslands in previous $CL_{emp}N$ revisions, but new studies have recently been published.

One of the main communities of Mediterranean xeric grasslands in the Iberian Peninsula are the annual pastures constituting the understory of Dehesas/Montados, a traditional agroforestry system, with a high biodiversity of species and high economic and ecological value. Nitrogen fertilisation experiments have been performed on the herbaceous stratum of a Dehesa in Central Spain (Migliavacca et al., 2017; Martini et al., 2019). The ecosystem is dominated by an annual grassland with low density of oak trees, mostly *Quercus ilex* (20% cover, 25 trees ha^{-1}). The herbaceous stratum is dominated by species of three main plant functional groups (grasses, forbs and legumes) whose proportion varied seasonally according to their phenological status. The plants are generally active from October to end of May. A first experiment assessed the effects of N or P alone or in combination at a small scale (9 x 9 m plots). In spring, N was applied as potassium nitrate (KNO_3) and ammonium nitrate (NH_4NO_3) at a rate of 100 kg N ha^{-1} and P was added as monopotassium phosphate (KH_2PO_4) in one application of 50 kg P ha^{-1}. The total doses of N were approximately 10 times higher than the current N deposition rate in the area (Morris et al., 2019). One of the challenges found in this experiment is that response variables showed wide variations over time. Additions of N changed the abundance of different plants affecting the community architecture and biochemistry (Migliavacca et al., 2017; Martini et al., 2019). In particular, higher N increased forbs while graminoid cover decreased. Relative abundance of legumes was marginal and did not show significant changes with nutrient additions (Martini et al., 2019). Changes in the abundance of different plant functional groups modified ecosystem processes such as increasing gross primary productivity (GPP), transpiration and albedo (Martini et al., 2019). The results of P addition alone showed negligible response on vegetation structure and soil properties (Pérez-Priego et al., 2015).

An additional experiment was performed at large scale (24 ha) in the same area using eddy covariance and intense monitoring to analyse carbon and water fluxes and their response to climate variability at ecosystem level. In this case only three treatments were established: control, nitrogen and nitrogen + phosphorus (NP). Nitrogen was applied as calcium ammonium nitrate ($Ca(NO_3)_2NH_4NO_3$) in the N treatment and as ammonium nitrate (NH_4NO_3) in the NP treatment in one application of 100 kg N ha^{-1} in the spring of first year and 20 kg N ha^{-1} the following year. Phosphorus was added as triple superphosphate ($Ca(H_2PO_4)_2$) in one application of 50 kg P ha^{-1} the first year and 10 kg P ha^{-1} the second year. A P only treatment was not applied this time since the previous study indicated that the ecosystem is N-limited and did not respond to P fertilisation alone (Migliavacca et al., 2017; Nair et al., 2019). Results were collected throughout four years showing a large inter-annual variability with, for example, larger differences in Gross Primary Productivity (GPP) in those years with precipitation above average rainfall compared to dry years. Nitrogen fertilisation increased root biomass and root length density (Nair et al., 2019) favouring nutrient absorption and increases of N pools in leaves.

These changes caused a rise in the photosynthetic capacity and higher GPP and leaf area index in fertilized areas (Nair et al., 2019; Luo et al., 2020). As a result, there was a higher evapotranspiration and consumption of water that accelerated grass senescence (Luo et al., 2020). The structure of the grassland community also varied with N fertilisation decreasing the abundance of forbs in some years (Luo et al., 2020), similar to the results found in the experiment at small scale. The results of these experiments highlight that the interaction between the availability of nutrients and water determine the functioning, composition and structure of Mediterranean xeric grasslands linked to Montado/Dehesas. The projected climate warming and increase in regional drought could compensate the effects of N enrichment due to atmospheric deposition (Luo et al., 2020).

N effects on annual grasslands have also shown a significant interaction with ozone (O_3), the most important air pollutant in the Mediterranean region. Two experiments were performed in open top chambers in Spain with mesocosm simplified communities exposed to four treatments with increasing O_3 concentrations and three treatments of N enrichment representing background (around 5 kg N ha^{-1}), 20 and 40 kg N ha^{-1}. Nitrogen supplementation was applied biweekly as ammonium nitrate (NH_4NO_3) in four equal applications during the experiment covering the active growing season. Ozone exposure reduced the fertilisation effect of enhanced N availability, while N could counteract pernicious O_3 effects on plant biomass production, but only at moderate O_3 levels (Calvete-Sogo et al., 2014, 2017). Grass species were more responsive to N enrichment compared with legumes but O_3 was a more important factor than N in explaining plant responses (Calvete-Sogo et al., 2016).

A N deposition gradient has been studied in semiarid ecosystems in central, southern and eastern Spain covering a range from 4.3 to 7.3 kg N ha^{-1} yr^{-1} (Ochoa-Hueso et al., 2013). The gradient included 16 sites dominated by grasslands (10 sites), shrublands (4 sites) or woodlands (2 sites). Nitrogen deposition affected soil nutrient cycling and fertility, and altered the functioning of biological soil crusts (Ochoa-Hueso et al., 2013). Soil bacteria and cyanobacteria and fungi abundance decreased with N deposition while green algae and cyanobacteria richness increased, contributing to ecosystem eutrophication (Ochoa-Hueso et al., 2013, 2016). Biological soil crusts formed by terricolous lichen and bryophyte communities are important for ecosystem functioning and nutrient cycling in these semiarid ecosystems. Based on the linear responses found in most soil indicators, a threshold of 4.3 kg N ha^{-1} yr^{-1} was proposed (Ochoa-Hueso et al., 2013).

In the previous update of $CL_{emp}N$ (Bobbink and Hettelingh, 2011), a N fertilisation experiment in a species-poor grassland dominated by *B. rupestre* performed in central Italy was included as Mediterranean xeric grassland (Bonamoni et al., 2006). The site represented an abandoned area 15-20 years after it had last been in agricultural use (rotating crops with legumes). An additional experiment performed in an area nearby in a stable species-rich mowed grassland has been added since then (Bonamoni et al., 2009). In this case, the community was a calcareous grassland ascribed to the association *Brizo mediae–Brometum erecti*. When this area was abandoned, the perennial grasses *B. rupestre* and *Dactylis glomerata* become dominant indicating this experiment should be considered in the calcareous grasslands section.

Summary for Mediterranean closely grazed dry grassland (R1D) or Mediterranean tall perennial dry grassland (R1E) or Mediterranean annual-rich dry grassland (R1F)

In the U.S., a $CL_{emp}N$ for similar Mediterranean grasslands of 6 kg N ha^{-1} yr^{-1} has been proposed, based on annual grasses replacing native herbs in a nutrient-poor serpentine grassland (Pardo et al., 2011). In the previous $CL_{emp}N$ revision in Europe, a first estimate of the $CL_{emp}N$ for the EUNIS categories R1D, R1E and R1F was 15 to 25 kg N ha^{-1} yr^{-1}, based on expert judgement that included information from an experiment in Italy that has now been transferred to the calcareous grasslands section R1A-R1B. Based on the gradient study outlined above and on the $CL_{emp}N$ available for scrublands (S5, S6) and forest (T21) communities, with which xeric grasslands are commonly associated, a lower range of 5-15 kg N ha^{-1} yr^{-1} is proposed as $CL_{emp}N$ for Mediterranean xeric grasslands based on expert judgement. However, further research is needed to disentangle the fate of N deposition effects in Mediterranean grasslands while interacting with a warmer and drier climate and high O_3 concentrations.

7.2.3 Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta* (R1M)

This EUNIS category groups all dry to mesic, base-deficient grasslands on acidic and neutral, weakly buffered, often sandy soils with closed vegetation in Atlantic or sub-Atlantic lowland and montane regions of northern and middle Europe and the western part of the Iberian Peninsula (R1M). Typical phytosociological units are *Violion caninae, Nardetalia strictae* and *Agrostion curtisii*. Species that are rare in parts of Europe, such as *Arnica montana*, *Antennaria dioica*, *Thymus vulgaris* and *Dactylorhiza maculata,* have been observed to disappear from these grasslands before tall and dense growing grasses started to dominate the vegetation in the Netherlands (e.g. Bobbink et al., 1996). In the Netherlands, these species are rare and endangered species are extremely sensitive to acidification and ammonium accumulation (e.g. Roelofs et al., 1996; De Graaf et al., 1998, 2009; Van den Berg et al., 2005; Kleijn et al., 2008). The input of acidifying nitrogenous deposition decreases the acid neutralising capacity (ANC) and subsequently the soil pH in these grasslands, which have weakly buffered soils. The deposited ammonium starts to accumulate once the pH significantly hampers nitrification (pH < 4.5). Thus, for these systems, species changes and loss of diversity are likely to be strongly associated with soil acidification inputs and/or changes in N form as well as the direct effects of N as a result of N.

Experimental N applications were carried out over three years on an *Agrostis capillaris* and a *Festuca ovina* grassland, respectively, both with a different initial fertility, in the province of Småland, in southern Sweden. This resulted in increased above-ground biomass as well as proportionately greater graminoid biomass following additions of 19 kg N ha^{-1} yr^{-1} for the low fertility *Festuca ovina* grassland (atmospheric load 13 kg N ha^{-1} yr^{-1}). No significant response was found for additions of 37 kg N ha^{-1} yr^{-1} on the more fertile *Agrostis capillaris* grassland area (with atmospheric deposition of 15 kg N ha^{-1} yr^{-1}), within three years of N additions (Berlin, 1998).

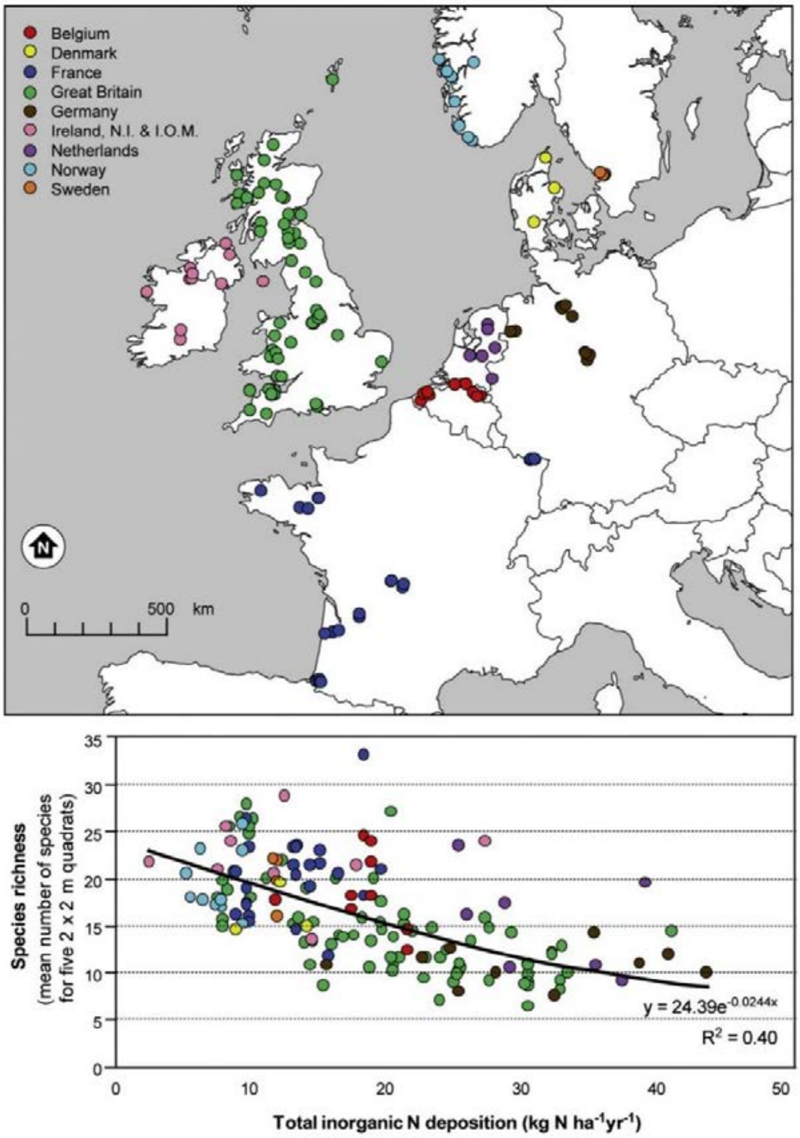
The effects of N additions (35, 70, 140 kg N ha^{-1} yr^{-1} in the form of ammonium nitrate) have been studied in a *Festuca*-*Agrostis*-*Galium* grassland in Derbyshire (UK) (atmospheric deposition approximately 25 kg N ha^{-1} yr^{-1}) (Morecroft et al., 1994; Johnson et al., 1999; Lee *et al*., 2000; Carroll et al., 2003; Phoenix et al., 2003; Horswill et al., 2008; Arróniz-Crespo et al., 2008). During the second year of the experiment a significant decline in bryophytes (especially *Rhytidiadelphus squarrosus*) was found following all levels of N treatment. Although this may partly have been an artefact, resulting from the relatively high N concentration applied, it indicates a high sensitivity to N. Moreover, *R. squarrosus* collected from the plots showed an increasingly higher N concentration with each N addition (Carroll et al., 1997, 2000). Nitrate reductase activities and soil N mineralisation rates clearly increased with increasing inputs of N during the first three years (≥ 35 kg N ha^{-1} yr^{-1}), this did not significantly alter the cover of vascular plant species, diversity or species composition, during the first four years (Morecroft et al., 1994; Carroll et al., 1997). From 1995 (sixth year of treatment) onwards, there has been a clear trend of decreased overall cover of the vegetation as well as decreased herb cover with increasing N additions. In 1995, an additional experiment with a factorial N x P design was initiated with N additions of 35 and 140 kg N ha^{-1} yr^{-1}. The results from the N only treatments corroborated the results found in the longer-term experiment, with a clear decrease in *Festuca ovina*, *Luzula campestris* and *Potentilla erecta*, and an increase in *Nardus stricta* (Lee and Caporn, 2001). This experiment also confirmed that this grassland was strongly limited by P rather than N (Lee and Caporn, 2001). N accumulation in the soil was not significant in any of the treatment areas, and N leaching was only significant at addition rates of ≥ 35 kg N ha^{-1} yr^{-1}.

Phosphomonoesterase activity (plays a critical role in controlling P cycling), increased in the soil during the long-term experiment at the lowest N addition (35 kg N ha^{-1} yr^{-1}) within three to four years (Carroll et al., 1997). Additionally, microcosm studies on soils from this field experiment (after seven years of N addition) showed increased root-surface phosphomonoesterase activity on the roots of *Agrostis stolonifera* seedlings, at N inputs of 35 kg N ha^{-1} yr^{-1} and higher. Thus, the increased N addition eventually also affected the P cycle in this severely P-limited system. A similar experiment with soils that had received N for only one and a half years showed no effect, indicating that the effect was the result of long-term changes in the soil (Johnson et al., 1999). After 12 years of N addition in the form of ammonium nitrate, ammonium-N concentrations in the soil showed strong dose-related treatment effects on the mineralisation-immobilisation N cycle in both organic and mineral horizons that were strongly dose related (O'Sullivan et al., 2011). Basto et al. (2015) studied the effect of N additions on the seed bank in the acidic grassland in the same experiment. They found that the seed bank size (i.e. number of seeds) was reduced by 61% in the 140 kg N ha^{-1} yr^{-1} N treatment and by 34% in the 35 kg N ha^{-1} yr^{-1} treatment. Seed bank richness was also reduced by 41% and 29%, respectively. Seed bank composition was also significantly changed with the largest reduction seen in forbs (73% at 140 kg N). Seed bank shifts were not the same as species composition changes observed in above-ground vegetation, resulting in an increased dissimilarity between above- and below ground species composition.

A field survey carried out by Stevens et al. (2004) revealed the relationship between atmospheric N deposition and plant species richness (including bryophytes) in acidic *Agrostis-Festuca* grasslands (*Violion caninae*) across the United Kingdom. The authors sampled 68 sites of high nature conservation interest in 2002 and 2003, with N deposition ranging from just above 5 kg N to 35 kg N ha^{-1} yr^{-1}. Of the 20 variables measured, total N deposition was the most important predictor of the variability in species richness. A clear negative linear (or negative exponential, see Emmett (2007)) relationship between species richness of these grassland communities and N deposition was found (Figure 7.4). This dataset was further analysed by Field et al. (2014) who used a subset of 22 sites with deposition between 7.8 and 30.3 kg N ha^{-1} yr^{-1} showing a consistent rate of species loss with N deposition. Species composition changed and richness of bryophytes, lichens, forbs and graminoids declined due to N. Payne et al. (2011, 2013) investigated changes in individual species abundance. The moss *Hylocomium splendens*, hemi-parasitic forb *Euphrasia officinalis* and forb *Plantago lanceolata* were the species most strongly negatively associated with N deposition, whereas the moss *Hypnum cupressiforme* responded positively to N deposition. TITAN analysis was used to reveal change points in individual species abundances. The lowest possible change point could be identified at 7 kg N ha^{-1} yr^{-1}. Further analysis provided by Payne et al. (2020) supported this with change points emerging below 10 kg N ha^{-1} yr^{-1}. A community level threshold of 14.2 kg N ha^{-1} yr^{-1} was identified where the community shifts to a more N tolerant assemblage, however, one third of individual species, mostly forbs, had change points below 10 kg N ha^{-1} yr^{-1}. Very few change points were noticed above 25 kg N ha^{-1} yr^{-1}, suggesting that changes have already occurred by this level. It should be noted though that some of the grasslands would also be slightly wetter than those found in other parts of Europe.

To investigate if this relationship holds for the whole Atlantic range of these acidic grasslands, and to gain more insights on the lower and higher ends of the N deposition ranges, the European Science Foundation (ESF) funded a programme named BEGIN (Biodiversity of European Grasslands: The Impact of Atmospheric Nitrogen Deposition), that carried out a survey identical to the UK study of Stevens et al. (2004) in nine countries, from Norway to the south of France, in the 2007-2009 period (Stevens et al., 2010b). This survey has shown that on this scale, species richness is again negatively correlated with total N deposition (Figure 7.4). Across the region, 9.8% of variation in species composition was driven by N deposition (Stevens et al., 2011a) with the proportion of grasses increasing with increasing N deposition (Stevens et al., 2011b). Helsen et al. (2014) combined this data with an additional data set (Ceulemans et al., 2011), on 297 semi-natural *Nardus* grasslands with a N deposition from 2 to 43 kg N ha^{-1} yr^{-1}. They found high soil N was associated with higher proportion of graminoids and long-lived, clonal species and a decrease in therophytes and forbs. Among other changes there was also a decrease in N_2-fixers, and orchids.

Figure 7.4. Species richness of acidic grasslands across a gradient of N deposition in the Atlantic region of Europe.



Source: Stevens et al., 2010b

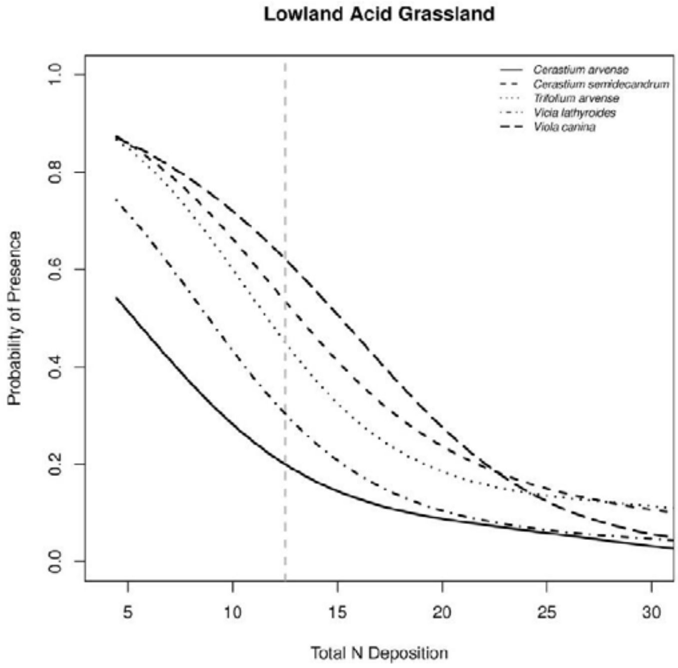
In addition to this, Maskell et al. (2010) reported results from the UK countryside survey using data collected in 1998 which broadly support these findings. Analysis of this data using non-parametric quantile regression with a breakpoint identified a threshold at 7.8 kg N ha^{-1} yr^{-1} (Tipping et al., 2013). Analysis of data from the same survey but using data collected in 2007 also showed a negative relationship between species richness and N deposition, reporting an increase in grass:forb ratio (Van den Berg et al., 2016). Results from both these reports confirm the findings in Stevens et al. (2004). Furthermore, an analysis of the changes in species composition across this gradient demonstrated that soil acidification is likely to be the most important driving factor of the observed decline in species alongside the other effects of N (Stevens et al., 2010a). Based on an evaluation with a Bayesian approach, Pescott and Jitlal (2020) suggested that the estimated negative effects of N deposition have been over-estimated. However, additional analyses showed their results were unreliable (Smart et al., 2021).

Other surveys in different countries have similarly confirmed the relationships identified by Stevens et al. (2004). Data from the Danish National habitat monitoring programme were adjusted to allow comparison with the Stevens et al. (2010) data. There was a significant correlation between the two data sets and the decrease in species richness along a N deposition gradient was evident. Whilst the range of N deposition in Denmark was too narrow to see this impact on its own, when combined with the larger European survey, it was apparent (Damgaard et al., 2011). Wilkins et al. (2016) examined data from the Irish National Parks and Wildlife Service. They examined 108 relevés for Natura 2000 community 6230 with a range of N deposition of 2.4 to 22.3 kg N ha^{-1} yr^{-1}. Results suggested loss of species richness with N deposition and indicated a community change point between 3.9 and 6.5 kg N ha^{-1} yr^{-1}.

A number of surveys have also investigated changes in species composition over time. Duprè et al. (2010) conducted a temporal analysis utilising data sets published between 1939-2007 from Great Britain, the Netherlands, Germany, Denmark, and Sweden. The analyses of 1114 plots belonging to the *Violion caninae* showed that Ellenberg R (soil reaction) scores increased up to 1980 and then stabilised or even slightly declined. Cumulative N deposition showed a negative relationship with species richness in all countries. Pannek et al. (2015) used these data combined with Stevens et al. (2010b) to show that 16 species (55%) responded significantly to N deposition, 75% of those responded negatively. Change in species frequency over time was related to N deposition, suggesting declining species were predominantly sensitive to N deposition. Species with high relative growth rate tended to respond positively to N deposition. Gaudnik et al. (2011) collated 162 published records over a period of 25 years, 1980-90 and 1995-2005 and some additional data collected in 2007 from nutrient poor, acidic grassland in Atlantic region of France. Ordinations suggested that N deposition was an influential driver of species composition at local scales although there were difficulties in disentangling the effects of N deposition and climate change. In a repeated survey of *Nardus* grasslands in central Germany between 1971-87 and 2012-2015, significant increases in soil pH and Ellenberg R were observed, indicating recovery from acidification. Increases in Ellenberg N and high nutrient indicators together with a decrease in low nutrient indicator species, and *Nardus* grassland specialist species indicated eutrophication. In contrast to other studies, forbs increased and graminoids declined (Peppler-Lisbach et al., 2019), which could be related to species identity or interaction with other driver variables.

Spatial analysis using data collected from the UK Vascular Plant Database, which gives vascular plant species at 10 km resolution between 1987 and 1999 was analysed as described in Chapter 7.2.1. Eleven species were analysed for lowland acidic grassland with five showing significant negative relationships with N, including declines below 10 kg N ha^{-1} yr^{-1} (Figure 7.5). Only one species could be analysed for upland acidic grassland, but this did not show a significant relationship with N deposition. Lowland acidic grasslands showed a clear increase in Ellenberg N with increasing N deposition but there was no significant relationship for upland acidic grasslands in either database (Henrys et al., 2014). In a similar study using 10 km resolution data on presence /absence from the British Lichen Society over the last 50 years (methods as described in Chapter 7.2.1) 16 taxa were analysed for acidic grasslands, three showed significant relationships with N deposition. *Cladonia foliacea* occurred less frequently above 10 kg N ha^{-1} yr^{-1}. The two other species showed variable responses (Stevens et al., 2012).

Figure 7.5. Relationships between probability of presence for species showing statistically significant relationships to N deposition in lowland acidic grasslands in the Vascular Plant database against increasing total inorganic N deposition (kg N ha^{-1} yr^{-1}).



Source: Henrys et al., 2011

Summary for Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta* (R1M)

These studies indicate that many dry to mesic acidic grasslands are sensitive to N loads. The effects of N deposition became apparent through the use of spatial and temporal national and regional surveys (gradient studies). All of these identified impacts of N from low levels of deposition. Four of these studies (Payne et al., 2020, 2013; Tipping et al., 2013; Wilkins et al., 2016) specifically estimate change points for individual species and the community as a whole. These results provide change points ranging from 3.9 to 6.6 in Ireland (Wilkins et al., 2016) to the highest of 14.2 (Payne et al., 2013) but with large numbers of individual species showing change below this point. It is clear from these numbers that the $CL_{emp}N$ range of 10 to 15 kg N ha^{-1} yr^{-1} should be reduced. Since there are few sites with deposition at and below 5 kg N ha^{-1} yr^{-1} uncertainty regarding the effects of N is high. The range is thus reduced to 6 to 10 kg N ha^{-1} yr^{-1}, quantified as reliable. There remains a need for more field studies in areas with very low atmospheric deposition.

7.2.4 Oceanic to subcontinental inland sand grassland on dry acid and neutral soils (R1P) or Inland sanddrift and dune with siliceous grassland (R1Q)

These habitats contain a special flora (red list species and a high proportion of cryptogams, particularly lichens) and fauna (insects, birds). The impacts of N loads have been studied for a pioneer community on sandy grassland (*Koelerion glauca*) in the Upper Rhine valley in Germany (Storm and Süss, 2008), with a background N deposition of 17 kg N ha^{-1} yr^{-1}. The vegetation was treated with two levels of N (25 and 100 kg N ha^{-1} yr^{-1}; in the form of ammonium nitrate), between 2000 and the summer of 2004. The above-ground biomass of the vascular plants significantly increased following the high N addition, whereas the biomass of the cryptogams declined during the last two years of the experiment. Other nutrients did not cause additional increases in above-ground productivity, thus indicating that this vegetation was clearly N limited. The cover of 10 species increased after high N addition. No significant changes in diversity were found between treatments, although effects in this originally very open community could occur in the long term.

Other investigations in the Netherlands have demonstrated that areas receiving high levels of N deposition (41 kg N ha^{-1} yr^{-1}) have reduced microbial biomass, increased net N mineralisation, higher soil N and higher microbial N:P ratios compared to lower deposition (24 kg N ha^{-1} yr^{-1}) sites (Sparrius and Kooijman, 2013). A gradient study spanning deposition between 17 and 50 kg N ha^{-1} yr^{-1} showed evidence of invasion of *Campylopus introflexus*, a neophytic moss species, at higher deposition (Sparrius et al., 2011). A N addition experiment incorporating 21 dunes in the Netherlands with deposition between 21 and 47 kg N ha^{-1} yr^{-1} showed that high deposition induced higher algal cover, reduced vascular species richness, reduced lichen richness and reduced occurrence of some plant species as well as reductions in soil pH (Sparrius et al., 2012). Comparing dunes from high (34-44.8 kg N ha^{-1} yr^{-1}) and lower (22.6-33.7 kg N ha^{-1} yr^{-1}) deposition areas, Sparrius et al. (2013a) showed that there was an increased vegetation cover resulting in greater loss of bare sand in high deposition areas. Furthermore, 2.5 years of N addition at a rate of 42.9 kg N ha^{-1} yr^{-1} at two sites (background deposition 25 kg N ha^{-1} yr^{-1} and 34 kg N ha^{-1} yr^{-1}) had a range of impacts including increased grass cover, reduced lichen cover and increased water extractable N and higher tissue N content of grasses and in lichen dominated vegetation (Sparrius et al., 2013b). However, whilst these investigations show that this habitat is sensitive to N deposition, all these studies have been carried out at high background levels of N deposition.

Summary for Oceanic to subcontinental inland sand grassland on dry acid and neutral soils (R1P) or Inland sanddrift and dune with siliceous grassland (R1Q)

Defining a $CL_{emp}N$ based on these findings is difficult, since a high proportion of the investigations have taken place at levels of deposition far exceeding the $CL_{emp}N$. These inland dune grasslands have a species composition and ecological functioning comparable with that of coastal dune grasslands (see Chapter 4 for details). Because of this similarity, the $CL_{emp}N$ for inland sand grassland on dry acid and neutral soils (R1P) or inland sanddrift and dune with siliceous grassland (R1Q) is set at the same level as that of coastal dune grasslands (see Chapter 4, 5-15 kg N ha^{-1} yr^{-1}). Inland dunes are acidic or strongly decalcified and so would likely lie to the more sensitive lower end of this range. However, this estimation is based completely on expert judgement and there is thus a significant need for long-term research on these ecosystems to substantiate the effects.

7.3 Mesic grasslands (R2)

7.3.1 Low and medium altitude hay meadows (R22)

In semi-natural grasslands situated at low or medium elevation that are managed for making hay (R22) field experiments have been conducted with N-only treatments in realistic doses. The famous Park Grass experiment at Rothamsted (UK) has been running since 1856 (Williams, 1978; Dodd et al., 1994; Crawley et al., 2005; Silvertown et al., 2006). Nitrogen has been annually applied (single dose) as ammonium sulphate or sodium nitrate (48 kg N ha^{-1} yr^{-1}) to plots of mesic, low altitude hay meadow (R22). On N-treated plots, within five to ten years the vegetation became dominated by a few grasses, such as *Alopecurus pratensis*, *Arrhenatherum elatius*, *Holcus lanatus* or *Agrostis* species. For species diversity a negative correlation was found with total biomass and soil acidity. Ammonium sulphate, through its acidifying effects, reduced the diversity of higher plant and bryophyte species significantly more than other forms of N within ten years, and this difference was still present after more than 150 years (Goulding et al., 1998; Virtanen et al., 2000; Silvertown et al., 2006). The experiment shows a positive response of biodiversity to reducing N addition from either atmospheric pollution or fertilisers. As atmospheric N deposition declined from 45 kg ha^{-1} yr^{-1} in 1996 to 21 kg ha^{-1} yr^{-1} in 2010, the proportion of legumes, plant species richness and species diversity increased. An important driver for the development was the increase in soil pH. Biodiversity has also partly recovered on plots formerly fertilised with 96 kg N ha^{-1} yr^{-1}. However, biodiversity is still below that of the control plots. As biodiversity has only partly recovered within the $CL_{emp}N$ range (20-30 kg N ha^{-1} yr^{-1}) this may be an indication that the $CL_{emp}N$ is currently set too high (Storkey et al., 2015).

In addition to an increase in biomass, a 25% reduction in species diversity was observed after relatively long-term (>4 years) additions of N (100 kg N ha^{-1} yr^{-1}), applied to a hay meadow along the river Rhine in the Netherlands (Beltman and Barendregt, 2002; Beltman et al., 2007). The effects of N inputs became less pronounced because of a large flooding event after eight years, although differences between N treatments and the controls remained significant. P additions did not affect the vegetation within the timeframe of this experiment.

Wilkins et al. (2016) analysed data from the Irish National Parks and Wildlife Service. They examined 125 relevés belonging to Annex I habitat type 6510 of the EU habitat directive with a range of N deposition between 2 and 22 kg N ha^{-1} yr^{-1}. Results pointed toward a loss of species richness with higher N deposition and indicated a community change point of between 7.5 and 8.7 kg N ha^{-1} yr^{-1}. In an update, the analyses were revised, additional habitats were added and deposition mapping was improved (Aherne et al., 2020). For the Atlantic region they suggested a $CL_{emp}N$ range for low and medium altitude hay meadows (R22) of 5-15 kg ha^{-1} yr^{-1}.

Summary for low and medium altitude hay meadows (R22)

Previously, the $CL_{emp}N$ for low and medium altitude hay meadows was set at 20-30 kg N ha^{-1} yr^{-1} based on expert judgement. Although new findings from a field experiment and gradient studies have been published since the last review, the data basis is still uncertain. Therefore, the $CL_{emp}N$ range for low and medium altitude hay meadows is as expert judgement specified as 10-20 kg N ha^{-1} yr^{-1}. There is, however, still a need for field addition studies in different countries, especially in regions with low atmospheric deposition.

7.3.2 Mountain hay meadows (R23)

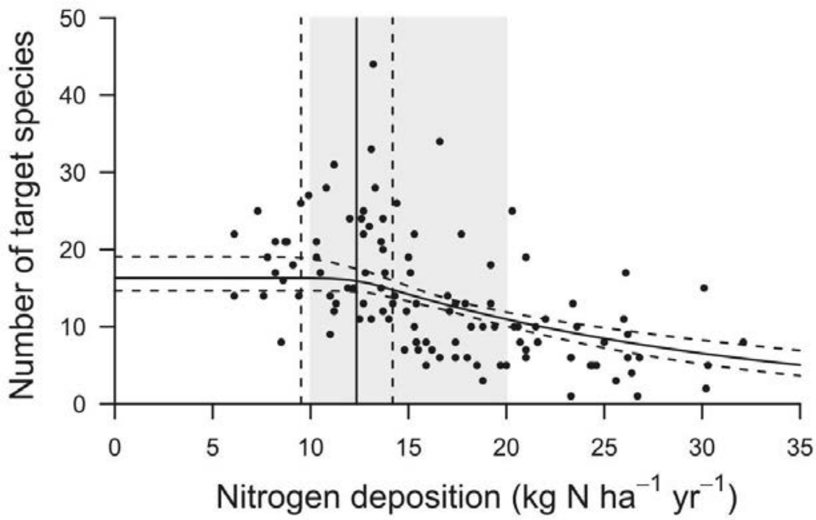
Many semi-natural grassland types occur in montane regions across Europe, containing many rare and endangered plant and animal species (e.g. Ellenberg, 1996). However, experimental studies with low doses of N are not available for this grassland type. The Rengen Grassland

Experiment in Germany has fertiliser treatments in a mountainous hay meadow but, however, N was applied in combination with Ca and Mg, thus preventing soil acidification after N inputs and also any significant impact of N deposition on these acidic meadows (Hejcman et al., 2007).

The previous $CL_{emp}N$ range (10-20 kg N ha^{-1} yr^{-1}) for mountain hay meadows was based on expert judgement only. In a gradient study using data on vascular plants and bryophytes from the Biodiversity Monitoring of Switzerland (2006-2010), Roth et al. (2013) inferred whether N deposition is negatively related to species richness and Simpson diversity in mountain hay meadows. The relationship between N deposition and species richness and Simpson diversity was analysed for all species together, but also for high and low nutrient indicators and for species of conservation concern. Non-linear effects of confounding variables such as elevation and mean or annual precipitation were accounted for in the generalised additive models (GAM) used to analyse effects of N deposition. The average (±SD) background N deposition on the 122 plots was 18 (±6) kg N $ha^{-1}yr^{-1}$. Species richness and diversity of vascular plants and bryophytes were negatively related to N deposition. Furthermore, a negative relationship between the number of low nutrient indicator species and N deposition was found. As low nutrient indicator species are generally rare, the species composition became more similar with increasing N deposition because the rare species disappeared at higher level of N deposition.

Using data from the Biodiversity Monitoring of Switzerland, but with recordings from 2010-2014, the $CL_{emp}N$ for mountain hay meadows could be directly estimated by a change-point model. Seven co-variables (elevation, inclination, precipitation, calcium carbonate content, aspect and mean indicator values for humidity and light) were taken in account. The results of this study suggest that the 2011 $CL_{emp}N$ range is too broad (Roth et al., 2017). The number of target species of this meadow type starts declining at a N deposition rate of 13 kg N ha^{-1} yr^{-1} (Fig. 7.6). Roth et al. (2017) thus proposed to reduce the $CL_{emp}N$ upper limit to 15 kg N ha^{-1} yr^{-1}.

Figure 7.6. Number of species of conservation concern in mountain hay meadows (EUNIS R23) across a gradient of nitrogen (N) deposition in Swiss mountains. The dots represent the raw counts of species numbers in the sampling sites of 10 m². The black line represents the change-point regression of species richness on N deposition that simultaneously accounted for seven confounding variables (see text). The vertical line represents the position of the change-point, the dashed lines the corresponding 95% credible intervals. The grey shaded area represents the expert-based empirical critical load range from 2011.



Source: Roth et al., 2017

Data from three Monitoring programs in Switzerland were recently pooled to check the $CL_{emp}N$ given above (Roth et al., unpublished data). The same change-point model was applied as by Roth et al. (2017) including elevation, inclination, precipitation, calcium carbonate content, aspect as well as mean indicator values for humidity and light as confounding variables. The gradient of N deposition ranged from 4 to 37 kg ha^{-1} yr^{-1}. When data from three Swiss monitoring programmes are combined, the change-point in mountain hay meadows for the target species was estimated at 7-9 kg N ha^{-1} yr^{-1} (Roth et al., unpublished data).

An experimental NPK fertiliser study on Swiss mountain hay meadows supports this finding. The number of vascular plant species decreased significantly at a nitrogen input of 16 kg N ha^{-1} yr^{-1} and the forb species richness at 15 kg N ha^{-1} yr^{-1} (Boch et al., 2021). However, the results may not be used directly for the revision of the $CL_{emp}N$, as the background N load was not accounted for, and the number of cuts increased from one to two cuts during the experiment.

Summary for mountain hay meadows (R23)

Since 2010, the $CL_{emp}N$ range can be delineated more precisely with data from gradient studies. The $CL_{emp}N$ for mountain hay meadows (R23) is set at 10-15 ha^{-1} yr^{-1} (previously 10-20 ha^{-1} yr^{-1}) and classified as reliable.

7.4 Seasonally wet and wet grasslands (R3)

7.4.1 Moist or wet mesotrophic to eutrophic hay meadow (R35) or Temperate and boreal moist and wet oligotrophic grasslands (R37)

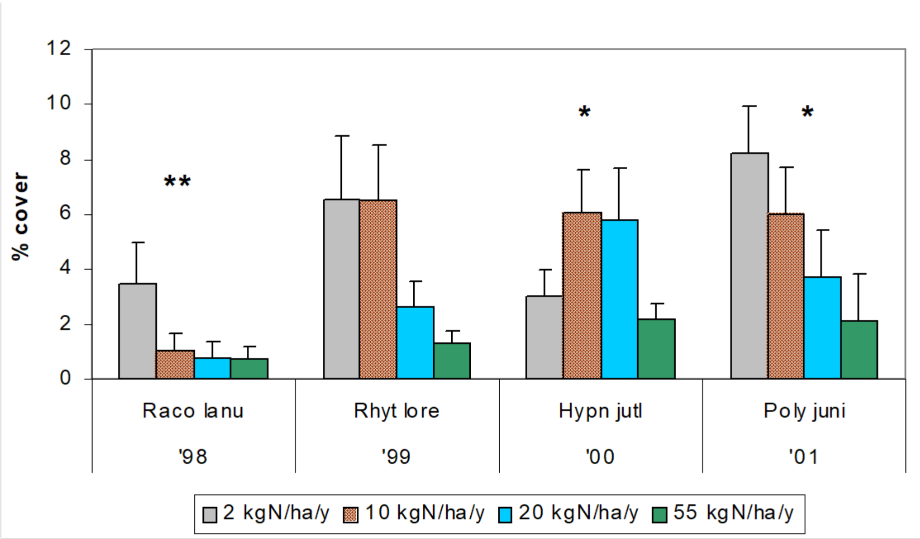
Temperate and boreal moist and wet oligotrophic grasslands (R37) are characterised by oligotrophic and moist to wet peaty soil conditions. This EUNIS category consists mostly of hay meadows under original agricultural management that are especially rich in typical plant and animal species. R37 combines two subcategories that were distinguished previously, namely (i) *Molinia caerulea* meadows (previously E3.51; 'litter meadows' or 'fen meadows') and (ii) heath meadows and humid *Nardus stricta* swards (previously E3.52). R35 covers moist or wet mesotrophic to eutrophic hay meadows. Because of their long history of traditional land use with low additional inputs of nutrients, these grassland communities are likely to be sensitive to extra nutrient inputs. Several fertilisation experiments in these wet oligotrophic grasslands have demonstrated limitation by either N or P or even co-limitation by these elements (e.g. Vermeer, 1986; Egloff, 1987; Spink et al., 1998; Van Duren et al., 1998; Olde Venterink et al., 2001). In the case of N limitation, grass productivity, especially of the dominant *Molinia caerulea*, increased and species diversity declined (e.g. Vermeer, 1986). However, almost all of the studies that have been performed in moist or wet oligotrophic grasslands were either carried out with high to very high loads (> 100 kg N ha^{-1} yr^{-1}) or had a time span that was too short to be used for setting a $CL_{emp}N$ range.

Fortunately, the impacts of N additions on species richness have been quantified in flower-rich, oligotrophic wet hay meadows (R35) in Somerset (UK) (Mountford et al., 1994; Tallowin et al., 1994; Kirkham et al., 1996). Nitrogen additions of 25 kg N ha^{-1} yr^{-1} and more (with an estimated background atmospheric load of 15-25 kg N ha^{-1} yr^{-1}) for six years significantly reduced the number of species, while several grasses increased in dominance (*Lolium perenne*, *Holcus lanatus* and *Bromus hordeaceus*). The forbs, characteristic of these meadows, declined sharply in number, and some, for example, *Cirsium dissectum*, *Lychnis flos-cuculi* and *Lotus pedunculatus* disappeared from N treated plots altogether.

More recently, a N manipulation experiment examined the effects of additions of ammonium sulphate (10 and 20 kg N ha^{-1} yr^{-1}) or sodium nitrate (20 kg N ha^{-1} yr^{-1} only) to an upland grass heath in Wales (R37) (Emmett et al., 2001, 2007). This site, at an altitude of 600 m, had been overgrazed in the 1970s and 1980s, resulting in degradation of *Calluna*-dominated moorland to a sward dominated by *Nardus stricta*, *Vaccinium myrtillus* and *Festuca*. The treatments were applied to paddocks which had different rates of experimental sheep grazing from 1989 up to the start of the experiment in 1997. After four years of treatment, relatively small effects were observed on the vascular plants, although there was evidence of increased *Festuca* cover on the lightly grazed paddock, and greater frost injury to *Vaccinium* (only on the plot treated with nitrate), which may have been linked to earlier bud break in the spring. The observed lack of response in palatable grasses in the heavily grazed paddock may reflect selective grazing by sheep. In later years of this experiment, high N addition significantly reduced the cover and species richness of bryophytes, although grazing intensities modified this response; it only became obvious at a low grazing intensity, while no effect could be observed when the grazing intensity was higher (Emmett et al., 2007).

Measurements of soil water chemistry at this site (lightly grazed plots) showed significant leaching losses in the control plots, representing 25% of inorganic inputs, which increased from 5 to 7 kg N ha^{-1} yr^{-1}, during the 20 kg N ha^{-1} yr^{-1} treatment. These high leaching rates suggested that N levels at this site were already above the $CL_{emp}N$. Only for the ammonium treatments, there was also increased base cation leaching and decreased pH. However, there were no significant treatment effects on mineralisation or nitrification rates.

Figure 7.7. Cover of moss species in *Nardus stricta* grassland mesocosms, exposed since 1997 to four N treatments (2, 10, 20, and 55 kg N ha^{-1} yr^{-1}; from left to right). The mosses are *Racomitrium lanuginosum, Rhytidiadelphus loreus, Hypnum jutlandicum* and *Polytrichum juniperinum*.



Source: Jones et al., 2002

The atmospheric deposition at the grass heath site in Wales was estimated at 20 kg N ha^{-1} yr^{-1}. To assess the impacts of lower deposition rates, Jones and Ashenden (2000) applied a range of N deposition treatments above and below that of the existing site estimate (2, 10, 20 and 55 kg N ha^{-1} yr^{-1} in the form of ammonium nitrate) to mesocosms that were taken from the site and grown in a greenhouse. To assess possible interactions with grazing pressure, three levels of simulated grazing (clipping) were also applied. Within one to two years, there were strong effects of N treatments of below 20 kg N ha^{-1} yr^{-1}, increasing the cover of certain moss and lichen species, but only in combination with heavy clipping, presumably because of the lower competition from vascular plants. Subsequent data (Jones et al., 2002) have shown the emergence of different optima for bryophyte species – that for *Racomitrium lanuginosum* and *Polytrichum juniperinum* lying below 10 kg N ha^{-1} yr^{-1} – while for *Hypnum jutlandicum* this would be around 20 kg N ha^{-1} yr^{-1} (Figure 7.7). Although results have shown an increase in fine grass cover and a decrease in *Nardus* following increasing N additions, these effects occurred primarily at between 20 and 55 kg N ha^{-1} yr^{-1}. In contrast to results from the parallel experiment on the calcareous mesocosms and results from the field site, there was no evidence of effects of nitrate leaching in the first two years of the experiment (Jones et al., 2002). More recent work at these experimental sites has indicated that whilst plant species composition varied strongly between treatments, N did not increase vegetation height compared to control plots (Stiles et al., 2017). Nitrogen treatments also had lower CO_2 fluxes and lower CH_4 uptake than in the controls (Stiles et al., 2018).

Summary for Moist or wet mesotrophic to eutrophic hay meadow (R35) or Temperate and boreal moist and wet oligotrophic grasslands (R37)

In summary, several moist or wet oligotrophic grasslands of high conservation value have been shown to be sensitive to N eutrophication. Increases in dominant grasses and decreases in species richness have been observed following increasing levels of N inputs. The study of a degraded upland heath meadow (Emmett et al., 2001, 2007) has provided evidence of response in bryophyte cover to relatively low levels of N deposition, and suggested increases in leaching and acidification at levels above 20 kg N ha^{-1} yr^{-1}. In view of these UK studies, the $CL_{emp}N$ for R37 moist to wet oligotrophic grasslands is set at 10-20 kg N ha^{-1} yr^{-1} specifically for *Nardus stricta* swards and considered to be 'quite reliable'. Studies for other parts of Europe are needed, especially as some studies included in the section on closed non-Mediterranean dry acid and neutral grassland span a gradient from dry to moist.

The $CL_{emp}N$ for moist or wet mesotrophic to eutrophic hay meadow (R35) however, is based on expert judgement only, and estimated to be somewhat higher (15-25 kg N ha^{-1} yr^{-1}), because, to date, this habitat has barely been studied and the one experiment for R35 grasslands has high levels of N addition relative to the $CL_{emp}N$ range. The $CL_{emp}N$ for both of these categories have not changed from those in the 2010 document. Base status is likely to be a significant modifying factor as systems with low base status are likely to be more sensitive to N deposition, while fluctuations in the water table may cause habitats in the wet hay meadows to be less sensitive.

7.5 Alpine and subalpine grasslands (R4)

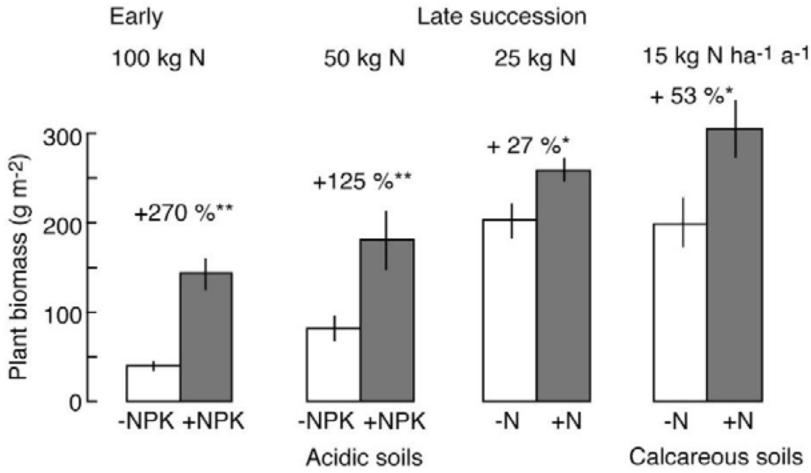
7.5.1 Temperate acidophilous alpine grasslands (R43) or Arctic-alpine calcareous grassland (R44)

Many natural grassland types are found in the alpine and subalpine regions of Europe and other mountains systems on both acidic (R43) and calcareous (R44) soils. These grasslands often encompass an outstandingly high biodiversity with many rare and endemic plant species (Körner, 2021). Alpine is defined as the vegetation zone above the upper natural treeline. The alpine vegetation zone is the only vegetation zone which can be clearly defined globally (Körner, 2021). As subalpine is not a clearly defined term, the term montane or mountainous areas below the treeline should be used.

Land use often contributes as a source of N emissions to increased atmospheric N deposition, although sustainable land use by mowing and grazing contributes to preservation of montane grasslands by keeping them open. Complete abandonment of land use leads to encroachment of woody taxa and biodiversity losses in the montane vegetation zone. Land use within the alpine vegetation zone is commonly less intensive and patchy (Körner, 2021).

There is clear evidence of impacts of high rates of N addition (> 40 kg N ha^{-1} yr^{-1}) in alpine grasslands. Late successional acidic grassland at a 2,500 m elevation in the central Swiss Alps (Furkapass area) showed a rapid and large response to additions of 40 to 50 kg N ha^{-1} yr^{-1} (Körner et al., 1997; Figure 7.8). Part of this early experiment was conducted with a multi-nutrient fertiliser, but there is little other evidence from these habitats. Biomass was doubled already in the second year, with sedges (*Carex curvula*) profiting most.

Figure 7.8. Responses of vegetation of high elevation (> 2450 m) grassland and glacier foreland (most left) to two to four years of nutrient addition in early and late successional habitats, control plots are shown as -NPK or -N.



Source: Heer and Körner, 2002; Körner et al., 1997; Hiltbrunner, unpublished data

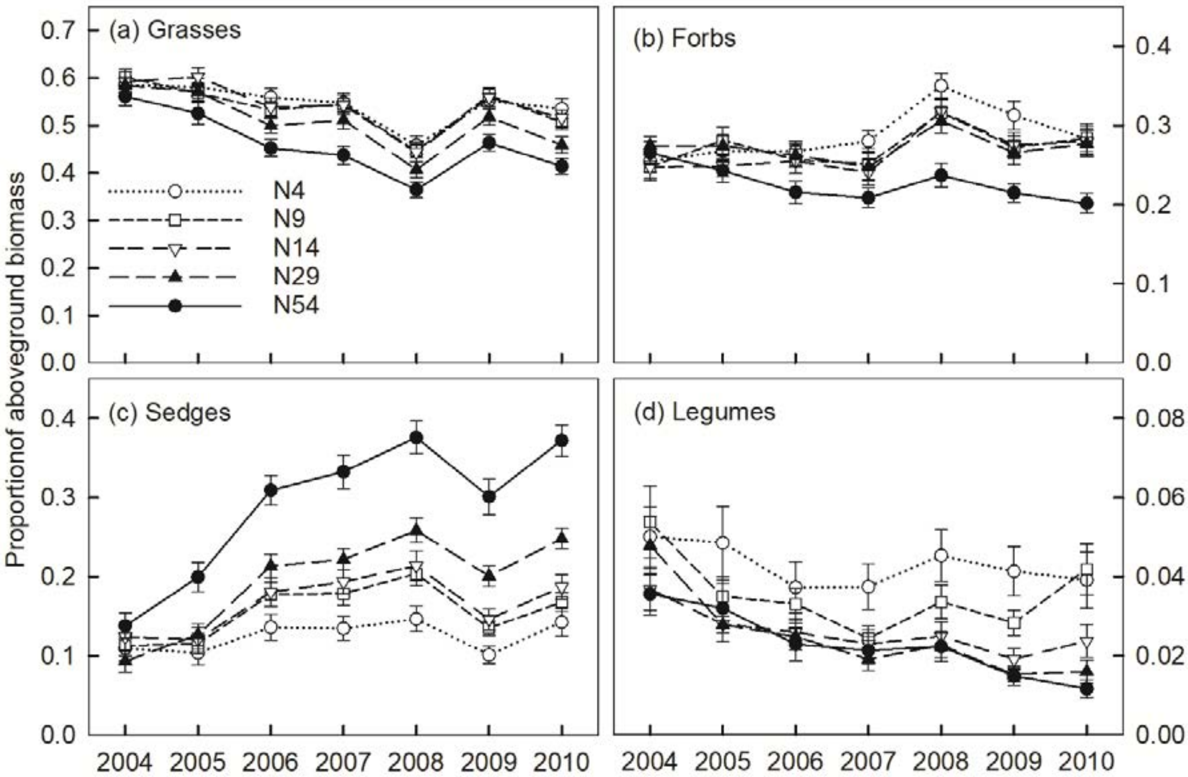
A 100 kg N ha^{-1} yr^{1} addition converted an open vegetation with cushion plants (< 10% cover) into a lush grassland (100% cover) within two years only (Heer and Körner, 2002). Earlier experiments with similarly high doses of N applied to alpine dwarf-shrub heath confirmed sensitivity of alpine vegetation to nutrient addition. These experiments ended with a rather unexpected response after four years of nutrient addition: a complete collapse of *Loiseleuria (Kalmia) procumbens* and *Calluna vulgaris* stands due to snow mould infestation exclusive to the nutrient addition area (Körner, 1984). Hence, there is no doubt that cold-climate, alpine vegetation is responsive to an elevated N supply. Several longer-term studies on N additions have been set up in Switzerland in alpine acidic (R43) and alpine calcareous grasslands (R44), to quantify the impacts of lower N loads.

The effects of N addition on plant production in acidic alpine grasslands (R43; *Caricietum curvulae*) was studied from 2002 to 2005 in the central Alps (Furka) in Switzerland, at two elevations (2450 and 2650 m a.s.l) (Hiltbrunner and Körner, 2004; Hiltbrunner et al., 2005). In the fourth year of this experiment, the addition of 25 kg N ha^{-1} yr^{-1}, in the form of ammonium nitrate, resulted in a significant increase (27-45%) in the above-ground biomass, compared to the control vegetation and irrespective of elevation. Background deposition at the research site was around 4 to 5 kg N ha^{-1} yr^{-1}.

In a seven-year N addition experiment, in a species-rich acidic grassland area (Geo-montani Nardetum), at 2000 m a.s.l. in the Swiss Alps, 5, 10, 25 and 50 kg N ha^{-1} yr^{-1} were applied to grassland monoliths, with an ambient background of approximately 4 kg N ha^{-1} yr^{-1} (Bassin et al., 2007, 2009). The above-ground biomass of the vegetation significantly increased with the added

N levels. The sedges *Carex sempervirens* and *Carex ornithopoda* tripled their fractional biomass at the expense of legumes (*Trifolium alpinum*), grasses (*Agrostis capillaris*, *Briza media*, *Festuca* spp.), and forbs, the latter of which responded inconsistently. Compositional changes were significant with +5 kg N ha^{-1} y^{-1}; at all levels of N, but changes ceased after five years (Bassin et al., 2013, Figure 7.9).

Figure 7.9. Relative contribution (means ± SE) of the four functional groups (a-d): grasses, forbs, sedges, and legumes to the total aboveground biomass following N additions (incl. background N deposition) of 4 (N4 - control), 9, 14, 29, and 54 kg N ha^{-1} y^{-1} (background plus N additions) during the experimental years 2004-2010.



Source: Bassin et al., 2013

In this same experiment, on plots receiving 50 kg N ha^{-1} yr^{-1}, the net ecosystem productivity (NEP) has been shown to yield losses of 54 g C m^{-2} per season from the grassland compared to the control area (Volk et al., 2010). However, after seven years of N addition, cumulative NEP was not significantly altered due to N addition, and NEP was slightly higher at 10 kg N ha^{-1} yr^{-1} than at 50 kg N ha^{-1} yr^{-1} (Volk et al., 2016).

In a fully factorial three-way transplant experiment with three-levels of N addition (0, 3, 15 kg N ha^{-1} year^{-1}), water treatment and transplants along an elevation gradient to expose grassland monoliths to temperature differences in the range of -1.4 to +3.0 °C revealed that the stimulating effect of increased N on sedge cover was dampened by increased temperatures. Unexpectedly, no changes in above-ground biomass formation were found due to N addition after four years (Wüst-Galley et al., 2021; Volk et al., 2021).

Based on the evidence provided, alpine acidic grasslands (R43) are likely to be sensitive to N loads. The $CL_{emp}N$ for these habitats remains unchanged at 5 to 10 kg N ha^{-1} yr^{-1}, which is considered to be ‘quite reliable’.

Arctic-alpine calcareous grasslands (R44) are especially species-rich communities. The impacts of N enrichment have been investigated in a calcareous alpine grassland area in Switzerland, at approximately 2500 m a.s.l. Background deposition at the experimental site ranged between 3 and 5 kg N ha^{-1} yr^{-1}, and six N application levels, of 2.5, 5.0, 10, 15, 20 and 25 kg N ha^{-1} yr^{-1} were applied. In the fourth year of this N addition experiment, the dominant sedge species *Carex firma*, showed a positive response in cover following additions of 5 kg N ha^{-1} yr^{-1} and more. Total above-ground biomass increased by 53 % at N addition of 15 kg N ha^{-1} yr^{-1}. However, after seven years of N addition, above-ground biomass was significantly stimulated at 25 kg N ha^{-1} yr^{-1} only, indicating a declining responsiveness to N addition over time (Hiltbrunner, unpublished data), similar to the observation on weaker compositional changes in the acidic grassland after five years of N addition (Bassin et al., 2013).

There is no additional evidence for alpine calcareous grasslands (R44) since the $CL_{emp}N$ was set in 2011. So, based on the results of this experiment, the $CL_{emp}N$ remains at 5 to 10 kg N ha^{-1} yr^{-1}**,** which is considered to be 'quite reliable'. There is a clear need for further investigation in this habitat.

7.5.2 Moss- and lichen-dominated mountain summits, ridges and exposed slopes

Within the old EUNIS system, an important sub-category of alpine and subalpine grasslands was communities without continuous snow cover which are dominated by moss and lichen species. They previously formed category E4.2, but no allocation exists for these habitats as a separate category for the new EUNIS classification (Chytrý et al., 2020). However, best represented is E4.2 in the new category R42 (Boreal and arctic acidophilous alpine grassland).

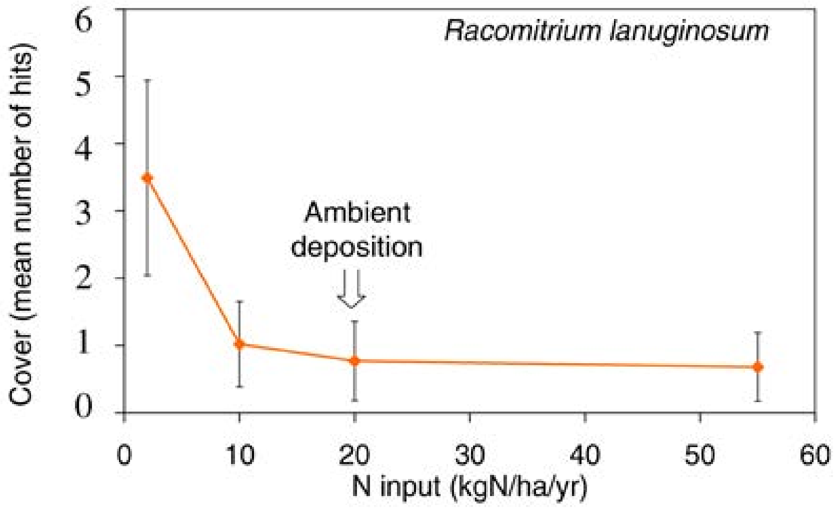
Since these communities are nutrient limited and many moss and lichen species are highly responsive to increased N deposition, it is likely that they are sensitive and should be assigned a low $CL_{emp}N$. The majority of N addition experiments and more recent gradient studies on this habitat type have been carried out in the *Racomitrium*-moss dominated heath, which is found on mountain summits in Britain and in montane areas of arctic and sub-arctic zones.

In the United Kingdom, there has been a serious decline in *Racomitrium* heath, over recent decades, with them being replaced by grass communities. Increasing rates of N deposition may be one of the main factors involved in the deterioration of *Racomitrium* heath (Thompson and Baddeley, 1991). However, evidence from experimental manipulation studies suggests that changes in grazing pressure also contribute to these changes. Pearce and Van der Wal (2002) set up an experiment in the north-east Scottish Highlands with montane *Racomitrium lanuginosum-Carex bigelowii* heath. In this experiment, plots on the summit were subject to low (10 kg ha^{-1} yr^{-1}) and high (40 kg ha^{-1} yr^{-1}) additions of N in two separate forms (NO_3^- and NH_4^+), during two consecutive summer seasons. Background deposition was estimated at 15 to 18 kg N ha^{-1} yr^{-1}. *Racomitrium* was shown to be extremely sensitive to even low N addition rates, responding with a raised tissue N concentration, and shoot growth rates which were less than 50% of those on the control plot. After only two growing seasons, Pearce and Van der Wal (2002) also demonstrated how quickly *Racomitrium* was replaced by graminoid species; *Racomitrium* cover was reduced by 31% at 10 kg ha^{-1} yr^{-1}, while graminoid cover increased by 57%. These results could reflect both a toxic effect and increased competition for light by graminoids, which utilised excess N.

The high sensitivity of *Racomitrium lanuginosum* to N deposition is also supported by the results from a glasshouse experiment that used monoliths taken from montane grassland in central Wales. In this experiment, N deposition and simulated grazing were manipulated over a four-year period (Jones, 2005). The applied N treatments were both above and below 20 kg N ha^{-1} yr^{-}

[1]. *Racomitrium* only persisted under heavy simulated grazing, confirming its sensitivity to competition for light by grasses. In these monoliths, a significant effect of N application became apparent within one year, with the cover of *Racomitrium* reducing from 10% at 2 kg ha^{-1} yr^{-1} to 2% or less at 10 kg ha^{-1} yr^{-1} and more (Figure 7.10).

Figure 7.10. Change in cover, detected by the number of hits using the pinpoint method, of the moss *Racomitrium lanuginosum* following N additions (> 20 kg N ha^{-1} yr^{-1}) as well as N reductions (< 20 kg N ha^{-1} yr^{-1}) on acidic grassland mesocosms in an experimental misting facility that excluded ambient N deposition.



Source: Jones, 2005; Emmett, 2007

However, a three-year manipulation study by Jónsdóttir et al. (1995), in which very low levels of N addition (4 kg N ha^{-1} yr^{-1}) were applied to a *Racomitrium-Carex* heath in Iceland with a background deposition of about 2 kg ha^{-1} yr^{-1}, found small non-significant increases in *Racomitrium* growth and shoot density. It was assumed that the small response to the low deposition rates used in this experiment was associated with growth limitations due to other factors.

The impacts of N deposition relative to those of climatic and grazing conditions on the composition of *Racomitrium* heath were examined along a N deposition gradient of 0.6-39.6 kg N ha^{-1} y^{-1}, and combined with climatic conditions across 36 European sites (Armitage et al., 2014). Besides climatic conditions, N deposition was the second most important driver, explaining 15% of variability, and it was more important than soil factors or current grazing. Along the N gradient, species richness declined by five species m^{-2} and there was a 30% shift in cover from mosses to graminoids. Changes in community composition were noticed across the whole range of N deposition, thus, there was no evidence for a threshold below which no change in composition occurred. Further analysis of this data suggests a substantial change in species richness between 0-5 and 5-10 kg N ha^{-1} yr^{-1} deposition categories (Wamelink et al., 2021).

Across 15 sites in the UK with N deposition ranging from 6.4-35.4 kg N ha^{-1} yr^{-1}, Britton et al. (2018) assessed the effects of N deposition on bryophyte litter quality, decomposition and C and N stocks in *Racomitrium* moss–sedge heath. Increasing N deposition reduced C:N in bryophyte litter, which in turn enhanced its decomposition. Thus, N-induced decomposition accelerated the depletion of the moss mat and allowed the graminoids to take over rapidly, pointing to an additional mechanism in addition to the light competition by graminoids.

At higher elevation and higher latitude, mountain summits are often dominated by the occurrence and abundance of lichens. Specific studies on lichens should therefore be accounted for. In a ten-year experiment, Fremstad et al. (2005) studied cover changes of different lichen species in low alpine and mid-alpine vegetation communities in south-central Norway. Nitrogen was added at 7, 35, and 70 kg ha^{-1} yr^{-1}; background N deposition was 2 to 4 kg ha^{-1} yr^{-1}. The most sensitive species at the warmer, low alpine site were the lichens *Alectoria nigricans* and *Cetraria ericetorum*, decreasing in cover at the lowest N application rate of 7 kg ha^{-1} yr^{-1}. Cover of another six lichens species declined at an N rate of 35 kg ha^{-1} yr^{-1}. Beside declines in cover, lichens developed discoloured and smaller thalli. Interestingly, the only lichen unaffected by any level of N application was the N_2-fixing lichen *Stereocaulon paschale*. However, at the colder, mid-alpine site, the lichen *Cetrariella delisei* was the only species in which cover changes over ten years due to N application of 70 kg ha^{-1} yr^{-1} were found and authors assumed a slower response time in colder climates.

In a meta-analysis referring to 39 articles and 31 experimental sites, Gutierrez-Larruga et al. (2020) found that N addition (in most studies between 10 and 56 kg N kg ha^{-1} yr^{-1}) accelerates lichen metabolism in both terricolous and epiphytic lichens in the short-term and decreases their abundance in the longer term. Early senescence of lichens is proposed as a possible mechanism linking the two observed responses. Currently, long-term effects of low N addition rates on lichens have not yet been adequately assessed in these habitats.

Summary for moss- and lichen-dominated mountain summits, ridges and exposed slopes

The experiments and more recent gradient studies described above and suggest that moss- and lichen-dominated mountain summits, ridges and exposed slopes habitats where background deposition is currently low have considerable potential for changes in the abundance of sensitive species and a decrease in lichen and bryophyte cover with even small increases in N. The 2011 $CL_{emp}N$ for moss and lichen dominated mountain summits, ridges and exposed slopes was set at 5-10 kg N ha^{-1} yr^{-1}. The more recent evidence supports that $CL_{emp}N$ and it should remain 5-10 kg N ha^{-1} yr^{-1} and is be considered ‘quite reliable’.

7.6 Overall summary for grasslands and lands dominated by forbs, mosses or lichens (EUNIS class R)

A summary of the $CL_{emp}N$ and their reliability is included in Table 7.1. Six $CL_{emp}N$ (presented in bold) have changed from the 2010 recommendations with all showing reductions either through a lowering of the whole $CL_{emp}N$ range or a narrowing of the range.

Referring to the entire grassland class R, there are some biogeographical regions in Europe, where no information is yet available on the effects of increased N deposition. This is the case of Pannonian, Steppic and Black Sea regions. Also in the Mediterranean region, regarded as a global biodiversity hotspot for conservation priorities, there are many grassland communities those sensitivity to N deposition has not been assessed. In particular, future attention should be paid to Mediterranean mountain areas as well as to alpine vegetation belts in other mountain systems (Pyrenees, Carpathians) where there are high levels of plant diversity and endemism.

Table 7.1. $CL_{emp}N$ and effects of exceedances on grasslands and lands dominated by forbs, mosses and lichens (R). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Semi-dry perennial calcareous grassland (meadow steppe)	R1A	15-25	##	**10-20**	##	Increase in tall grasses; decline in diversity; change in species composition; increased mineralisation; N leaching; surface acidification
Mediterranean closely grazed dry grasslands or Mediterranean tall perennial dry grassland or Mediterranean annual-rich dry grassland	R1D or R1E or R1F	15-25	(#)	**5-15**	(#)	Increased production; dominance by graminoids; changes to soil crusts; changes to soil nutrient cycling
Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta*	R1M	10-15	##	**6-10**	##	Increase in graminoids; decline of typical species; decrease in total species richness
Oceanic to subcontinental inland sand grassland on dry acid and neutral soils or Inland sanddrift and dune with siliceous grassland	R1P or R1Q	8-15	(#)	**5**-15	(#)	Decrease in lichens; increase in biomass
Low- and medium altitude hay meadows	R22	20-30	(#)	**10-20**	(#)	Increase in tall grasses; decrease in diversity; decline of typical species
Mountain hay meadows	R23	10-20	(#)	10-**15**	**#**	Increase in nitrophilous graminoids; changes in diversity; decline of typical species
Moist or wet mesotrophic to eutrophic hay meadow	R35	15-25	(#)	15-25	(#)	Increase in tall graminoids, decreased diversity; decrease in bryophytes

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Temperate and boreal moist and wet oligotrophic grasslands	R37	10-20	#	10-20	#	Increase in tall graminoids, decreased diversity; decrease in bryophytes
Temperate acidophilous alpine grasslands	R43	5-10	#	5-10	#	Changes in species composition; increase in plant production
Arctic-alpine calcareous grassland	R44	5-10	#	5-10	#	Changes in species composition; increase in plant production
Moss and lichen dominated mountain summits	(Earlier E4.2)	5-10	#	5-10	#	Change in species composition; effects on bryophytes or lichens

7.7 References

Aherne, J., Wilkins, K. and Cathcart, H. (2021). *Nitrogen-sulphur critical loads: assessment of the impacts of air pollution on habitats* (2016-CCRP-MS.43). EPA Research Programme 2014-2020. 42 p. Environmental protection agency, Wexford, Ireland.

Armitage, H.F., Britton, A.J., Van der Wal, R. and Woodin, S.J. (2014). The relative importance of nitrogen deposition as a driver of *Racomitrium* heath species composition and richness across Europe. *Biological Conservation* **171**, 224-231.

Arróniz-Crespo, M., Leake, J.R., Horton, P. and Phoenix, G.K. (2008). Bryophyte physiological responses to, and recovery from, long-term nitrogen deposition and phosphorus fertilisation in acidic grassland. *New Phytologist* **180**, 864 - 874.

Baker, M. E. and King, R. S. (2010). A new method for detecting and interpreting biodiversity and ecological community thresholds. *Methods in Ecology and Evolution* **1**, 25-37.

Bassin, S., Volk, M., Suter, M., Buchmann, N. and Fuhrer, J. (2007). Nitrogen deposition but not ozone affects productivity and community composition of subalpine grassland after 3 years of treatment. *New Phytologist* **175,** 523-534.

Bassin, S., Werner, R.A., Sörgel, K., Volk, M., Buchmann, N. and Fuhrer, J. (2009). Effects of combined ozone and nitrogen deposition on the in-situ properties of eleven key plant species of a subalpine pasture. *Oecologia* **58**, 474-756.

Bassin, S., Volk, M. and Fuhrer, J. (2013). Species composition of subalpine grassland is sensitive to nitrogen deposition, but not to ozone, after seven years of treatment. *Ecosystems* **16**, 1105-1117

Basto, S., Thompson, K., Phoenix, G.K., Sloan, V., Leake, J.R. and Rees, M. (2015). Long-term nitrogen deposition depletes grassland seed banks. *Nature Communications* **6**, 6185.

Beltman, B. and Barendregt, A. (2002). *Nature conservation and restoration ecology*. Report Landscape Ecology, Utrecht University.

Beltman, B., Willems, J.H. and Güsewell, S. (2007). Flood events overrule fertiliser effects on biomass production and species richness in riverine grasslands. *Journal of Vegetation Science* **18**, 625-634.

Berlin, G. (1998). *Semi-natural meadows in southern Sweden - changes over time and the relationship between nitrogen supply and management.* PhD thesis, Lund University.

Bobbink, R. and Willems, J.H. (1987). Increasing dominance of *Brachypodium pinnatum* (L.) Beauv. in chalk grasslands; a threat to a species-rich ecosystem. *Biological Conservation* **40**, 301-14.

Bobbink, R., Bik, L. and Willems, J.H. (1988). Effects of nitrogen fertilization on vegetation structure and dominance of *Brachypodium pinnatum* (L.) Beauv. in chalk grassland. *Acta Botanica Neerlandica* **37**, 231-242.

Bobbink, R., Den Dubbelden, K.C. and Willems, J.H. (1989). Seasonal dynamics of phytomass and nutrients in chalk grassland. *Oikos* **55**, 216-224.

Bobbink, R. (1991). Effects of nutrient enrichment in Dutch chalk grassland. *Journal of Applied Ecology* **28**, 28-41.

Bobbink, R., Boxman, D., Fremstad, E., Heil, G., Houdijk, A. and Roelofs, J. (1992). Critical loads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna. In: Grennfelt, P. and Thörnelöf, E. (eds.). *Critical loads for nitrogen*, p. 111. Nord 41, Nordic Council of Ministers, Copenhagen.

Bobbink, R., Hornung, M. and Roelofs, J.G M. (1996). *Empirical nitrogen critical loads for natural and semi-natural ecosystems*. Texte 71 - 96, III-1. Federal Environmental Agency, Berlin.

Bobbink, R., Ashmore, M., Braun, S., Flückiger, W. and Van den Wyngaert, I.J.J. (2003). Empirical critical loads for natural and semi-natural ecosystems: 2002 update. In: Achermann, B. and Bobbink, R. (eds.). *Empirical Critical Loads of Nitrogen*, SAEFL Report 164, Swiss Agency for Environment Forests and Landscape, Bern, pp. 43-169.

Bobbink, R. and Hettelingh, J.P. (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships.* Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), Bilthoven, The Netherlands.

Boch, S., Kurtogullari, Y., Allan, E., Lessard-Therrien, M., Rieder, N.S., Fischer, M., Martínez De León, G., Arlettaz, R. and Humbert, J.-Y. (2021). Effects of fertilization and irrigation on vascular plant species richness, functional composition and yield in mountain grasslands. *Journal of Environmental Management* **279**, e111629

Bonanomi, G., Caporaso, S. and Allegressa, M. (2006). Short-term effects of nitrogen enrichment, litter removal and cutting on a Mediterranean grassland. *Acta Oecologica* **30,** 419-425.

Bonanomi, G., Caporaso, S. and Allegrezza, M. (2009). Effects of nitrogen enrichment, plant litter removal and cutting on a species-rich Mediterranean calcareous grassland. *Plant Biosystems*, **143**, 443- 455.

Britton, A.J., Mitchell, R.J., Fisher, J.M., Riach, D.J., and Taylor, A.F.S. (2018). Nitrogen deposition drives loss of moss cover in alpine moss-sedge heath via lowered C:N ratio and accelerated decomposition. *New Phytologist* **218**, 470-478.

Calvete-Sogo, H., Elvira, S., Sanz, J., González-Fernández, I., Sánchez-Martín, L., Alonso, R. and Bermejo-Bermejo, V. (2014). Current ozone levels threaten gross primary production and yield of Mediterranean annual pastures and nitrogen modulates the response. *Atmospheric Environment* **95**, 197-206.

Calvete-Sogo, H., González-Fernández, I., Sanz, J., Elvira, S., Alonso, R., García-Gómez, H., Ibáñez-Ruiz, MA. and Bermejo-Bermejo, V. (2016). Heterogeneous responses of component species to ozone and nitrogen deposition shift the structure of Mediterranean annual pastures. *Oecologia* **181**, 1055-1067.

Calvete-Sogo, H., González-Fernández, I., García-Gómez, H., Alonso, R., Elvira, S., Sanz, J., and Bermejo-Bermejo, V. (2017). Developing ozone critical levels for multi-species canopies of Mediterranean annual pastures. *Environmental Pollution* **220**, 186-195.

Carroll, J.A., Caporn, S.J.M., Morecroft, M.D., Lee, J.A., Johnson, D., Taylor, A., Leake, J.R., Campbell, C.D., Cawley, L., Lei, Y. and Read, D.J. (1997). *Natural vegetation responses to atmospheric nitrogen deposition - critical levels and loads of nitrogen for vegetation growing on contrasting native soils*. Report University of Sheffield, Sheffield, 101.

Carroll, J.A., Johnson, D., Morecroft, M., Taylor, A., Caporn, S.J.M. and Lee, J.A. (2000). The effect of long-term nitrogen additions on the bryophyte cover of upland acidic grasslands. *Journal of* Bryology **22**, 83-89.

Carroll, J.A., Caporn, S.J.M., Johnson, D., Morecroft, M.D. and Lee, J.A. (2003). The interactions between plant growth, vegetation structure and soil processes in semi-natural acidic and calcareous grasslands receiving long-term inputs of simulated pollutant nitrogen deposition. *Environmental Pollution* **121**, 363-376.

Ceulemans, T., Merckx, R., Hens, M. and Honnay, O. (2011). A trait-based analysis of the role of phosphorus vs nitrogen enrichment in plant species loss across North-west European. *Journal of Applied Ecology* **48**, 1145-1163.

Ceulemans, T., Stevens, C.J., Duchateau, L., Gowing, D.J.G., Jaquemyn, H., Wallace, H., vans Rooijen, N., Goethem, T., Bobbink, R., Dorland, E., Gaudnik, C., Alard, D., Corcket, E., Muller, S., Dise, N., Duprè, C., Diekmann, M., Honnay, O. (2014) Soil phosphorus constrains biodiversity across European grasslands. *Global Change Biology* **20**, 3814-3822.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**, 648-675

Crawley, M.J., Johnston, A.E., Silvertown, J., Dodd, M., De Mazancourt, C., Heard, M.S., Henman, D.F. and Edwards, G.R. (2005). Determinants of Species Richness in the Park Grass Experiment. *The American Naturalist* **165**, 179-192.

Damgaard, C., Jensen, L., Frohn, L.M., Borchsenius, F., Nielsen, K.E., Ejrnæs, R. and Stevens, C.J. (2011). The effect of nitrogen deposition on the species richness of acid grasslands in Denmark: A comparison with a study performed on a European scale. *Environmental Pollution* **159**, 1778-1782.

Davies, C.E., Moss, D. and Hill, M.O. (2004). *EUNIS habitat classification revised 2004.* Winfrith Technology Centre, Dorian Ecological Information Ltd. and Monks Wood, UK.

De Graaf, M.C.C., Bobbink, R., Roelofs, J.G.M. and Verbeek, P.J.M. (1998). Differential effects of ammonium and nitrate on three heathland species. *Plant Ecology* **135**, 185-196.

De Graaf, M.C.C., Bobbink, R., Smits, N.A.C., Van Diggelen, R. and Roelofs, J.G.M. (2009). Biodiversity, vegetation gradients and key biogeochemical processes in the heathland landscape. *Biological Conservation* **10,** 2191-2201.

De Kroon, H. and Bobbink, R. (1997). Clonal plant dominance under elevated nitrogen deposition, with special reference to *Brachypodium pinnatum* in chalk grassland. In: De Kroon, H. and Van Groenendael, J.M. (eds.). *The ecology and evolution of clonal plants*. Backhuys Publishers, Leiden, pp. 359-379.

De Vries, W., Klijn, J.A. and Kros, J. (1994). Simulation of the long-term impact of atmospheric deposition on dune ecosystems in the Netherlands. *Journal of Applied Ecology* **31**, 59-73.

Dengler, J., Biurrun, I., Boch, S., Dembicz, I. and Török, P. (2020). Grasslands of the Palaearctic biogeographic realm: Introduction and synthesis. In: Goldstein, M. and DellaSala, D. (eds.) *Encyclopedia of the World's Biomes*. Elsevier

Diekmann, M., Jandt, U., Alard, D., Bleeker, A., Corcket, E., Gowing, D.J.G., Stevens, C.J. and Duprè, C. (2014). Long-term changes in calcareous grassland vegetation in North-western Germany - No decline in species richness, but a shift in species composition. *Biological Conservation* **172**, 170-179.

Dierschke, H. (1985). Experimentelle Untersuchungen zur Bestandesdynamik von Kalkmagerrasen (Mesobromion) in Südniedersachsen, I. Vegetationsentwicklung auf Dauerflächen 1972-1984. *Münstersche Geographische Arbeiten* **20**, 9-24.

Dodd, M.E., Silvertown, J., McConway, K., Potts, J. and Crawley, M. (1994). Application of the British national vegetation classification to the communities of the Park Grass experiment through time. *Folia Geobotanica and Phytotaxonomica* **29**, 321-334.

Duprè, C., Stevens, C.J., Ranke, T., Bleeker, A., Peppler-Lisbach, C., Gowing, D.J.G., Dise, N.B., Dorland, E., Bobbink, R. and Diekmann, M. (2010). Changes in species richness and composition in European acidic grasslands over the past 70 years: the contribution of cumulative atmospheric nitrogen deposition. *Global Change Biology* **16**, 344-357.

During, H.J. and Willems, J.H. (1986). The impoverishment of the bryophyte and lichen flora of the Dutch chalk grasslands in the thirty years 1953-1983. *Biological Conservation* **36**, 143-158.

Egloff, D.A. (1987). Food and growth relations of the marine microzooplankter, *Synchaeta cecilia* (Rotifera). *Hydrobiologia* **157,** 129-141

Ellenberg, H. (1988). *Vegetation of Central Europe.* Cambridge University Press, Cambridge

Ellenberg, H. (1996). Vegetation Mitteleuropas mit den Alpen in ökologischer, dynamischer und historischer Sicht. UTB, Ulmer, Stuttgart.

Emmett, B.A., Gordon, C., Williams, D.L., Woods, C., Norris, D., Bell, S.A. and Pughm, B. (2001). Grazing/nitrogen deposition interactions in upland acid grassland. Report to the UK Department of the Environment, Transport and the Regions, Centre for Ecology and Hydrology, Bangor, 53 pp.

Emmett, B.A. (2007). Nitrogen saturation of terrestrial ecosystems: some recent findings and their implications for our conceptual framework. *Water, Air and Soil Pollution: Focus* **7,** 99-109.

Emmett, B., Ashmore, M., Britton, A., Broadmeadow, M., Bullock, J., Cape, N., Caporn, S.J.M., Carroll, J.A., Cooper, J.R. Cresser, M.S., Crossley, A., d'Hooghe, P., De Lange, I., Edmondson, J., Evans, C.D., Field, C., Fowler, D., Grant, H., Green, E., Griffiths, B., Haworth, B., Helliwell, R., Hicks, K., Hinton, C., Holding, H., Hughes, S., James, M., Jones, A., Jones, M., Jones, M.L.M., Leake, J., Leith, I., Maskell, L., McNamara, N., Moy, I., Oakley, S., Ostle, N., Pilkington, M., Power, S., Prendergast, M., Ray, N., Reynolds, B., Rowe, E., Roy, D., Scott, A., Sheppard, L., Smart, S., Sowerby, A., Sutton, M., Terry, A., Tipping, E., Van den Berg, L., Van Dijk, N., Van Zetten, E., Vanguelova, E., Williams, B., Williams, D. and Williams, W. (2007). Terrestrial Umbrella: effects of eutrophication and acidification on terrestrial ecosystems. Final report. NERC/Centre for Ecology & Hydrology, 288 pp. (CEH Project Number: C02613, Defra Contract No. CPEA 18).

Field, C., Dise, N.B., Payne, R.J., Britton, A., Emmett, B.A., Helliwell, R., Hughes, S., Jones, L.M., Leake, J.R., Phoenix, G., Power, S., Sheppard, L., Southon, G., Stevens, C. and Caporn, S.J.M. (2014). Nitrogen drives plant community change across semi-natural habitats. *Ecosystems* **17**, 864-877.

Fremstad, E., Paal, J. and Mols, T. (2005). Impacts of increased nitrogen supply on Norwegian lichen rich alpine communities: a 10-year experiment. *Journal of Ecology* **93**, 471-481.

Gaudnik, C., Corcket, E., Clement, B., Delmas, C.E.L., Gombert-Courvoisier, S., Muller, S., Stevens, C.J. and Alard, D. (2011). Detecting the footprint of changing atmospheric nitrogen deposition loads on acid grasslands in the context of climate change. *Global Change Biology* **17**, 3351-3365.

Goulding, K.W.T., Bailey, N.J., Bradbury, N.J., Hargreaves, P., Howe, M., Murphy, D.V., Poulton, P.R. and Willison T.W. (1998). Nitrogen deposition and its contribution to nitrogen cycling and associated soil processes. *New Phytologist* **139**, 49-58

Gutiérrez-Larruga, B., Estébanez-Pérez, B. and Ochoa-Hueso, R. (2020). Effects of nitrogen deposition on the abundance and metabolism of lichens: a meta-analysis. *Ecosystems* **23**, 783-797.

Haworth, B.J., Ashmore, M.R. and Headley, A.D. (2007). Effects of nitrogen deposition on Bryophyte species composition of calcareous grasslands. *Water Air and Soil Pollution: Focus* **7,** 111-117.

Heer C. and Körner C.H. (2002). High elevation pioneer plants are sensitive to mineral nutrient addition. *Basic and Applied Ecology* **3,** 39-47.

Hejcman, M., Klaudisova, M., Schellberg, J and Honsova, D. (2007). The Rengen Grassland Experiment: Plant species composition after 64 years of fertilizer application. *Ariculture, Ecosystems and Environment* **122,** 259-266.

Helsen, K., Ceulemans, T., Stevens, C.J. and Honnay, O. (2014). Increasing Soil Nutrient Loads of European Semi-natural Grasslands Strongly Alter Plant Functional Diversity Independently of Species Loss. *Ecosystems* **17**, 169-181.

Henrys, P.A., Stevens, C.J., Smart, S.M., Maskell, L.C., Walker, K.J., Preston, C.D., Crowe, A., Rowe, E.C., Gowing, D.J. and Emmett, B.A. (2011). Impacts of nitrogen deposition on vascular plants in Britain: an analysis of two national observation networks. *Biogeosciences* **8**, 3501-3518.

Hiltbrunner, E., and Körner, C. (2004). Sheep grazing in the high alpine under global change. *Grassland Science in Europe*, **9**, 305-307.

Hiltbrunner, E., Schwikowiski, M and Korner, C. (2005). Inorganic nitrogen storage in alpine snow pack in the Central Alps (Switzerland). *Atmospheric Environment* **12,** 2249-2259.

Horswill, P., O'Sullivan, O., Phoenix, G.K., Lee, J.A. and Leake, J.R. (2008). Base cation depletion, eutrophication and acidification of species-rich grasslands in response to long-term simulated nitrogen deposition. *Environmental Pollution* **155**, 336-349.

Jacquemyn, H., Butaye, J. and Hermy, M. (2003). Short-term effects of different management regimes on the response of calcareous grassland vegetation to increased nitrogen. *Biological Conservation* **111**, 137-147.

Jeffrey, D.W. and Pigott, C.D. (1973). The response of grasslands on sugar limestone in Teesdale to application of phosphorus and nitrogen. *Journal of Ecology* **61**, 85-92.

Johnson, D., Leake, J.R., Lee, J.A. and Campbell, C.D. (1998). Changes in soil microbial biomass and microbial activities in response to 7 years simulated pollutant nitrogen deposition on a heathland and two grasslands. *Environmental Pollution* **103**, 239-250.

Johnson, D., Leake, J.R. and Lee, J.A. (1999). The effects of quantity and duration of simulated pollutant nitrogen deposition on root-surface phosphatase activities in calcareous and acid grasslands: a bioassay approach. *New Phytologist* **141**, 433-442.

Jones, M.L.M. and Ashenden, T.W. (2000). Critical loads of nitrogen for acidic and calcareous grasslands in relation to management by grazing. Centre for Ecology and Hydrology, Bangor.

Jones, M.L.M., Emmett, B.A. and Ashenden, T.W. (2002). *Grazing/nitrogen deposition interactions in upland acid moorland*. Contract Report, Centre for Ecology and Hydrology Bangor, Bangor Gwynedd LL57 2UP, U.K.

Jones, M.L.M. (2005). *Nitrogen deposition in upland grasslands: Critical loads, management and recovery*. PhD Thesis, University of Sheffield.

Jonsdottir, I.S., Fagerstroem, T. and Augner, M. (1995). *Variations in the population dynamics and local adaptations of some plant species along a circumpolar sector in relation to the population size of potential herbivores and climate*. Swedish Polar Research Secretariat, Stockholm.

Kirkham, F.W., Mountford, J.O. and Wilkins, R.J. (1996). The effects of nitrogen, potassium and phosphorus addition on the vegetation of a Somerset peat moor under cutting management. *Journal of Applied Ecology* **33**, 1013-1029.

Kleijn, D., Bekker R.M., Bobbink, R., De Graaf, M.C.C. and Roelofs, J.G.M. (2008). In search for key biogeochemical factors affecting plant species persistence in heathland and acidic grasslands: a comparison of common and rare species. *Journal of Applied Ecology* **45**, 680-687.

Körner, C.H. (1984). Auswirkungen von Mineraldünger auf alpine Zwergsträucher. *Verhandlungen der Gesellschaft für Ökologie* **12,** 123-136.

Körner, C.H, Diemer, M., Schäppi, B., Niklaus, P. and Arnone, J. (1997). The responses of alpine grassland to four seasons of CO_2 enrichment: a synthesis. *Acta Oecologica* **18,** 165-175.

Körner, C. (2021). *Alpine Plant Life - Functional plant ecology of high mountain ecosystems*. 3rd ed. Springer, Heidelberg.

Lee, J.A. and Caporn, S.J.M. (1999). Natural vegetation responses to atmospheric nitrogen deposition. Department of the Environment, Transport and the Regions, UK.

Lee, J.A., Caporn, S.J.M., Pilkington, M., Johnson, D. and Phoenix, G. (2000). *Natural vegetation responses to atmospheric nitrogen deposition - Critical levels and loads of nitrogen for vegetation growing on contrasting native soils*. Progress report, contract EPG 1/3/111, Department of the Environment, Transport and the Regions. Department of Animal and Plant Sciences, University of Sheffield, Sheffield S10 2TN.

Lee, J.A. and Caporn, S.J.M. (2001). Effects of enhanced atmospheric nitrogen deposition on semi-natural ecosystems. Progress report, 2000-01. Department of Animal and Plant sciences, University of Sheffield, Sheffield S10 2TN.

Luo, Y., El-Madany, T., Ma, X., Nair, R., Jung, M., Weber, U., Filippa, G., Bucher, SF., Moreno, G., Cremonese, E., Carrara, A., Gonzalez-Cascon, R., Cáceres Escudero, Y., Galvagno, M., Pacheco-Labrador, J., Martín, MP., Perez-Priego, O:, Reichstein, M., Richardson, AD., Menzel, A., Römermann, C., and Migliavacca, M. (2020). Nutrients and water availability constrain the seasonality of vegetation activity in a Mediterranean ecosystem. *Global Change Biology* **26**, 4379-4400.

Martini, D., Pacheco-Labrador, J., Perez-Priego, O., Van der Tol, C., El- Madany, T. S., Julitta, T., Rossini, M., Reichstein, M., Christiansen, R., Rascher, U., Moreno, G., Martín, P., Yang, P., Carrara, A., Guan, J., González-Cascón, R. and Moreno, G. (2019). Nitrogen and phosphorus effect on sun-induced fluorescence and gross primary productivity in Mediterranean grassland. *Remote Sensing* **11**, 2562.

Maskell, L.C., Smart, S.M., Bullock, J.M., Thompson, K. and Stevens, C.J. (2010). Nitrogen deposition causes widespread loss of species richness in British habitats. *Global Change Biology* **16**, 671-679.

Migliavacca, M., Perez-Priego, O., Rossini, M., El-Madany, T. S., Moreno, G., Van der Tol, C., Rascher, U., Berninger, A., Bessenbacher, V., Burkart, A., Carrara, A., Fava,F., Guan, J., Hammer, T.W., Henkel, K., Juarez-Alcalde, E., Julitta, T., Kolle, O., Martín, M.P., Musavi, T., Pacheco-Labrador, J., Pérez-Burgueño, A., Wutzler, T., Zaehle, S. and Reichstein, M. (2017). Plant functional traits and canopy structure control the relationship between photosynthetic CO_2 uptake and far-red sun-induced fluorescence in a Mediterranean grassland under different nutrient availability. *New Phytologist* **214**, 1078-1091.

Morecroft, M.D., Sellers, E.K. and Lee, J.A. (1994). An experimental investigation into the effects of atmospheric nitrogen deposition on two semi-natural grasslands. *Journal of Ecology* **82**, 475-483.

Morris, K. A., Nair, R. K. F., Moreno, G., Schrumpf, M. and Migliavacca, M. (2019). Fate of N additions in a multiple resource-limited Mediterranean oak savanna. *Ecosphere* **10**, e02921

Mosier, A.R., Stillwel, M., Paton, W.J. and Woodmansee, R.G. (1981). Nitrous oxide emissions from a native shortgrass prairie. *Journal of the American Soil Science Society* **45**, 617-619.

Mountford, J.O., Lakhani, K.H. and Holland, R.J. (1994). The effects of nitrogen on species diversity and agricultural production on the Somerset Moors, Phase II: *a. After seven years of fertiliser application. b. After cessation of fertiliser input for three years.* English Nature Research Report 86, 1-106. English Nature, Peterborough.

Nair, R. K. F., Morris, K. A., Hertel, M., Luo, Y., Moreno, G., Reichstein, M., Schrumpf, M., and Migliavacca, M. (2019). N:P stoichiometry and habitat effects on Mediterranean savanna seasonal root dynamics. *Biogeosciences* **16**, 1883-1901.

Neitzke, M. (1998). Changes in nitrogen supply along transects from farmland to calcareous grassland. *Zeitschrift für Pflanzenernährung und Bodenkunde* **161**, 639-646.

Neitzke, M. (2001). Analysis of vegetation and nutrient supply in calcareous grassland border zones to determine critical loads for nitrogen. *Flora* **196**, 292-303.

Ochoa-Hueso, R., Maestre, F.T., De los Ríos, A., Valea, S., Theobald, M.R., Vivanco, M.G., Manrique, E. and Bowker, M.A. (2013). Nitrogen deposition alters nitrogen cycling and reduces soil carbon content in low-productivity semiarid Mediterranean ecosystems. *Environmental Pollution* **179**, 185-193.

Ochoa-Hueso, R., Delgado-Baquerizo M., Gallardo, A., Bowker, M.A., and Maestre, F.T. (2016). Climatic conditions, soil fertility and atmospheric nitrogen deposition largely determine the structure and functioning of microbial communities in biocrust-dominated Mediterranean drylands. *Plant and Soil* **399**, 271- 282.

Olde Venterink, H., Van der Vliet, R.E. and Wassen, M.J. (2001). Nutrient limitation along a productivity gradient in wet meadows. *Plant and Soil* **234**, 171-179.

O'Sullivan, O.S., Horswill, P., Phoenix, G.K., Lee, J.A. and Leake, J.R. (2011). Recovery of soil nitrogen pools in species-rich grasslands after 12 years of simulated pollutant nitrogen deposition: a 6-year experimental analysis. *Global Change Biology* **17**, 2615-2628.

Pannak, A., Duprè, C., Gowing, D.J.G., Stevens, C.J. and Diekmann, M. (2015). Spatial gradient in nitrogen deposition affects plant species frequency in acidic grasslands. *Oecologia* **177**, 39-51.

Pardo, L.H., Fenn, M.E., Goodale, C.L., Geiser, L.H., Driscoll, C.T., Allen, E.B., Baron, J.S., Bobbink, R., Bowman, W.D., Clark, C.M., Emmett, B., Gilliam, F.S., Greaver, T.L., Hall, S.J., Lilleskov, E.A., Liu, L., Lynch, J.A., Nadelhoffer, K.J., Perakis, S.S., Robin- Abbott, M.J., Stoddard, J.L., Weathers, K.C. and Dennis, R.L. (2011). Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. *Ecological Applications*. **21**, 3049-3082.

Payne, R.J., Stevens, C.J., Dise, N.B., Gowing, C.J., Pilkington, M.G., Phoenix, G.K., Emmett, B.A. and Ashmore, M.R. (2011). Impacts of atmospheric pollution on the plant communities of British acid grasslands. *Environmental Pollution* **159**, 2602-2608.

Payne, R.J., Dise, N.B., Stevens, C.J. and Gowing, C.J. (2013). Impact of nitrogen deposition at the species level. *Proceedings of the National Academy of Sciences of the United States of America* **110**, 984-987.

Payne, R.J., Campbell, C., Stevens, C.J., Pakeman, R.J., Ross, L.C., Britton, A.J., Mitchell, R.J., Jones, L., Field, C., Caporn, S.J.M., Carroll, J.A., Edmondson, J., Carnell, E.J., Tomlinson, S., Dore, A.J., Dragosits, U. and Dise, N.B. (2020). Disparities between plant community responses to nitrogen deposition and critical loads in UK semi-natural habitats *Atmospheric Environment* **239**, 117478.

Pearce, I.S.K. and Van der Wal, R. (2002). Effects of nitrogen deposition on growth and survival of montane *Racomitrium lanuginosum* heath. *Biological Conservation* **104**, 83-89.

Peppler-Lisbach, C., Stanik, N., Könitz, N. and Rosenthal, G. (2019). Long-term vegetation changes in *Nardus* grasslands indicate eutrophication, recovery from acidification, and management change as the main drivers. *Applied Vegetation Science* **23**, 508-521.

Pérez-Priego, O., Guan, J., Rossini, M., Fava, F., Wutzler, T., Moreno, G., Carvalhais, N., Carrara, A., Kolle, O., Julitta, T., Schrumpf, M., Reichstein, M. and Migliavacca, M. (2015). Sun-induced chlorophyll fluorescence and photochemical reflectance index improve remote-sensing gross primary production estimates under varying nutrient availability in a typical Mediterranean savanna ecosystem. *Biogeosciences* **12**, 6351-6367.

Phoenix, G.K., Booth, R.E., Leake, J.R., Read, D.J., Grime, J.P. and Lee, J.A. (2003). Effects of enhanced nitrogen deposition and phosphorus limitation on nitrogen budgets of semi-natural grasslands. *Global Change Biology* **9,** 1309-1321.

Phoenix, G.K., Booth, R.E., Leake, J.R., Read, D.J., Grime, J.P. and Lee, J.A. (2004). Simulated pollutant nitrogen deposition increases P demand and enhances root-surface phosphatase activities of three plant functional types in a calcareous grassland. *New Phytologist* **161**, 279-289.

Pitcairn, C.E.R., Fowler, D. and Grace, J. (1991). Changes in species composition of semi-natural vegetation associated with the increase in atmospheric inputs of nitrogen. Institute of Terrestrial Ecology/NERC, Penicuik.

Pescott, O.L. and Jitlal, M. (2020). Reassessing the observational evidence for nitrogen deposition impacts in acid grassland: spatial Bayesian linear models indicate small and ambiguous effects on species richness. *PeerJ* **8**, e9070.

Ratcliffe, D.A. (1984). Post-medieval and recent changes in British vegetation: the culmination of human influence. *New Phytologist* **98**, 73-100.

Roelofs, J.G.M., Bobbink, R., Brouwer, E. and De Graaf, M.C.C. (1996). Restoration ecology of aquatic and terrestrial vegetation of non-calcareous sandy soils in the Netherlands. *Acta Botanica Neerlandica* **45**, 517-541.

Roth, T., Kohli, L., Rihm, B., and Achermann, B. (2013). Nitrogen deposition is negatively related to species richness and species composition of vascular plants and bryophytes in Swiss mountain grassland. *Agriculture Ecosystems and Environment* **178**, 121-126.

Roth, T., Kohli, L., Rihm, B., Meier, R. and Achermann, B. (2017). Using change-point models to estimate empirical critical loads for nitrogen in mountain ecosystems. *Environmental Pollution* **220**, 1480-1487.

Silvertown, J., Poultin, P., Johnston, E., Edwards, G., Heard, M. and Biss, P.M. (2006). The Park Grass Experiment 1856-2006: It's contribution to ecology. *Journal of Ecology* **94,** 801-814.

Smart, S.M., Stevens, C.J., Tomlinson, S.J., Maskell, L.C. and Henrys, P.A. (2021). Comment on Pescott & Jitlal 2020: Failure to account for measurement error undermines their conclusion of a weak impact of nitrogen deposition on plant species richness. *PeerJ* **9**, e10632

Smith, C.T., Elston, J. and Bunting, A.H. (1971). The effects of cutting and fertilizer treatment on the yield and botanical composition of chalk turfs. *Journal of the British Grassland Society* **26**, 213-219.

Sparrius, L.B. and Kooijman, A.M. (2011). Invasiveness of *Campylopus introflexus* in drift sands depends on nitrogen deposition and soil organic matter. *Applied Vegetation Science* **14,** 221-229.

Sparrius, L.B. and Kooijman, A.M. (2013). Nitrogen deposition and soil carbon content affect nitrogen mineralization during primary succession in acid inland drift sand vegetation. *Plant and Soil* **364**, 219-228.

Sparrius, L.B., Sevink, J. and Kooijman, A.M. (2012). Effects of nitrogen deposition on soil and vegetation in primary succession stages in inland drift sands. *Plant and Soil* **353**, 261-272.

Sparrius, L.B., Kooijman, A.M., Riksen, M.P.J.M., and Sevink, J. (2013a). Effect of geomorphology and nitrogen deposition on rate of vegetation succession in inland drift sands. *Applied Vegetation Science* **16**, 379-389.

Sparrius, L.B., Kooijman, A.M., and Sevink, J. (2013b). Response of inland dune vegetation to increased nitrogen and phosphorus levels. *Applied Vegetation Science* **16**, 40-50.

Spink, A., Sparks, R.E., Van Oorschot, M. and Verhoeven, J.T.A. (1998). Nutrient dynamics of large river floodplains. *Regulated Rivers: Research and Management* **14**, 203-216.

Stevens, C.J., Dise, N.B., Mountford, J.O. and Gowing, D.J. (2004). Impact of nitrogen deposition on the species richness of grasslands. *Science* **303**, 1876-1879.

Stevens, C.J., Maskell, L.C., Smart, S.M., Caporn, S.J.M., Dise, N.B. and Gowing, D.J.G. (2009). Identifying indicators of atmospheric nitrogen deposition impacts in acid grasslands. *Biological Conservation* **142**, 2069-2075.

Stevens, C.J., Thompson, K., Grime, P., Long, C.J. and Gowing, D.J.G. (2010a). Contribution of acidification and eutrophication to declines in species richness of calcifuge grasslands along a gradient of atmospheric nitrogen deposition. *Functional Ecology*, **24**, 478-484.

Stevens, C.J., Duprè, C. Dorland, E., Gaudnik, C., Gowing, D.J.G., Bleeker, A., Diekmann, M., Alard, D., Bobbink, R., Fowler, D., Corcket, E., Mountford, J.O., Vandvik, V., Arild Aarrestad, P., Muller, S. and Dise, N.B. (2010b). Nitrogen deposition threatens species richness of grasslands across Europe. *Environmental Pollution* **158**, 2940-2945.

Stevens, C.J., Dupre, C., Dorland, E., Gaudnik, C., Gowing, D.J.G., Bleeker, A., Diekmann, M., Alard, D., Bobbink, R., Fowler, D., Corcket, E., Mountford, J.O., Vandvik, V., Aarrestad, P.A., Muller, S. and Dise, N.B. (2011a). The impact of nitrogen deposition on acid grasslands in the Atlantic region of Europe. *Environmental Pollution* **159**(10), 2243-2250.

Stevens, C.J., Dupre, C., Gaudnik, C., Dorland, E., Dise, N.B., Gowing, D.J., Bleeker, A., Alard, D., Bobbink, R., Fowler, D., Corcket, E., Vandvik, V., Mountford, J.O., Aarrestad, P.A., Muller, S. and Diekmann, M. (2011b). Changes in species composition of European acid grasslands observed along a gradient of nitrogen deposition. *Journal of Vegetation Science* **22**, 207-215.

Stevens, C.J., Smart, S.M., Henrys, P., Maskell, L.C., Crowe, A., Simkin, J., Walker, K., Preston, C.D., Cheffings, C., Whitfield, C., Rowe, E., Gowing, D.J. and Emmett, B.A. (2012). Terricolous lichens as indicators of nitrogen deposition: Evidence from national records. *Ecological Indicators* **20**, 196-203.

Stiles, W.A.V., Rowe, E.C. and Dennis, P. (2017). Long-term nitrogen and phosphorus enrichment alters vegetation species composition and reduces carbon storage in upland soil. *Science of the Total Environment* **593-594**, 688-694.

Stiles, W.A.V., Rowe, E.C. and Dennis, P. (2018). Nitrogen and phosphorus enrichment effects on CO_2 and methane fluxes from an upland ecosystem. *Science of the Total Environment* **618**, 1199-1209.

Storkey, J., Macdonald, A.J., Poulton, P.R., Scott, T., Köhler, I.H., Schnyder, H., Goulding, K.W.T., and Crawley, M.J. (2015). Grassland biodiversity bounces back from long-term nitrogen addition. *Nature.* **528**, 401-404.

Storm, C. and Süss, K. (2008). Are low-productive plant communities responsive to nutrient addition? Evidence from sand pioneer grassland. *Journal of Vegetation Science* **19**, 343-354.

Tallowin, J.R. and Smith, R.E.N. (1994). *The effects of inorganic fertilisers in flower-rich hay meadows on the Somerset Levels*. English Nature Research Report 87. Peterborough, English Nature, 1-27.

Thompson, D.B.A. and Baddeley, J.A. (1991). Some effects of acidic deposition on montane *Racomitrium lanuginosum* heaths. In: Woodin, S.J. and Farmer, A.M. (eds.) *The Effects of Acid Deposition on Nature Conservation in Great Britain*, NCC Peterborough, Cambridgeshire, UK, pp. 17-28.

Tipping, E., Henrys, P., Maskell, L. and Smart, S. (2013). Nitrogen deposition effects on plant species diversity; threshold loads from field data. *Environmental Pollution* **179**, 218-223.

Tomassen, H., Bobbink, R., Peters, R., Van der Ven, P. and Roelofs, J. (1999). *Kritische stikstofdepositie in heischrale graslanden, droge duingraslanden en hoogvenen: op weg naar meer zekerheid*. Eindrapport in het kader van het Stikstof Onderzoek Programma (STOP), 1997-1999. Nijmegen and Utrecht, Katholieke Universiteit Nijmegen en Universiteit Utrecht, 1-46.

Unkovich, M., Jamieson, J., Monaghan, R. and Barraclough, D. (1998). Nitrogen mineralisation and plant nitrogen acquisition in a nitrogen-limited calcareous grassland. *Environmental and Experimental Botany* **40**, 209-219.

Van den Berg, L.J.L., Tomassen, H.B.M., Roelofs, J.G.M. and Bobbink, R. (2005). Effects of nitrogen enrichment on coastal dune grassland: A mesocosm study. *Environmental Pollution* **138,** 77-85.

Van den Berg, L.J.L., Vergeer, P., Rich, T.C.G., Smart, S.M., Guest, D. and Ashmore, M.R. (2011). Direct and indirect effects of nitrogen deposition on species composition change in calcareous grasslands. *Global Change Biology* **17**, 1871-1883.

Van den Berg, L.J.L., Jones, L., Sheppard, L.J., Smart, S.M., Bobbink, R., Dise, N.B. and Ashmore, M.R. (2016). Evidence for differential effects of reduced and oxidised nitrogen deposition on vegetation independent of nitrogen load. *Environmental Pollution* **208**, 890-897.

Van Duren, I.C., Strykstra, R.J., Grootjans, A.P., Ter Heerdt, G.N.J. and Pegtel, D.M. (1998). A multidisciplinary evaluation of restoration measures in a degraded *Cirsio-Molinietum* fen meadow. *Applied Vegetation Science* **1**, 115-130.

Van Tooren, B.F., Od, B., During, H.J. and Bobbink, R. (1990). Regeneration of species richness of the bryophyte layer of Dutch chalk grasslands. *Lindbergia* **16**, 153-160.

Vermeer, J.G. (1986). The effects of nutrients on shoot biomass and species composition of wetland and hayfield communities. *Acta Oecologica, Oecologia Plantarum* **7**, 31-41.

Virtanen, R., Johnston, A.E., Crawley, M.J. and Edwards, G.R. (2000). Bryophyte biomass and species richness on the Park Grass Experiment, Rothamsted, UK. *Plant Ecology* **151**, 129-141.

Volk, M., Enderle, J. and Bassin, S. (2016). Subalpine grassland carbon balance during 7 years of increased atmospheric N deposition. *Biogeosciences.* 13, 3807-3817.

Volk, M., Obrist, D., Novak, K., Giger, R., Bassin, S. and Fuhrer, J. (2010). Subalpine grassland carbon dioxide fluxes indicate substantial carbon losses under increased nitrogen deposition, but not at elevated ozone concentration. *Global Change Biology* **17**, 366-376.

Volk, M., Suter, M., Wahl, A.-L. and Bassin, S. (2021). Subalpine grassland productivity increased with warmer and drier conditions, but not with higher N deposition, in an altitudinal transplantation experiment. *Biogeosciences* **18**, 2075-2090.

Wamelink, G.W.W., Goedhart, P.W., Roelofsen, H.D., Bobbink, R., Posch, M., Van Dobben, H.F. and Data providers (2021). *Relaties tussen de hoeveelheid stikstofdepositie en de kwaliteit van habitattypen*. Wageningen Environmental Research, Wageningen, Rapport 3089.

Wells, T.C.E. (1974). Some concepts of grassland management. In: Duffey, E. (ed.). Grassland ecology and wildlife management. Chapman and Hall, London, 163-74.

Wells, T.C.E., Sparks, T.H., Cox, R. and Frost, A. (1993). *Critical loads for nitrogen assessment and effects on southern heathlands and grasslands*. Report to National Power, Institute of Terrestrial Ecology, Monks Wood, UK.

Wilkins, K., Aherne, J. and Bleasdale, A. (2016). Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high *Atmospheric Environment* **164**, 324-331.

Williams, E.D. (1978). Botanical composition of the Park Grass plots at Rothamsted 1856-1976. Rothamsted Experimental Station Internal Report, Harpenden.

Wolkinger, F. and Plank, S. (1981). *Dry grasslands of Europe*. Council of Europe, Strasbourg.

Wüst-Galley, C., Volk, M. and Bassin, S. (2021). Interaction of climate change and nitrogen deposition on subalpine pastures. *Journal of Vegetation Science* **32**, e12946.

8 Effects of nitrogen deposition on heathland, scrub and tundra habitats (EUNIS class S, formerly F)

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Alpine heath (S2) in Scotland. Photo: Andrea Britton.

Summary

The amount of new (long-term) experimental evidence for N impacts on heathland, scrub and tundra habitats published since the review of 2010 is limited. However, a large number of gradient studies have become available, covering many of these habitats.

From the experimental and gradient studies, a clear picture is emerging that deposition rates of as low as 5 kg N ha^{-1} yr^{-1} can affect tundra ecosystems (S1) and arctic, alpine and subalpine scrub habitats (S2). Therefore, empirical N critical loads ($CL_{emp}N$) for these habitats were maintained or narrowed to 3 to 5 kg ha^{-1} yr^{-1} and 5 to 10 kg ha^{-1} yr^{-1} respectively.

Gradient studies also allowed a first $CL_{emp}N$ of 5 to 15 kg N ha^{-1} yr^{-1} to be set for lowland to montane temperate and submediterranean Juniperus scrub (S31). In addition, a number of gradient studies provided evidence for N deposition impacts on vascular plant and bryophyte species composition at or below the lower end of the previous $CL_{emp}N$ range for upland Calluna-dominated wet heath (S411), lowland Erica tetralix -dominated wet heath (S411) and dry heaths (S42). Therefore, the $CL_{emp}N$ ranges for these habitats were lowered to 5 to 15 kg N ha^{-1} yr^{-1}.

Maquis, arborescent matorral and thermo-Mediterranean scrub (S5) are less well studied but are expected to show similar N responses to related Garrique and woodland habitats. Therefore, the $CL_{emp}N$ range for Maquis, arborescent matorral and thermo-Mediterranean scrub (S5) was lowered to 5 to 15 kg N ha^{-1} yr^{-1}, classifying it as 'expert judgement'. New experimental evidence on the Garrique habitat (S6) allowed a first estimate of $CL_{emp}N$ of 5 to 15 kg N ha^{-1} yr^{-1}.

8.1 Introduction

Historically, heathlands have played an important role in the western European landscape. The term heath generally describes various types of plant communities, but here the term is being applied to those plant communities for which the dominant life form is that of small-leaved dwarf shrubs that form a canopy at one metre or less above soil surface. Dwarf shrub canopies may be discontinuous and interspersed with grasses and forbs, and a ground cover of mosses or lichens is often present (Gimingham et al., 1979; De Smidt, 1979). Heathlands are classified together with scrub and tundra habitats in class S of the European Nature Information System (EUNIS). This class includes all dry and seasonally wet inland vegetation (cover > 30%) that is dominated by shrubs, dwarf shrubs or scrubs (Davies and Moss, 2002; Davies et al., 2004). In some subcategories of this class, the vegetation is determined by climate. Here, succession towards woodland is inhibited by drought, low temperature, or growing season length (e.g. categories S1, and S2). In contrast, the extensive inland, lowland dwarf-shrub heathlands in Atlantic and sub-Atlantic Europe are man-made and have existed for several centuries. In these heaths, the development towards woodland is prevented by regular mowing, burning, sheep grazing or sod removal. They are dominated by *Ericaceae*, especially *Calluna vulgaris* in the dry heathlands and *Erica tetralix* in the wet heathlands, or *Erica cinerea* in the western Atlantic heathlands (e.g. Gimingham et al., 1979). These communities are found on nutrient-poor mineral soils with a low pH (3.5-4.5), which makes them sensitive to the effects of both eutrophication and acidification caused by elevated nitrogen (N) deposition. Shrublands in the Mediterranean (e.g. categories S3, S5, S6) occur on a wide variety of soil types, including limestone and sandy soils. Succession towards woodland in these categories is often inhibited by severe drought, occasional fires and grazing by livestock. Because of their high nature conservation value, many scrub and heathlands have been designated as nature reserves.

In accordance with the EUNIS habitat classification, this chapter distinguishes the following categories and subcategories. Tundra (S1), arctic, alpine and subalpine scrub habitats (S2), temperate and Mediterranean montane scrub (S3), temperate shrub heathland habitats (S4), with subcategories of wet (S41) and dry (S42) heaths. In view of their functional differences, wet heaths are subdivided, according to climate, into northern (S411) and southern (S412) wet heaths. For southern wet heaths, no data are available to assign them a $CL_{emp}N$. For northern wet heaths a separate $CL_{emp}N$ was assigned when dominated by *Calluna vulgaris* (upland *Calluna* moorlands) or *Erica tetralix*. Coastal dune heaths have been categorised as coastal habitats (Chapter 4.2.3; N18, N19), and acidic grasslands with some heather species as grassland habitats (Chapter 7.2; R1M). Lowland Mediterranean scrub lands are covered in the maquis, arborescent matorral and thermo-Mediterranean scrubs (S5) and Garrigue (S6). Both habitats are characterised by temperate to Mediterranean climate and consist of evergreen and sclerophyllous shrublands and heathlands. For other EUNIS categories in class S (S7, 8 and 9), including other heathland types, no $CL_{emp}N$ have been determined, due to a lack of data availability.

8.2 Tundra (S1) and arctic, alpine and subalpine scrub habitats (S2)

Alpine and arctic habitats have many ecological characteristics in common, although climatic conditions are more severe in arctic regions than in most alpine regions. The growing season is short, temperatures are low, winds are frequent and strong, and the distribution of plant communities depends on the distribution of snow during winter and spring. Most alpine and all arctic zones are influenced by frost or solifluction. A continuous snowpack in winter insulates the soils from freezing temperatures. Although soil organic matter contents are typically high, decomposition and nutrient cycling are frequently slow, and the low nutrient supply limits primary production (Robinson and Wookey, 1997). Despite these constraints, there are a number of plant species growing in the tundra and arctic and sub-arctic, including herbaceous species, small dwarf shrubs, sedges and tussock grasses, reindeer and other lichens (crustose and foliose) and bryophytes (mosses and liverworts).

In classifying these communities under the EUNIS system, it is necessary to distinguish between tundra (S1) and arctic, alpine and subalpine scrub habitats (S2). Tundra is defined as vegetated land with graminoids, shrubs, mosses and lichens, overlying permafrost (Davies et al., 2004). The presence of permafrost prevents root penetration and often keeps the ground waterlogged in summer. European tundras are limited to Spitsbergen, Norway and northern Russia. A similar vegetation type occurs on boreal mountains and in the low arctic region, far away from the main permafrost region, for instance in Fennoscandia and Iceland. These habitats are listed under alpine and subalpine grasslands (R4), or arctic, alpine and subalpine scrub habitats (S2). The latter comprise scrub habitats that occur north of or above the climatic tree line, but outside the permafrost zone, and scrub occurring close to but below the climatic tree line where trees are locally suppressed by late lying snow, wind or repeated grazing.

8.2.1 Tundra (S1)

Tundra habitats are divided into two subcategories: shrub tundra (S11) and moss and lichen tundra (S12). Shrub tundras have extensive cover of small dwarf-shrubs over herbs, mosses, and lichens. They are sporadically found on permafrost soils of the southern arctic and subarctic zones, often grazed into mosaics dominated by grasses. Dominant species are typically dwarf shrubs such as *Empetrum nigrum*, *Rubus chamaemorus, and Cassiope tetragona* with mosses such as *Pleurozium schreberi*. Moss and lichen tundras are found in the middle and northern high arctic zone where permafrost soils dominate. These have sparse cover of mosses, lichens, and low-stature herbs. Dominating species are dwarf shrubs such as *Empetrum nigrum* and *Betula nana* with lichens and mosses including *Cladonia stellaris*, *Racomitrium lanuginosum* and *Cetraria nivalis* (Schaminée et al., 2014). There are not enough data at the sub-category level available to assign a $CL_{emp}N$ to these tundra subcategories. However, studies are available that provide information at the category level.

Numerous nutrient addition field studies have been conducted in manipulating tundra ecosystems. Most of the early studies added NPK fertiliser (e.g. Robinson et al., 1998; Press et al., 1998; Schmidt et al., 2000). There are also studies that used single and large applications of N (50 and 250 kg ha^{-1} yr^{-1}) (see Henry et al., 1986; Shaver and Chapin, 1995). Unfortunately, such high application rates in a single dose do not realistically simulate atmospheric N depositions and, since they do not allow conclusions regarding $CL_{emp}N$, they are excluded from the following review.

In an experimental study on dwarf shrub tundra, significant effects have been reported at much lower N application rates (Baddeley et al., 1994). In that mixed tundra heath near Svalbard, Spitsbergen (with an estimated background deposition of 1.5 kg N ha^{-1} yr^{-1}) plots were located in three different tundra heath vegetation types, which received factorial combinations of N (10 and 50 kg N ha^{-1} yr^{-1}) and phosphorus (5 kg P ha^{-1} yr^{-1}) in four to five applications during the growing seasons. Plots were treated for specific periods of time; those dominated by *Dryas octopetala* were treated from 1991 to 1998, those dominated by *Salix polaris* from 1991 to 1997, and those dominated by *Cassiope tetragona* from 1991 to 1993. Baddeley et al. (1994) reported early responses to the N additions (after one year of treatment); *Salix polaris* had increased levels of foliar N, increased leaf biomass and an increased photosynthetic rate at both N levels. *Cassiope tetragona* showed no response to the N addition, whilst *Dryas octopetala* showed an intermediate response in these measured variables.

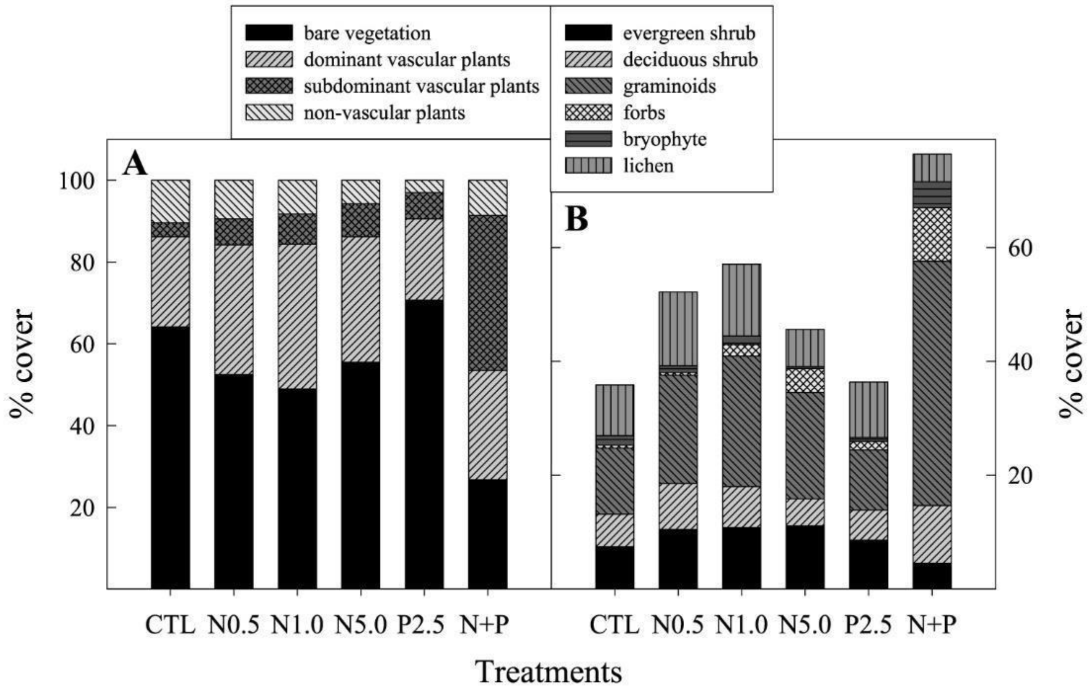
After cessation of the treatment applications (in1993, 1997 and 1998, respectively), Gordon et al. (2001) re-examined the impacts of N on these plots with particular attention to the bryophyte communities. Overall bryophyte cover was unaffected by increased N addition, although this was the net result of different individual species responses. For example, *Polytrichum juniperinum* increased in cover, whilst *Dicranum scoparium* cover declined. Tissue N concentration increased with increasing addition. Importantly, a number of significant persistent effects were observed at additions of 10 kg N ha^{-1} yr^{-1}. Nitrate reductase activity was clearly inhibited in *Polytrichum juniperinum*, suggesting N saturation at 10 kg N ha^{-1} yr^{-1}. This N saturation of bryophytes is of ecosystem importance, since N inputs may pass through the bryophyte layer (e.g. Britton et al., 2008; Britton et al., 2019), becoming available for soil microbes and higher plants. This may lead to increased growth of vascular plant species and hence, changes in species composition over time. Nitrogen addition also increased the proportion of green bryophyte shoots to a small extent, thus, apparently increasing bryophyte biomass production. The increased 'greenness' of the bryophyte cover on the fertilised *Cassiope* heath plots was independent of the P additions and was observed five years after N additions had ceased. This suggests that the added N was retained within the bryophyte layer of the *Cassiope* heath.

A subsequent revisit to this experiment 18 years after cessation of the additions showed that the effects of N treatments persisted and were P-dependent (Street et al., 2015). In plots where P was added, there were still effects of low N addition on community composition and nutrient dynamics. Further, N + P additions caused increased moss abundance, which influenced nutrient dynamics. These results show a lack of recovery in tundra and show that even small amounts of N deposition may potentially cause long-term ecological changes. Similar conclusions were reached by Liu et al. (2020) who examined the effects of continued nutrient additions or recovery (no further additions) in plots in northern Sweden that had previously been treated with high levels of N (50 kg N ha^{-1} yr^{-1}) for eight to ten years. After the initial nutrient additions (eight to ten years), no further responses to additional N treatments (four years) were observed, suggesting that these subarctic habitats were resilient to further changes six years after cessation of additions. They concluded that recovery of a subarctic ecosystem to conditions prior to nutrient amendment may be very slow due to the slow responsiveness and possibly the high levels of N retention. Choudhary et al. (2016) assessed the fate of N deposition (0, 4, 40, and 120 kg N ha^{-1} yr^{-1}) to high arctic bryophyte dominated tundra in a ^{15}N labelling experiment. They found that more than 95% of the total ^{15}N applied was recovered after one growing season, demonstrating the considerable capacity of this ecosystem to retain N from deposition events in arctic tundra. Regardless of the application rate or form, the following sinks were found (in order of magnitude): non-vascular plants > vascular plants > organic soil > litter > mineral soil.

In many experiments in tundra ecosystems, N additions were studied in relation to P additions or climatic variables. Co-limitation by N and P was clearly demonstrated on tundra heath in Svalbard (Street et al., 2015). Hence, $CL_{emp}N$ for tundra ecosystems may be dependent on factors such as P availability. Shaver et al. (1998) suggested that wet tundra sites are more likely to be P limited than moist sites with a thinner peat layer, while dry tundra deserts are primarily N limited. Cornelissen et al. (2001) examined relationships between lichen and vascular plant abundances in arctic manipulation experiments (manipulated factors were temperature and nutrient availability). They concluded that negative correlations were greater at sites in milder climates with a greater above-ground biomass, where increased shading and litter production negatively affected the lichens.

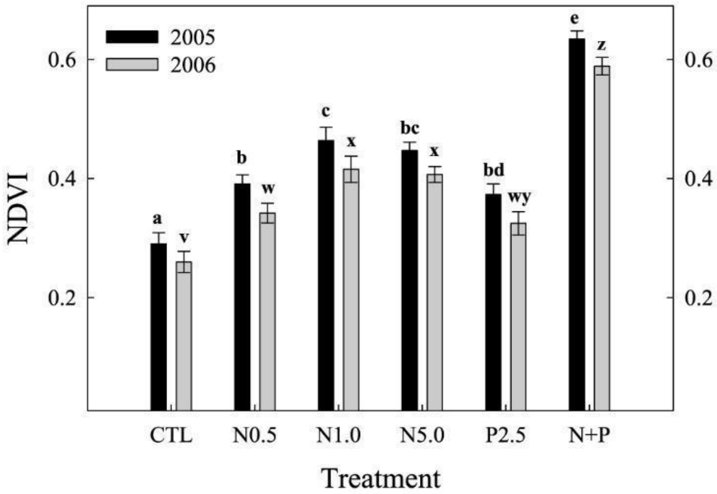
Arens et al. (2008) studied the effects of additions of N (5, 10 and 50 kg N ha^{-1} yr^{-1}) on vegetation characteristics and CO_2 exchange in a high arctic prostrate dwarf-shrub, herb tundra in north-western Greenland (background deposition < 1 kg N ha^{-1} yr^{-1}). They used factorial additions of N and P (25 kg P ha^{-1} yr^{-1}) to test for potential co-limitations. Dry ammonium nitrate and/or commercial phosphate fertiliser were applied twice during the growing seasons of three consecutive years. At the study site, approximately 40% of the area was covered by vascular plants, of which *Salix arctica*, *Carex rupestris* and *Dryas integrifolia* were the dominant species, and 60% of the area was unvegetated. Vegetation cover and composition and ecosystem CO_2 exchange appeared to be very sensitive to low rates of N inputs (5 kg ha^{-1} yr^{-1}). Additions of 5 kg N ha^{-1} yr^{-1} led to a significant increase in *Salix arctica*, 10 kg N ha^{-1} yr^{-1} almost doubled the cover of graminoids, and 50 kg N ha^{-1} yr^{-1} resulted in a more than seven-fold increase in the cover of forbs (Figure 8.1). The mean NDVI (Normalised Difference Vegetation Index = cover of green biomass) was calculated for each plot (20x40cm) and showed a saturation response to increasing levels of N addition, such that the largest NDVI response occurred at 10 kg N ha^{-1} yr^{-1}, while no further increase in NDVI was observed at 50 kg N ha^{-1} yr^{-1} (Figure 8.2). Next to a decrease in ecosystem respiration and photosynthesis in the plots receiving 50 kg N ha^{-1} yr^{-1}, the results suggest N saturation in the ecosystem between 10 and 50 kg N ha^{-1} yr^{-1} (Arens et al., 2008). Combined additions of both N (50 kg N ha^{-1} yr^{-1}) and P dramatically increased ecosystem photosynthesis and respiration, leading to a drastic increase in the cover of graminoids (especially *Festuca brachyphylla*; Figure 8.1).

Figure 8.1. Percentage of cover per (a) cover type and (b) functional group, in 2006, following three years of N additions in a prostrate dwarf-shrub, herb tundra (north-western Greenland). Each bar represents the mean of six plots for each treatment. N+P treatment = 2.5 g P m^{-2} yr^{-1} + 5.0 g N m^{-2} yr^{-1} Numbers behind the type of treatment indicate nutrient load in g m^{-2} yr^{-1} (times 10 for kg N ha^{-1} yr^{-1}).



Source: Arens et al., 2008

Figure 8.2. Mean NDVI (Normalised Difference Vegetation Index; a vegetation index that is correlated with the presence of photosynthetically active vegetation), in 2005 and 2006, representing the second and third growing season, respectively. Values represent the treatment mean (n = 6) ± 1.0 SE. Bars with the same letter are not significantly different at alpha = 0.05. N+P treatment =2.5 g P m^{-2} yr^{-1} + 5.0 g N m^{-2} yr^{-1} Numbers behind the type of treatment indicate nutrient load in g m^{-2} yr^{-1} (times 10 for kg N ha^{-1} yr^{-1}).



Source: Arens et al., 2008

Summary Tundra (S1)

Despite the limited number of long-term experiments, a clear picture is emerging of the potential impact of long-term N deposition on tundra ecosystems. Ecosystem response to N has been observed at deposition rates of as low as 5 kg N ha^{-1} yr^{-1}. To a large extent, however, the response to atmospheric N within tundra ecosystems may well depend on other factors, such as P status or climate.

In the $CL_{emp}N$ revision by Bobbink and Hettelingh (2011), the experiment by Arens et al. (2008) confirmed that tundra ecosystems are very sensitive to additional loads of N. Since significant effects were already seen at additions of as low as 5 kg N ha^{-1} yr^{-1}, a new $CL_{emp}N$ was set at 3 to 5 kg ha^{-1} yr^{-1} and considered as 'quite reliable'. The increasing number of studies documenting effects of N deposition within this critical load, strengthen the arguments for setting the $CL_{emp}N$ at 3 to 5 kg ha^{-1} yr^{-1}. The $CL_{emp}N$ for the North American tundra ecoregion has been suggested to be set at 1 to 3 kg ha^{-1} yr^{-1} (Nadelhoffer and Geiser, 2011; Pardo et al., 2011). However, to do so for Europe, studies documenting effects of N additions of 3 kg ha^{-1} yr^{-1} and below are needed.

The strong responses to N in situations where P was also applied are an indication of N and P co-limitation, identifying P as an important modifier of the $CL_{emp}N$. Thus, higher critical loads should be applied to systems that are limited by P, and lower critical loads to systems that are not.

8.2.2 Arctic, alpine and subalpine scrub habitats (S2)

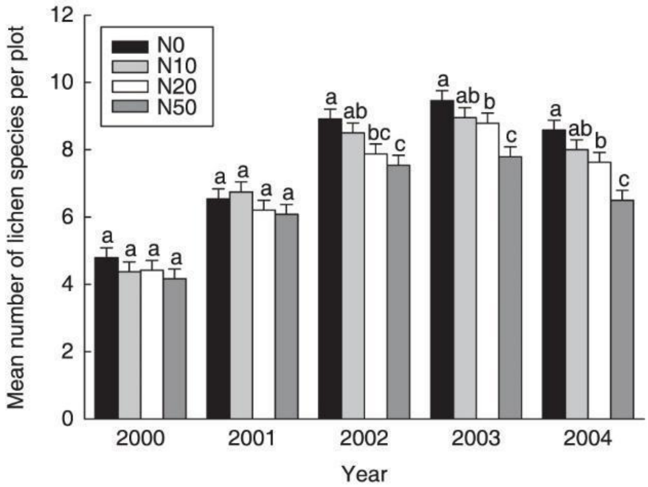
The EUNIS class of arctic, alpine and subalpine scrub habitats is subdivided into seven categories: subarctic and alpine dwarf willow scrubs (S21), alpine and subalpine ericoid heath (S22), alpine and subalpine juniper scrub (S23), subalpine *Genista* scrub (S24), subalpine and subarctic deciduous scrubs (S25), subalpine *Pinus mugo* scrub close to tree line (S26) and scrub close to the tree line with conifers other than *Pinus mugo* (S27) (Chytrý et al., 2020). Dwarf willow scrubs (S21) are well developed in boreal and arctic mountains and in subarctic lowlands. Alpine and subalpine ericoid heaths (S22) are dwarf or prostrate shrub formations in alpine and subalpine mountainous zones, dominated by ericaceous species or other woody species such as *Dryas octopetala, Kalmia procumbens or Rhododendron* species*.* Alpine and subalpine juniper scrub (S23) is dominated by dwarf junipers and may occur as either primary vegetation or as a result of deforestation and long-term grazing at high elevations. Subalpine *Genista* scrub (S24) occurs on high mountains around the Adriatic region. Subalpine deciduous scrubs (S25) include the subalpine scrubs of *Alnus*, *Betula*, *Salix* and *Rosaceae* (*Amelanchier, Potentilla, Rubus, Sorbus*), which are less than five metres tall, often accompanied by tall herbs. The last categories of conifer scrub close to the tree line (S26 and S27) relate to scrubland with dwarf conifers (krummholz) shaped by strong winds, often with incomplete canopy cover. Treelines dominated by *Pinus mugo* are classified as S26, and those dominated by other conifers as S27. The tree species at the arctic tree line can grow to large stature under favourable conditions. Despite the distinction of seven subcategories, most of the studies for this class have focussed on the alpine and subalpine ericoid heath (S22). Only a few studies have incorporated subalpine and subarctic deciduous scrubs (S25).

Experimental studies

A study in the Dovre mountains in Norway at 1000 to 1400 m above sea level, investigated the effects of three years of fertilisation of a *Betula nana* dominated community (S25) with 12 and 61 kg N ha^{-1} yr^{-1}, at a site with an estimated background deposition of 2 to 4 kg N ha^{-1} yr^{-1} (Paal et al*.,* 1997). There was no significant effect on plant growth, and no evidence of increased N content in vegetation or soils. In two other studies (Möls et al*.,* 2001; Fremstad et al*.,* 2005), two lichen-dominated communities were investigated, one in low-alpine and the other in middle-alpine regions. After ten years of applications of 7, 35 and 70 kg ha^{-1} yr^{-1}, there was no significant effect on vascular plants. Lichens proved to be more sensitive; the cover of the lichens *Alectoria ochroleuca* and *Cetraria nivalis* had already decreased at the lowest dose of 7 kg N ha^{-1} yr^{-1}. A possible reason for the limited effects of N on vascular plants and soils in this long-term experiment could be that other factors such as climate, soil properties and community structure may have been more important for determining species composition and cover (Fremstad et al*.,* 2005).

Britton and Fisher (2007a) studied the effects of N deposition on low-alpine *Calluna* – *Cladonia* heath (S22) in Scotland (background deposition of 10 kg N ha^{-1} yr^{-1}). Three levels of N addition (10, 20 and 50 kg N ha^{-1} yr^{-1}) were applied over a five-year period. After five years, *Calluna vulgaris* shoot extension was stimulated by N additions of 10 kg ha^{-1} yr^{-1}, indicating that low-alpine heathlands are very sensitive to low levels of N deposition (Britton and Fisher, 2008). Diversity of lichens was significantly reduced at additions of more than 10 kg N ha^{-1} yr^{-1} (Figure 8.3). Nitrogen addition caused rapid and significant increases in N content and N:P and N:K ratios of *Calluna vulgaris* following the two highest N treatments, suggesting increasing P and potassium limitation of growth. Soil C:N ratios declined significantly with N addition (only 50 kg N ha^{-1} yr^{-1}), indicating N saturation and increasing likelihood of N leakage (Britton et al*.,* 2008). A gradient study in low-alpine heathlands across Scotland suggested, based on N:P ratios, that growth of *Calluna vulgaris* on most sites is usually co-limited by N and P or P limited, due to the accumulated long-term N deposition in these mountain regions (Britton and Fisher, 2007b).

Figure 8.3. Effect on mean lichen species richness (+ 1 SE) of N addition treatments of 10, 20 and 50 kg ha^{-1} yr^{-1} (background deposition of 10 kg N ha^{-1} yr^{-1}). Means for years not sharing the same letter are significantly different (P < 0.05); error bars show the standard error of the mean.



Source: Britton and Fisher, 2007a

In the N addition experiment on low-alpine *Calluna* heath described above (Britton and Fisher, 2007a), Nitrogen addition was also combined with burning and grazing (clipping) management treatments. Burning had a large effect on plant diversity and vegetation composition, but both recovered quickly. Nitrogen addition interacted with burning; burned plots showed no significant effect of N on species diversity, while the diversity on unburned plots was significantly reduced following the 10 kg N ha^{-1} yr^{-1} treatment (Britton and Fisher, 2007a; 2008). Clipping had no effect on plant diversity. This suggests that burning mitigates the impacts of low-dose N addition on species diversity.

The effects of 14 kg N ha^{-1} yr^{-1} addition on the emergence and survival of seedlings in subalpine heath dominated by *Empetrum hermaphroditum* and *Vaccinium uliginosum* was studied in Swedish Lapland (Milbau et al., 2017). Nitrogen addition significantly enhanced seedling emergence and survival for *Betula nana*, had no effect on *Solidago vigaurea* and reduced seedling establishment in *Vaccinium myrtillus*. Also utilising N additions at the lower limit of the critical load, Britton and Fisher (2010) tested the effects of both N load and concentration on thallus chemistry and growth of five terricolous alpine lichen species in a laboratory study. Responses to N addition varied between species; thallus N concentration was positively related to N load in *Cetraria islandica*, *Cladonia rangiferina* and *Flavocetraria nivalis*, with the greatest change occurring between 2.5 and 12.5 kg N ha^{-1} yr^{-1}, but there was no relationship for *Platismatia glauca*. In this short (three month) study, impacts of the N additions on growth were only seen in two species (*Alectoria nigricans* and *Cetraria islandica*) both of which exhibited negative, linear relationships between growth rate and N concentration. In the Pacific Northwest of the USA, Simpson et al. (2019) tested the effects of low levels of N addition (3, 5 and 10 kg N ha^{-1} yr^{-1}) on soil chemistry in alpine heaths dominated by *Phyllodoce*, *Cassiope* and *Vaccinium* species with a background deposition of 0.2-2 kg N ha^{-1} yr^{-1}. This three-year study showed increases in soil NO_3^- and NH_4^+ availability in response to additions of 5 kg N ha^{-1} yr^{-1} and declines in extractable soil organic carbon and increases in extractable soil NO_3^- and microbial N in response to additions of 10 kg N ha^{-1} yr^{-1}.

New experimental studies also contribute further evidence of the impacts of N deposition above the current $CL_{emp}N$ range. Papanikolaou et al. (2010) studied the effects of N additions of 10, 20 and 50 kg N ha^{-1} yr^{-1} on litter decomposition and associated enzyme activities and microbial communities in a Scottish *Calluna*-dominated alpine heath with a background deposition of 10 kg N ha^{-1} yr^{-1}. They found no effect of N addition on litter mass loss over a two-year period, but activity of phosphomonoesterase in the litter was significantly increased by the 10 kg ha^{-1} yr^{-1} N addition. Bacterial community richness in litter harvested after two years increased with N addition and bacterial and fungal community composition (as determined by terminal restriction fragment length polymorphism) were also altered by N additions, although the magnitude of these effects was small. The effects of N additions on multiple ecosystem parameters over an eleven-year period at the same Scottish alpine heath are summarised by Phoenix et al. (2012). Additions of 10 kg N ha^{-1} yr^{-1} above background significantly increased *Calluna* productivity and flowering, increased availability of NO_3^- and NH_4^+ in the soil and increased leaching of NH_4^+. With larger additions of 20 and 50 kg N ha^{-1} yr^{-1} there was also evidence of increased frost damage of *Calluna*, enhanced N mineralisation in the soil and leaching of NO_3^- into soil water (Helliwell et al., 2010; Phoenix et al., 2012).

Gradient studies

In addition to the experimental studies, gradient studies including areas of low background N deposition provide some of the best evidence of N deposition impacts on vegetation composition at or below the current critical load. Three studies, which used vegetation composition data to identify community change points (thresholds) associated with N deposition, have been published since the previous critical loads update. In Swiss alpine heaths (S22) with data spanning a deposition range of approximately 5-19 kg N ha^{-1} yr^{-1} (modelled total N deposition at 0.1 × 0.1 km resolution), Roth et al. (2017) reported a community change point of 10.7-10.8 kg N ha^{-1} yr^{-1} above which total vascular plant richness, the richness of species associated with oligotrophic conditions and richness of conservation target species all declined. A study of alpine heaths in the UK (Payne et al., 2020) over a deposition range of 4.9-19.4 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution) that included data on lichens and bryophytes, found individual species change points occurring from the lowest deposition levels in the dataset, with negatively responding taxa clustered into two groups responding around 5-7 and 10-14 kg N ha^{-1} yr^{-1}. A third study from Ireland (Wilkins et al., 2016) included slightly lower N deposition levels (range of 4.0-19.9 kg N ha^{-1} yr^{-1} for alpine heaths, observation-based total N deposition at 5 × 5 km resolution). In this study, change points for the complete vegetation community (including bryophytes and lichens) were identified and considered to be evidence of 'significant harmful effects' if the number of positive indicator species (plant species indicative of good habitat status) that declined in cover was greater than the number that increased in cover. In alpine heaths (S2) twenty species declined in response to N deposition, including nine positive indicator species, while only five species (including one positive indicator) increased. Eleven of the twenty declining species were bryophytes or lichens. The community change point for declining species was estimated at 5.5 kg N ha^{-1} yr^{-1}. Additional support for the impacts of low levels of N deposition on terricolous lichen species in heathland is provided by Stevens et al. (2012). In this study, records of lichen presence on a 10 km grid across the UK were used to model probability of presence at a given level of N deposition (CBED model, total N deposition at 5 × 5 km resolution). Several heathland lichen taxa were found to decline in response to N deposition, with declines in prevalence occurring from the lowest levels of N deposition (approximately 5 kg N ha^{-1} yr^{-1}) and many taxa reaching a very low probability of presence by 20 kg N ha^{-1} yr^{-1}.

Summary Arctic, Alpine and subalpine scrub (S2)

Studies on the effects of N addition to the vegetation types within the S2 class are scarce and most use N additions which fall above or within the $CL_{emp}N$ range when background deposition is incorporated (Aerts, 2010, Phoenix et al., 2012, Manninen and Tolvanen, 2013). A few gradient studies that include areas of low background N deposition have provided evidence for impacts around the lower $CL_{emp}N$ limit (Wilkins et al., 2016; Payne et al., 2020) but almost all evidence for N impacts in the EUNIS S2 category relates to alpine and subalpine ericoid heaths (S22). Since there is very limited evidence for impacts in other alpine and subalpine heath and scrub habitats, critical loads are only provided for the S2 level. Previously, the $CL_{emp}N$ for arctic, alpine and subalpine scrubs was set at 5 to 15 kg ha^{-1} yr^{-1} (Bobbink and Hettelingh, 2011). Data from new experimental studies with low background deposition provide additional evidence for biological and biogeochemical impacts occurring within this range. In addition, survey studies encompassing low background deposition areas provide evidence for N deposition impacts at the lower end of the current $CL_{emp}N$ range, particularly affecting bryophytes and lichens. Therefore, we propose to narrow the $CL_{emp}N$ range, setting it at 5 to 10 kg ha^{-1} yr^{-1} for arctic, alpine and subalpine scrub habitats (S2), classifying it as 'quite reliable'. For this habitat, similarly to S1, responses to N deposition depend on N and P co-limitation. Higher critical loads should be applied to systems that are limited by P, and lower critical loads to systems that are not.

8.3 Temperate and Mediterranean-montane scrub (S3)

Temperate and Mediterranean-montane scrub (S3) habitats are subdivided into eight categories. However, few of these habitats have published studies relevant to $CL_{emp}N$, with the exception of two gradient studies in lowland to montane temperate and submediterranean *Juniperus* scrub (S31). Gruwez and colleagues (Gruwez et al., 2014) collected seeds from 42 populations of *Juniperus communis* throughout its distribution in Europe to assess the effects of climate and atmospheric depositions on seed viability. Seed viability was determined using seed dissection. Nitrogen deposition, which ranged from 1.8 to 36.0 kg ha^{-1} yr^{-1} across the study area (EMEP total N deposition at 50 × 50 km resolution), was negatively related to seed ripening time, with the proportion of seeds ripening in two vs. three years rapidly increasing across the deposition range of ~2 to 10 kg N ha^{-1} yr^{-1}. Potentially acidifying deposition, including N deposition was also associated with a reduction in seed viability. The authors suggested that the failure of natural regeneration in many European juniper populations might be attributed to enhanced atmospheric deposition of N. Wilkins et al. (2016), with updates in Aherne et al. (2021), identified vegetation community change points for *Juniperus* scrub using species abundance data from 191 relevés in Ireland covering a deposition gradient of ~3 to 25 kg N ha^{-1} yr^{-1} (observation-based total N deposition at 5 × 5 km resolution). The community change point for declining species was estimated at 6.2 kg N ha^{-1} yr^{-1} with 19 species decreasing in abundance (16 of which were positive indicator species considered typical for this habitat).

Summary Temperate and Mediterranean-montane scrub (S3)

The gradient studies support the possible sensitivity of *Juniperus* scrub to N deposition. Here we propose a $CL_{emp}N$ range of 5 to 15 kg N ha^{-1} yr^{-1} for lowland to montane temperate and submediterranean *Juniperus* scrub (S31), classifying it as 'expert judgement' given the limited studies.

8.4 Temperate shrub heathlands (S4)

As discussed above, both wet and dry heathlands (S41 and S42) have been placed within EUNIS class S4 (temperate shrub heathlands), because they occur in the Atlantic climate region and are dominated by ericoid shrubs. This EUNIS class has been divided into subcategories of wet heaths (S41), which are characterised by wet, organic-rich to peat soils, and dry heaths (S42) characterised by free-draining sandy-podzol soils. Both upland *Calluna* moorlands and lowland wet heaths dominated by *Erica tetralix* fall within the category of 'northern' wet heaths (S411). However, since these communities are clearly ecologically different, it is important that this distinction is retained. Therefore, each habitat has been assigned a different critical load. Given that there is no elevation-based cut-off that can be recommended to distinguish the two habitats, the primary criterion must be that of species dominance.

Since Bobbink and Hettelingh (2011), the number of new relevant experimental studies is limited, with many using high N additions, e.g. 50+ kg N ha^{-1} yr^{-1} or in regions with high background deposition, e.g. 20+ kg N ha^{-1} yr^{-1}. However, continuation of some of the original studies provide valuable data on long term effects. Several publications on the effects of N on upland *Calluna vulgaris* heath have focused on management measures to counteract the negative effects of N on locations where critical loads are being exceeded. In contrast, a notable number of new gradient studies have been published since the Bobbink and Hettelingh (2011) update. These studies have generally considered plant species data from large-scale surveys and have primarily focused on changes in species richness and plant community composition along a N deposition gradient either in the UK (Maskell et al., 2010; Henrys et al., 2011; Tipping et al., 2013; Field et al., 2014; Van den Berg et al., 2016), Denmark (Strandberg et al., 2012; Damgaard et al., 2014), or Ireland (Wilkins et al., 2016; Aherne et al., 2021). A number of these studies have used change-point analysis, such as TITAN, or similar statistical analysis to identify a deposition threshold where change occurs (Strandberg et al., 2012; Tipping et al., 2013; Wilkins et al., 2016; Aherne et al., 2021). Further, these gradient studies typically include regions with low N deposition, e.g. less than 5 or 10 kg N ha^{-1} yr^{-1}, allowing for the assessment of impacts at lower N inputs compared with experimental additions.

8.4.1 Upland Calluna-dominated wet heath (upland moorland) (S411)

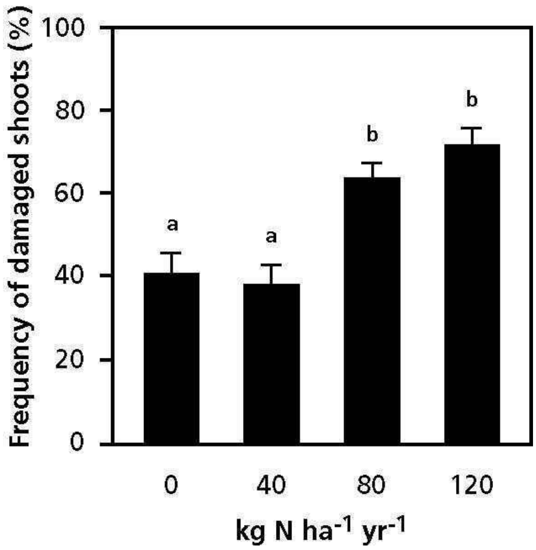
Calluna vulgaris dominated, upland moorland heaths (S411) are generally developed on acidic, peaty organic soils, and are characterised by a dominance of dwarf shrubs (in particular *Calluna vulgaris*) and a high abundance of bryophyte species. The effects of N deposition on upland *Calluna vulgaris* heaths have been studied in the UK using both field surveys and experiments. The $CL_{emp}N$ range of 10 to 20 kg N ha^{-1} yr^{-1} that was recommended by Bobbink et al. (1996; 2003) and Bobbink and Hettelingh (2011) for this community, was based on three types of evidence from these UK studies: effects on growth and species composition, effects on shoot nutrient concentration, and effects on soils and root characteristics. Experimental studies reported after 2010 support this range (described below), while gradient studies encompassing low background deposition regions provide evidence for N deposition impacts at the lower end of the current $CL_{emp}N$ range.

Effects on growth and species composition

The longest running N manipulation experiment in *Calluna vulgaris* moorland is at Ruabon, north Wales, where experiments were established in May 1989 on moorland at an altitude of 470 metres (Caporn et al., 1994). This site was estimated recently to receive an atmospheric N deposition of approximately 19 kg N ha^{-1} yr^{-1} (APIS 2021). In this experiment, additions of N as ammonium nitrate at 40, 80 and 120 kg N ha^{-1} yr^{-1}, have been applied since 1989, at monthly intervals (Caporn et al., 1994; Lee and Caporn, 1998; Carroll et al., 1999; Pilkington et al., 2007a). The initial period from 1989 to 1993 was characterised by positive effects of N on *Calluna vulgaris* in terms of increased shoot growth, N concentration and flowering, with no indication that the dose applied exceeded the capacity of the plants for uptake and subsequent growth. The following three years of the study, however, showed a much-reduced effect of the treatment on shoot extension, and no clear dose response to increasing N inputs. The 1996 data, in particular, showed no effect at all of N on shoot extension after seven years (Carroll et al., 1999). One interpretation of the *Calluna vulgaris* growth responses in this experiment is that additions of N accelerated the natural *Calluna vulgaris* cycle, with earlier ageing and opening of the canopy in the plots that received the highest doses.

One factor causing increased canopy opening, with potential for grass invasion, may be greater damage in winter to *Calluna vulgaris* shoots. Detailed experimental studies on frost tolerance in *Calluna vulgaris* shoots collected in the early years of the study (1989-1994) demonstrated that N addition had actually improved frost tolerance in autumn (Caporn et al., 1994). However, field surveys in 1996 and 1998 showed large increases in 'winter browning' of heather shoots in the late winter, most notably following additions of 80 and 120 kg N ha^{-1} yr^{-1} (Figure 8.4) (Carroll et al., 1999; Lee et al., 2000). The mechanism behind this damage may have been frost injury.

Figure 8.4. Effects of seven years of ammonium nitrate additions (kg N ha^{-1} yr^{-1}) on the frequency (means ± SE) of damaged *Calluna vulgaris* shoots in northern wet heath (S411) in North Wales. Columns sharing a letter are statistically not significantly different.

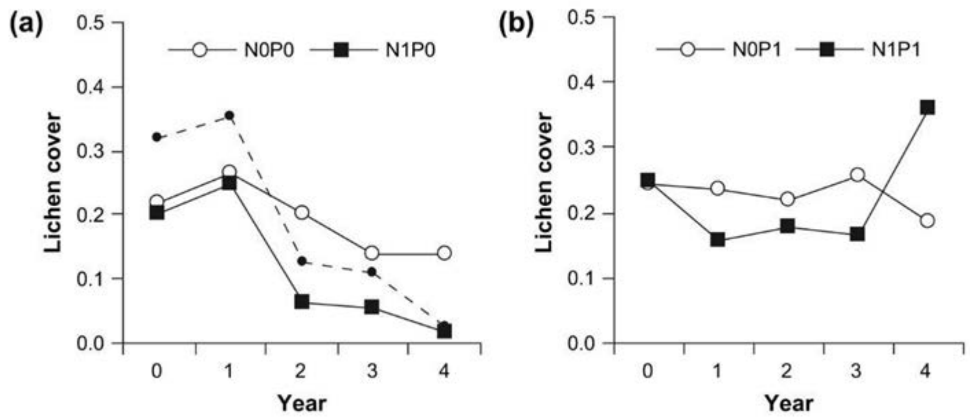


Source: Carroll et al., 1999

In the earlier years, both bryophytes and lichens had disappeared from below the *Calluna vulgaris* canopy following all N addition treatments, although *Vaccinium myrtillus* persisted (Carroll et al., 1999). It is not certain, however, whether this was a direct effect of N addition or a response to changes in *Calluna vulgaris* canopy cover and increased litter production resulting in reduced light penetration. A later survey, following ten years of treatment, showed not only that bryophytes, notably *Hypnum jutlandicum,* had returned to the N treated plots, but also their cover had actually increased with increasing N depositions, probably as a result of the accelerated ageing and opening up of the *Calluna vulgaris* canopy (Lee et al., 2000). No such response was found in lichen species.

Adjacent to the above original experiment, Pilkington et al., (2007a) treated experimental plots in a upland mature growth phase *Calluna* heath with factorial combinations of N (0 and 20 kg N ha^{-1} yr^{-1}) and P (0 and 5 kg ha^{-1} yr^{-1}). They found that lichen cover had virtually disappeared within four years from plots that had received 20 kg N ha^{-1} yr^{-1} and from separate plots that had received 10 kg N ha^{-1} yr^{-1} (background N deposition of 16.4 kg N ha^{-1} yr^{-1}). However, this effect was reversed by the addition of P (Figure 8.5). Continued research on the same experiment by Edmondson (2007) demonstrated similar effects of N and P treatments on the mosses and liverworts as well as lichens. The adverse effects of N were evident for species such as the liverwort *Lophozia ventricosa* but less so for the more nutrient tolerating *Hypnum jutlandicum* (Edmondson et al., 2013).

Figure 8.5. Effect of factorial combinations of N and P additions on the cover of lichens in an upland *Calluna vulgaris* heath (background N deposition of 16.4 kg N ha^{-1} yr^{-1}). N0 = 0 kg N ha^{-1} yr^{-1}, N1 = 20 kg N ha^{-1} yr^{-1}, P0 = 0 kg P ha^{-1} yr^{-1} and P1 = 5 kg P ha^{-1} yr^{-1}. The dotted line in Figure (a) indicates a separate N addition of 10 kg N ha^{-1} yr^{-1}. The decline in Lichen cover in the control (N0P0) reflects the increased growth of the *Calluna* canopy.



Source: Pilkington et al., 2007a

The experimental plots at Ruabon were subject to a controlled burn in 2000 in order to simulate traditional management of these systems. Subsequent post-burn regrowth, primarily from stem bases, was significantly lower in the plots with higher N treatments (Lee and Caporn, 2001). Pilkington et al. (2005) completed an N budget for the site based on harvesting data. Significant increases in green tissue, wood and litter biomass, and litter depth were found for all levels of N treatments. Although most of the added N in the 40 kg N ha^{-1} yr^{-1} treatment was found in green tissue and litter, increasing amounts of added N were found in the peat horizons in the plots with higher N treatments (Pilkington et al., 2005).

Effects on shoot nutrient content

Several studies found that N content in shoots of *Calluna vulgaris* and bryophytes was significantly higher in areas with higher N inputs (> 10-15 kg N ha^{-1} yr^{-1}), substantially higher compared with measurements of historical plant material (Pitcairn et al., 1995). Indeed, the N concentration of *Calluna* shoots, common mosses (e.g. *Hypnum jutlandicum*) and leaf litter were suggested to be reliable indicators of atmospheric N deposition across UK heathlands (Caporn et al., 2014). Additionally, increases in N concentration in shoots have been linked to increased performance of winter moths (*Operophtera brumata*); infestations of which have led to extensive damage to heather moorlands in Scotland over the last decade (Kerslake et al., 1998).

In the long-term experiment in north Wales, analysis of foliar nutrients between 1989 and 1992 showed increased N concentrations with increasing doses of N, and measurements taken in 1996 still showed a significant increase in response to the earlier N additions (Carroll et al., 1999). By 1996, N:P ratios had clearly increased as a result of the earlier treatments, with values of 23:1 in the plots that had received the highest N treatment (120 kg N ha^{-1} yr^{-1}), compared to values of 16:1 in control plots (Carroll et al., 1999). Carroll et al. (1999) compared these values with the critical threshold for the N:P ratio of 14:1 to 16:1, as proposed by Koerselman and Meuleman (1996), to indicate a switch from N to P limitation, and suggested that the onset of P limitation might explain the declining response to N in shoot extensions, which occurred at this stage of the experiment. This interpretation is supported by evidence of increased phosphomonoesterase activity in peat and root surfaces (Johnson et al., 1998; Lee et al., 2000).

This observation is also consistent with Kirkham (2001), who sampled *Calluna vulgaris* shoots from a number of sites in England and Wales. The N:P ratios in shoots of *Calluna vulgaris* were above 16:1 at about half of the sampled sites, suggesting that N deposition had changed a substantial proportion of *Calluna*-dominated uplands in the UK from N-limited ecosystems into P-limited or N:P co-limited heaths, again (see also tundra) stressing the modulating effects of P.

Effects on soil and root characteristics

Calluna vulgaris roots characteristically exhibit a substantial degree of ericoid mycorrhizal infection (Yesmin et al., 1996), which is important for the degradation of complex organic substances in order to give plants access to N sources that would otherwise be unavailable to them. The N addition experiment in North Wales consistently showed little effect on mycorrhizal infection levels (Caporn et al., 1995; Lee et al., 2000), using either the ergosterol method or visual assessments. In contrast, Yesmin et al. (1996) reported a negative correlation between N deposition and mycorrhizal infection rate at five remote Scottish sites with a total deposition in the range of 2 to 10 kg N ha^{-1} yr^{-1}. In addition, a separate greenhouse study showed a small but significant decrease in infection rate when deposition rates were increased from 12 to 24 kg N ha^{-1} yr^{-1} for one year.

Soil studies at Ruabon, north Wales, found a number of responses to N additions. Leaching rates, although showing a small response to the higher N addition treatments, account for only a very small percentage of the added N (Pilkington et al., 2005). There was also evidence of increased mineralisation and decreased C:N ratio in the litter and rhizosphere with increasing N treatments, although this effect was not found below two centimetres in the soil (Lee and Caporn, 2001). Recent work at this site showed that N addition of 20 kg N ha^{-1} yr^{-1} significantly increased storage of C and N in the litter layer and surface soil horizons (Field et al., 2017). Overall, results implied a high retention of the added nitrate and ammonium, probably through microbial immobilisation; Johnson et al. (1998) demonstrated that the long-term application of N at a site increased soil microbial biomass. Curtis et al. (2005) used a stable isotope tracer (^{15}N) to determine the fate of N inputs in a gradient study (varying from 6.4 to 30.7 kg N ha^{-1} yr^{-1}) applied in small doses over a one-year period. The purpose of the study was to determine the fate of ^{15}N-labelled N additions at four sites selected from the UK Acid Waters Monitoring Network. The sites represented gradients of total N deposition and leaching losses of inorganic N (in north-east Scotland, mid-Wales and north-west England) measured as part of a larger N budget study. Mosses and lichens showed far greater ^{15}N recovery per unit of biomass than grasses or ericaceous shrubs. High N deposition rates reduced the biomass of mosses and lichens and thereby the absorption capacity of the cryptogams and the proportion of N recovered; this may lead to increased nitrate leaching.

N deposition and management measures

These upland *Calluna vulgaris* heaths are actively managed by cutting or burning. In the UK, rotational burning of upland moorland (once every 7 to 20 years) is a commonly used management practice to maintain *Calluna vulgaris* stands. Pilkington et al. (2007b) studied the effects of moorland burning on N pools and leaching in a long-term N manipulation experiment in North Wales. Burning increased leaching of total dissolved inorganic N and dissolved organic N from organic and mineral soil horizons. Nitrogen additions magnified the effect of burning on leaching losses but reduced this effect on the N pools in the mineral layer. Pilkington et al. (2007b) concluded that burning approximately every ten years may be effective in removing N retained in the system, at N deposition rates of up to 56 kg N ha^{-1} yr^{-1}, although burning exacerbates the threat of N loading to groundwater in heavily N-polluted areas.

In addition, grazing is likely to be an important regulating factor. In general, active management of *Calluna vulgaris* moorlands is thought to reduce the impact of increased N deposition and allows for higher critical loads but the effect strongly depends on the grazing levels (Alonso et al., 2001; Hartley et al., 2003; Hartley and Mitchell, 2005).

Gradient studies

Since Bobbink and Hettelingh (2011), a large number of broad-scale gradient studies in the UK have assessed the impacts of N deposition on upland and lowland heaths (Edmondson et al., 2010; Field et al., 2014; Henrys et al., 2011; Stevens et al., 2012; Southon et al., 2013; Tipping et al., 2013; Van den Berg et al., 2016). These studies provide some of the best evidence of N impacts on vegetation composition at or below the current critical load. In general, upland and lowland heath habitats have primarily been categorised as dry heaths (S42) in these studies but they likely include wet heaths (S411) given their underlying organic soil; only one of the broad-scale studies directly identified S411 (Tipping et al., 2013). Field et al. (2014) and Southon et al. (2013) studied 25 upland heaths along a deposition gradient of 5.9 to 32.4 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution). Here, heathlands were ascribed to S411 given their underlying organic soil, dominance by *Calluna vulgaris* and high rainfall. Field et al. (2014) found reduced species richness and changed species composition associated with higher N deposition. Species richness declined by about 40% of maximum species richness from the lowest to the highest N deposition sites (32 kg N ha^{-1} yr^{-1}), with a steeper decline in species richness in the lower deposition range < ~11 kg N ha^{-1} yr^{-1}. Similarly, Southon et al. (2013) found that the abundance of nitrophilous species increased with increasing N deposition (CBED model, total N deposition at 5 × 5 km resolution). The number of forb and graminoid species also decreased with increasing total N deposition. The biggest losses were seen at the lower end of the N deposition gradient, with an average of 13 species lost per site between 5 to 10 kg N ha^{-1} yr^{-1}, compared to an average of only three species lost as N increased from 10 to 20 kg N ha^{-1} yr^{-1}.

Summary Upland Calluna-dominated wet heath (S411)

Since the last update (Bobbink and Hettelingh, 2011), the number of new addition experiments with N loads within the range of the current $CL_{emp}N$ is limited. In general, the available experimental data are based on total N inputs (background plus treatments) that are significantly above the previously established $CL_{emp}N$ range of 10 to 20 kg N ha^{-1} yr^{-1}. In contrast, a growing number of gradient studies have incorporated regions with low N deposition (< 10 kg N ha^{-1} yr^{-1}); they provide evidence for N deposition impacts on plant species composition at or below the lower end of the current $CL_{emp}N$ range. Therefore, we propose to revise the $CL_{emp}N$ range, setting it at 5 to 15 kg N ha^{-1} yr^{-1} for upland *Calluna*-dominated wet heath (S411), classifying it as 'reliable'. This $CL_{emp}N$ range is dependent on management practices, with the high end of the range applying to wet *Calluna*-dominated heath with high intensity management, and the low end of the range to wet *Calluna*-dominated heath with low intensity management.

8.4.2 Lowland *Erica tetralix*-dominated wet heath (S411)

The wet habitats in western European lowland heathlands are dominated by the dwarf-shrub *Erica tetralix* (Ellenberg, 1988) and classified within EUNIS as northern wet heath (S411). A drastic change in species composition has been observed in Dutch wet heathlands, from *Erica tetralix* dominated dwarf shrub vegetation to monospecific stands of the grass *Molinia caerulea*. Alongside *Erica tetralix,* almost all of the rare plant species have disappeared from this habitat type. It has been hypothesised that this change was caused by eutrophication induced by elevated atmospheric N deposition. This favours the highly competitive grass *Molinia caerulea*, which is able to outcompete *Erica tetralix* and other key species from this habitat (Berendse and Aerts, 1984; Aerts and Berendse, 1988).

Using the competitive relationship between *Molinia caerulea* and *Erica tetralix*, the biomass production of these species and taking into account management measures, Berendse (1988) suggested that 17 to 22 kg N ha^{-1} yr^{-1} is the $CL_{emp}N$ range for the transition of lowland wet heath towards a grass-dominated sward. This $CL_{emp}N$ range was also the value recommended by Bobbink et al. (1996). However, because of the lack of natural variation in the modelling approach, the range of 17 to 22 kg N ha^{-1} yr^{-1} was considered too narrow. The model by Berendse (1990) was based on the intensive management of sod (turf) with cutting every 50 years, in combination with grazing. Sod cutting is a common restoration measure in the Netherlands that effectively removes the N-rich and acidified top layer of the soil in order to decrease eutrophication and eliminate thick *Molinia* tussocks. It is a very intensive measure, setting back succession of the vegetation. In the UK, Allchin et al. (2001) applied a similar model to dry heaths with less intensive management regimes, such as mowing every 15 years, which gave a threshold for changes in species composition of about 10 kg N ha^{-1} yr^{-1}. Similar effects of management regimes were expected for wet heaths and hence the lower end of the $CL_{emp}N$ range was reduced to account for the effects of N under less intensive management.

There is no clear evidence of a differential response in *Calluna vulgaris* and *Erica tetralix* to simulated N depositions; Smart et al. (2004) found similar spatial relationships between changes in cover and N deposition in the UK for the two species. Therefore, Bobbink and Hettelingh (2011) recommended that the lower end of the $CL_{emp}N$ range should be 10 kg N ha^{-1} yr^{-1}, the same as that for *Calluna*-dominated wet and dry heaths. The upper end of the $CL_{emp}N$ was also reduced, to make it equivalent with that for other heathland (S411 (upland) and S42) habitats. Bobbink and Hettelingh (2011) proposed a revised $CL_{emp}N$ of 10-20 kg N ha^{-1} yr^{-1}, based on expert judgment.

Gradient studies

Since Bobbink and Hettelingh (2011), gradient studies have provided new evidence with respect to the $CL_{emp}N$ for *Erica tetralix*-dominated wet heaths. A number of gradient studies from the UK (e.g. Maskell et al., 2010; Henrys et al., 2011; Van den Berg et al., 2016) have repeatedly shown reduced species richness and changed species composition in heathlands associated with higher N deposition. Tipping et al. (2013) used a broken stick median regression to estimate the threshold above which N deposition had an effect on plant species richness data from the 1998 Countryside Survey in the UK. The analysis included 457 heathlands sites (classified as S411 and S42) spanning a deposition gradient of 4.9 to 40 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution). They estimated a threshold of 8.8 kg N ha^{-1} yr^{-1} (4.7-10.1 kg N ha^{-1} yr^{-1}) with an average relative loss of species with increasing N deposition of 2.3% per kg N ha^{-1} yr^{-1}.

In Denmark, Damgaard et al. (2014) applied a structural equation modelling (SEM) approach to 89 wet heathland sites (spanning a deposition range of 8 to 22 kg N; NOVANA model, total N deposition) to understand the underlying causes for the decrease in cover of *E. tetralix*. The most important causal effect revealed by the SEM was a significant negative effect of N deposition on the cover of *E. tetralix*. Strandberg et al. (2012) analysed *E. tetralix* cover data from 500 wet heathland plots (S411) in the Danish National Monitoring Programme. They found a significant negative relation between the cover of *E. tetralix* and N deposition, indicating a threshold at a deposition between 8.3 and 13.2 kg N ha^{-1} yr^{-1}. Wilkins et al. (2016), with updates in Aherne et al. (2021), identified a vegetation community change point (threshold) for northern Atlantic wet heaths in Ireland using species abundance data from 231 relevés spanning a deposition gradient of ~3 to 24 kg N ha^{-1} yr^{-1} (observation-based total N deposition at 5 × 5 km resolution). The community change point for declining species was estimated at 4.7 kg N ha^{-1} yr^{-1} with eleven species decreasing in abundance (four identified as positive indicator species).

Summary Lowland Erica-dominated wet heath (S411)

Gradient studies incorporating regions with low N deposition provide strong evidence of N deposition impacts on vegetation composition at or below the current critical load. Therefore, we propose to revise the $CL_{emp}N$ range, setting it at 5 to 15 kg N ha^{-1} yr^{-1} for lowland *Erica tetralix*-dominated wet heath (S411), classifying it as 'reliable'. This CLempN range is dependent on management practices, with the high end of the range applying to wet *Calluna*-dominated heath with high intensity management, and the low end of the range to wet *Calluna*-dominated heath with low intensity management.

8.4.3 Dry heaths (S42) (mostly sub-Atlantic *Calluna-Genista* heaths (S422))

Despite the conservation and management efforts in nature reserves, many lowland heaths (S42) in western Europe have become dominated by grass species. By using aerial photographs, it was demonstrated that more than 35% of Dutch heaths developed into grasslands during the 1980s (Van Kootwijk and Van der Voet, 1989). It was suggested that a strong increase in atmospheric N deposition contributed to this transition towards grassland. Similar patterns were found in the UK over the past 20 to 50 years. Pitcairn et al. (1991) showed declines in abundance of *Calluna vulgaris* at three heathland areas in East Anglia and an increase in grasses; the authors concluded that increased N deposition (up to 30 to 40 kg N ha^{-1} yr^{-1}) had at least been partly responsible for these changes, but also noted that the management had changed. A wider assessment of heathlands in eastern England showed that in some cases *Calluna vulgaris* had declined and subsequently been outcompeted by grasses, while other areas were still dominated by dwarf shrubs (Marrs, 1993). Although a move away from traditional management practices, such as grazing, burning or sod cutting, may be partly responsible, the decline in British heathlands has been linked with the historic increase in N deposition during the past 50 years. Furthermore, it has been hypothesised that, besides important changes in land use, increased N deposition is an additional cause of the decline in heaths in the southern parts of the Nordic countries (e.g. Fremstad, 1992; Tybirk et al., 1995).

Plant productivity and nutrient limitation

In N-limited systems, one of the first effects of increased N availability through atmospheric deposition is an increase in biomass production of the vegetation. Many studies found increased productivity of dwarf shrubs following experimental N enrichment in dry heathlands in several north-western European countries (e.g. Heil and Diemont, 1983; Van der Eerden et al., 1991; Aerts and Heil, 1993; Power et al., 1995; 1998a; Lee and Caporn, 2001). Gaining reliable estimates of critical loads is difficult in many European regions where background N deposition is already moderate to high (10 to 20 kg N ha^{-1} yr^{-1}). However, Bahring et al. (2017) studied lowland coastal heaths on the Baltic island of Fehmarn where the background deposition was 9 kg N ha^{-1} yr^{-1}. In this relatively clean environment, the addition of 5 kg N ha^{-1} yr^{-1} significantly increased *Calluna* shoot extension. This indicates that most of these lowland dry heath ecosystems are primarily limited by N, although some inland dry heaths are limited by P (Riis-Nielsen, 1997; Nielsen et al., 2000).

An illustrative example of the growth stimulation of *Calluna vulgaris* was found in a field experiment in the UK. The experiment was set up in 1989 to assess long-term impacts of realistic N loads on lowland dry heaths (S422) in southern Britain (Uren, 1992; Uren et al., 1997; Power et al., 1995; 1998a; 2001). After seven years of applications of ammonium sulphate (7.7

and 15.4 kg N ha^{-1} yr^{-1} with a background deposition of 8 kg N ha^{-1} yr^{-1}, Power and Barker (2003) found no negative effects on *Calluna vulgaris*. Indeed, a significant stimulation of flower production, shoot density, and litter production occurred and, after six to seven years, the canopy was 50% taller at 15.4 kg N ha^{-1} yr^{-1} than in the control plots (Power et al., 1995; 1998a). The increased shoot growth for the N-treated vegetation was not reflected by root growth, and an increased shoot:root ratio was inferred. Nitrogen concentrations in shoots also increased, with significant effects found in the months of July and/or October of several of the years that were assessed (Power et al., 1995; 1998a). In 1998, a parallel experiment was set up at the same site involving the addition of N (30 kg ha^{-1} yr^{-1} in two-weekly additions of ammonium sulphate) over a 12-year period. Similar to the earlier study, this experiment also demonstrated large and sustained increases in above-ground productivity, increased foliar N concentrations and an acceleration in the rate of N cycling and storage within the ecosystem (Barker, 2001; Jones, 2009). Similar growth stimulation of *Calluna vulgaris* was observed in a dry lowland heath in northern England. Since 1996, N has been applied in the form of ammonium nitrate (20, 60 and 120 kg N ha^{-1} yr^{-1}) in Cheshire; the atmospheric deposition at this site was estimated at 20 kg N ha^{-1} yr^{-1}. Within two years, shoot growth and flowering in *Calluna vulgaris* increased following the two highest N addition rates; after five years of N addition these effects were maintained in terms of canopy density, while the canopy height of *Calluna vulgaris* increased by ~20 centimetres (Cawley, 2001; Lee and Caporn, 2001). Collectively, these experiments demonstrated continued N limitation of this lowland heathland under prolonged N inputs.

Nitrogen accumulation and mineralisation

During secondary dry heath succession, an increase in the amount of organic material and N in the soil has been observed (Chapman et al., 1975; Gimingham et al., 1979). The accumulation of organic matter and N after sod removal was quantified in dry heaths in the Netherlands by Berendse (1990). A large increase was reported in plant biomass, soil organic matter and total N storage in the first 20 to 30 years of succession suggesting an annual increase in N in the system of ~33 kg N ha^{-1} yr^{-1}. These values are in good agreement with the measured N inputs in Dutch heathlands (Bobbink et al., 1992b).

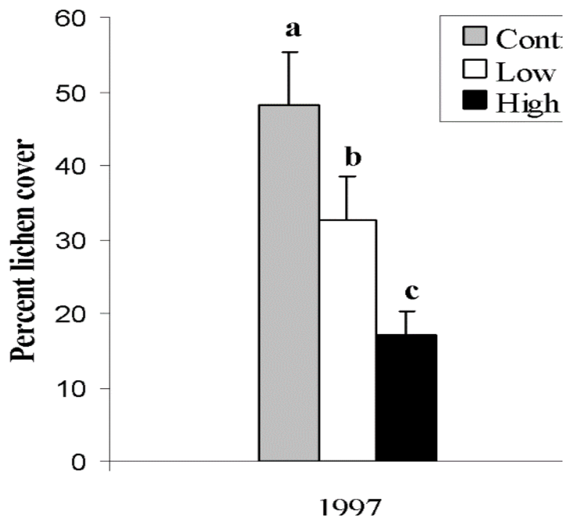
Owing to their high organic matter and microbial biomass content, ammonium (the dominant form of mineral N in these systems) immobilisation in the soil is high, and hardly any leaching losses to deeper layers have been measured in Dutch, British or Danish dry non-coastal heaths even under high N inputs (De Boer, 1989; Van der Maas, 1990; Power et al., 1998a; Kristensen and McCarty, 1999; Kristensen, 2001; Nielsen et al., 2000), very similar to wet heaths S411. This means that almost no N is lost from these systems. Indeed, the N content of the soil (upper 10 cm) significantly increased from 35.0 g N m^{-2} to 45.5 g N m^{-2}, following seven years of 15.4 kg N ha^{-1} yr^{-1} addition applied to Thursley dry heath, while N leaching remained very low (Power et al., 1998a, 1998b; Barker, 2001). Significant relationships between N deposition and concentrations of extractable N and total soil N (in addition to foliar N) have also been found in an English lowland heath survey (Jones, 2010, Jones and Power, 2012), providing field-based evidence of an accumulation of N in lowland heathlands in response to N inputs. At the Budworth lowland heath site, Caporn et al. (2002) reported leaching only at additions of 120 kg N ha^{-1} yr^{-1}, and constituting less than 10% of the added N. At the same experimental plots but following several years more N additions, Field et al. (2013) found leaching even at the lowest 20 kg N addition and observed that dissolved N concentrations in the soil solution were much higher than in the upland moorland experiment at Ruabon, north Wales. They suggested that the much lower N concentrations in the latter were due to immobilisation in the *Calluna* litter layer and more organic surface soils in the moorland. Only after severe damage to the *Calluna vulgaris* canopy, caused by heather beetles, was significant N leaching to the subsoil observed at lower rates of deposition (Van der Maas, 1990; Nielsen et al., 2000).

The accumulation of N in the soil and litter layers affects decomposition and soil N mineralisation. Power et al. (1998a) found that adding 15.4 kg N ha^{-1} yr^{-1} resulted in faster rates of cotton strip degradation, a clear indication of stimulated decomposer activity. Furthermore, the estimated time for incorporation of litter into the soil humus pool, based on measurements of annual litter production and the size of the litter pool, decreased from 8.6 years in control areas to 6.3 years in plots with low additions of N (7.7 kg N ha^{-1} yr^{-1}), and to 6.1 years in plots with high additions of N (15.4 kg N ha^{-1} yr^{-1}). The increased mineralisation will lead to enhanced availability of N. This will alleviate the N limitation on plant production and may lead to a shift to more nitrophilous species.

Changes in species composition

Competition experiments in containers and in the field have clearly demonstrated an important effect of increased N availability on the competitive interactions between *Calluna vulgaris* and grasses in the early phase of secondary succession in dry lowland heath. In the Netherlands, following experimental N additions (7 and 28 kg N ha^{-1} yr^{-1}) over four years during the 1980s (background deposition of approximately 25-35 N ha^{-1} yr^{-1}), grasses (*Festuca ovina*) strongly outcompeted *Calluna vulgaris*, under low initial vegetation cover (e.g. Heil and Diemont, 1983). However, under dense vegetation *Calluna vulgaris* clearly is a stronger competitor than grass species in mature heath vegetation, even at high N loads (Aerts et al., 1990; Aerts, 1993).

Figure 8.6. Lichen cover (%) following seven years of N additions applied to dry heath (S42) at plots in Thursley, Surrey (UK) with low (7.7 kg N ha^{-1} yr^{-1}) and high (15.4 kg N ha^{-1} yr^{-1}) additions of N.



Source: Barker, 2001

Understory species, especially the typical and frequently occurring lichen and moss species, can be negatively affected by increased growth of vascular species. On the Baltic island of Fehmarn, Bahring et al. (2017) found that the increased N accumulation in the system and falling C:N ratios were linked to an increase in graminoid cover at additions of 10 kg N ha^{-1} yr^{-1} and subsequent decline in bryophytes and *Cladonia* lichens at 50 kg N ha^{-1} yr^{-1}. After seven years of N additions in Surrey (UK), the cover of lichens and lichen diversity (*Cladonia* species; *Parmelia*) significantly decreased, following additions of 7.7 and 15.4 kg N ha^{-1} yr^{-1} (background deposition 8 kg N ha^{-1} yr^{-1}) (Barker, 2001; Figure 8.6). Because of weekly applications of relatively low concentrations of additional N, this decline was almost certainly not caused by the direct toxic effects of N, but probably by increased shading through the greater canopy density of *Calluna vulgaris*. The same was found for moss understorey in the N experiment in Cheshire (UK). The cover of *Hypnum* species, a nitrophilous moss, was also considerably lower following four or five years of N additions (60 and 120 kg N ha^{-1} yr^{-1}) (Lee and Caporn, 2001).

Opening of the *Calluna vulgaris* canopy and heather beetle infestations

When the productivity of *Calluna vulgaris* is no longer primarily limited by N, the growth response is less or absent, and luxury consumption will lead to increased N concentrations in the plant. In an experiment on a P-limited heathland in Denmark (background deposition of 18 kg N ha^{-1} yr^{-1}), N concentrations in shoots increased significantly following N additions of 15, 35 and 70 kg N ha^{-1} yr^{-1} in the form of ammonium nitrate (Johansson, 2000), while there was no significant growth response in this plant species (Riis-Nielsen, 1997). However, a relationship has been established between increased foliar N concentrations and pathogenic infestations as well as sensitivity to secondary stresses such as frost and drought.

Infestations of heather beetles (*Lochmaea suturalis)*, a chrysomelid beetle, occur frequently in dry lowland heaths. These beetles forage exclusively on the green parts of *Calluna vulgaris.* Insect herbivory is generally affected by the nutritive value of the plant material, with N concentration being especially important (e.g. Crawley, 1983; Brunsting and Heil, 1985; Van der Eerden et al., 1990, 1991). Increased *Calluna* tissue N concentrations have been found to be associated with faster development of heather beetle larvae and increased adult weights (Van der Eerden et al., 1990, 1991; Power et al., 1998b). Infestations of these beetles may lead to opening of the closed *Calluna vulgaris* canopy over large areas, greatly reducing light interception (Berdowski, 1987; 1993), thus leading to enhanced growth of understorey grasses, such as *Deschampsia flexuosa (Avenella flexuosa)* or *Molinia caerulea*. It is thus likely that the frequency and intensity of insect infestations are stimulated by increased atmospheric N loads. This is supported by the observations by Blankwaardt (1977), who reported that, from 1915 onwards, heather beetle infestations occurred at ca 20-year intervals, in the Netherlands, whereas in the last 15 years of the observation period, this interval decreased to less than eight years. In addition, it has been observed that *Calluna vulgaris* plants are more severely damaged in N-fertilised vegetation during a heather beetle infestation, in the Netherlands (Heil and Diemont, 1983), in Denmark (Tybirk et al., 1995; Riis-Nielsen, 1997) and in the UK (Lee and Caporn, 2001).

Opening of the *Calluna vulgaris* canopy: secondary stresses

Similar to upland *Calluna*-dominated wet heath (S411), impacts of N deposition on the frost sensitivity of *Calluna vulgaris* have been suggested as the cause of observed die-back of *Calluna vulgaris* shoots in successive severe winters of the mid-1980s in dry heathlands in the Netherlands (Van der Eerden et al., 1990). Nitrogen addition experiments with ammonia (fumigation) or ammonium sulphate suggest that N addition may increase frost sensitivity in *Calluna vulgaris* during late winter (Van der Eerden et al., 1991; Uren, 1992). Van der Eerden et al. (1991) studied the frost sensitivity of *Calluna vulgaris* vegetation that was artificially sprayed with different levels of ammonium sulphate (3-91 kg N ha^{-1} yr^{-1}). After five months the frost sensitivity of *Calluna vulgaris* had already increased significantly in vegetation treated with the highest level of ammonium sulphate (400 µmol l^{-1}; 91 kg N ha^{-1} yr^{-1}), compared to the control vegetation. However, another study that measured the effects of low levels of N addition (7.7 and 15.4 kg N ha^{-1} yr^{-1}) on frost sensitivity of *Calluna vulgaris* showed only limited effects after seven years of treatment (Power et al., 1998b). Hence, the significance of increased frost sensitivity at ambient N loads is very uncertain.

In addition to effects during winter, summer 'browning' of *Calluna vulgaris* canopies in the Netherlands was frequently seen in dry summers of the 1980s— the decade with the highest N loads. It was suggested that N enrichment increased the sensitivity of *Calluna vulgaris* to periods of drought due to reduced root growth and greater shoot:root ratio or as a result of a decrease in mycorrhizal infection. Indeed, the root weight ratio (RWR) of *Calluna* significantly decreased with increasing N additions (Aerts et al., 1991) and a small reduction in root to shoot ratio was found after seven years of N additions (15.4 kg N ha^{-1} yr^{-1}) in a dry lowland heathland in the UK (Power et al., 1998a). However, experimentally imposed drought on roofed split plots in an N-enriched lowland heath in the UK showed that increased sensitivity in *Calluna* and grass encroachment may be the result of a number of interacting effects such as root:shoot ratio and heather beetle infestation (Cawley et al., 1998; Lee and Caporn, 2001; Green, 2005). In addition, ericoid mycorrhizal infection of heather roots could also be influenced by increased N load but studies on this subject report highly variable results (Caporn et al., 1995; Yesmin et al., 1996; Aerts and Bobbink, 1999; Johansson, 2000). It is obvious that the sensitivity of *Calluna vulgaris* to drought stress might be increased by a shift in root:shoot ratio, and that grasses might profit from this damage to the heather canopy, but the precise importance of this process has yet to be clarified.

Gradient studies

Edmondson et al. (2010) assessed potential bio-indicators of N deposition (e.g. litter phenol oxidase activity, bryophyte species richness, N:P ratios, litter extractable N) in dry heaths (S42); they investigated 18 managed upland heather moorland sites throughout the UK spanning an N deposition gradient from ~7 to 31 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution). They found that litter phenol oxidase activity had a significant negative association with N deposition, notable at the lower end of the deposition range, ~7 to 11 kg N ha^{-1} yr^{-1}. Field et al. (2014) assessed species richness and plant community composition along gradients of climate and pollution in upland heaths (n = 25) with N deposition from 7.4 to 32.4 kg N ha^{-1} yr^{-1} and lowland heaths (n = 27) with N deposition from 5.9 to 29.4 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution). They found reduced species richness and changed species composition (more grasses) associated with higher N deposition. Species richness declined by about 40% of maximum species richness from the lowest to the highest N deposition sites in both types of heathlands, with a steeper decline in species richness in the lower deposition range < ~11 kg N ha^{-1} yr^{-1}. Maskell et al. (2010) observed a significant reduction in species richness with N deposition (5 to 50 kg N ha^{-1} yr^{-1}) in 459 heathland sites from the 1998 UK Countryside Survey even after fitting covarying factors. Similarly, Van den Berg et al. (2016) assessed species richness of vascular plants as a measure of biodiversity in upland (n = 267) and lowland (n = 182) dry heaths from the 2007 UK Countryside Survey; total N deposition ranged from 5.1 to 54.2 kg N ha^{-1} yr^{-1} (CBED model, total N deposition at 5 × 5 km resolution). Their results provide clear evidence that N deposition affects species richness, after factoring out correlated explanatory variables such as climate and sulphur deposition. The strongest negative coefficient was found for dry lowland heath followed by dry upland. Henrys et al. (2011) examined the response of individual vascular plant species to N in heathlands using the Vascular Plant Database and Botanical Society of the British Isles Local Change survey data. In lowland heaths, *Viola canina* had a negative relationship with N deposition declining considerably in its probability of presence between 10 and 25 kg N ha^{-1} yr^{-1}. In upland heaths, *Arctostaphylos uva-ursi* had a negative relationship with N deposition declining in probability of presence between 5 and 15 kg N ha^{-1} yr^{-1}, and *Vaccinium vitis-idaea* showed a clear decline in response to N deposition from 5 to 25 kg N ha^{-1} yr^{-1}.

Stevens et al. (2012) analysed the probability of presence of individual lichen taxa (n = 26) in heathlands at a given level of N deposition together with driver data for climate, change in sulphur deposition, and land-use using generalised additive models. Nine species showed a significant relationship with N deposition, with the majority starting from the lowest levels of deposition (< 5 kg N ha^{-1} yr^{-1}). Southon et al. (2013) assessed plant species richness in 52 heathlands (25 upland and 27 lowland) across an N deposition gradient of 5.9 to 32.4 kg N ha^{-1} yr^{-1}. Plant species richness declined with increasing temperature and N deposition, and the abundance of nitrophilous species increased with increasing N. The number of forb and graminoid species decreased with increasing total N deposition. The relationships were broadly similar between upland and lowland sites, with the biggest reductions in species number associated with increasing N inputs at the low end of the deposition range, with an average of 13 species lost per site between 5-10 kg N ha^{-1} yr^{-1}. Tipping et al. (2013) estimated the thresholds above which N deposition definitely had an effect on plant species richness in (dry and wet) heathlands (n = 457) from the 1998 UK Countryside Survey under an N deposition gradient of 4.9 to 40 kg N ha^{-1} yr^{-1}. The threshold N deposition was estimated to be 8.8 kg N ha^{-1} yr^{-1} (4.7 to 10.1 kg N ha^{-1} yr^{-1}) with an average relative loss of species of 2.3% per kg N ha^{-1} yr^{-1}.

Outside of the UK, Wilkins et al. (2016), with updates in Aherne et al. (2021), identified a vegetation community change point (threshold) for European dry heaths in Ireland using species abundance data from 161 relevés spanning a deposition gradient of ~3 to 24 kg N ha^{-1} yr^{-1} (observation-based total N deposition at 5 × 5 km resolution). The community change point for declining species was estimated at 5.6 kg N ha^{-1} yr^{-1} with 17 species decreasing in abundance.

8.4.4 Dry heaths (S42) (Submontane *Vaccinium* – *Calluna* heaths (S421))

The effects of fertilisation and experimental cutting were intensively studied in three heathlands in Spain where the dominant species were *Calluna vulgaris, Vaccinium myrtillus* and *Erica tetralix* (Marcos et al., 2003; Calvo et al., 2005, 2007; Cuesta et al., 2008). Additions of 56 kg N ha^{-1} yr^{-1} did not significantly alter soil characteristics. At these sites, background N deposition ranged between 7.5 and 15 kg N ha^{-1} yr^{-1}, according to the EMEP and CHIMERE models for Spain (García-Gómez et al., 2014). Nitrogen addition led to increased plant N concentration in *Calluna* and, to a lesser extent, in *Erica*. Nitrogen addition favoured perennial herbaceous graminoid species (e.g. *Nardus stricta, Festuca rubra* and *Deschampsia flexuosa*). Calvo et al. (2005) concluded that, in the short term, increased nutrients at twice the estimated current atmospheric deposition for the area, would not significantly alter the composition of the mountain heathlands. However, once stands matured, the capacity of the community to regenerate after a severe disturbance would diminish. A drastic impact such as cutting may not result in re-growth of the same shrub species, but in replacement by herbaceous species, which would also benefit from the increased nutrients. In the *Calluna* and *Erica* heathlands studied, cutting plus fertilisation led to an increase in plant diversity over time. Cutting patches of heathland was recommended as a mechanism for maintaining high vegetation diversity, when grazing is not possible (Calvo et al., 2007). Recent research at this site highlighted the sensitivity of these montane heathlands to N deposition. Calvo-Fernandez et al. (2018) reported on the ten-year impact of 56 kg N ha^{-1} yr^{-1} and also a three-year experiment using lower N treatments of 0, 10, 20 and 50 kg N ha^{-1} yr^{-1}. While several biogeochemical measures were not significantly affected by the high ten-year treatment, a number of variables were significantly changed by addition of only 10 kg N ha^{-1} yr^{-1} including soil available NH_4^+ and *Calluna* tissue N and P concentration. Taboada et al. (2018) reported results of plant and community changes from the same experiments at the Cantabrian mountain location. The most sensitive responses were increased shoot extension in *Calluna* and number of flowers, similar to findings from other European heathland experiments. Community species changes were only significant following the highest treatment (56 kg N ha^{-1} yr^{-1}) over nine years, and these were linked to reductions in bryophyte and lichen cover.

Summary Dry heaths (S42)

The impacts of increased N inputs to dry inland heaths (S42) are complex and occur at different time scales. Firstly, increased N availability stimulates biomass and litter production of *Calluna vulgaris* in most situations. This N is strongly retained in the system, gradually leading to higher N mineralisation rates in the soil. However, *Calluna* can be a strong competitor with respect to grasses, even at very high N availability if its canopy is closed. A shift from dwarf shrub towards grass dominance is clearly triggered by opening of the canopy caused by heather beetle attacks, winter injury or drought. If *Calluna* canopy cover is reduced, grasses quickly profit from the increased light availability, together with the high N availability. Within a few years, this may lead to a drastic increase in grass cover. Because of the random nature of several processes (e.g. heather beetle infestations, winter injury, drought) and the many long-term processes that interact with them, it is very difficult to model and clarify all these stochastic relationships without results from long-term (10-20 years) and large-scale experiments.

The $CL_{emp}N$ range for dry inland heaths was previously set at 10 to 20 kg N ha^{-1} yr^{-1}(Bobbink and Hettelingh, 2011). This range, based primarily on a long-term field experiment in the UK, was also supported by the results from simulation modelling using low intensity management regimes. Since the last update, no experimental data have become available that would warrant an adjustment of this. However, gradient studies encompassing low N deposition areas provide evidence for impacts at the lower end of the current $CL_{emp}N$ range. Given the strong evidence of N deposition impacts on vegetation composition at or below the current critical load, we propose to revise the $CL_{emp}N$ range, setting it at 5 to 15 kg N ha^{-1} yr^{-1} for dry heaths (S42), classifying it as 'reliable'. It should be stated that most N addition studies and gradient studies have been conducted on a subcategory of dry heaths and sub-Atlantic *Calluna-Genista* heaths (S422), but it seems reasonable for this $CL_{emp}N$ to be applied to all habitats in the S42 category. The intensity of management of *Calluna* heathlands may affect the impact of increased N deposition (Power et al., 2001). The high end of the CLempN range applies to dry *Calluna*-dominated heath with high intensity management, and the low end of the range to dry *Calluna*-dominated heath with low intensity management. The relative importance of P availability in some dry heath areas and habitat management as modifiers of dry heath response to increased N deposition needs further investigation.

8.5 Maquis, arborescent matorral and thermo-Mediterranean scrub (S5)

Maquis, arborescent matorral and thermo-Mediterranean scrub (S5; in short Mediterranean scrub) are important habitats in terms of diversity and cover in Mediterranean areas of Europe. Maquis is dominated by deep-rooting small evergreen shrubs and occasionally dense oak vegetation. This class generally consists of complex mosaics of herbaceous, shrub and even arborescent strata with a high diversity that hinders a detailed categorisation in habitat classification systems when using floristic composition.

The first N manipulation field study on these habitats was started in 2007, in southern Portugal, investigating the effects of N doses (40 and 80 kg N ha^{-1} yr^{-1}) and forms (ammonium as a 1:1 NH_4Cl to $(NH_4)_2SO_4$ mixture or ammonium nitrate (NH_4NO_3)) on maquis vegetation at the Natura 2000 Arrábida/Espichel site. Nitrogen additions were applied in three equal doses throughout the year (spring, summer and between autumn and winter) on top of a low background deposition of <4 kg N ha^{-1} yr^{-1} (Dias et al., 2014b). This study was performed in an area that was burnt four years prior to the start of the experiment and therefore showed the early stages of post-fire succession.

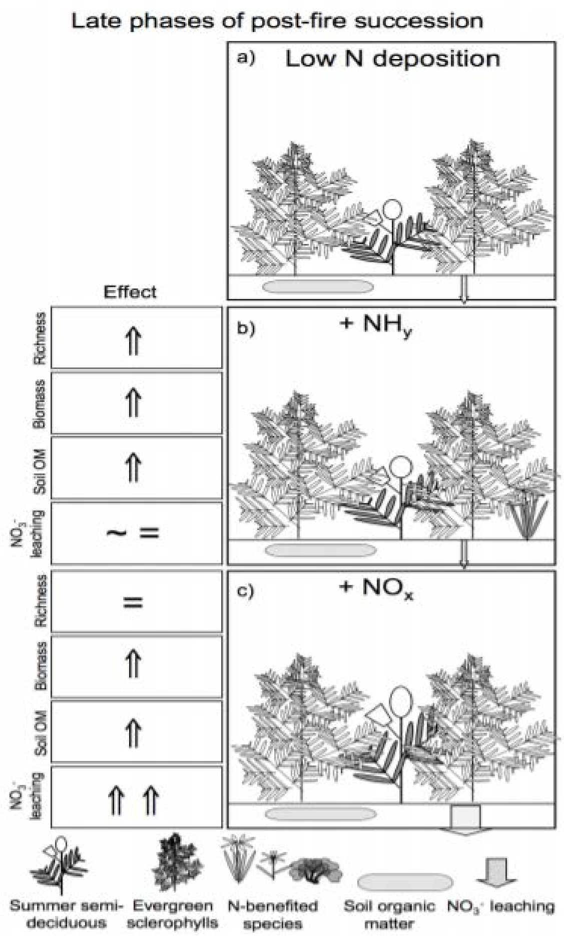
In contrast to most N addition studies (see Bobbink et al., 2010), the results suggested that one year of N enrichment had already caused shifts in the soil bacterial community structure, arbuscular mycorrhizal fungal community and plant composition (Dias et al., 2014a) Plant species richness increased within one year, with ruderal and herbaceous Maquis species particularly benefitting from the N addition.

Perennial shrubs such as evergreen *Cistus ladanifer* showed a negative effect of N addition; after seven years *C. ladanifer* had declined in cover with N applications of 40 kg N $ha^{-1}yr^{-1}$ (Dias et al., 2017). In addition, lower mineral weathering and lower N fixation were observed with applications of 40 kg N due to impacts on ectomycorrhizal fungi and N-fixing bacteria. The form in which N was applied to these ecosystems strongly affected the responses of the shrubs. *C. ladanifer* showed low tolerance to NH_4^+ application, which led to a lower photosynthetic N use efficiency for this species and reduced cover (Dias et al., 2014b, 2017). As a result, N addition at 40 kg N ha^{-1} yr^{-1} resulted in decreased soil protection (increased patches of bare soil), which may lead to increased erosion. In these naturally N limited systems, added N was largely retained via recycling of N in the biomass (Dias et al., 2012). Based on these findings and considering the importance of ammonium as a driving force in Mediterranean ecosystems, Dias et al. (2017) suggested a critical threshold at 20-40 kg ammonium-N ha^{-1} yr^{-1}.

In addition to the Arrábida/Espichel N-application experiment, a gradient study in central, southern and eastern semiarid Mediterranean Spanish shrublands, showed that increased N deposition above 4.4 kg N ha^{-1} yr^{-1} resulted in soil acidification with potential to affect the N-cycle (Ochoa-Hueso et al., 2014a). Additionally, studies of a Mediterranean habitat in the USA (Chaparral), very similar to the EUNIS category S5, provide additional information on the sensitivity of this habitat type to N deposition (Fenn et al., 2010). Chaparral showed elevated nitrate leaching to stream water with increased N deposition (above 10 kg N $ha^{-1}yr^{-1}$), which contrasts with the N retention described by Dias et al. (2012). However, the soils at the Portuguese experimental site were low in nutrients compared to the Californian soils, potentially muting the effects of N through P co-limitation. Moreover, the Portuguese experimental site was in an early successional state, while the longer N-deposition history in Chaparral systems in the USA may already have altered their structure and composition in the past. It has been suggested that in later phases of post-fire succession in European Mediterranean scrub, increased N deposition would be expected to cause increased nitrate leaching, particularly under elevated nitrate deposition (Figure 8.7; Dias et al*.,* 2012).

Changes in the epiphytic lichen community towards dominance by eutrophic lichen species has also been linked to elevated N deposition (Fenn et al., 2010). Based on these responses, a $CL_{emp}N$ range for Chaparral was set between 5.5 and 10 kg N ha^{-1} yr^{-1}. Chaparral habitats and coastal sage scrubs (see Dias et al*.,* 2012) were estimated to have lower critical loads than the Mediterranean scrubs (Pardo et al., 2011, 2015). This difference between American and European habitats could be attributed to several factors, including methodological reasons (e.g. lack of N deposition gradient studies and long-term addition experiments) but also differences between land use, N-deposition history, soil fertility and plant communities.

Figure 8.7. Effects of increased N deposition in the form of ammonium or nitrate (respectively b and c) on Mediterranean Maquis.



Source: Dias et al., 2012

Summary Maquis, arborescent matorral and thermo-Mediterranean scrub (S5)

The habitat Maquis, arborescent matorral and thermo-Mediterranean scrub is spatially and ecologically related to other Mediterranean habitat like Mediterranean evergreen (*Quercus*) woodland, Mediterranean xeric grasslands and Garrigue. The $CL_{emp}N$ for Mediterranean evergreen (*Quercus*) woodland is 10-15 kg N ha^{-1} yr^{-1} (see Chapter 9), Mediterranean xeric grasslands 5-10 kg N ha^{-1} yr^{-1} (see Chapter 7) and Garrigue 5-15 kg N ha^{-1} yr^{-1} (this chapter). It is expected that the Maquis habitat responds in a similar way to nitrogen. In addition, the $CL_{emp}N$ for the related Mediterranean scrubs in USA (Chaparral 3.1-14 kg N ha^{-1} yr^{-1}; Coastal sage scrub 7.8-10 kg N ha^{-1} yr^{-1}) correspond with these ranges. Therefore, we propose a $CL_{emp}N$ range for Maquis, arborescent matorral and thermo-Mediterranean scrub (S5) of 5 to 15 kg N ha^{-1} yr^{-1}, classifying it as 'expert judgement'. This value should be applied with caution in mature habitats and in habitats that are not on calcareous soils.

8.6 Garrigue (S6)

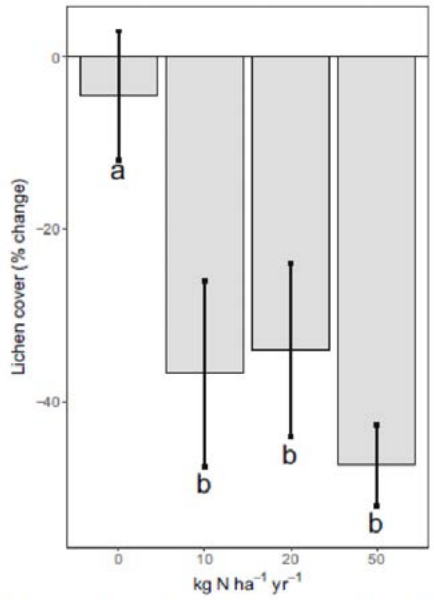
Garrigue is a Mediterranean shrub habitat that is generally considered to be a drier and more open habitat than Maquis. Garrigue consists of sclerophyllous shrubs on shallow ground with generally > 10% bare soil or biocrust.

A long-term N addition study in Spain run by R. Ochoa-Hueso and colleagues has improved our knowledge and understanding of Mediterranean shrub vegetation responses to N deposition. In the Nature Reserve “El Regajal-Mar de Ontígola”, N was applied as NH_4NO_3 at levels of 0, 10, 20, 50 over background (~6 kg N ha^{-1} yr^{-1}) on a mosaic of open and dense shrubland habitat dominated by the sclerophyllous scrub Kermes oak (*Quercus coccifera*) and *Rosmarinus officinalis* with bare soil and a diverse and well-developed biological soil crust with short-lived therophytes. The denser parts of this study site could be designated as Mediterranean maquis (EUNIS classification S5) but because of its structure and location, the plant species description and the open, low canopy structure in the fertilisation experimental area, we suggest that this habitat is classified as S61: Basiphilous Garrigue. This semi-arid calcareous shrubland could be considered highly representative of other shrublands from the Mediterranean basin (Ochoa-Hueso et al., 2017), and here we consider that the $CL_{emp}N$ derived from this long-term experiment might be applied to other Mediterranean shrublands where canopy openness, dry conditions and basic soils are the dominant characteristics (i.e. Garrigue S6), regardless of their *a priori* habitat categorisation.

In the open areas of the study site, Ochoa-Hueso and Stevens (2015) showed that the characteristically dominant annual forbs responded negatively to N addition (0, 10, 20 and 50 kg N ha^{-1} yr^{-1}) after 2.5 years. Annual species such as *Limonium echioides* and small chamaephytes such as *Helianthemum violaceum* decreased in biomass and cover with increasing N application. In contrast, the nitrophilous forbs (mainly crucifers) increased with N after 2.5 years, but this response was conditioned by between-year variations in rainfall and the heterogeneous distribution of P availability, which limited growth of these species. These ecosystems proved to be highly sensitive, as responses to N were recorded at all N addition levels above background for biomass of *L. echioides*. Additionally, soil macrofauna abundance (*Collembola* and *Pauropoda*) changed due to the eutrophying effects, acidification and increased NH_4^+ concentrations in soils (Ochoa-Hueso et al., 2014a). In addition to acidification, potassium became less available after four years as a result of increased nitrate leaching (Ochoa-Hueso et al., 2013). Although the experiment was executed on calcareous soils, these slight but significant effects were evident in the experimental plots that received 20 or 50 kg N ha^{-1} yr^{-1}. It was found that N deposition resulted in changes in pigment ratios in the Mediterranean bryophyte *Pleurochaete squarrosa* and the lichen *Cladonia foliacea*, which may lead to changes in physiological priorities either for photosynthesis or protection against photooxidation (Ochoa-Hueso et al., 2014b). Responses in physiological parameters such as PME/NR ratios are widely used as bioindicators to identify effects of atmospheric N deposition (Arróniz-Crespo et al., 2008). In the Kermes oak vegetation, PME (phosphomonoesterase) enzyme activity increased and nitrate reductase (NR) activity respectively increased in both *P. squarrosa* and *C. foliacea* and decreased in *P. squarrosa* following N addition.

In a follow up study, Cabal et al. (2017) showed that N application reduced the cover and leaf lifespan (measured as increased defoliation) of shrubs (*Rosmarinus officinalis*) at additions of 10 kg N ha^{-1} yr^{-1} and higher. Shrubs in these semiarid ecosystems provide shelter, soil nutrient input (via litter accumulation) and shading, affecting soil moisture, colonisation potential of other plant species and protection against erosion. Nitrogen deposition also gradually reduced the cover of the lichen *Cladonia foliacea*, an important component of biocrusts (Ochoa-Hueso et al., 2017; Benvenutto-Vargas and Ochoa-Hueso, 2020), with significant effects observed at the lowest level of N addition (Figure 8.8). In contrast, moss cover did not change in response to N. The plots treated with 50 kg N ha^{-1} yr^{-1} showed a 50% reduction in lichen cover compared to the control plots after ten years (from 41% to 20%).

Figure 8.8. Effects of N addition on percentage of change of lichen cover (mean ± SE) between the years 2008 and 2012. The different letters indicate the statistically significant differences (P<0.05).



Source: Ochoa-Hueso et al., 2017

Summary Garrigue (S6)

The effects of additional N deposition to these semiarid N-limited ecosystems are complex, and clear, direct effects of N manipulations were not found on a single ecosystem response variable. However, when multiple (and interacting) ecosystem responses were evaluated in European studies, it was clear that at 10 kg N ha^{-1} yr^{-1}, ecosystem disruption was visible after four years of application (Ochoa-Hueso, 2016). Based on these results, and the fact that they correspond well with similar habitats from the USA that show similar critical loads (Ochoa-Hueso et al., 2017), the CL$_{emp}$N for the Garrigue (S6) is set between 5 and 15 kg N ha^{-1} yr^{-1}, classifying it as 'quite reliable'.

8.7 Overall summary of CL$_{emp}$N for heathland, scrub and tundra habitats (S)

An overview of the CL$_{emp}$N for heathland, scrub and tundra habitats (S) is presented in Table 8.1.

Table 8.1. CL$_{emp}$N and effects of exceedances on heathland, scrub and tundra habitats (S). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Tundra	S1	3-5	#	3-5 [a]	#	Changes in biomass; physiological effects; changes in bryophyte species composition; decrease in lichen species richness
Arctic, alpine and subalpine scrub habitats	S2	5-15	#	5-**10** [a]	#	Decline in lichens; bryophytes and evergreen shrubs

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Lowland to montane temperate and submediterranean *Juniperus* scrub	S31			**5-15**	**(#)**	Shift in vegetation community composition; reduced seed viability
Northern wet heath	S411					
▶ ‘U’ *Calluna*-dominated wet heath (upland)	S411	10-20	#	**5-15** [b]	**##**	Decreased heather dominance; decline in lichens and mosses; increased N leaching
▶ ‘L’ *Erica tetralix*-dominated wet heath (lowland)	S411	10-20	#	**5-15** [b]	**##**	Transition from heather to grass dominance; decrease in heather cover; shift in vegetation community composition
Dry heaths	S42	10-20	##	**5-15** [b]	##	Transition from heather to grass dominance; decline in lichens; changes in plant biochemistry; increased sensitivity to abiotic stress
Maquis, arborescent matorral and thermo-Mediterranean scrub	S5	20-30	(#)	**5-15**	(#)	Change in plant species richness and community composition; nitrate leaching and acidification of soil.
Garrigue	S6			**5-15**	**#**	Changes in species composition; decline in shrub cover and increased invasion of annual herbs

a) use towards high end of range if phosphorus limited, and towards lower end if phosphorus is not limiting.

b) use towards high end of range with high intensity management, and use towards lower end of range with low intensity management.

8.8 Recommendations and knowledge gaps

Based on the number of studies that were published we can conclude that long-term experimental studies with low N dose additions at a low N deposition background are particularly needed for Mediterranean, alpine and arctic habitats. In addition, the inclusion of the differential effects of reduced and oxidised N is needed to further disentangle mechanisms of effect.

The arctic, alpine and subalpine scrub habitats category (S2) is particularly varied, encompassing a range of habitats variously dominated by evergreen and deciduous shrub species and with varying importance of bryophytes and lichens. The sensitivity of these habitats to N deposition may well vary and more low-dose experimental studies and well-designed gradient studies which include low deposition areas are needed to refine critical loads for these communities.

New experimental or gradient evidence is also needed for the major heath and scrub categories for which no $CL_{emp}N$ is currently defined. These include Spiny Mediterranean heaths (S7), Thermo-Atlantic xerophytic scrub (S8) and riverine and fen scrub habitats (S9).

As well as vascular plants, new studies should always include bryophytes and lichens where relevant to the habitat since these are often the most sensitive component of the vegetation and are also important to ecosystem functioning. New studies addressing biodiversity impacts of N deposition beyond vegetation, should also be a high priority. Soil biodiversity (fungi, bacteria, micro- and meso-fauna) and invertebrate biodiversity are particularly important for ecosystem functioning.

Climate change and nitrogen deposition are likely to have strong interactive effects on ecosystem functioning and climate change may alter ecosystem responses to nitrogen deposition and vice versa. More experimental studies are needed to examine these interactions and also more gradient studies which explicitly examine the impacts of nitrogen deposition in combination with climatic gradients.

At present there are only a few studies which have looked at both impact of and recovery from nitrogen deposition. More studies on this topic are needed to understand the reversibility of nitrogen deposition effects and the long-term prognosis for N impacted ecosystems.

8.9 References

Aerts, R. and Berendse, F. (1988). The effect of increased nutrient availability on vegetation dynamics in wet heathlands. *Vegetatio* **76**, 63-69.

Aerts, R., Berendse, F., De Caluwe, H. and Schmitz, M. (1990). Competition in heathland along an experimental gradient of nutrient availability. *Oikos* **57**, 310-318.

Aerts, R., Boot, R.G.A. and Van der Aart, P.J.M. (1991). The relation between above- and belowground biomass allocation patterns and competitive ability. *Oecologia* **87**, 551-559.

Aerts, R. (1993). Competition between dominant plant species in heathlands. In: Aerts, R. and Heil, G.W. (eds.). *Heathlands: patterns and processes in a changing environment*. Kluwer, Dordrecht, 125-151.

Aerts, R. and Heil, G.W. (eds.) (1993). *Heathlands: Patterns and processes in a changing environment*. Kluwer Academic Publishers, Dordrecht.

Aerts, R. and Bobbink, R. (1999). The impact of atmospheric nitrogen deposition on vegetation processes in terrestrial, non-forest ecosystems. In: Langan, S.J. (ed.). *The impact of nitrogen deposition on natural and semi-natural ecosystems.* Kluwer Academic Publishers, Dordrecht, 85-122.

Aerts, R. (2010). Nitrogen-dependent recovery of subarctic tundra vegetation after simulation of extreme winter warming damage to *Empetrum hermaphroditum*. *Global Change Biology* **16**, 1071-1081.

Aherne, J., Wilkins, K. and Cathcart, H. (2021). *Nitrogen–Sulfur Critical Loads: Assessment of the Impacts of Air Pollution on Habitats*. Environmental Protection Agency, Johnstown Castle, Ireland. URL: www.epa.ie/publications/research/air/Research_Report_390.pdf

Allchin, E.A., Power, S.A. and Ashmore, M.R. (2001). *Impacts of enhanced inputs of N to Calluna systems: integration, synthesis and modelling.* Final contract report to UK Department of Environment Transport and the Regions.

Alonso, I., Hartley, S.E. and Thurlow, M. (2001). Competition between heather and grasses on Scottish moorlands: interacting effects of nutrient enrichment and grazing regime. *Journal of Vegetation Science* **12**, 249-260.

APIS (2021). http://www.apis.ac.uk/

Arens, S.J.T., Sullivan, P.F. and Welker, J.M. (2008). Nonlinear responses to nitrogen and strong interactions with nitrogen and phosphorus additions drastically alter the structure and function of a high Arctic ecosystem. *Journal of Geophysical Research* **113**, 1-10

Arróniz-Crespo, M., J.R. Leake, P. Horton and G.K. Phoenix (2008). Bryophyte physiological responses to, and recovery from, long-term nitrogen deposition and phosphorus fertilisation in acidic grassland. *New Phytologist* **180**, 864-874.

Baddeley, J.A., Woodin, S.J. and Alexander, I.J. (1994). Effects of increased nitrogen and phosphorus availability on the photosynthesis and nutrient relations of three Arctic dwarf shrubs from Svalbard. *Functional Ecology* **8**, 676-685.

Bahring, A., Fichtner, A., Ibe, K., Schutze, G., Temperton, V.M., von Oheimb, G. and Hardtle, W. (2017). Ecosystem functions as indicators for heathland responses to nitrogen fertilisation. *Ecological Indicators* **72**, 185-193.

Barker, C.G. (2001). *The impact of management on heathland response to increased nitrogen deposition*. PhD Thesis, University of London.

Benvenutto-Vargas, V.P. and Ochoa-Hueso, R. (2020). Effects of nitrogen deposition on the spatial pattern of biocrusts and soil microbial activity in a semi-arid Mediterranean shrubland. *Functional Ecology* **34**, 923-937.

Berdowski, J.J.M. (1987). *The catastrophic death of* Calluna vulgaris *in Dutch heathlands.* PhD thesis, Utrecht University.

Berdowski, J.J.M. (1993). The effect of external stress and disturbance factors on *Calluna*-dominated heathland vegetation. In: Aerts, R. and Heil, G.W. (eds.). *Heathlands: patterns and processes in a changing environment*. Kluwer, Dordrecht, 85-124.

Berendse, F. and Aerts, R. (1984). Competition between *Erica tetralix* L. and *Molinia caerulea* (L.) Moench. as affected by the availability of nutrients. *Acta Oecologia/Oecologia Plantarum* **5**, 3-14.

Berendse, F. (1988). *De nutriëntenbalans van droge zandgrondvegetaties in verband met eutrofiëring via de lucht. I: Een simulatiemodel als hulpmiddel bij het beheer van vochtige heidevelden.* Report CABO, Wageningen (in Dutch).

Berendse, F. (1990). Organic matter accumulation and nitrogen mineralization during secondary succession in heathland ecosystems. *Journal of Ecology* **78**, 413-427.

Blankwaardt, H.F.H. (1977). Het optreden van de heidekever (*Lochmaea suturalis* Thomson) in Nederland sedert 1915. *Entomologische Berichten* **37**, 34-40.

Bobbink, R., Heil, G.W. and Raessen, M.B.A.G. (1992b). Atmospheric deposition and canopy exchange processes in heathland ecosystems. *Environmental Pollution* **75**, 29-37.

Bobbink, R., Hornung, M. and Roelofs, J.G.M. (1996). Empirical nitrogen critical loads for natural and semi-natural ecosystems. In: *Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded,* UN ECE Convention on long-range transboundary air pollution, Federal Environmental Agency, Berlin.

Bobbink, R., Ashmore, M., Braun, S., Flückiger, W. and Van den Wyngaert, I.J.J. (2003). Empirical nitrogen loads for natural and semi-natural ecosystems: 2002 update. In: *Empirical critical loads for nitrogen, Expert Workshop Convention on long-range transboundary air pollution UNECE)*, Swiss Agency for the Environment, Forests and Landscape SAEFL, Berne.

Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., Emmett, B., Erisman, J.-W., Fenn, M., Gilliam, F., Nordin, A., Pardo, L. and De Vries,

W. (2010). Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. *Ecological Applications* **20**, 30-59.

Bobbink, R. and Hettelingh, J. (2011). *Review and revision of empirical critical loads and dose-response relationships: Proceedings of an expert workshop*, Noordwijkerhout, 23-25 June 2010. Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM), RIVM report 680359002/2011 (244 pp).

Britton, A.J. and Fisher, J.M. (2007a). Interactive effects of nitrogen deposition, fire, grazing on diversity and composition of low-alpine prostrate *Calluna vulgaris* heathland. *Journal of Applied Ecology* **44**, 125-135.

Britton, A.J. and Fisher, J.M. (2007b). NP stoichiometry of low-alpine heathland: Usefulness for bio-monitoring and prediction of pollution impacts. *Biological Conservation* **138**, 100-108.

Britton, A.J. and Fisher, J.M. (2008). Growth responses of low-alpine dwarf-shrub heath species to nitrogen deposition and management. *Environmental Pollution* **153**, 564-573.

Britton, A.J., Helliwell, R.C., Fisher, J.M. and Gibbs, S. (2008). Interactive effect of nitrogen deposition and fire on plant and soil chemistry in an alpine heathland. *Environmental Pollution* **156**, 409-416.

Britton, A.J. and Fisher, J.M. (2010). Terricolous alpine lichens are sensitive to both load and concentration of applied nitrogen and have potential as bioindicators of nitrogen deposition. *Environmental Pollution* **158**, 1296-1302.

Britton, A.J., Gibbs, S., Fisher, J.M. and Helliwell, R.C. (2019). Impacts of nitrogen deposition on carbon and nitrogen cycling in alpine Racomitrium heath in the UK and prospects for recovery. *Environmental Pollution* **254**, 112986.

Brunsting, A.M.H. and Heil, G.W. (1985). The role of nutrients in the interactions between a herbivorous beetle and some competing plant species in heathlands. *Oikos* **44**, 23-26.

Cabal, C., Ochoa-Hueso, R., Pérez-Corona, M.E. and Manrique, E. (2017). Long-term simulated nitrogen deposition alters the plant cover dynamics of a Mediterranean rosemary shrubland in Central Spain through defoliation. *Environmental Science and Pollution Research* **24**, 26227-26237.

Calvo, L., Alonso, I., Fernàndez, A.J. and De Luis, E. (2005). Short-term study effects of fertilisation and cutting treatments on the vegetation dynamics of mountain heathlands in Spain. *Plant Ecology* **179**, 181-191.

Calvo, L., Alonso, I., Marcos, E. and De Luis, E. (2007). Effects of cutting and nitrogen deposition on biodiversity in Cantabrian heathlands. *Applied Vegetation Science* **10**, 43-52.

Calvo-Fernandez, J., Taboada, A., Fichtner, A., Hardtle, W., Calvo, L. and Marcos, E. (2018). Time- and age-related effects of experimentally simulated nitrogen deposition on the functioning of montane heathland ecosystems. *Science of the Total Environment* **613**, 149-159.

Caporn, S.J.M., Risager, M. and Lee, J.A. (1994). Effect of nitrogen supply on frost hardiness in *Calluna vulgaris* (L.) Hull. *New Phytologist* **128**, 461-468.

Caporn, S.J.M., Song, W., Read, D.J. and Lee, J.A. (1995). The effect of repeated nitrogen fertilization on mycorrhizal infection in heather (*Calluna vulgaris* (L.) Hull). *New Phytologist* **129**, 605-609.

Caporn, S., Wilson, D., Pilkington, M., Carroll, J., Cresswell, N. and Ray, N. (2002). Long term impacts of enhanced and reduced nitrogen deposition on semi-natural vegetation. In: *Progress Report of UK, DEFRA terrestrial umbrella-eutrophication and acidification of terrestrial ecosystems in the UK.*

Caporn, S.J., Carroll, J.A., Dise, N.B. and Payne, R.J. (2014). Impacts and indicators of nitrogen deposition in moorlands: Results from a national pollution gradient study. *Ecological Indicators* **45**, 227-234.

Carroll, J.A., Caporn, S.J.M., Cawley, L., Read, D.J. and Lee, J.A. (1999). The effect of increased deposition of atmospheric nitrogen on *Calluna vulgaris* in upland Britain. *New Phytologist* **141**, 423-431.

Cawley, L.E., Caporn, S.J.M., Carroll, J.A., Cresswell, N. and Stronach, I.M. (1998). Influence of elevated nitrogen on drought tolerance in lowland heath. *Book of abstracts* CAPER, Rothamsted.

Cawley, L.E. (2001). *Pollutant nitrogen and drought tolerance in heathland plants*. PhD thesis. The Manchester Metropolitan University. UK.

Chapman, S.B., Hibble, J. and Rafael, C.R. (1975). Net aerial production by *Calluna vulgaris* on lowland heath in Britain. *Journal of Ecology* **63**, 233-258.

Choudhary, S., Blaud, A., Osborn, A.M., Press, M.C. and Phoenix, G.K. (2016). Nitrogen accumulation and partitioning in a High Arctic tundra ecosystem from extreme atmospheric N deposition events. *Science of the Total Environment* **554**, 303-310.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**, 648-675.

Cornelissen, J.H.C., Callaghan, T.V., Alatalo, J.M., Michelsen, A., Graglia, E., Hartley, A.E., Hik, D.S., Hobbie, S.E., Press, M.C., Robinson, C.H., Henry, G.H.R., Shaver, G.R., Phoenix, G.K., Gwynn Jones, D., Jonasson, S., Chapin, F.S. III, Molau, U., Neill, C., Lee, J.A., Melillo, J.M., Sveinbjonsson, B. and Aerts, R. (2001). Global change and Arctic ecosystems: is lichen decline a function of increases in vascular plant biomass? *Journal of Ecology* **89**, 984-994.

Crawley, M.J. (1983). *Herbivory the dynamics of animal/plant interactions*. Blackwell, Oxford.

Cuesta, D., Taboada, A., Calvo, L. and Salgado, J.M. (2008). Short- and medium-term effects of experimental nitrogen fertilization on arthropods associated with *Calluna vulgaris* heatlands in north-west Spain. *Environmental Pollution* **152**, 394-402.

Curtis, C.J., Emmett, B.A., Grant, H., Kernan, M., Reynolds, B. and Shilland, E. (2005). Nitrogen saturation in UK moorlands: the critical role of bryophytes and lichens in determining retention of atmospheric N deposition. *Journal of Applied Ecology* **42**, 507-517.

Damgaard, C., Strandberg, M., Kristiansen, S.M., Nielsen, K.E. and Bak, J.L. (2014). Is *Erica tetralix* abundance on wet heathlands controlled by nitrogen deposition or soil acidification? *Environmental Pollution* **184**, 1-8.

Davies, C.E. and Moss, D. (2002). *EUNIS habitat classification, Final report.* CEH Monks Wood, UK.

Davies, C.E., Moss, D. and Hill, M.O. (2004). EUNIS habitat classification revised 2004. Winfrith Technology Centre, Dorian Ecological Information Ltd. and Monks Wood, UK.

De Boer, W. (1989). *Nitrification in Dutch heathland soils.* PhD thesis, Wageningen University.

De Smidt, J.T. (1979). Origin and destruction of Northwest European heath vegetation. In: Wimanns, O. and Töxen, R. (eds.). *Werden und Vergehen von Pflanzengesellschaften*. J. Cramer, Vaduz, 411-43.

Dias, M.T.M. (2012). *Effects of increased nitrogen availability on the structure and functioning of a Mediterránean Basin maquis*, PhD Thesis. Universidade de Lisboa (Portugal). 186 pages.

Dias, T., Martins-Loucao, M.A., Sheppard, L. and Cruz, C. (2012). The strength of the biotic compartment in retaining nitrogen additions prevents nitrogen losses from a Mediterranean maquis. *Biogeosciences* **9**, 193-201.

Dias, T., Chaves, S. Tenreiro, R., Martins-Loução, M.-A., Sheppard, L.J. and Cruz, C. (2014a). Effects of increased nitrogen availability in Mediterranean ecosystems: a case study in a Natura 2000 site in Portugal. In: Sutton, M.A., Mason, K.E. Sheppard, L.J., Sverdrup, H., Haeuber, R. and Hicks, W.K. (eds.) *Nitrogen Deposition, Critical Loads and Biodiversity*. Springer, 251-258.

Dias, T., Clemente, A., Martins-Loucao, M.A., Sheppard, L., Bobbink, R. and Cruz, C. (2014b). Ammonium as a Driving Force of Plant Diversity and Ecosystem Functioning: Observations Based on 5 Years' Manipulation of N Dose and Form in a Mediterranean Ecosystem. *Plos One* **9**, e92517.

Dias, T., Crous, C.J., Liberati, D., Munzi, S., Gouveia, C., Ulm, F., Afonso, A.C., Ochoa-Hueso, R., Manrique E. and Sheppard, L. (2017). Alleviating nitrogen limitation in Mediterranean maquis vegetation leads to ecological degradation. *Land Degradation & Development* **28**, 2482-2492.

Edmondson, J.L. (2007). *Nitrogen pollution and the ecology of heather moorland*. PhD Thesis, Manchester Metropolitan University.

Edmondson, J., Carroll, J., Price, E. and Caporn, S. (2010). Bio-indicators of nitrogen pollution in heather moorland. *Science of the Total Environment* **408**, 6202-6209.

Edmondson, J., Terribile, E., Carroll, J., Price, E. and Caporn, S. (2013). The legacy of nitrogen pollution in heather moorlands: ecosystem response to simulated decline in nitrogen deposition over seven years. *Science of the Total Environment* **444**, 138-144.

Ellenberg, H. (1988). *Vegetation Ecology of Central Europe*. Cambridge Univ. Press, Cambridge.

Fenn, M.E., Allen, E.B., Weiss, S.B., Jovan, S., Geiser, L.H., Tonnesen, G.S., Johnson, R.F., Rao, L.E., Gimeno, B.S., Yuan, F., Meixner T. and Bytnerowicz, A. (2010). Nitrogen critical loads and management alternatives for N-impacted ecosystems in California. *Journal of Environmental Management* **91**, 2404-2423.

Field, C., Sheppard, L., Caporn, S. and Dise, N. (2013). The ability of contrasting ericaceous ecosystems to buffer nitrogen leaching. *Mires & Peat* 11, Art. 5.

Field, C.D., Dise N.B., Payne R.J., Britton A.J., Emmett B.A., Helliwell R.C., Hughes S., Jones L., Lees S., Leake J.R., Leith I.D., Phoenix G.K., Power S.A., Sheppard L.J., Southon G.E., Stevens C.J. and Caporn S.J.M. (2014). The Role of Nitrogen Deposition in widespread Plant Community Change Across Semi-natural Habitats. *Ecosystems* **17**, 864-877.

Field, C.D., Evans C.D., Dise N.B., Hall J.R. and Caporn S.J.M. (2017). Long-term nitrogen deposition increases heathland carbon sequestration. *Science of the Total Environment* **592**, 426-435.

Fremstad, E. (1992). *Virkninger av nitrogen på heivegetasjon*. En litteraturstudie. NINA Oppdragsmelding 124. Trondheim, Norsk Institut for Naturforskning, 1-44.

Fremstad, E., Paal, J. and Möls, T. (2005). Impact of increased nitrogen supply on Norwegian lichen-rich alpine communities: a ten-year experiment. *Journal of Ecology* **93**, 471-481.

Gimingham, C.H., Chapman, S.B. and Webb, N.R. (1979). European heathlands. In: Specht, R.L. (ed.). *Ecosystems of the world, 9A*. Elsevier, Amsterdam, 365-386.

García-Gómez, H., Garrido, J.L., Vivanco, M.G., Lassaletta, L., Rábago, I., Àvila, A., Tsyro, S., Sánchez, G., González Ortiz, A., González-Fernández, I. and Alonso, R. (2014). Nitrogen deposition in Spain: modelled patterns and threatened habitats within the Natura 2000 network. *Science of the Total Environment* **485-486**, 450-460.

Gordon, C., Wynn, J.M. and Woodin, S.J. (2001). Impacts of increased nitrogen supply on high Arctic heath: the importance of bryophytes and phosphorus availability. *New Phytologist* **149**, 461-471.

Green, E.R. (2005). *N enrichment and lowland heathland*. PhD Thesis, Imperial College.

Gruwez, R., De Frenne P., De Schrijver A., Leroux O., Vangansbeke P. and Verheyen K. (2014). Negative effects of temperature and atmospheric depositions on the seed viability of common juniper (*Juniperus communis*). Annals of Botany **113**, 489-500.

Hartley, S.E., Gardner, S.M. and Mitchell, R.J. (2003). Indirect effects of grazing and nutrient addition on the hemipteran community of heather moorlands. *Journal of Applied Ecology* **40**, 793-803.

Hartley, S.E. and Mitchell, R.J. (2005). Manipulation of nutrients and grazing levels on heather moorland: changes in *Calluna* dominance and consequences for community composition. *Journal of Ecology* **93**, 990-1004

Heil, G.W. and Diemont, W.H. (1983). Raised nutrient levels change heathland into grassland. *Vegetatio* **53**, 113-120.

Helliwell, R.C., Britton A.J., Gibbs S., Fisher J.M. and Potts J.M. (2010). Interactive Effects of N Deposition, Land Management and Weather Patterns on Soil Solution Chemistry in a Scottish Alpine Heath. *Ecosystems* **13**, 696-711.

Henry, G.H.R., Freedman, B. and Svoboda, J. (1986). Effects of fertilization on three tundra plant communities of a polar desert oasis. *Canadian Journal of Botany* **64**, 2502-2507.

Henrys, P.A., Stevens, C.J., Smart S.M., Maskell, L.C., Walker, K.J., Preston, C.D., Crowe, A., Rowe, E.C., Gowing, D. and Emmett, B.A. (2011). Impacts of nitrogen deposition on vascular plants in Britain: an analysis of two national observation networks. *Biogeosciences* **8**, 3501-3518.

Johansson, M. (2000). The influence of ammonium nitrate on the root growth and ericoid mycorrhizal colonization of *Calluna vulgaris* (L.) Hull from a Danish heathland. *Oecologia* **123**, 418-424.

Johnson, D., Leake, J.R., Lee, J.A. and Campbell, C.D. (1998). Changes in soil microbial biomass and microbial activities in response to 7 years simulated pollutant nitrogen deposition on a heathland and two grasslands. *Environmental Pollution* **103**, 239-250.

Jones, A.G. (2009). *Nitrogen deposition and heathland ecosystems* MSc Thesis Imperial College, London.

Jones, A.G. (2010). Heathland responses to nitrogen deposition: exploring the role of habitat management and soil biochemistry.

Jones, A.G. and Power S.A. (2012). Field-scale evaluation of effects of nitrogen deposition on the functioning of heathland ecosystems. *Journal of Ecology* **100**, 331-342.

Kerslake, J.E., Woodin, S.J. and Hartley, S.E. (1998). Effects of carbon dioxide and nitrogen enrichment on a plant-insect interaction: the quality of *Calluna vulgaris* as a host for *Operophtera brumata*. *New Phytologist* **140**, 43-53.

Kirkham, F.W. (2001). Nitrogen uptake and nutrient limitation in six hill moorland species in relation to atmospheric nitrogen deposition in England and Wales. *Journal of Ecology* **89**, 1041-1053.

Koerselman, W. and Meuleman, A.F. (1996). The vegetation N:P ratio: a new tool to detect the nature of nutrient limitation. *Journal of Applied Ecology* **33**, 1441-1450.

Kristensen, H.L. and McCarty, G.W. (1999). Mineralization and immobilization of nitrogen in heath soil under intact *Calluna*, after heather beetle infestation and nitrogen fertilization. *Applied Soil Ecology* **13**, 187-198.

Kristensen, H.L. (2001). High immobilization of NH_4 in Danish heath soil related to succession, soil and nutrients: implications for critical loads of N. *Water, Air and Soil Pollution: Focus* **1**, 211-230.

Lee, J.A. and Caporn, S.J.M. (1998). Ecological effects of atmospheric reactive nitrogen deposition on semi-natural terrestrial ecosystems. *New Phytologist* **139**, 127-134.

Lee, J.A., Caporn, S.J.M., Pilkington, M., Johnson, D. and Phoenix, G. (2000). *Natural vegetation responses to atmospheric nitrogen deposition – Critical levels and loads of nitrogen for vegetation growing on contrasting native soils*. Progress report, contract EPG 1/3/111, Department of the Environment, Transport and the Regions. Department of Animal and Plant Sciences, University of Sheffield, Sheffield S10 2TN.

Lee, J.A. and Caporn, S.J.M. (2001). *Effects of enhanced atmospheric nitrogen deposition on semi-natural ecosystems.* Progress report, 2000-01. Department of Animal and Plant sciences, University of Sheffield, Sheffield S10 2TN.

Liu, N., Michelsen A. and Rinnan R. (2020). Vegetation and soil responses to added carbon and nutrients remain six years after discontinuation of long-term treatments. *Science of the Total Environment* **722**, 137885.

Manninen, O.H. and Tolvanen A. (2013). N-fertilization and disturbance impacts and their interaction in forest-tundra vegetation. *Plant Ecology* **214**, 1505-1516.

Marcos, E., Calvo, L. and Luis-Calabuig, E. (2003). Effects of fertilization and cutting on the chemical composition of vegetation and soils of mountain heathlands in Spain. *Journal of Vegetation Science* **14**, 417-424.

Marrs, R.H. (1993). An assessment of change in *Calluna* heathland. *Biological Conservation* **65**, 133-139.

Maskell, L.C., Smart S.M., Bullock J.M., Thompson K. and Stevens C.J. (2010). Nitrogen deposition causes widespread loss of species richness in British habitats. *Global Change Biology* **16**, 671-679.

Milbau, A., Vandeplas N., Kockelbergh F. and Nijs I. (2017). Both seed germination and seedling mortality increase with experimental warming and fertilization in a subarctic tundra. *AoB Plants* **9**, plx040.

Möls, T., Paal, J. and Fremstad, E. (2001). Response of Norwegian alpine communities to nitrogen. *Nordic Journal of Botany* **20**, 705-712.

Nadelhoffer, K. and Geiser L. (2011). Tundra, Chapter 5. In: Pardo, L.H., Robin-Abbott, M.J., Driscoll, C.T. (eds.) *Assessment of Nitrogen deposition effects and empirical critical loads of Nitrogen for ecoregions of the United States* Gen Tech Rep NRS-80 Newtown Square, PA: US Department of Agriculture, Forest Service, Northern Research Station, 37-47.

Nielsen, K.E., Hansen, B., Ladekarl, U.L. and Nornberg, P. (2000). Effects of N-deposition on ion trapping by B-horizons of Danish heathlands. *Plant and Soil* **223**, 265-276.

Ochoa-Hueso, R., Stevens C.J., Ortiz-Llorente M.J. and Manrique E. (2013). Soil chemistry and fertility alterations in response to N application in a semiarid Mediterranean shrubland. *Science of The Total Environment* **452**: 78-86.

Ochoa-Hueso, R., Arróniz-Crespo M., Bowker M.A., Maestre F.T., Pérez-Corona M.E., Theobald M.R., Vivanco M.G. and Manrique E. (2014a). Biogeochemical indicators of elevated nitrogen deposition in semiarid Mediterranean ecosystems. *Environmental Monitoring and Assessment* **186**, 5831-5842.

Ochoa-Hueso, R., Paradela C., Pérez-Corona M.E. and Manrique E. (2014b). Pigment ratios of the Mediterranean bryophyte *Pleurochaete squarrosa* respond to simulated nitrogen deposition. In: *Nitrogen Deposition, Critical Loads and Biodiversity*, Springer, 207-216.

Ochoa-Hueso, R. and Stevens C.J. (2015). European semiarid Mediterranean ecosystems are sensitive to nitrogen deposition: impacts on plant communities and root phosphatase activity. *Water, Air, & Soil Pollution* **226**, 1-13.

Ochoa-Hueso, R. (2016). Nonlinear disruption of ecological interactions in response to nitrogen deposition. *Ecology* **97**, 2802-2814.

Ochoa-Hueso, R., Mondragon-Cortes T., Concostrina-Zubiri L., Serrano-Grijalva L. and Estebanez B. (2017). Nitrogen deposition reduces the cover of biocrust-forming lichens and soil pigment content in a semiarid Mediterranean shrubland. *Environmental Science and Pollution Research* **24**, 26172-26184.

Paal, J., Fremstad, E. and Möls, T. (1997). Responses of the Norwegian alpine *Betula nana* community to nitrogen fertilisation. *Canadian Journal of Botany* **75**, 108-120.

Papanikolaou, N., Britton A.J., Helliwell R.C. and Johnson D. (2010). Nitrogen deposition, vegetation burning and climate warming act independently on microbial community structure and enzyme activity associated with decomposing litter in low-alpine heath. *Global Change Biology* **16**, 3120-3132.

Pardo, L.H., Robin-Abbott M.J. and Driscoll C.T. (2011). *Assessment of nitrogen deposition effects and empirical critical loads of nitrogen for ecoregions of the United States*. U.S. Dept. of Agriculture, Forest Service, Northern Research Station. Newtown Square, PA.

Pardo, L.H., Robin-Abbott M.J., Fenn M.E., Goodale C.L., Geiser L.H., Driscoll C.T., Allen E.B., Baron J.S., Bobbink R., Bowman W.D., Clark C.M., Emmett B., Gilliam F.S., Greaver T.L., Hall S.J., Lilleskov E.A., Liu L.L., Lynch J.A., Nadelhoffer K.J., Perakis S.J., Stoddard J.L., Weathers K.C. and Dennis R.L. (2015). *Effects and Empirical Critical Loads of Nitrogen for Ecoregions of the United States*. Dordrecht, Springer.

Payne, R.J., Campbell C., Stevens C.J., Pakeman R.J., Ross L.C., Britton A.J., Mitchell R.J., Jones L., Field C. and Caporn S.J. (2020). Disparities between plant community responses to nitrogen deposition and critical loads in UK semi-natural habitats. *Atmospheric Environment* **239**, 117478.

Phoenix, G.K., Emmett B.A., Britton A.J., Caporn S.J.M., Dise N.B., Helliwell R., Jones L., Leake J.R., Leith I.D., Sheppard L.J., Sowerby A., Pilkington M.G., Rowe E.C., Ashmore M.R. and Power S.A. (2012). Impacts of atmospheric nitrogen deposition: responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments. *Global Change Biology* **18**, 1197-1215.

Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Cresswell, N., Lee, J.A., Reynolds, B. and Emmett, B.A. (2005). Effects of increased deposition of atmospheric nitrogen on an upland moor: nitrogen budgets and nutrient accumulation. *Environmental Pollution* **138**, 473-484.

Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Cresswell, N., Lee, J.A., Emmett, B.A. and Bagchi, R. (2007a). Phosphorus supply influences heathland responses to atmospheric nitrogen deposition. *Environmental Pollution* **148**, 191-200.

Pilkington, M.G., Caporn, S.J.M., Carroll, J.A., Cresswell, N., Phoenix, G.K., Lee, J.A., Emmett, B.A. and Sparks, T. (2007b). Impacts of burning and increased nitrogen deposition on nitrogen pools and leaching in an upland moor. *Journal of Ecology* **95**, 1195-1207.

Pitcairn, C.E.R., Fowler, D. and Grace, J. (1991). *Changes in species composition of semi-natural vegetation associated with the increase in atmospheric inputs of nitrogen*. Institute of Terrestrial Ecology/NERC, Penicuik.

Pitcairn, C.E.R., Fowler, D. and Grace, J. (1995). Deposition of fixed atmospheric nitrogen and foliar nitrogen content of bryophytes and *Calluna vulgaris* (L.) Hull. *Environmental Pollution* **88**, 193-205.

Power, S.A., Ashmore, M.R., Cousins, D.A. and Ainsworth, N. (1995). Long term effects of enhanced nitrogen deposition on a lowland dry heath in southern Britain. *Water Air and Soil Pollution* **85**, 1701-1706.

Power, S.A., Ashmore, M.R. and Cousins, D.A. (1998a). Impacts and fate of experimentally enhanced nitrogen deposition on a British lowland heath. *Environmental Pollution* **102**, 27-34.

Power, S.A., Ashmore, M R., Cousins, D.A. and Sheppard, L.J. (1998b). Effects of nitrogen addition on the stress sensitivity of *Calluna vulgaris*. *New Phytologist* **138**, 663-673.

Power, S.A., Barker, C.G., Allchin, E.A., Ashmore, M.R. and Bell, J.N.B. (2001). Habitat management - a tool to modify ecosystem impacts of nitrogen deposition? *Scientific World Journal,* **1** Suppl 2, 714-721.

Power, S.A. and Barker, C.G. (2003). Deposition measurements at Thursley Common Heathland Nature Reserve. In: *Emiprical Critical Loads for Nitrogen – Proceedings.* BUWAL, Berne.

Press, M.C., Potter, J.A., Burke, M.J.W., Callaghan, T.V. and Lee, J.A. (1998). Responses of a sub-Arctic dwarf shrub heath community to simulated environmental change. *Journal of Ecology* **86**, 315-327.

Riis-Nielsen, T. (1997). *Effects of nitrogen on the stability and dynamics of Danish heathland vegetation*. PhD thesis. University of Copenhagen.

Robinson, C.H. and Wookey, P.A. (1997). Microbial ecology, decomposition and nutrient cycling. In: Woodin, S.J and Marquiss, M. (eds.). *Ecology of Arctic Environments*. Blackwell Science, Oxford, UK, 41-68.

Robinson, C.H., Wookey, P.A., Lee, J.A., Callaghan, T.V. and Press, M.C. (1998). Plant community responses to simulated environmental change at a high Arctic polar semi-desert. *Ecology* **79**, 856-866.

Roth, T., L. Kohli, B. Rihm, R. Meier and B. Achermann (2017). Using change-point models to estimate empirical critical loads for nitrogen in mountain ecosystems. *Environmental Pollution* **220**, 1480-1487.

Schaminée, J.H., Chytrý M., Hennekens S.M., Janssen J.A., Jiménez-Alfaro B., Knollová I., Mucina L., Rodwell J.S. and Tichý L. (2014). *Vegetation analysis and distribution maps for EUNIS habitats. Task 1 & 2*. Report EEA/NSV/14/006.

Schmidt, I.K., Ruess, L., Baath, E., Michelsen, A., Ekelund, F. and Jonasson, S. (2000). Long-term manipulation of the microbes and microfauna of two sub-Arctic heaths by addition of fungicide, bactericide, carbon and fertilizer. *Soil Biology & Biochemistry* **32**, 707-720.

Shaver, G.R. and Chapin, F.S. (1995). Long-term responses to factorial, NPK fertilizer treatment by Alskan wet and moist tundra sedge species. *Ecography* **18**, 259-275.

Shaver, G.R., Johnson, L.C., Cades, D.H., Murray, G., Laundre, J.A., Rastetter, E.B., Nadelhoffer, K.J. and Giblin, A.E. (1998). Biomass and CO_2 flux in wet sedge tundras: responses to nutrients, temperature, and light. *Ecological Monographs* **68**, 75-97.

Simpson, A., Zabowski D., Rochefort R. and Edmonds R. (2019). Increased microbial uptake and plant nitrogen availability in response to simulated nitrogen deposition in alpine meadows. *Geoderma* **336**, 68-80.

Southon, G.E., Field C., Caporn S.J., Britton A.J. and Power S.A. (2013). Nitrogen deposition reduces plant diversity and alters ecosystem functioning: field-scale evidence from a nationwide survey of UK heathlands. *Plos One* **8**: e59031.

Smart, S., Ashmore, M. Hornung, M., Scott, W., Fowler, D., Dragosits, U., Howard, D., Sutton, M. and Famulari, D. (2004) Detecting the signal of atmospheric N deposition in recent national-scale vegetation change across Britain. *Water, Air and Soil Pollution*: Focus **4**, 269-278.

Stevens, C.J., Smart S.M., Henrys P.A., Maskell L.C., Crowe A., Simkin J., Cheffings C. M., Whitfield C., Gowing D.J.G., Rowe E.C., Dore A.J. and Emmett B.A. (2012). Terricolous lichens as indicators of nitrogen deposition: evidence from national records. *Ecological Indicators* **20**, 196-203.

Strandberg, M., Damgaard C., Degn H.J., Bak J. and Nielsen K.E. (2012). Evidence for acidification-driven ecosystem collapse of Danish *Erica tetralix* wet heathland. *Ambio* **41**, 393-401.

Street, L.E., Burns N.R. and Woodin S.J. (2015). Slow recovery of High Arctic heath communities from nitrogen enrichment. *New Phytologist* **206**, 682-695.

Taboada, A., Calvo-Fernandez J., Marcos E. and Calvo L. (2018). Plant and vegetation functional responses to cumulative high nitrogen deposition in rear-edge heathlands. *Science of the Total Environment* **637**, 980-990.

Tipping, E., Henrys P., Maskell L. and Smart S. (2013). Nitrogen deposition effects on plant species diversity; threshold loads from field data. *Environmental Pollution* **179**, 218-223.

Tybirk, K., Bak, J., and Henriksen, L.H. (1995). Basis for mapping of critical loads. *TemaNord* 1995: 610, 1-69. Copenhagen, Nordic Council of Ministers.

Uren, S.C. (1992). *The effects of wet and dry deposited ammonia on Calluna vulgaris*. PhD thesis, Imperial College, South Kensington.

Uren, S.C., Ainsworth, N., Power, S.A., Cousins, D.A., Huxedurp, L.M. and Ashmore, M.R. (1997). Long-term effects of ammonium sulphate on *Calluna vulgaris*. *Journal of Applied Ecology* **34**, 208-216.

Van den Berg, L.J., Jones L., Sheppard L.J., Smart S.M., Bobbink R., Dise N.B. and Ashmore M.R. (2016). Evidence for differential effects of reduced and oxidised nitrogen deposition on vegetation independent of nitrogen load. *Environmental Pollution* **208**, 890-897.

Van der Eerden, L.J., Dueck, Th.A., Elderson, J., Van Dobben, H.F., Berdowski, J.J.M. and Latuhihin M. (1990). *Effects of NH_3 and $(NH_4)_2SO_4$ deposition on terrestrial semi-natural vegetation on nutrient-poor soils*. Report IPO/RIN.

Van der Eerden, L.J., Dueck, T.A., Berdowski, J.J.M., Greven, H. and Van Dobben, H.F. (1991). Influence of NH_3 and $(NH_4)_2SO_4$ on heathland vegetation. *Acta Botanica Neerlandica* **40**, 281-296.

Van der Maas, M.P. (1990). *Hydrochemistry of two douglas fir ecosystems and a heather ecosystem in the Veluwe, the Netherlands.* Report Agricultural University of Wageningen.

Van Kootwijk, E.J. and Van der Voet, H. (1989). *De kartering van heidevergrassing in Nederland met de Landsat Thematic Mapper sattelietbeelden.* Report RIN 89/2, Arnhem (in Dutch).

Wilkins, K., Aherne J. and Bleasdale A. (2016). Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high. *Atmospheric Environment* **146**, 324-331.

Yesmin, L., Gammack, S.M. and Cresser, M.S. (1996). Effects of atmospheric nitrogen deposition on ericoid mycorrhizal infection of *Calluna vulgaris* growing in peat soils. *Applied Soil Ecology* **4**, 49-60.

9 Effects of nitrogen deposition on forests and other wooded land (EUNIS class T, formerly G)

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Signs of strong eutrophication in a mixed forest stand in Switzerland (beech/Norway spruce). Photo: Sabine Braun.

Summary

In this chapter, empirical N critical loads ($Cl_{emp}N$) for forests have been updated and revised based on current reviewed scientific evidence. Compared to the previous report, no new experimental results could be included, but a number of gradient studies using modelled total N or throughfall N deposition were reviewed. Recommendations for $Cl_{emp}N$ were derived using TITAN and change point analyses. A number of studies suggested that the upper level of the previous $Cl_{emp}N$ for deciduous broadleaved forests (T1) was too high. It has, therefore, now been set at 10-15 kg N ha^{-1} yr^{-1}. Furthermore, the lower level of the previous $Cl_{emp}N$ for coniferous forests (T3) has been reduced to 3-15 kg N ha^{-1} yr^{-1}. Information on N effects on lichens and on N leaching led to a lowering of the $Cl_{emp}N$ for northern forests to 3-5 kg N ha^{-1} yr^{-1} (Picea abies, dark taiga or spruce taiga woodlands, T3F) and to 2-5 kg N ha^{-1} yr^{-1} (Pinus sylvestris, light taiga or pine taiga woodlands T3G). The Mediterranean forests could now be included in more detail. This led to the inclusion of new forest types and the updating of the $Cl_{emp}N$ for broadleaved evergreen forests (T2) to 10-15 kg N ha^{-1} yr^{-1}.

9.1 Introduction

Woodland, forests and other wooded lands (i.e. class T ecosystems of the EUNIS) include habitats where the vegetation is, or was until very recently, dominated by trees with canopy cover of at least 10%.

According to the EUNIS classification, woodland and forest habitats (EUNIS level 2; T1, T2 and T3) are separated from other wooded habitats (T4), such as lines of trees, small anthropogenic woodlands (< 0.5 ha), recently felled woodlands, early-stage woodlands and coppice. In EUNIS forests are characterised by the dominant tree types, which may be mixtures of species within the categories *deciduous broadleaved forest* (T1), *broadleaved evergreen forest* (T2) or *coniferous* forest (T3). The EUNIS classification emphasises the dominant tree species, soil hydrology and management practices, more so than soil chemistry. For more details, see Chytrý et al. (2020) and the EUNIS website.

As in the previous updating procedure, non-forest ecosystems (Chapters 3 to 8) have been classified and listed according to the EUNIS habitat classification for Europe. In the previous background document, the then available empirical data on forest ecosystems did not allow for a differentiation below EUNIS level 2 (Bobbink and Hettelingh, 2011). Empirical critical loads of nitrogen ($CL_{emp}N$) were set in 2011 for T1 (broadleaved deciduous forests) and T3 (coniferous forests), with the latter being divided into boreal and temperate types.

One of the main aims of this background document was to achieve a more detailed differentiation (down to level 3) of the $CL_{emp}N$ for forest ecosystems (class T) by updating the literature. However, this approach has been restricted by the following major constraints:

- Several studies cannot be classified below EUNIS level 2 because of a lack of original data or a combination of different forest types within data sets;
- Lack of data from nitrogen (N) addition studies and/or gradient studies on major habitat types, such as all riparian forests, wet forest types and broadleaved evergreen woodland in the Mediterranean region (T2);
- Often, EUNIS classes referring to the dominant tree species do not represent the natural communities because the tree species composition is of anthropogenic origin. This is particularly the case in central Europe, where the native deciduous tree cover has often been replaced by conifers.

To address these constraints, the following structure was adopted in this chapter. First, Chapter 9.2 contains some general remarks on the quantification of N deposition in forests. The overall $CL_{emp}N$ for European forest ecosystems are discussed in Chapter 9.3. It summarises the main effects of N deposition on habitats in classes T1 and T3, without specification down to level 3 categories, as was the case in the 2011 document. A separate evaluation of the impact of N deposition on the boreal forest zone (Taiga woodlands, T3F and T3G) is presented in Chapter 9.4, as sufficient experimental data were now available to distinguish effects on this important subtype of coniferous forests in Europe (T3). The Mediterranean ecosystems are dealt with in Chapter9.5. Furthermore, an additional differentiation between EUNIS types is proposed in Chapter 9.6, based on overview tables that summarise the data from N addition studies and gradient studies across EUNIS types (if classification was possible). In this way, $CL_{emp}N$ values could be allocated for an additional six to eight level 3 categories. Finally, an overview of the $CL_{emp}N$ for class T are summarised in Chapter 9.7. As before, any studies based solely on plantation stands or short rotation forestry (e.g. EUNIS categories T1H, T29, T3M) were excluded. This is because critical loads of N for these intensively used systems were obtained via the steady-state mass balance method (UNECE Mapping Manual, 2004).

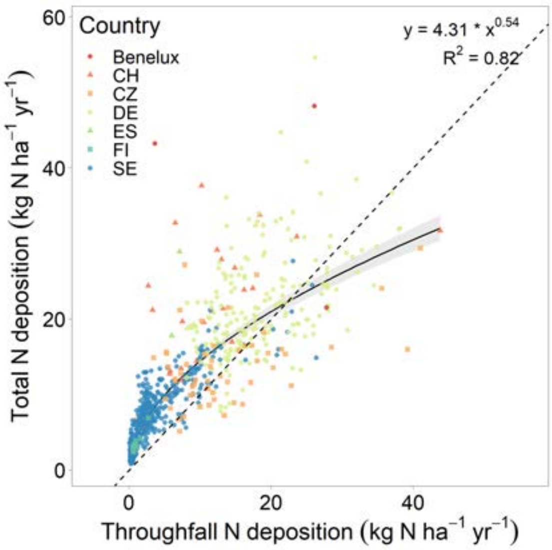
9.2 Atmospheric deposition of N in forest ecosystems

The atmospheric deposition of reactive N species in forest ecosystems occurs mainly as dry and wet deposition, with some "occult" deposition from fog and low clouds. Forests are particularly efficient at scavenging N via dry and occult deposition due to their aerodynamically rough canopies (Fowler et al., 1989). The type and form of N deposition (e.g. NH_4^+, NO_3^- or dissolved organic nitrogen (DON)) may be relevant particularly in terms of the impact on lichen and bryophyte communities. N deposition in forests is usually measured using throughfall (TF), i.e. precipitation collected below canopy plus stemflow (for trees with a smooth bark, such as beech). The deposition collected in the open field is called bulk deposition (BD) (UNECE, 2015). The BD samplers mainly collect wet deposition, while the TF samplers also collect a fraction of dry deposition which varies depending on the atmospheric species (Fenn et al., 2015). The collection of NH_3 may be incomplete as NH_3 is also taken up by stomata (Cape et al., 2009). Particulate NO_3^- and NH_4^+ can also be taken up directly in the canopies (Karlsson et al., 2019). This leads to the underestimation of total N deposition in forests measured as TF, especially in boreal forests where the leaf area index is high, and the precipitation is low (Esseen et al., 2016).

The canopy uptake of part of the deposited N leads to an underestimation of total N deposition by TF. A fraction of the deposited N is taken up directly by forest canopies, without passing through the soil, through the process of canopy exchange (Adriaenssens et al., 2012; Draaijers et al., 1996; Harrison et al., 2000; Karlsson et al., 2019). In Mediterranean areas dry deposition typically exceeds wet deposition in evergreen broadleaf and conifer forests (Aguillaume et al., 2017; Avila et al., 2017; Garcia-Gomez et al., 2018; Sanz et al., 2002). Consequently, TF cannot be used as a direct measure of total N deposition but is used, together with the BD, to calculate the total deposition using a canopy budget model (UNECE, 2015) or in combination with measurements with surrogate surfaces (Karlsson et al., 2019). In polluted areas, TF is higher than the BD alone (outside or above the canopy). This reflects the accumulated dry deposition and the wash-off from the canopy (Vanguelova et al., 2011). However, measurements of TF and BD in Sweden and Finland clearly demonstrated that this was the case only in the southern parts with higher N deposition, while in the northern parts of both Sweden and Finland the amount of N in TF can be lower than in BD (Karlsson et al., 2019; Mustajärvi et al., 2008; Salemaa et al., 2020). Similar observations were made in high elevation spruce forests in Switzerland. Thimonier et al. (2019) published comparisons of TF measurements and total deposition obtained from the inferential measurement of single components. An updated analysis of these comparisons is given in Figure 9.1. At 2 kg N ha^{-1} yr^{-1} throughfall deposition, 6.0 kg N ha^{-1} yr^{-1} total deposition is estimated which is in line with the results discussed previously (Karlsson et al., 2019). Above 20 kg N ha^{-1} yr^{-1} canopy uptake can no longer be observed. The relation can be used to estimate total deposition from TF deposition. This equation should not, however, be applied in northern Fennoscandia where wet deposition is a better estimate of total N deposition than TF (Karlsson et al., 2019; Figure 9.2).

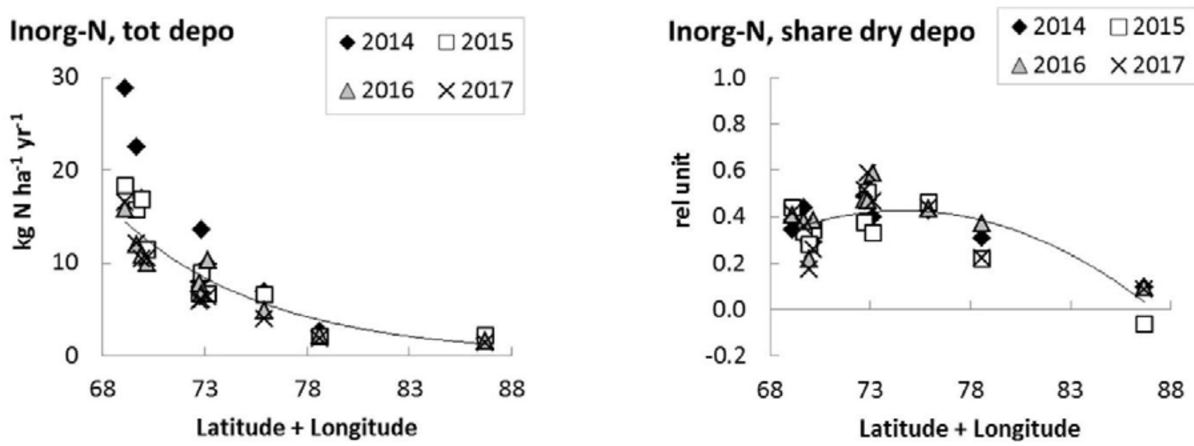
In Mediterranean broadleaf evergreen forests, canopy uptake and retention show characteristic seasonal variations linked to water availability for biological activity (Garcia-Gomez et al., 2016). Moreover, significant alterations of the N deposited when passing through tree canopy have been described in these forests due to microbial activity (Guerrieri et al., 2020). It is thus essential that gradient studies clearly state the fraction of N deposition used for the analysis: studies with only TF deposition and total deposition (either inferential, canopy budget models or modelled total deposition with a high spatial resolution) must be treated separately and, for the former, an estimate of canopy uptake should be added.

Figure 9.1. Relation between total N deposition (dry deposition and wet deposition) and throughfall deposition from coniferous (n = 103), deciduous (n = 33), broadleaved evergreen (n = 4) and mixed (n = 2) forests. Swiss data: coniferous stands from Thimonier et al. (2019) with one additional measurement site (Muri) from Braun et al. (2018). German data: Ahrends et al. (2020) and Schmitz (pers. Comm), Swedish data: Karlsson et al. (2019) and Pihl-Karlsson (pers. Comm), Spanish data: García-Gómez et al. (2018). Additional datasets included were from Zimmermann et al. (2006), Neirynck et al. (2007; 1 site each from Germany and Belgium), Flechard et al. (2011), Korhonen et al. (2013) as well as 36 plots from the Czech Republic (Hůnová et al., 2016).



Source: Braun et al., 2022a

Figure 9.2. The importance of dry deposition decreases towards northern latitudes. The geographical index used on the x-axis, latitude + longitude, decreases from south-west towards north-east. It strongly reflects the gradient in N deposition over Sweden.



Source: Karlsson et al., 2019

The relative importance of the different pathways by which deposited N enters the forest ecosystems has been discussed (Wang et al., 2017), but no overall conclusions have been drawn. More particularly, it is unclear whether and how much N is retained in canopies, re-emitted and/or altered by chemical or biological reactions and what portion and chemical form of deposited N eventually reaches the soil as washed-out N compounds. Some of the N taken up through canopy exchange is, however, likely to end up in the epiphytes growing on tree branches, leaves, needles, trunks (Dahlman et al., 2004; Woods et al., 2012) and on bryophytes in the forest floor layer (Liu et al., 2013; Meyer et al., 2015; Salemaa et al., 2020). Studies using multiple isotope tracers provided evidence of important canopy N transformation processes which should not be ignored and merit further exploration (Guerrieri et al., 2015, 2020). It will not be possible to draw any conclusions on whether it is necessary to provide differentiated CLempN for dry and wet N deposition until further research has been conducted.

Continuous N deposition results in an accumulation in the ecosystem (Aber et al., 1998; Emmett, 2002). Based on vegetation surveys in various ecosystems (grassland, heath, coastal, peatland), Payne et al. (2019) propose 30-year cumulative deposition as an optimum metric for changes in vegetation composition. Rowe et al. (2016) further developed this by proposing a habitat-specific, integrated exceedance of the critical load, over the preceding 30 years for soil-based habitats or three years for epiphytic/epilithic sub-habitats (see also the explicit application of this metric with ICP Integrated Monitoring data in Forsius et al. (2021)). However, the critical load is based on actual deposition data. Currently there is no measure of accumulation.

Meta-analyses suggest that reduced N produces effects at lower loads than the oxidised forms (Yan et al., 2019). In trees and forbs, a greater response of plant growth to NH_4^+-N (+6.3% per g N) than to NO_3^--N (+1.0% per g N) addition was detected. As noted above, any conclusions on whether it is necessary to provide critical loads for reduced and oxidised N species will require further research.

9.3 Effects of N deposition on temperate broadleaved and coniferous forests (T11, T31, T34, T35 and T37)

Forest ecosystems consist of different compartments which may be affected differently by increased N deposition. The soil may be acidified or eutrophied, both processes having consequences for microbiology, groundwater quality, soil fauna and vegetation. The species composition of the ground vegetation and of the mycorrhizal fungi may also be affected, with N-demanding or acid-tolerant species increasing and other species decreasing. Acidification and eutrophication also affect tree nutrition and growth. This may alter trees' resistance to abiotic and biotic stress factors. Additionally, forest ecosystems may release NO, N_2 and N_2O into the atmosphere, the last of which contributes to global warming and depletion of the stratospheric ozone layer. Furthermore, an ecosystem approach requires understanding of the interactions between different compartments that lead to an overall forest ecosystem response. The aim is to find threshold loads for N deposition that correspond to changes in system structure and functioning. This section describes the following indicators for the impacts of N deposition with respect to critical loads:

- **Soil processes:**
  Eutrophication, acidification, mineralisation, nitrification, leaching, N trace-gas emission, litter decomposition and nutrient cycling.
- **Trees:**
  Nutrition, physiology, phenology, recruitment and susceptibility to pest and pathogens.

- **Biodiversity:**
  Macrofungi and mycorrhiza, lichens, free-living green algae, cyanobacteria (i.e. blue-green algae in lichens and bryophytes), ground- and field-layer vegetation and fauna.

9.3.1 Effects on soil processes

Soil plays an important role in mediating N effects on the whole forest ecosystem. The following processes are important:

c) **Soil eutrophication.** A surplus of N, originating from deposition or enhanced nitrification due to accumulated N in the soil, will lead to eutrophication. Field-based ^{15}N studies demonstrated that a large proportion of incoming N (11-56%) was retained in the forest floor through biotic and abiotic processes within the first two years of N enrichment (Emmett et al., 1998; Tietema et al., 1998). An accumulation of ammonium on the soil ion exchange complex may occur in areas with high deposition of reduced N (Boxman et al., 1991; Roelofs et al., 1985; Schulze et al., 1989; Van Dijk and Roelofs, 1988). Ammonium is usually not detectable in forest soil solution, as evidenced by data from ICP Forests plots (De Vries et al., 2003). The need for a multiple indicator approach to monitor and detect forest eutrophication and recovery from N saturation was demonstrated by Verstraeten et al. (2017). The ratio between DON and dissolved inorganic nitrogen (DIN) in soil solution, DON:DIN, has been used as an indicator of N saturation in forests (Park and Matzner, 2006; Williams et al., 2001, 2004). Similarly, low ratios of DON to total dissolved nitrogen (TDN) in soil solution, DON:TDN, and of dissolved organic carbon (DOC) to NO_3^-, DOC: NO_3^-, are also often used as indicators of soil N saturation (Currie et al., 1996; Sleutel et al., 2009). To determine the stage of N saturation, Williams (2004) proposed critical limits of the DON:TDN ratio (stage 0: > 67% DON, stage 1: 33-67% DON, stage 2: < 33% DON).

d) **Nitrate leaching.** Nitrate that is not taken up by the plants or incorporated into microbial biomass or organic matter is leached. Nitrate leaching is an indicator of ecosystem N status. It increases with N deposition (Figure 9.6). Nitrate leaching depends on the C:N ratio in the organic matter (Augustin et al. 2005; Gundersen et al. 1998a; Dise et al., 2009), the tree species and the vegetation cover as well as disturbance of the forest stand (Braun et al., 2020b). Below a C:N ratio of approximately 25 and above an annual N deposition of 10 kg N ha^{-1} yr^{-1}, the level of nitrate leaching drastically increases and endangers groundwater quality (Borken and Matzner, 2004; UNECE, 2005). However, nitrate leaching could be site specific and depends on the N saturation of the ecosystem. Moldan et al. (2018). Tahovská et al. (2020) reported only limited leaching from the Gårdsjön catchment after more than 20 years of 40 kg N ha^{-1} yr^{-1} addition. As with N deposition, N leaching is influenced by forest type, age and soil type amongst other factors and also depends on the amount and fate of N deposition. Trends of N leaching are region and scale specific.

e) **Soil organic matter decomposition and carbon and nutrient cycling.** Generally, short-term and low-dose N inputs to N-poor forests tend to stimulate microbial activity, root autotrophic respiration and organic matter decomposition whereas long-term and high-dose N addition have obvious inhibition effects on soil decomposer activity and heterotrophic respiration in N-rich forests (DeForest et al., 2004; Janssens et al., 2010). Moreover, N addition accelerates the decomposition of soil organic matter (SOM) in high C:N soil but reduces decomposition of soil organic carbon (SOC) in low C:N soil (Lu et al., 2011). Thus, the varying effects of N inputs on soil carbon (C) dynamics observed in studies could be attributed to different responses of forests with differing in N status, suggesting an N input threshold for soil C storage increases or decreases. The C:N ratio is a good indicator of soil organic matter quality as it determines how much N can potentially be mineralised per unit of C respired (Lehtonen et al., 2016). Root trait (e.g. adsorptive root biomass, specific roots length, root tissues density, ectomycorrhizal root biomass) dynamics are strongly related to the soil C:N ratio across a European north to south gradient (Ostonen et al., 2017). For example, fine roots biomass per tree basal area decreased with decreasing soil C:N ratio suggesting lower C input to the soil from fine roots with increasing N accumulation in soils.

f) **Exchange of trace gases between soils and the atmosphere.** The production of N-trace gases in forest soils is mainly due to microbiological processes such as nitrification and denitrification (Bahl-Butterbach et al., 1997; Davidson, 1991). The uptake of atmospheric CH_4 by forest soils is also catalysed by soil microorganisms (Dunfield et al., 1999; King and Schnell, 1998). Various authors have shown a positive correlation between the magnitude of NO and N_2O emissions and the amount of N deposition, as well as a negative correlation between CH_4 uptake and the amount of N deposition for different temperate forest ecosystems (Butterbach-Bahl et al., 2002; Davidson and Kingerlee, 1997; Fenn et al., 1996; Gasche and Papen, 1999; Jenssen et al., 2002). Oulehle et al. (2021) found a strong correlation between the denitrification and precipitation to runoff ratio.

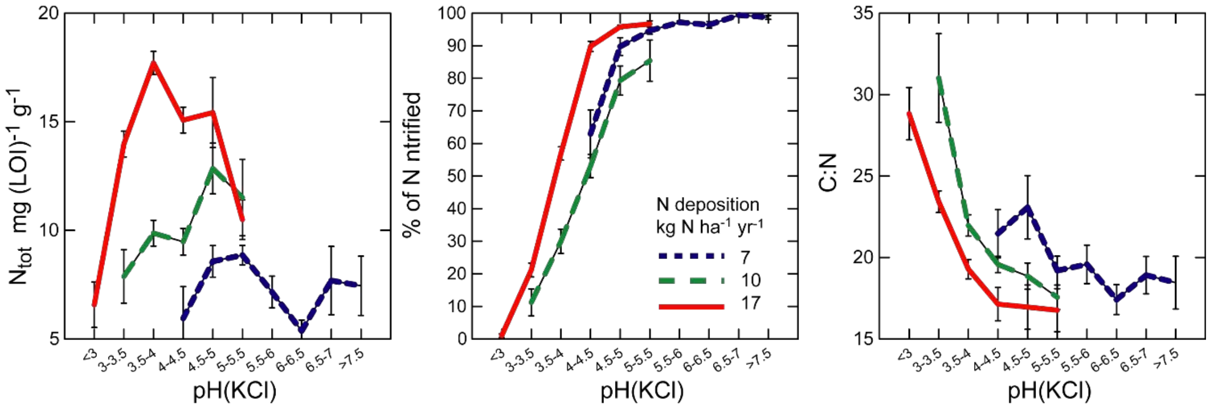
g) **Soil acidification**. This occurs as a result of the nitrification of ammonium and leaching of nitrate. This process leads to mobilisation and leaching of base cations. Below a pH of 4.5, aluminium is increasingly dissolved. This can damage fine root development and mycorrhiza, thereby reducing nutrient and water uptake (Ritter, 1990; Sverdrup and Warfvinge, 1993). Soil acidification and critical loads for acidification are addressed in a separate UNECE document (CLRTAP, 2017).

9.3.2 N mineralisation, nitrification, NO_3^- leaching, N accumulation

N mineralisation and nitrification

N mineralisation and nitrification rates may both be stimulated by N deposition. In a field study on 600 deciduous forests in four geographically separate regions of southern Sweden, the N mineralisation and nitrification rates were by far the highest in the region, with the highest N deposition (17 kg N ha^{-1} yr^{-1}), especially in the most acidic soils. Soil N mineralisation rates increased by 40 to 80%, nitrification rates increased by 20 to 90%, and the C:N ratio decreased by 10 to 25%, compared to the region with a deposition of 7 to 10 kg N ha^{-1} yr^{-1}. (Falkengren-Grerup et al., 1998; Falkengren-Grerup and Diekmann, 2003). Differences in N mineralisation, nitrification and the relationship between C:N ratio and pH of the soil were also observed between areas with 7 and 10 kg N ha^{-1} yr^{-1} (Figure 9.3). The effect on mineralisation may depend on deposition history and background N deposition. Cheng et al. (2020) suggested that the responses of gross rates of N mineralisation, nitrification, and NO_3 immobilisation to experimental N addition changed from positive to negative as background N deposition increased. N deposition of > 15 kg N ha^{-1} yr^{-1} significantly increased the soil nitrification rate in comparison to N inputs of 8-12 kg N $ha^{-1}year^{-1}$. NITREX experiments, for example, did not identify any effects on mineralisation (Emmet, 1999). Lovett et al. (2013) showed that elevated N deposition (after six years of 50 kg N ha^{-1} yr^{-1} addition) led to significant decline in potential N mineralisation and nitrification rates in the mineral horizon but not in the forest floor. The response varied from species to species. Heuck et al. (2018) reported that N addition did not affect N mineralisation in coniferous organic soil horizons, but N mineralisation rates significantly increased in deciduous organic soil horizons.

Figure 9.3. N mineralisation, nitrification and C:N ratio in 10 pH classes (topsoil) and three regions with varying N depositions. Means ± SE.



Source: Falkengren-Grerup and Diekmann, 2003

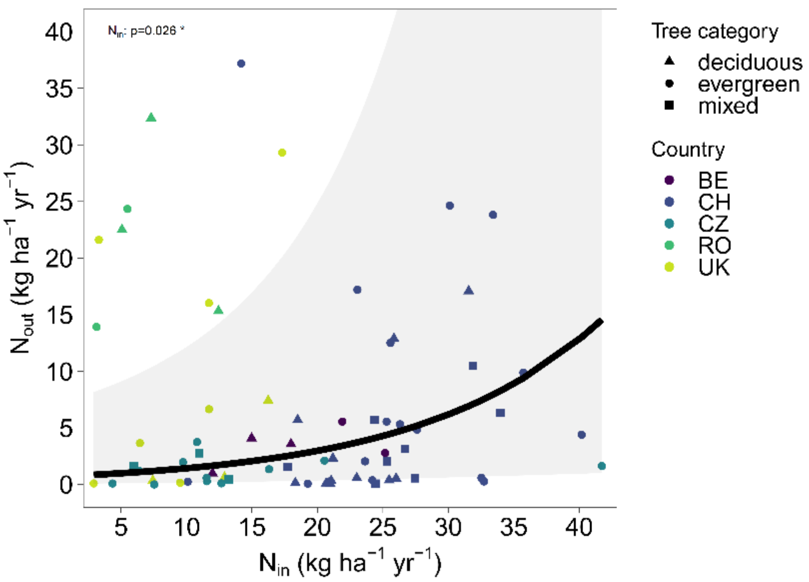
N accumulation

Nitrification is less important in acidic forest soils with raw humus cover and C:N ratios of more than approximately 25. In these soils, N losses are negligible and N is accumulated in all compartments of the ecosystem. The amount of soil C is very important in determining the fate of N in soils. Different soil types have a differing capacity for N retention. For example N stocks in the top 1 m of the soil could vary between 6 t N ha^{-1} in sandy soils and more than 15 t N ha^{-1} in organic soils (Vanguelova et al., 2018). Soil type and texture can also influence the risk of N leaching or N accumulation capacity of various N forms. For example, NH_4^+ can be fixed by some clay types (2:1 clay minerals), be bound to negatively charged soil particles or be bound firmly and irreversibly between clay layers. It can account for up to 20 to 40% of the total mineral soil N in the subsoil and 5%-10% in the surface soil (Weil and Brady, 2017). Regarding accumulation of N, it is worth noting that a high level of retention of airborne N is widespread in European catchments (Vuorenmaa et al., 2017), even in catchments where N deposition was > 20 kg N ha^{-1} yr^{-1} for many decades (Dirnböck et al., 2020). The accumulation of N as NH_4^+ in the soil may shift the ratio between NH_4^+ and base cations (K^+, Ca^{2+} or Mg^{2+}) in the soil with severe consequences for tree nutrition (see Chapter 9.3.2; Roelofs et al., 1985b; Boxman et al., 1988; Van Dijk and Roelofs, 1988) although usually little NH_4^+ can be detected in soil solution (De Vries et al., 2003). Forstner et al. (2019) reported a significant increase in extractable NH_4^+ in the forest floor in N addition plots. The harmful effects of N accumulation on biodiversity are discussed in Chapter 9.3.3.

N leaching

Excess N is leached out from the soil (Figure 9.4), especially at low C:N ratios (Gundersen et al., 2006). The relationship between N input and N leaching may depend on stand structure, tree age, the proportion of dry deposition in total N load (with coniferous stands having a higher potential dry deposition), the forest floor (usually with higher C:N ratios in coniferous stands) and the actual growth rate (which may be higher in coniferous stands than in deciduous ones, if there are no other factors limiting conifer growth). Different leaching patterns between broadleaved and coniferous forests do not necessarily imply a differentiation between $CL_{emp}N$. In forest soils with moder-like and mull-like humus and C:N ratios of below approximately 25, the nitrification effect of tree species on N leaching is not always clear. De Vries et al. (2003) observed that the relationship between N input and output was significantly steeper in deciduous stands than in coniferous ones, whereas Borken and Matzner (2004) and Rothe and Mellert (2004) found the opposite. After comparing directly adjacent stands of Norway spruce and European beech, Braun et al. (2020b) found significantly higher N leaching rates under spruce than under beech trees. The relations between N input and N output are confounded by disturbances and clearcutting which can increase soil mineralisation and NO_3^- leaching (Akselsson et al., 2004; Reynolds et al., 1995; Mannerkoski et al., 2005; Braun et al., 2020b).

Figure 9.4. **Europe-wide forest soil data analysis of N deposition in throughfall (BE, CZ, RO, UK) or total deposition (CH) (N_{in} kg N ha^{-1} yr^{-1}) and NO_3 leaching (N_{out} kg N ha^{-1} yr^{-1}) measured in the mineral soil solution (50-80 cm). Annual data from 70 forest sites averaged for the years 2015-2019 including six tree species (beech, oak, Norway spruce, Sitka spruce, Scots pine and Corsican pine) and tree species mixtures (beech/spruce and oak/beech) for UK, see Vanguelova et al. (2010) and for CH see Braun et al. (2020b). The linear model (LMEM) included besides N_{in} the C:N ratio and tree group (deciduous, evergreen or mixed) as confounding factors and the countries as random effect. The estimated explained variances are $R^2_{marginal}$ = 0.16, $R^2_{conditional}$ = 0.65. The line indicates the estimated effect based on the LMEM with the 95% confidence interval as the shaded area. Data analysis made for this background document by Simon Tresch.**

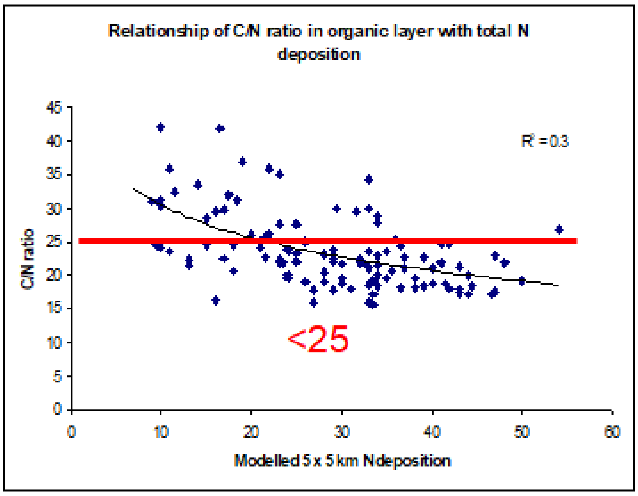


Source: Vanguelova et al., 2010; Braun et al., 2020b; Simon Tresch

Data from the UK's BioSoil network (Vanguelova et al., 2013) showed that the forest floor C:N ratio of conifer woodland is negatively related to modelled 5 x 5 km total N deposition and falls below the suggested critical C:N ratio of 25 when total N deposition is > 20 kg N ha^{-1} yr^{-1} (Figure 9.5). Tree species had an impact on the relationship between N deposition and forest floor C:N ratio (Villada et al., 2013). In areas with higher N loads this indicator is, however, no longer useful (Desie et al., 2020). Forest floor C:N decrease has been found at the UK's ICP forest Level I plots with C:N ratios of > 25 at 75 % of the conifer plots in 1995 (Kennedy, 2003) compared to 40% of conifer plots with C:N ratio > 25 in 2008 (RoTAP, 2012).

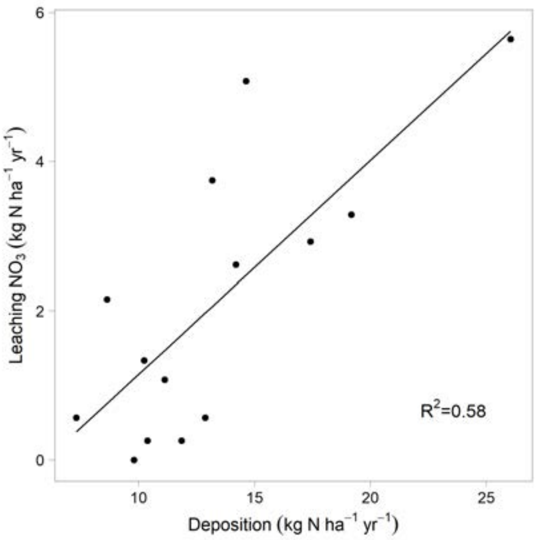
Data from the long-term monitoring GEOMON forested watersheds (Figure 9.6, Oulehle et al., 2021) also show that N leaching increases as the forest floor C:N ratio decreases and with increasing N:P ratio. This is an indication that N saturation might have shifted our forests toward phosphorus (P) limitation.

Figure 9.5. Relationship between forest soil organic layers C:N ratio and total N deposition. Data from 167 BioSoil sites across England, Scotland and Wales. Data from Vanguelova et al. (2013). The red line is the organic layer C:N threshold for NO_3 leaching as suggested by Gundersen (1998a).



Source: Vanguelova et al., 2013; Gundersen, 1998a

Figure 9.6. Runoff leaching losses of NO_3 related to N deposition. Linear regression coefficients shown (R^2) at α = 0.05.



Source: Oulehle et al., 2021

Summary N mineralisation, nitrification, NO_3 leaching, N accumulation

In summary, the available data on soil processes suggest a $CL_{emp}N$ range of 10 to 15 kg N ha^{-1} yr^{-1} for mineralisation, nitrification and nitrate leaching in both coniferous and broadleaved forests ('quite reliable').

Soils N and C trace-gas fluxes

In order to identify the effects of atmospheric N deposition on the magnitude of N and C trace-gas fluxes, a series of Scots pine forest sites with different loads of atmospheric N input was investigated in the north-east of the German Northern Lowland (Butterbach-Bahl et al., 2002;

Jenssen et al., 2002). These studies showed a strong increase in NO and N_2O emissions, both related to humus quality (transition from raw humus to moder, or C:N ratio of humus below 25). The N deposition varied from 10 to 25 kg N ha^{-1} yr^{-1} (throughfall deposition). Furthermore, a decrease in atmospheric CH_4 uptake by forest soils was measured with increasing N deposition.

Unmanaged forest sites were treated with simulated increased N deposition in the range of 10-30 kg N ha^{-1} yr^{-1} over two years. N_2O emissions increased three-fold in the coniferous sites in the first growing season in response to the low N treatment of 10 kg N ha^{-1}, although the response was barely significant ($p < 0.06$). In deciduous forests, increased levels of soil mineral N were observed during the second year of N fertilisation. However, the N_2O fluxes did not increase. Rates of methane oxidation were similar in all sites with no impact of field N application. No effects were observed in soil CO_2 efflux in response to N additions (Ambus and Robertson, 2006).

Litter decomposition, carbon sequestration, DOC leaching, CO_2 release

Litter decomposition is an important part of nutrient cycling in forests. Litter decomposition processes are affected by several drivers in time and space such as litter quality, climate, soil properties and soil biological activity. These drivers are, in turn, influenced by N deposition which means that decomposition rates can be affected as well. The effect of increased N deposition on litter decomposition seems to depend on the lignin concentration of the substrate (Carreiro et al., 2000; Frey et al., 2004; Knorr et al., 2005; Sinsabaugh et al., 2002) and on the decomposition stage (Magill and Aber, 1998). The activity of phenol oxidases, which decompose lignin, is sometimes decreased by N while cellulase activity is stimulated. Thus, the decomposition of lignin-rich litter and litter of the late decomposition stages are delayed. Moreover, in a meta-analysis of 106 long-term studies on litter decomposition, encompassing 21 litter types, the litter mass remaining after decomposition was significantly negatively related to N concentrations. The higher the N concentration in the litter (i.e. the lower the C:N ratio), the more organic matter was left when litter decomposition reached its limit value (Berg and Meentemeyer, 2002). Růžek et al. (2021) analysed the N addition effect (50 kg N ha^{-1} yr^{-1}) on spruce needles, beech leaves, green tea and rooibos tea for 24 months in a spruce and a beech forest in the Czech Republic. They reported only spruce needle decomposition reduction by N addition. Kwon et al. (2021) analysed the response of incubated green and rooibos tea across nine biomes and observed decreased litter decomposition with increasing N deposition in temperate biomes, where atmospheric N deposition rates were high (up to 22 kg N ha^{-1} yr^{-1}). Carreiro et al. (2000) observed significant effects of experimental NH_4NO_3 application on litter decomposition of dogwood (*Cornus florida*) and oak (*Quercus rukbra*) at N applications of 20 kg N ha^{-1} yr^{-1}, with an atmospheric deposition of 10 kg N ha^{-1} yr^{-1}. The mean litter residence time of oak litter was increased from 3.4 years to 4.0 years (20 kg N ha^{-1} yr^{-1}) and 4.5 years (80 kg N ha^{-1} yr^{-1}), respectively.

Long-term N fertilisation in the range of 4-75 kg N ha^{-1} yr^{-1} in northern temperate zones (including roof exclosures) has been estimated to enhance C storage by 0.25 Pg C yr^{-1} (Nadelhoffer et al., 1999). This estimate does not, however, include the effects on soil organic matter (SOM) processes such as the stability increase of soil with long-term elevated N deposition (Hyvönen et al., 2008; Swanston et al., 2004). Concomitant decreases in rates of microbial respiration (the release of CO_2) and decreases in C mineralisation in forest soil (Bowden et al., 2004; Sjöberg et al., 2003; Swanston et al., 2004) increase the potential for C sequestration. Forstner et al. (2019) concluded that the long term N addition of 22 and 35 kg N ha^{1} yr^{1}, respectively, increased SOC in the organic horizon but decreased it in the mineral soil. Thus, it led to vertical redistribution of SOC pools but the overall SOC storage within the topmost 30 cm of soil was unaffected.

Up to now, no consistent DOC leaching response to N addition has been documented. Evans et al. (2008) reviewed 17 field N manipulation studies across northern Europe and north-eastern United States and found that DOC concentrations depended on the form of N used for manipulation: increases (9 experiments) were documented with $NaNO_3$ additions or gaseous NH_3 exposure, and decreases (8 experiments) with most NH_4 additions. Lovett et al. (2013) reported that DOC in soil solution was unaffected by the six years of 50 kg N ha^1 yr^1 addition but did correlate with the C stock in the forest floor. However, although northern temperate forests might now function as significant CO_2 sinks, N deposition only accounted for < 20% of the sink. Predicting the future role of forests in the global carbon budget requires the identification of the mechanisms behind changes in C sequestration (Nadelhoffer et al., 1999).

Soil acidification

Soil acidification is only briefly reviewed in this document because critical loads for acidity, set on the basis of base cation to aluminium ratios and tree growth, are well established and addressed in separate guidelines (Nilsson et al., 1988; Sverdrup and Warfvinge, 1993, CLRTAP, 2017). However, the significance of N compounds in acidification is increasing as sulphur emissions are decreasing. In western Europe, their contribution increased from 53% (1990) to 72% (1999) (Vigdis, 2001). N addition significantly decreased soil exchangeable Ca^{2+}, Mg^{2+} and K^+ in forest ecosystems and significantly increased free Al^{3+} (Tian et al., 2015; Braun et al., 2020b).

Conclusions

The effect of N deposition on soil processes in forest ecosystems is regional- and scale-specific; it depends on the age of ecosystems or soil conditions. Some studies show only a limited effect of N deposition on soil processes, even at doses around 40-50 kg N ha^{-1} yr^{-1}. However, we consider the leaching of nitrates that affects water quality, causes acidification and potentially shifts ecosystems from N limitation to limitation by other nutrients, for instance, base cations or P, to be an essential indicator for setting the critical load. Nitrate leaching increases significantly with N deposition over 10-15 kg N ha^{-1} yr^{-1} throughout European forests. The results from Falkengren-Grerup et al. (1998) and Falkengren-Grerup and Diekmann (2003a) from 600 Swedish hardwood forests on N mineralisation rates, nitrification rates and the C:N ratio also suggested a $CL_{emp}N$ < 17 kg N ha^{-1} yr^{-1}. The results of Thorpe (2011) for nitrification rates suggested a $CL_{emp}N$ of < 15. Therefore, a range of 10-15 kg N ha^{-1} yr^{-1} is proposed as $CL_{emp}N$ for soil processes (N mineralisation and nitrification classified as 'quite reliable', and NO_3^- leaching classified as 'reliable').

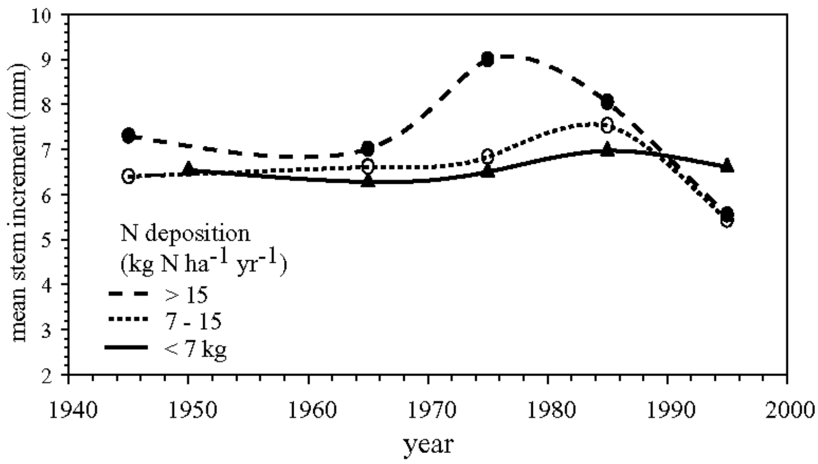
9.3.3 Effects of N deposition on growth, nutrition, physiology and parasite attacks on trees

The growth of a vast majority of the forest tree species in the northern hemisphere was originally limited by N (Tamm, 1991). An increase in the supply of any essential nutrient, including N, will stimulate tree growth as long as growth is not limited by other factors. The initial impact of increased N deposition, therefore, most often has a fertilising effect. However, chronic N deposition may result in 'N saturation'. At this point, increased N inputs no longer stimulate tree growth but start to disrupt ecosystem structure and function (Aber et al., 1989; Agren, 1983; Tamm, 1991).

9.3.4 Growth of aboveground plant parts

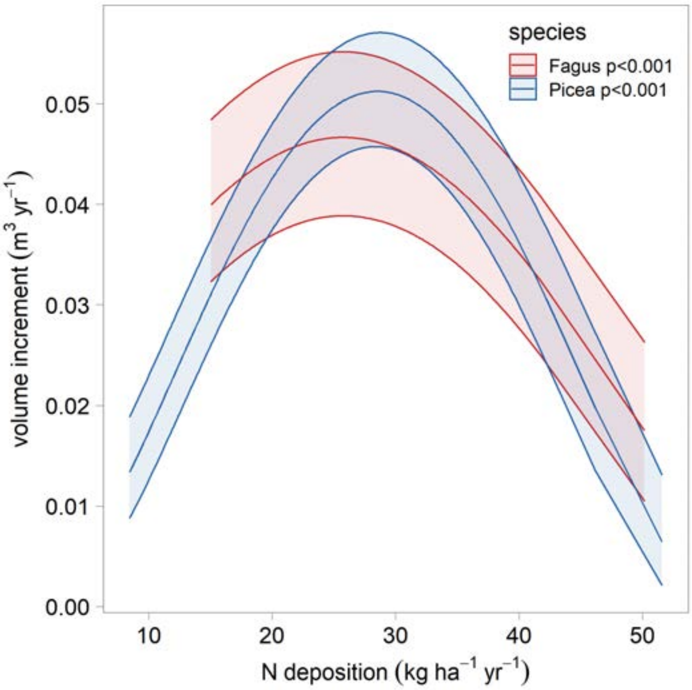
Based on the definition of critical load (Grennfelt and Thörnelöf, 1992), all changes in growth due to anthropogenic N input have to be regarded as undesirable effects, and this includes increased growth. The promoting effect of N on forest growth in temperate European regions has been demonstrated in the past 40 to 50 years where Spiecker et al. (1996) showed growth increases and EFI (2002) concluded that increased N deposition was the main cause. In the meantime, this increase has, however, changed to a growth decrease especially in regions with high N deposition (Kint et al., 2012; Nabuurs et al., 2013). Such a reverse of the response is in line with an increment core study from southern Norway where an initial stem growth increase shifted to a decrease in regions with a modelled wet N deposition of 7 to 15 kg N ha^{-1} yr^{-1} in the 1990s. No change was observed in plots with a modelled wet N deposition below 7 kg N ha^{-1} yr^{-1} (Figure 9.7; Nellemann and Thomsen, 2001). Relationships between growth and N deposition often show a growth maximum at 20-40 kg N ha^{-1} yr^{-1} (e.g. Braun et al. (2017): gradient with beech and Norway spruce from 10 to 42 kg N ha^{-1} yr^{-1} (Figure 9.8), and Etzold et al. (2020): gradient with spruce, pine, beech and oak from 2-48 kg N ha^{-1} yr^{-1}).

Figure 9.7. Stem increment of spruce in Norway, grouped in relation to the amount of wet N deposition. Growth increase in the highest deposition class, as well as growth decrease in the two highest classes are significant at $p < 0.01$. R. Wright (personal communication) estimated dry deposition in southern Norway to add another 10 to 20% to these N deposition rates.



Source: Nellemann and Thomsen, 2001

Figure 9.8. Volume increment of beech and Norway spruce in Switzerland in relation to total N deposition. Dataset described in Braun et al. (2022b).



Source: Braun et al., 2022b

The gradient studies with mature *Fagus sylvatica* presented by Braun et al. (2017) are similar in the type of change to the ones derived from experimental N addition to a young beech forest on calcareous and acidic soils, with modelled background deposition of 15 or 30 kg N ha^{-1} yr^{-1}, respectively (Flückiger and Braun, 2011). But the responses in the gradient study are more sensitive, suggesting that gradient studies, with a number of observations at low N depositions and long-term exposures, may predict more sensitive reactions. Growth responses seem to be similar on acidic and calcareous soils (Flückiger and Braun, 2011).

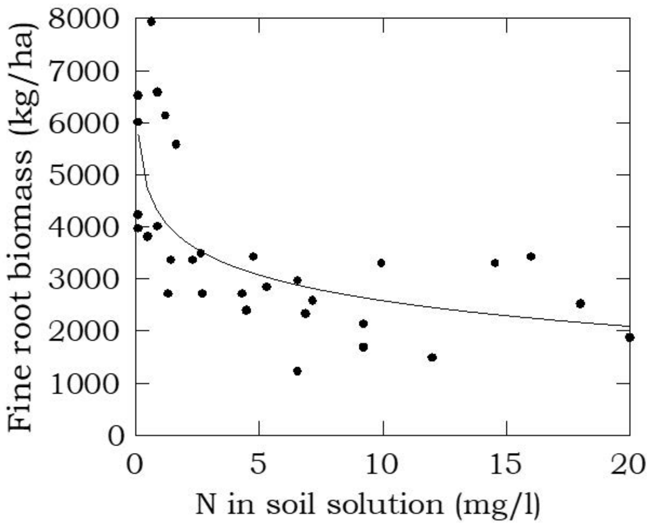
A growth reduction due to N was also shown on the NITREX experimental plot in the Netherlands, where ambient N deposition was reduced from 56 to 4 kg N ha^{-1} yr^{-1}. Trees in a roofed environment with low N, grew better than in the roofed control environment with high N (Boxman et al., 1998). In the Swedish Skogaby experimental plot, Norway spruce fertilised with 100 kg N ha^{-1} yr^{-1} (in the form of $(NH_4)_2SO_4$, background deposition 16 kg N ha^{-1} yr^{-1}) grew better for the first three years than those on the control plots, but after ten years their growth fell below that of the control plot (Jönsson et al., 2004b).

A study using a unique dataset of 80 forest FLUXNET sites (Fleischer et al., 2013) showed that forest canopy photosynthetic capacity relates positively to N deposition for evergreen needleleaf forests below an observed critical load of ~ 8 kg N ha^{-1} yr^{-1}, with a slope of 2.0 ± 0.4 (S.E.) μmol CO_2 m^{-2} s^{-1} per kg N ha^{-1} yr^{-1}. Above this threshold canopy photosynthetic capacity levels off, exhibiting a saturating response in line with the N saturation hypothesis. Climate effects on canopy photosynthetic capacity cannot be separated from the effect of N deposition due to considerable covariation. For deciduous broadleaf forests and forests in the temperate (continental) climate zones, the analysis shows the N deposition effect to be either small or absent (Fleischer et al., 2013).

Root growth

N may also stimulate root growth (Heinsdorf and Schulzke, 1969; Persson, 1980) in ecosystems which are not N saturated. For example, fine root biomass increased significantly after 20 years of N addition (35 kg N ha^{-1} yr^{-1} at the Danish site Klosterhede; Forstner et al., 2019). However, some studies showed that root growth is inhibited by excessive N supply. There is strong evidence of increased N deposition causing reduced fine-root biomass and root length. Increasing root biomass and root vitality in Scots pine, Douglas-fir and Norway spruce were reported when trees were protected from N deposition in the NITREX roofed experiments (Boxman et al., 1995; Murach and Parth, 1999; Persson and Ahlström, 2002). The treatments consisted of a reduction in N deposition from 56 to 4 kg N ha^{-1} yr^{-1} in The Netherlands (Scots pine) (Gundersen et al., 1998b), from 36 to less than 5 kg N ha^{-1} yr^{-1} in Germany (Douglas-fir, Norway spruce) and from 13 to less than 4 kg N ha^{-1} yr^{-1} in Sweden (Norway spruce). In a gradient study, Matzner and Murach (1995) observed a relation between total fine-root biomass of Norway spruce saplings and N concentrations: it decreased significantly when NO_3^- and NH_4^+ in soil water were more than 2 mg N l^{-1} (Figure 9.9). From the relationship between N deposition and NO_3^- concentration as published by De Vries et al. (2001), this concentration may be attributed in coniferous stands to an average N throughfall load of 25 kg N ha^{-1} yr^{-1} (range 13-33). Magill et al. (2004) also provided evidence of declining fine-root biomass although they found only a trend of a 20 to 25% fine-root biomass reduction in the organic horizons after a 15-year N addition experiment at 50 and 150 kg N ha^{-1} yr^{-1}, on pine and hardwood stands, respectively (Harvard Forest, Massachusetts (USA), background deposition 8 kg N ha^{-1} yr^{-1}). Fenn et al. (2008) reported a 26% reduction in fine-root biomass of *Pinus ponderosa* at a 17 kg ha^{-1} yr^{-1} N deposition from throughfall. Additionally, Braun et al. (2005) observed decreasing fine-root lengths (< 0.25 mm ⌀) of young beech in a gradient study in Switzerland, which was related to modelled N deposition, although in this study a confounding effect from soil acidification could not be excluded (range of modelled N deposition of 18 to 35 kg N ha^{-1} yr^{-1}). Altogether, increased N deposition will lead to a less developed fine-root system, possibly resulting in reduced tree stability. Forests experiencing high N depositions may become more vulnerable to storms.

Figure 9.9. Fine-root biomass of Norway spruce in relation to N in soil solution.



Source: Matzner and Murach, 1995

Fine-root density and specific root length were found to be either reduced at increased N availability in several gradient studies and N experiments focussing on ectomycorrhizal fungal communities (Almeida et al., 2019; De Witte et al., 2017; Kjøller et al., 2012) or unchanged (Morrison et al., 2016).

Stem increment data are more difficult to interpret as the relative change depends on current nutrition status. The dataset by Braun et al. (2017) showed a significant increase of volume increment of Norway spruce at N deposition of < 15 kg N ha^{1} yr^{1}. Root growth data are more relevant for tree vitality, but they are less frequently collected. The root biomass results from the NITREX roof clean experiments (Gundersen et al., 1998b) suggested a $CL_{emp}N$ range of 10 to 15 kg N ha^{-1} yr^{-1}. The $CL_{emp}N$ can be set for both coniferous and broadleaved temperate forests and is considered as 'quite reliable'. This is lower than the currently valid $CL_{emp}N$ for broadleaved temperate forests of 10-20 kg N ha^{-1} yr^{-1}.

Nutrition of trees

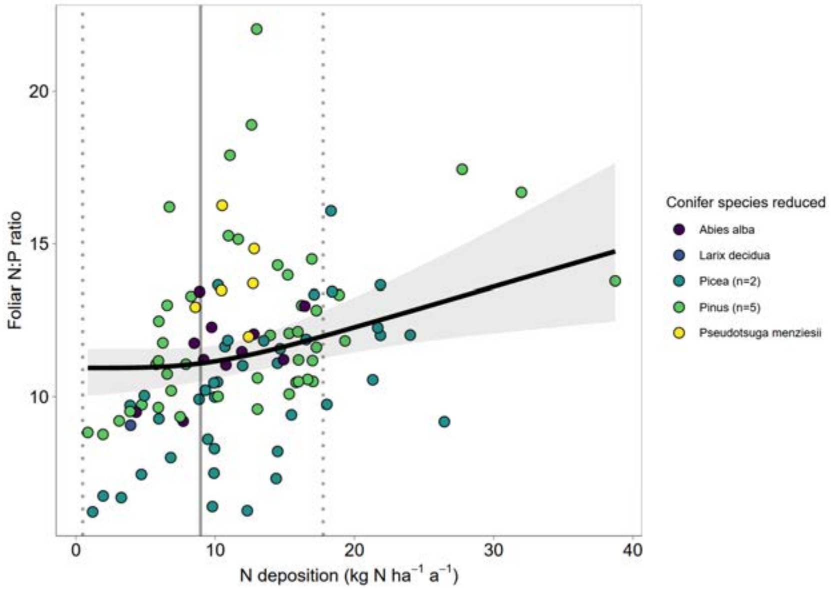
It is a well-known fact that P and N are known to be limiting nutrients in forests (Johnson and Taylor, 1989). However, increasing N deposition rates during the last decades of the 20th century could have exacerbated P deficiency in forest ecosystems that already had a low P supply (Tamm, 1991). Increased N deposition may change the nutrition for trees by increasing the N concentration in the foliage and/or decreasing the (relative) uptake of other nutrients. Usually, P concentrations and – depending on the soil – also K and/or Mg concentrations are lowered in parallel. Thus, the ratios between N on the one hand and P, K and Mg on the other, tend to increase. These changes were found in both field observations and experimental N additions. The mechanisms behind the changes may differ from element to element. In the case of Mg, soil acidification and leaching of nutrients seem to play an important role (Braun et al., 2020b; Cape et al., 1990; Elling et al., 2007). Competition from NH_4^+ in root uptake may be important in areas with very high NH_y deposition (Roelofs et al., 1985). Nonetheless, the main reason is probably the result of N impacts on mycorrhizal fungi (see Chapter 9.3.3). In ecosystems where N is still promoting growth, a decrease in nutrient contents may also be the result of a dilution effect (Thelin et al., 1998).

An increase in the N:P ratio seems to be a quite general response as shown in the meta-analysis by Peng et al. (2019), either by increasing the N or decreasing the P concentration. While earlier studies described rising foliar N concentrations with increasing N deposition (Duquesnay et al., 2000; De Vries et al., 2003), this relationship seems to have disappeared as N concentrations decreased between 1992 and 2009 (Jonard et al., 2015). While the studies of Duquesnay and Jonard compared time series with no direct relationship to N deposition, Braun et al. (2020a) observed a changing relationship in the foliar N concentrations of *Fagus sylvatica* in Switzerland between 1984 and 2015. Whereas the relationship with N current deposition was positive in the 1980s, it disappeared later, suggesting a saturation process. At the same time, the relationships between foliar P concentrations and N deposition became more negative. The N deposition gradient over the 1984-2015 period was between 12 and 45 kg N ha^{1} yr^{1}.

Experimental N addition to saplings of beech and Norway spruce in young stands on both acidic and calcareous soils induced nutrient imbalances and deficiencies. They were significant at added N loads of more than 10 to 20 kg N ha^{-1} yr^{-1}, after four and six years of N treatment (with modelled atmospheric depositions of 15 and 20 kg N ha^{-1} yr^{-1}, respectively). On acidic soil, N treatment led to acute Mg deficiency, whereas on calcareous soil, K and P became limiting (Flückiger and Braun, 1999). N concentrations remained unchanged. Foliar nutrient concentrations in mature forest trees showed significant relationships of P and K concentrations with N deposition, in a range of 10-50 kg N ha^{-1} yr^{-1} (Braun et al., 2020a). These relationships observed in the field suggested a more sensitive response than in the experiment. Additions of 35 kg N ha^{-1} yr^{-1} to an ambient N input of 15 to 20 kg N ha^{-1} yr^{-1}, in a spruce forest at Klosterhede (Denmark), led to increased concentrations of N in the needles, and to decreased P and Mg concentrations in the foliage, during the three years of treatment (Gundersen, 1998).

With high N inputs, concentrations of organic N in needles may increase to levels above the optimum range (De Kam et al., 1991; Van Dijk and Roelofs, 1988). N-rich free amino acids, especially arginine, were found to have increased significantly in needles with high tissue N concentrations (Hällgren and Näsholm, 1988; Näsholm and Ericsson, 1990; Påhlsson, 1992; Pietilä et al., 1991; Van Dijk and Roelofs, 1988). It has been suggested that arginine concentrations in foliage are a sensitive indicator of N input (Edfast et al., 1990; Huhn and Schulz, 1996). In Sweden, arginine concentrations in coniferous foliage of more than 5 µmol g dw^{-1} have been linked to NO_3^- leaching, because the arginine accumulation in trees correlated with decreased uptake rates of NH_4^+, leaving NH_4^+ ions available for nitrification. This was subsequently followed by NO_3^- leaching (Näsholm et al., 1997). In permanent observation plots in Sweden, arginine concentrations in Norway spruce were higher in areas with high inputs of N (Ericsson et al., 1995). In Swiss plots, arginine concentrations in spruce foliage, although not in beech leaves, correlated with modelled N depositions in the range of 14 to 37 kg N ha^{-1} yr^{-1} (Braun et al., 2010; Quiring et al., 1997). The strongest correlation, however, was observed between arginine and P concentrations in the foliage of both tree species, suggesting P limitation. By decreasing N deposition in the NITREX roofed experiment, arginine concentrations in needles of Scots pine decreased significantly (Boxman et al., 1995; Boxman and Van Dijk, 1994). Similarly, cessation of N additions resulted in a rapid decrease in arginine in Scots pine growing in central Sweden (Edfast et al., 1996) and northern Sweden (Quist et al., 1999). In 109 ICP Forest plots, median N deposition for plots with a balanced nutrition (ratio between N and the other macronutrients) was 9.6 kg N ha^{-1} yr^{-1}, and for the unbalanced plots it was 21 kg N ha^{-1} yr^{-1} (De Vries et al., 2003). N deposition was total deposition derived from throughfall and bulk deposition using a canopy exchange model (ICP Forests, 2001). The old critical load for nutrition imbalances was based on this analysis. An update has been published by Du et al. (2021). These authors showed a relation between foliar N:P and N deposition from EMEP (0.1°grid resolution). An analysis of these data using change-point regression according to Roth et al. (2017) by Tresch, Braun and Roth yielded a $CL_{emp}N$ of 14.3 kg N ha^{-1} yr^{-1} (Figure 9.10).

Figure 9.10. Relationship between foliar N:P ratio of European conifer tree species (103 sites, 10 conifer species from 1995-2017) and N deposition (EMEP data spatial resolution 0.1° (~ 11.1 km)) provided by Du et al. (2021). The estimated change point from the non-linear Bayesian change-point regression is given with 95% CI as shaded grey areas (R^2=0.42). The dashed lines are the 95% CrI from the change-point model, with an estimated CL of N of 9.0 (SD = 4.3) kg N ha^{-1} yr^{-1}. Analysis made for this background report by Roth et al. (2022).



Source: Du et al., 2021; Roth et al., 2022

From the data analysis on N:P ratio by Du et al. (2021), a $CL_{emp}N$ of 15 g N ha^{-1} yr^{-1} is proposed. The gradient study presented by Braun et al. (2020a) suggested a $CL_{emp}N$ for coniferous temperate forests of 10-15 kg N ha^{-1} yr^{-1} and for deciduous temperate forests of 15-20 kg N ha^{-1} yr^{-1}. Both ranges are considered 'quite reliable'. Although calcareous and acidic soils may have different types of nutrient imbalances, there is no fundamental difference in the sensitivity of the response.

Tree physiology

Winter injury

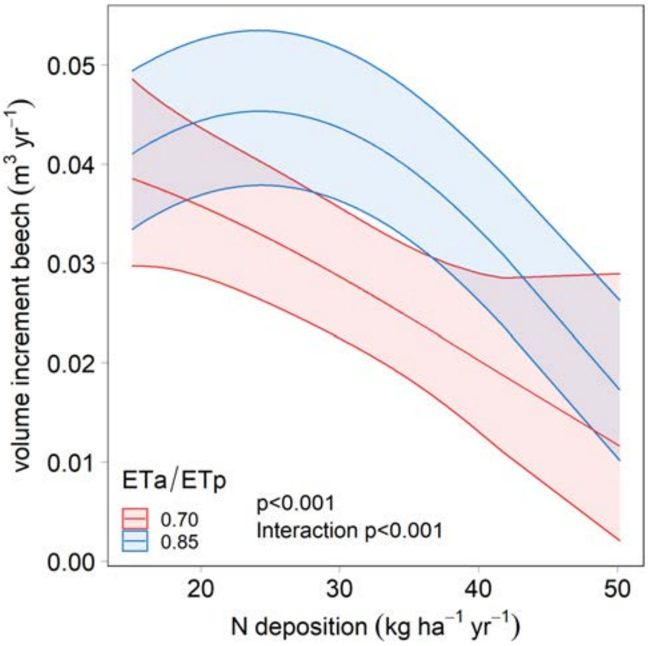
Changes in nutrient status may influence frost hardiness by affecting carbon production, respiration and allocation, as well as via changes in membrane properties and osmotic potential (Bigras et al., 2001). Increased N concentrations in foliage may increase respiration rates and thereby reduce non-structural carbohydrate reserves, including the sugars that protect against frost during the winter. Winter injury may be caused by either low temperatures (frost sensitivity) or drought stress (winter desiccation). Most studies found a decreased sensitivity of needles to frost, after N addition (DeHayes et al., 1989; Klein et al., 1989; L'Hirondelle et al., 1992). Sensitivity to drought also seemed to increase. For example, long-term fertilisation with various loads of N (15.7, 19.8, 25.6 and 31.4 kg N ha^{-1} yr^{-1}, atmospheric bulk precipitation 5.4 kg N ha^{-1} yr^{-1}) between 1988 and 1996, in the eastern United States, significantly increased winter injury in montane Red Spruce (*Picea rubens*) foliage at N additions of more than 15.7 kg N ha^{-1} yr^{-1}, although cold tolerance was not affected and N treatment decreased dehardening (Perkins et al., 1999). Jönsson et al. (2004a) observed increased frost sensitivity of the inner bark of Norway spruce after eleven years of continuous application of ammonium sulphate (Skogaby experimental plot, 100 kg N ha^{-1} yr^{-1}). They attributed this observation mainly to nutrient deficiency with Mg of the N treated trees in the deficient range. In another experiment, Jönsson et al. (2004a) found that spruce needles with a lower K and P status were more sensitive to frost. This may be of importance as an N surplus often lowers the supply of these elements (see section on tree nutrition). Moreover, in field fertilisation experiments it is often observed that tree growth starts earlier in the season, which may increase damage by late frosts (Jönsson et al., 2004a).

Drought tolerance

There are a number of reports indicating that N addition increases drought sensitivity. These interactions are especially important in the light of climate change. Increased water loss, a consequence of increased growth, has often been mentioned as a reason for increased drought sensitivity in response to N addition (Valliere et al., 2017). But there may be other reasons: increased competition from grass vegetation (Anders et al., 2002; Valliere et al., 2017), increased stomatal conductance (Liang et al., 2020; Nilsen, 1995), decreased root growth (see Chapter 9.3.2), decrease of the mycorrhizal symbionts (De Witte et al., 2017; Van der Linde et al., 2018) and unbalanced nutrition as a consequence of N deposition (see Chapter 9.3.2). The effect of an interaction between drought and increased N on water relations at the leaf level, which is often analysed with carbon isotopes (intrinsic water use efficiency), seems to depend very much on species. Increased water use efficiency has been observed in temperate China for *Quercus variabilis* (Hu et al., 2019) while the contrary was found in California for Mediterranean-type shrubs including *Artemisia califonica* (Valliere et al., 2017) or for European beech (*Fagus sylvatica*) (Braun et al., 2009). Several dendrochronological studies with temperate European tree species observed stronger drought effects when N deposition was high (Hess et al., 2018; Kint et al., 2012; Latte et al., 2016; Maes et al., 2019). In a forest monitoring study in Switzerland, N deposition enhanced the drought effect on the stem increment of *Fagus sylvatica* (Braun et al., 2017, Figure 9.11).

Spatial and temporal (over 30 years) changes in tree-ring $\delta^{13}C$-derived intrinsic water-use efficiency (iWUE), $\delta^{18}O$ and $\delta^{15}N$ were studied for four species in twelve forests across climate and atmospheric deposition gradients in Britain (Guerrieri et al., 2020). Six of the sites underwent N deposition > 10-12 kg N ha^{-1} yr^{-1} and six sites deposition < 10-12 kg N ha^{-1} yr^{-1}. There was an increase in iWUE but this was not uniform across sites and species-specific underlying physiological mechanisms reflected the interactions between climate and atmospheric drivers (oak and Scots pine), but also an age effect (Sitka spruce). Most species showed no significant trends for tree-ring $\delta^{15}N$, suggesting no changes in N availability. An increase in iWUE was mostly associated with an increase in temperature and a decrease in moisture conditions across the south-north gradient and over a period of 30 years. Overall, climate had the prevailing effect on changes in iWUE across the investigated sites, while detection of N deposition signals was partially confounded by structural changes during stand development (Guerrieri et al., 2020).

Figure 9.11. Volume increment of beech in Swiss monitoring plots in relation to N deposition. The interaction with drought is shown as regression estimates for two levels of drought (ratio between actual and potential evapotranspiration, Eta/Etp). 0.85 is moist, 0.7 is dry. Dataset described in Braun et al. (2022b). Data: 7400 trees, 8 increment periods, 91 sites.



Source: Braun et al., 2022b

Herbivores and pathogens

With increasing N deposition, the susceptibility of trees to fungal pathogens and insects may change. Increased drought sensitivity in response to N, as outlined in the previous section, may play a role as many pathogens and herbivores require weakened hosts. Decreased rates of mycorrhizal infection (see Chapter 9.3.3) may increase susceptibility to root pathogens (Branzanti et al., 1999; Marx, 1969). Furthermore, altered concentrations of phenolic compounds and soluble N compounds, such as free amino acids, may also play a role (Bolsinger and Flückiger, 1989; Huber, 1980; McClure, 1980; Nordin et al., 2005; Nybakken et al., 2018). A fertilisation experiment involving additions of 10 kg N ha^{-1} yr^{-1} at two Swiss sites (with an atmospheric background deposition of 12 and 20 kg N ha^{-1} yr^{-1}, respectively) showed that such additions were enough to alter the concentrations of fungistatic phenolic compounds in the fine roots of young beech and spruce after seven years of N treatment, with mostly decreasing levels (Tomova et al., 2005). In the same experiment, two fungistatic phenolic compounds in the leaves correlated negatively with the N:P ratio in leaves. The total amount of phenolic compounds in *Fagus* leaves in a 120-year stand in southern Sweden also decreased by more than 30% after four years of fertilisation with approximately 45 kg N ha^{-1} yr^{-1}, compared with the control trees (Balsberg-Påhlsson, 1992).

Fungal pathogens

In the Netherlands, an epidemic of the pathogenic fungi *Brunchorstia pineae* and *Sphaeropsis sapinea* on *Pinus nigra* in coniferous forests was observed in the 1980s, especially in the south-eastern part of the country with high levels of atmospheric N deposition (Roelofs et al., 1985). Affected trees in the infested stand had significantly higher foliar N concentrations and higher soil ammonium levels than uninfected trees. Most of the additional N in the needles of the affected stands was stored as free amino acids, especially arginine, but proline concentrations were also increased in the infected trees, indicating an enhanced degree of water stress (Van Dijk et al., 1992). Both high N supply and water stress increased the trees' susceptibility to attacks from *Sphaeropsis sapinea* (Blodgett et al., 1997).

In permanent beech monitoring plots in Switzerland (modelled N deposition of 15-35 kg N ha^{-1} yr^{-1}) and N fertilisation experiments, a significant positive correlation was found between the N:K ratio in leaves and the necroses caused by the beech canker *Nectria ditissima* (Flückiger et al., 1986; Flückiger and Braun, 1998). In addition, beech bark lesions caused by the fungal pathogen *Nectria coccinea,* var. *faginata,* were found in 25 out of 48 studied sites in Scania (south Sweden). They were more frequent at sites with higher N deposition (20-25 kg N ha^{-1} yr^{-1}) than those with lower N deposition (15-20 kg N ha^{-1} yr^{-1}) (Westling et al., 1992). In a gradient study in two US regions, Latty et al. (2003) observed a positive correlation between the incidence of beech bark disease on *Fagus grandifolia* with bark N content and tree size. In two N fertilisation experiments, one on acidic and the other on calcareous soil, with additions of 10, 20, 40, 80 and 160 kg N ha^{-1} yr^{-1}, damage to young beech by the pathogenic fungi *Apiognomonia errabunda* and *Phomopsis* species, was found to be significantly increased after treatments of > 10 kg N ha^{-1} yr^{-1} (atmospheric N depositions of 20 and 15 kg N ha^{-1} yr^{-1}). In addition, a strong positive correlation was found between the extent of twig necroses and both N:P and N:K ratios in leaves (Flückiger and Braun, 1999). The effects in the experiments on acidic and calcareous soils were similar.

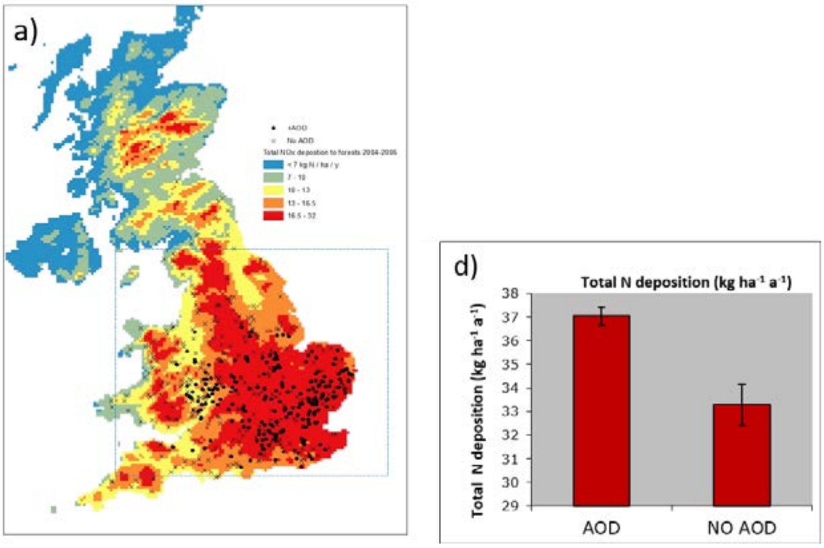
Effects of N on pathogen and insect infestations have also been observed in understory shrubs. They are addressed in Chapter 9.3.3.

Insects

The Ellenberg N indicator value of the host plant and, by extension soil eutrophication, was positively related to the body size of butterflies and moths (Pöyry et al., 2017). Thus, soil N enrichment amplifies the diverging trends of herbivorous insects feeding on nitrophilous versus nitrophobous plants through differential plant-herbivore interactions. N deposition-induced changes in food quality, quantity and phenology may alter herbivore population dynamics. For insect herbivores, the N concentration of the host plants very much controls processes such as growth, survivorship, population levels and outbreak frequency. Changes to these processes result from both the direct effects of N on host plant quality and its influences on plant defensive chemistry. N deposition may also affect plant-herbivore interactions by altering relationships between herbivores and their natural enemies, leading to changes in herbivore survival and population dynamics (Throop and Lerdau, 2004). Many of the insects have been found to respond to N fertilisation feed by sucking. Infestation by the beech aphid *Phyllaphis fagi,* in an N fertilisation experiment, increased significantly with increasing foliar N concentrations and N:P ratios (Flückiger and Braun, 1998). On Swiss permanent observation plots, damage to beech nuts by the tortricid *Cydia amplana*, a non-sucking insect, was also found to have increased significantly with increasing foliar N:P ratios (N deposition 15-60 kg N ha^{-1} yr^{-1}) (Flückiger and Braun, 2004).

The occurrence of insect damage to pine needles in permanent observation plots in the UK correlated positively with modelled N deposition (range 7-22 kg N ha^{-1} yr^{-1}), but only within Scotland. For these plots, a negative relationship between needle retention and modelled N deposition was also reported (NEGTAP, 2001). Another study revealed a significant influence of total and dry N deposition on the occurrence of Acute Oak Decline syndrome (AOD), across England and Wales (Denman et al., 2014). It suggested that low rainfall, high temperature and high N deposition (> 8 kg dry NO_x ha^{-1} yr^{-1}) and > 33 kg total N ha^{-1} yr^{-1}) could increase the oak's predisposition to bacterial and pest attacks (Figure 9.12; Brown et al., 2018).

Figure 9.12. Map of modelled total NO_x deposition (5 x 5 km grid; data from Centre of Ecology and Hydrology, UK) is shown with black dots representing oak forests with Acute Oak Decline syndrome (AOD, n = 241), grey crosses representing healthy oak forests (NO AOD, n = 291) (a) and annual mean modelled total N deposition in both AOD and healthy forests (d).



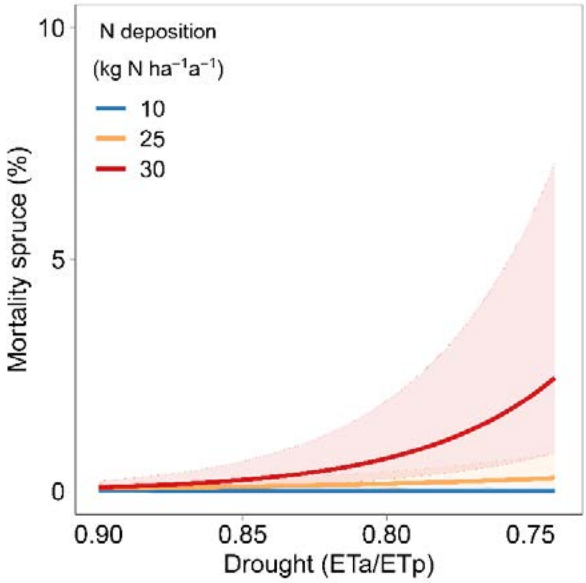
Source: Brown et al., 2018

While the greater presence of the insects listed above may be explained by higher concentrations of soluble N compounds in the leaves or needles, the situation is more complicated in the case of *Haematoloma dorsatum,* a cicade*.* Originating from the Mediterranean region, this insect causes severe needle damage in pine stands in the Netherlands. Nymphs of this bug suck exclusively on the roots or basal stem parts of *Deschampsia flexuosa*. Only the adults cause damage to trees. *Deschampsia flexuosa* seems to be crucial for nymph development. As wintergreen grass, it is a food source in wintertime and early spring. The abundance of this grass in pine forests is thus an important ecological factor for the development of *Haematoloma dorsatum* (Moraal, 1996). Various studies have shown that the abundance of *Deschampsia (Avenella) flexuosa* increases significantly when N deposition is more than 10 to 15 kg N ha^{-1} yr^{-1} (see Chapter 9.3.3). Although this is a correlation study, it may explain the mechanism behind vegetation changes.

Tree mortality

Tree mortality is often the result of infestation by an insect or a pathogen. In a US-wide study with 71 tree species, Horn et al. (2018) observed a positive relationship between N deposition and mortality in 29 species, with effect peaks as low as < 2 kg N ha^{-1} yr^{-1} (average 6.9). Only in six species did mortality decrease as N deposition increased. In a gradient study in Switzerland (N deposition range 8-47 kg N ha^{-1} yr^{-1}), N deposition increased the mortality of Norway spruce (*Picea abies*) by the bark beetle *Ips typographus*(Tresch et al., 2022). In a drought situation, the effect of N deposition on mortality was much stronger (Figure 9.13). Mortality also increased when K nutrition was out of balance which may itself be the result of increased N deposition (Braun et al., 2020a). An increased bark beetle attack was also observed by Eatough Jones et al. (2004) in pine stands in California in response to elevated N deposition. The incidence of bark beetle activity on pines at the site with low ambient N throughfall deposition (18 kg N ha^{-1} yr^{-1}) was 20% higher in stands receiving N additions of 50 and 150 kg N ha^{-1} yr^{-1} as slow release urea-formaldehyde. Bark beetle activity at the site with high atmospheric N input (94 kg N ha^{-1} yr^{-1}) was generally high, with 30 to 57% of trees affected, regardless of the amount of additional N. Between 1999 and 2003, severe drought throughout the region was a major factor in decreased tree resistance. It was found that both ozone exposure and N deposition further increased pine susceptibility to beetle attacks.

Figure 9.13. Mortality of Norway spruce in Switzerland in relation to drought (quantified as ratio between actual and potential evapotranspiration averaged over the season, average lagged effect over three previous years), with the interaction effect of N deposition. Number of dead trees: 1132, 76 sites, 131,819 tree observations for 36 years, gradient of total N deposition 8.5-81 kg N ha^{-1} yr^{-1}. Change point regression yields critical load estimate of 10.1 ± 3.5 (SD) kg N ha^{-1} yr^{-1}.



Source: Tresch et al., 2022

N addition of 10 kg N ha^{-1} yr^{-1} (background deposition of 15-20 kg N ha^{-1} yr^{-1}) sufficed to increase an attack on *Fagus sylvatica* by two pathogenic fungi and change the phenolic concentration in the fine roots (background deposition of 14 kg N ha^{-1} yr^{-1}).

Summary tree physiology

Given the importance of drought interactions on beech stem increments (Figure 9.11) and on Norway spruce mortality (Figure 9.13), a $CL_{emp}N$ range of 10 to 15 kg N ha^{-1} yr^{-1} is recommended, based on 'expert judgement'. However, to date, not enough data are available to permit differentiation between forest types.

9.3.5 Effects of N deposition on the biodiversity of temperate forests

Effects on soil microbiota

It is well known that N deposition and N fertilisation (Treseder, 2004; Wallenda and Kottke, 1998) can influence the growth of soil microbiota. Mycorrhizal fungi are still the most studied group of soil organisms in relation to N deposition because of the close symbiosis between many tree species and their fungal partners. All the same, research on rhizosphere bacteria, which are also essential for plant health and growth, has gained considerably in importance.

Early studies focused on fungal fruit body formation aboveground (Treseder, 2008; Van Strien et al., 2018) and still offer opportunities for long-term evaluation (Andrew et al., 2018). These fungi include species with both types of nutritional modes, saprophytes and mycorrhiza. However, belowground mycorrhizal communities are most relevant for the trees, as the external mycelium's surface area increase is essential to the trees' uptake of nutrients and water, and provides protection against root pathogens. As molecular approaches became affordable and methods to study bacterial and fungal communities in soil have improved, there have been reports of more and more belowground changes in biomass, community composition and species richness with increasing N availability (Lilleskov et al., 2018; Morrison et al., 2016; Ochoa-Hueso, 2016). Most recently, partial community recovery was also observed after reducing deposition or halting fertilisation experiments for conifer and birch forests in the Netherlands (Van Strien et al., 2018) and at a boreal conifer forest site in Sweden (Choma et al., 2017).

A very important finding throughout these studies is that responses are species-specific in both fungal and bacterial communities and that this can have an important impact on the magnitude of effects. For example, saprophytic and mycorrhizal fungi might show contrasting responses as their nutritional mode differs and mycorrhizal fungi are more involved in the nutrient cycling of trees (Gillet et al., 2010; Maaroufi et al., 2019; Morrison et al., 2016)

Effects on aboveground fungal fruit-body formation

Data from long-term N-deposition studies on the formation of fruit bodies indicate that there are prominent effects discernible aboveground. Fruit-body formation in 'generalist' species, forming a symbiosis with a wide range of tree species, seems to be less affected by increased N availability than in 'specialist' species. *Laccaria, Paxillus, Theleophora, Scleroderma* and *Lactarius* are examples of the less sensitive group, whereas *Tricholoma, Cortinarius* and *Suillus* have been found to be more sensitive (Arnolds, 1991). Fruit-body formation increased in *Paxillus involutus*, *Lactarius rufus* and *Laccaria bicolor* after N fertilisation with up to 240 kg N ha^{-1} yr^{-1} (Hora, 1959; Laiho, 1970; Ohenoja, 1988). The easily cultivated species that are used in culture experiments are adapted to higher N concentrations. This makes it difficult to derive a critical load from this type of experiment (Wallenda and Kottke, 1998).

There are numerous reports of decreases in species diversity and an abundance of mycorrhizal fungi in forests based on long-term monitoring and gradient studies of fruit-body formation. In the Netherlands, the average number of ectomycorrhizal species declined significantly, from 71 to 38, between 1912 and 1954 and between 1973 and 1983, while wood-colonising saprophytic and parasitic fungi increased from 38 to 50 (Arnolds, 1985, 1991). Similar observations were made by Rücker and Peer (1988) in forests in the Salzburg region (Austria). Data collected in 1937 showed 110 and in 1987 48 species of ectomycorrhizal fungi. The number of wood-colonising saprophytic and parasitic species increased from 17 to 19. Grosse-Branckmann and Grosse-Branckmann (1978) compared the occurrence of sporocarps in the Darmstadt area of Germany between 1970 and 1976 with data collected between 1918 and 1942. From the 236 species that were encountered during the first period, only 137 were still found to be there in the second period, corresponding to a loss of 99 species, including many ectomycorrhizal fungi. Termorshuizen and Schaffers (1987) found a negative correlation between the total N input in mature *Pinus sylvestris* stands in the Netherlands and the abundance of fruit bodies of ectomycorrhizal fungi (EMF). Schlechte (1986) compared two *Picea abies* sites in the Göttingen area in Germany. He found a negative relationship between N deposition and ectomycorrhizal species. At the site with N depositions of 23 kg N ha^{-1} yr^{-1}, 85 basidiomycetes were found, including 21 ectomycorrhiza (25%), while at the site with 42 kg N ha^{-1}yr^{-1}, 55 basidiomycetes were recorded, including 3 ectomycorrhiza (5%).

Effects on belowground fungal communities

Many studies on the belowground effects of N deposition focus on the formation of mycorrhiza on root tips (Suz et al., 2014). Others aim to investigate the production and distribution of mycorrhizal mycelium in the soil (e.g. Bahr et al., 2013). The former type of studies cannot easily draw conclusions about the actual fungal growth or biomass production because many mycorrhiza are expected to outlast adverse conditions as persistent tissue on root tips (fungal mantle). Both types of studies are usually based on measurements in the upper soil horizons (usually 10-15 cm). However, root tip colonisation and fungal mycelium growth can reach deep soil horizons (Dickie et al., 2002; Preusser et al., 2017; Rosling et al., 2004). Consequently, the effects of N deposition on fungal growth and microbial biomass production in deeper soils may be underestimated. Studies on rhizosphere fungi and bacteria became popular over the last ten years and first results indicate that these microbiotas may also be affected by N availability. Soil microbial communities have been studied in experiments and in gradient studies. The major differences observed in sensitivities between fungal species inhibited or promoted by N deposition may be caused by differences in their enzymatic capability to acquire N directly from complex soil organic compounds. Indeed, fungi that use organic N tended to be negative indicators of N deposition, and fungi that use inorganic N tended to be positive (Van der Linde et al., 2018).

Experimental N addition

N experiments also showed reduced fruit-body production of mycorrhizal fungi. Termorshuizen (1990) applied 30 and 60 kg N ha^{-1} yr^{-1} over a three-year period to young *Pinus sylvestris* stands, in the form of $(NH_4)_2SO_4$ and NH_4NO_3. In general, fruit-body production was more negatively influenced by the higher ammonium treatment than by the ammonium-nitrate mixture. In N addition experiments in both pine and hardwood stands of the Harvard Forest Long Term Ecological Research Program (US), fertilisation with 50 and 150 kg N ha^{-1} yr^{-1} on top of 8-15 kg N ha^{-1} yr^{-1}, background deposition reduced species diversity and changed the species composition of mycorrhizal fungi, while ascomycetes and saprotrophs responded positively to N enrichment (Frey et al., 2004; Morrison et al., 2016). Avis et al. (2008) observed a decrease of approximately 20% in ectomycorrhizal fungal richness, with only a three-fold increase in experimental N deposition in two North American oak forests. The amount of N applied (in the form of KNO_3 and $(NH_4)_2SO_4$) was 21 kg N ha^{-1} yr^{-1}, the ambient deposition approximately 7 kg N ha^{-1} yr^{-1}.

Arbuscular mycorrhiza (AMF) play a less important role in forest ecosystems, but effects on this type of mycorrhiza have also been described. Phospholipid analysis of fine roots identified a significant decline in AMF fungi in 2 out of 4 *Acer species* stands after 12 years of additions of 12 kg N ha^{-1} yr^{-1} (background deposition of 4.8-8.3 kg N ha^{-1} yr^{-1}) (Van Diepen et al., 2007, 2011).

Gradient studies

In a gradient study from less than 1 to up to 18 kg N ha^{-1} yr^{-1} (bulk deposition) near an industrial ammonia production facility in Alaska, which had operated for almost 30 years, sporocarps of 14 mycorrhizal fungi species were found at plots with the highest N loads, compared to 144 mycorrhizal species at the six plots with the lowest N loads (Lilleskov et al., 2001). The authors hypothesised that N-efficient species, which prevail under N-limiting conditions, were replaced by species that best functioned in nutrient-rich soils and, subsequently, by P-efficient species under high N conditions. However, no information was provided about the importance of dry (and therefore total) deposition at the investigated site. It is, therefore, difficult to use these data to derive a critical load.

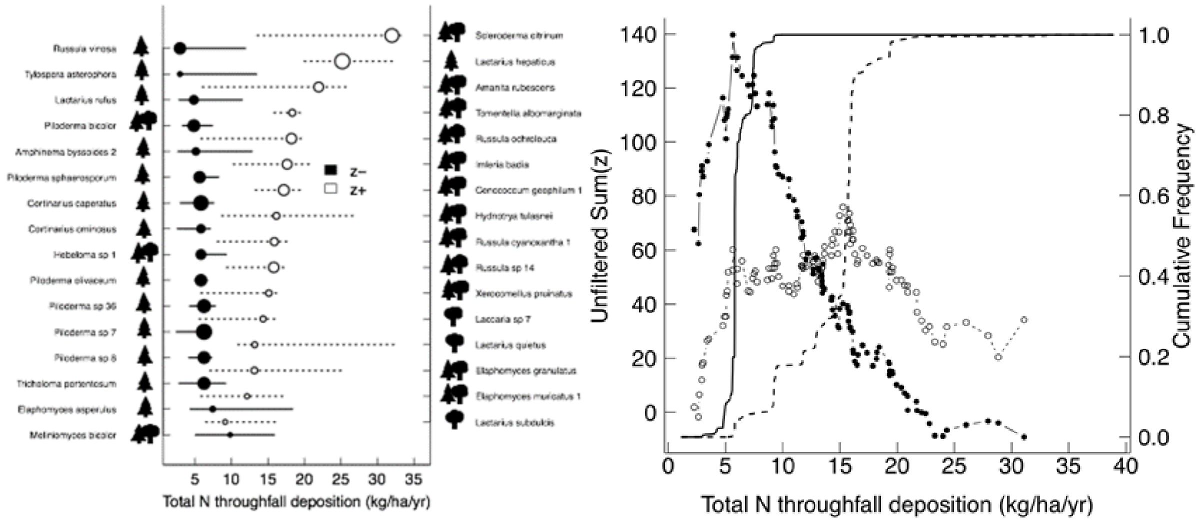
Erland and Taylor (2001) used a gradient from low deposition in northern Europe (< 2 kg N ha^{-1} yr^{-1}) to higher deposition in southern Europe (25 kg N ha^{-1} yr^{-1}). They did not find any apparent negative effects on ectomycorrhizal fungal diversity in beech forests. However, ectomycorrhizal root tips in spruce forests appeared to be more sensitive to high levels of N deposition. The diversity in root morphotypes decreased with increasing deposition from north to south. Interestingly, the proportion of species that could take up organic N declined as mineral N availability increased. In two similar 60-year old *Picea abies* forests in south Sweden, with different rates of N deposition (Vedby, 14-15 kg N ha^{-1} yr^{-1} and Skrylle, 24-29 kg N ha^{-1} yr^{-1}), the level of mycorrhizal colonisation was almost 100%. However, the total number of mycorrhizal species was 30 to 42% higher at the site with low N deposition. The total number of mycorrhizal roots and the number of mycorrhizal morphotypes were also significantly lower at Skrylle than at Vedby.

In temperate oak forests on a broad N deposition gradient in Central Europe (5 to 36 kg N ha^{-1} yr^{-1}), variable sensitivities were detected for ectomycorrhizal fungi (EMF) species belowground but species with large mycelial networks tended to be more sensitive (Suz et al., 2014). This study also found a moderate (9.5-13.5 kg N ha^{-1} yr^{-1}) and drastic (17 kg N ha^{-1} yr^{-1}) effect of N throughfall deposition on EMF community composition. In a gradient study in oak-dominated deciduous forests in southern Sweden, a trend was observed towards reduced ectomycorrhizal mycelial growth at higher N depositions (20 kg N ha^{-1} yr^{-1}), compared to depositions of 10 kg N ha^{-1} yr^{-1} (Nilsson et al., 2007). In a gradient study on forests dominated by European beech across Switzerland with N deposition levels between 16 and 33 kg N ha^{-1} yr^{-1}, fungal mycelial growth and species diversity were found to be reduced along the gradient. Most indicator taxa that decreased in abundance as N deposition increased showed response thresholds between 22 and 23 kg N ha^{-1} yr^{-1} (De Witte et al., 2017).

In a European scale gradient study covering Mediterranean and northern ecosystems too, threshold indicator taxa analyses were carried out to identify distinct thresholds of ectomycorrhizal responses to key environmental variables (Van der Linde et al., 2018). A threshold was detected for fungi in coniferous forests negatively affected by N throughfall deposition at 5.8 kg N ha^{-1} yr^{-1} and in deciduous forests at 9.2 (Figure 9.14). Based on the relationship between throughfall and total deposition shown in Figure 9.1, a critical load derived from this number was estimated of 10.6 kg N ha^{-1} yr^{-1} for conifer forests and of 13.7 kg N ha^{-1} yr^{-1} for deciduous forests. Fungal species identified as indicator taxa were mainly conifer specialists which thrive in poor soils and at pre-industrial N levels (approximately < 2 kg N ha^{-1} yr^{-1}). Potentially, however, they are unable to adapt to currently far higher N deposition levels. Positively affected fungi have a much broader response range. A less defined peak at 15.5 kg N ha^{-1} yr^{-1} for these fungi suggests that adaptation by positively affected fungi to increased N deposition varies greatly. Smaller-scale studies in Norway spruce forests in Sweden confirm a low N throughfall deposition threshold around 5-6 kg N ha^{-1} yr^{-1} (Bahr et al., 2013; Cox et al., 2010a). In the UK, Jarvis et al., (2013) concluded a $CL_{emp}N$ of 5-10 kg N ha^{-1} yr^{-1} for Scots pine, using a gradient of total N deposition from 3.1-9.9 kg N ha^{-1} yr^{-1}.

A recent review by Lilleskov et al. (2019) concluded that EMF communities respond much more strongly to N availability than AMF communities with the disappearance of a key taxa. However, estimates of critical loads for AMF in forests are still sparse. And within EMF communities, it is the conifer forest communities that are more sensitive to N deposition than broadleaf-associated EMF (Cox et al., 2010b; Van der Linde et al., 2018). Responses of saprophytic fungi to changing N availability seem to be less pronounced (e.g. Arnolds, 1991; Gillet et al., 2010; Morrison et al., 2016) but need further confirmation. Congruent species-specific changes for ectomycorrhizal fungi in response to N availability were found for *Cenococcum geophilum* (decrease; De Witte et al., 2017; Morrison et al., 2016); *Boletus, Tricholoma, Cortinarius and Piloderma* species (decrease : De Witte et al., 2017; Morrison et al., 2016; Suz et al., 2014; Van der Linde et al., 2018); and *Russula ochroleuca, Tylospora asterophora, Elaphomyces* and *Laccaria* species (increase; De Witte et al., 2017; Morrison et al., 2016; Van der Linde et al., 2018).

Figure 9.14. ECM threshold indicator taxa analysis results reproduced from Van der Linde et al. (2018). Left) Individual OTU abundances in response to N throughfall deposition. Black symbols show taxa declining with increasing N deposition (z−), open symbols depict increasing taxa (z+). Symbol size is proportional to magnitude of response (z-score). Horizontal lines represent the 5th and 95th quantiles of values resulting in the largest change in taxon z-scores among 1,000 bootstrap replicates. Tree shapes indicate host generalist, conifer-specific or broadleaf-specific. Right) Community-level output of accumulated z-scores per plot is shown in response to N deposition.



Source: Van der Linde et al., 2018

After correction from throughfall to total N deposition, the European-wide results from Van der Linde et al. (2018) suggested a $CL_{emp}N$ of 11 kg N ha^{-1} yr^{-1} for coniferous and of 14 kg N ha^{-1} yr^{-1} for deciduous forests, while the study of Jarvis et al. (2013) from UK proposed a $CL_{emp}N$ of 5-10 kg N ha^{-1} yr^{-1} for Scots pine.

Critical loads for soil microbiota

Mainly based on the gradient study by Van der Linde et al. (2018) in temperate ecosystems, it is concluded that conifer ectomycorrhizal fungal communities are more sensitive than broadleaf ectomycorrhizal fungal communities. Total deposition was estimated from throughfall deposition using the relation presented in Figure 9.1. For conifer forests, a $CL_{emp}N$ of 10-13 kg N ha^{-1} yr^{-1} and for broadleaf forests a $CL_{emp}N$ of 10-15 kg N ha^{-1} yr^{-1} are proposed. In semiarid Mediterranean ecosystems, current $CL_{emp}N$ are estimated at 5-10 kg N ha^{-1} yr^{-1}.

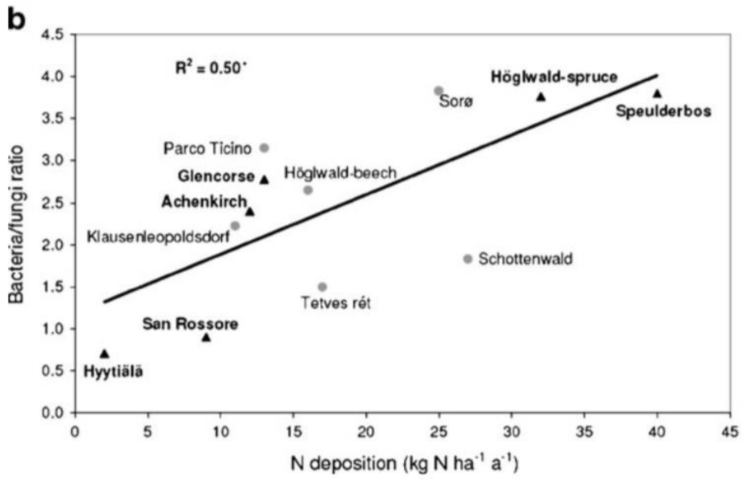
Effects on soil microbial communities

According to a review of N effects on soil bacteria by Lladó et al. (2017) and references therein, biomass and activity of soil microbial communities in many different soil environments, including temperate and boreal forests, changed as N deposition increased. Changes in abundance, however, seemed to depend on the level of background N deposition and N addition.

Gradient studies

In a N gradient study across Europe and different forest types from northern boreal, temperate and Mediterranean ecosystems, Zechmeister-Boltenstern et al. (2011) observed, in addition to a decrease in fungal species richness and changes in ratios of AM to EM fungi, an increase in the bacteria to fungi ratio especially in Central European forest stands with N deposition between 30 and 40 kg N ha^{-1} yr^{-1} (Figure 9.15).

Figure 9.15. Ratio of bacteria to fungi on a wide N gradient across Europe.



Source: Zechmeister-Boltenstern et al., 2011

Experimental N addition

Frey et al. (2014), for example, observed a shift to greater dominance by bacteria in the N amended plots and a parallel reduction of fungal abundance after 25 years of N addition (50 kg N ha^{-1} yr^{-1}) in mixed temperate forest stands in northern America with 8 kg N ha^{-1} yr^{-1} background deposition.

After 20 years of N addition (40 kg N ha^{-1} yr^{-1}) in boreal conifer forests in central Sweden, soil microbial biomass increased, while the fungi to bacteria ratio did not change (Tahovská et al., 2020). N in open field precipitation at this site was 9 kg N ha^{-1} yr^{-1}. Similarly, Choma et al. (2020) reported that the addition of 50 kg N ha^{-1} yr^{-1} had no significant effect on fungal and bacterial responses after four years. Background deposition at the experimental site was 3.2 N ha^{-1} yr^{-1}. After 20 years of N addition (30 kg N ha^{-1} yr^{-1}) in maple forests in Michigan, comparisons of soil bacterial and fungal communities did not reveal any changes in abundance or species richness in these communities. However, the enzyme activity profiles did indicate negative responses to N addition (Hesse et al., 2015). The background throughfall deposition at the two experimental sites was 7.1 and 6.4 kg N ha^{-1} yr^{-1}, respectively. These studies in temperate and boreal forests suggested that, while fungal communities respond to N addition far below 30 kg N ha^{-1} yr^{-1}, bacterial communities seemed to respond only to higher-level N inputs. However, in a 16-year N addition experiment (12.5 and 50 kg N ha^{-1} yr^{-1}) in boreal conifer forests in northern Sweden (background deposition 2 kg N ha^{-1} yr^{-1}), Maaroufi et al. (2015) found that total fungal and microbial biomass decreased already at the lower-level N input. This was in line with a study on an N deposition gradient in temperate forests in England (Thorpe, 2011). In this case, N deposition of > 15 kg N ha^{-1} yr^{-1} (versus 8-12 kg N ha^{-1} yr^{-1}) increased the soil nitrification rate while the diversity of soil microbiota was reduced. In a broadleaf forest in Belgium (background deposition of 25.3 kg N ha^{-1} yr^{-1}), that combined increased light, N (10 kg N ha^{-1} yr^{-1}) and warming treatments (Ma et al., 2018), N addition and warming did not significantly affect the soil microbial biomass and plant community composition. Warming did, however, significantly alter the composition of the soil bacterial community. Yet, the number of unique operational taxonomic units of plants was higher in plots with N addition, and there were significant interactive effects of light and N addition.

Critical loads for soil microbial communities

Soil bacteria show a potential response to N deposition, but it depends heavily on other factors. Based on the results presented, it is not possible to propose a critical load for this receptor.

Effect on ground-living and epiphytic lichens and algae

Since the end of the 18th century, epiphytic or tree-bark-inhabiting lichens have been used in air pollution mapping studies. In the Netherlands, the forest vegetation of one Scots pine stand in the central part of the country, with a deposition of around 20 kg N ha^{-1} yr^{-1}, was investigated in 1958, and then re-investigated in 1981 when the deposition was around 40 kg N ha^{1} yr^{1}. In the intervening period, all lichens had disappeared (Dirkse and Van Dobben, 1989). This could also have been a sulphur (S) or acidity effect, but results from fertilisation experiments in northern Sweden with low background depositions of both N and S showed that all ground-living *Cladina*-species had disappeared after 28 years of N additions (34 kg N ha^{-1} yr^{-1}). They were, however present on the unfertilised control plots (background deposition 2 kg N ha^{-1} yr^{-1}) (Strengbom et al., 2001). This is an indication that N deposition may be partly responsible for observed reductions in the abundance of ground-living lichens, although other factors such as changes in forestry practices must also be considered (Sandström et al., 2016). A reduction of air pollution does not guarantee the recovery of lichen communities. Many epiphytic lichen species have a limited ability to colonise new sites (Kiebacher et al., 2017). In previously polluted areas, the presence of sensitive species can depend as much on their dispersal abilities as on current air quality (Hawksworth and McManus, 1989). While much of the research into recovery has focused on acid-sensitive species, the same problem of recolonisation applies where N sensitive species have become locally extinct.

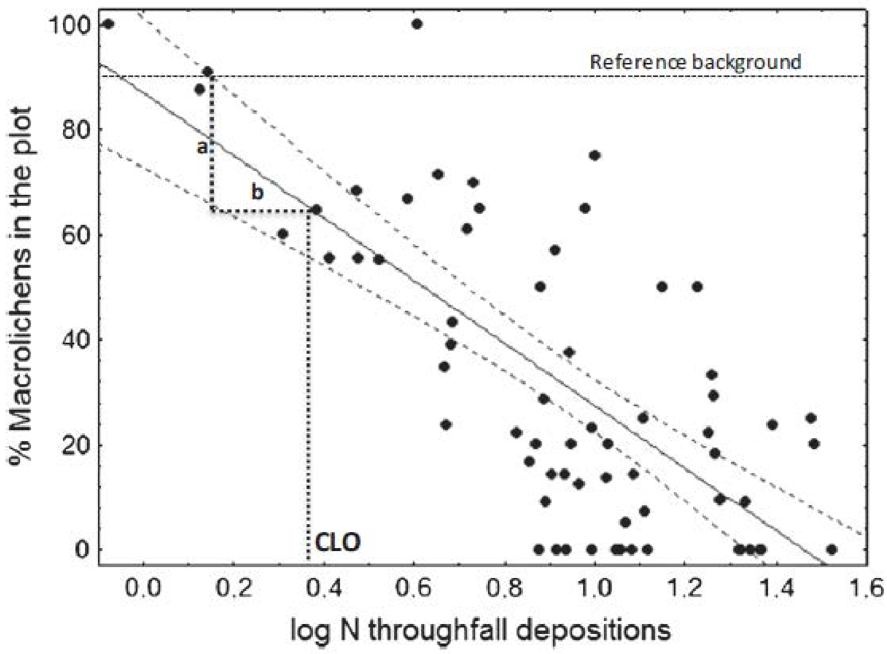
Around 10 percent of all lichen species in the world have blue-green algae (cyanobacteria) as the photobiont (Insarova et al., 1992). These blue-green algae lichens are negatively affected by both acidity and N deposition. In an international survey that stretched from the Netherlands via Denmark to Sweden, the decline in lichens with blue-green algae was found to correlate significantly with N deposition rates of over 5 to 10 kg N ha^{-1} yr^{-1} (Göransson, 1990). Instead of being caused by N deposition, the negative effects on ground-living lichens may have been an indirect effect of competition with N-favouring vascular plants (Cornelissen et al., 2001). Free-living green algae, especially of the genus *Pleurococcus* (syn. *Protococcus*, *Desmococcus*), are strongly stimulated by enhanced N deposition. They cover outdoor surfaces which are not subject to frequent desiccation in regions with high N deposition, above approximately 10 kg N ha^{-1} yr^{-1} (Bobbink et al., 1996).

Epiphytic lichens, such as *Lobaria pulmonaria* and *Lobaria amplissima,* have been shown to be effective indicators of N pollution, for instance in Atlantic oak woods in Scotland and north-western England. In a comparison of sites with low (9.6-17.6 kg N ha^{-1} yr^{-1}) and high N depositions (11.2-53 kg N ha^{-1} yr^{-1}), these epiphytes were only found at sites with low N (Mitchell et al., 2003). Earlier data on epiphytic lichens also suggested a $CL_{emp}N$ range of 5 to 10 kg N ha^{-1} yr^{-1}, which was considered 'reliable'. Although most of these data referred to boreal forests, results from Bobbink et al. (1996) confirmed the range.

In a recent study, Geiser et al. (2019) assessed ecological risks from atmospheric N and S deposition in TF to the community composition of epiphytic macrolichens in US forests. They used 20%, 50%, and 80% declines in selected biodiversity responses as cut-offs for low, moderate and high ecological risk from total deposition of N or S. The low ecological risk (20% decline) critical loads for total species richness, sensitive species (oligotroph) richness, forage lichen abundance and cyanolichen abundance were 3.5, 3.1, 1.9 and 1.3 kg N ha^{-1} yr^{-1}, respectively. High ecological risk (80% decline), excluding total species richness, occurred at 14.8, 10.4 and 6.6 kg N ha^{-1} yr^{-1}. The 'low-risk' critical loads proposed by Geiser et al. (2019) are supported by the fact that N deposition ranges from 1-4 kg N ha^{-1} yr^{-1} in many ecoregions in the USA (Pardo et al., 2011).

The critical load of 2.4 kg N ha^{-1} yr^{-1} (based on N deposition in throughfall) by Giordani et al. (2014) was linked to a significant decrease in the percentage of oligotrophic macrolichens, (Figure 9.16). An N dose of 4 kg N ha^{-1} yr^{-1} in TF was related to an approximate decrease > 90-50% in microlichens. The results were consistent when considering all forest types and for coniferous forests only. According to Figure 9.1, total N deposition for these two throughfall deposition estimates is 8.0 and 9.4 kg N ha^{-1} yr^{-1}, respectively. The analysed dataset of 286 epiphytic lichen species was collected from 83 plots of the ForestBIOTA project (Forest Biodiversity Test Phase Assessments) covering the whole of Europe from the Mediterranean to Finnish Lapland.

Figure 9.16. Critical loads of N deposition on European ICP Forest plots considering the % macrolichens. Plots with the lowest depositions for which the response of % macrolichens was significantly different in the model was set as the background reference (horizontal dotted line). Their lower 95% confidence limit is calculated (line a) and the point corresponding to where this is met on the regression line is identified (line b). The x coordinate (logN deposition) of this latter represents the critical load.



Source: Giordani et al., 2014

Critical load for lichens and algae

Given the effect of canopy retention of inorganic N in TF, the results of Giordani et al. (2014) from Europe and those of Geiser et al. (2019) from the USA support a $CL_{emp}N$ range of 3-5 kg N ha^{-1} yr^{-1}.

Effect on forest field-layer vegetation

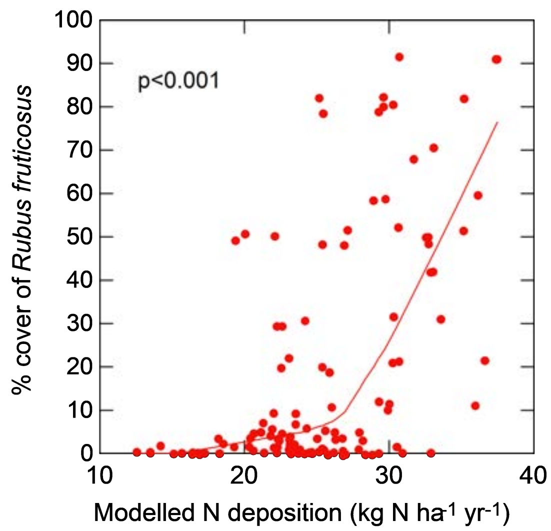
Introduction

Since the last report in 2011, more case studies have examined the impacts of N deposition on forest understorey species (or the forest field-layer vegetation; defined here as: herbaceous and woody species, typically less than 1 to 2 m in height, including tree and shrub seedlings). They looked at time series, gradient studies and, less frequently, fertilisation experiments. Unfortunately, the experiments tended to be relatively short-term and often used higher levels of N addition than are found in the field, with designs hampering extrapolation of results to a regional (or global) level (Hettelingh et al., 2015). Case studies have typically shown increases in the abundance of nitrophilous species. Many of the local to regional studies were recently supplemented by synthetic analyses that consolidated datasets across temperate deciduous European forests and allowed spatio-temporal analyses across scales (e.g. Bernhardt-Römermann et al., 2015). Alternatively, they used innovative experimental platforms (e.g. Blondeel et al., 2020). Synthetic analyses also used ICP Forests and ICP Integrated Monitoring data, as well as independent initiatives, for instance, forestREplot (www.forestreplot.ugent.be) or LTER Europe (www.lter-europe.net/lter-europe). They covered a broader set of temperate forest types. Using TITAN analysis in a gradient study with total N deposition in a 5x5 km grid, Wilkins et al. (2016) found community change points in Atlantic oak woodland of 15.3 kg N ha^{-1} yr^{-1}, and Wilkins and Aherne (2016) in sessile oak woodland of 8.8 kg N ha^{-1} yr^{-1}. We used these findings to suggest that the $CL_{emp}N$ for temperate forest field-layer vegetation in Europe remained in the range 10-15 kg N ha^{-1} yr^{-1}. We emphasise that synthetic analyses have brought new insight into the context dependency of critical loads. It has always been important to stress this context dependency in critical load assessments. It means that forest understorey vegetation can be more or less sensitive to instantaneous rates of N deposition, depending on the underlying abiotic environmental conditions, the overstorey composition, current and prior management actions and previous atmospheric deposition (and therefore the cumulative amounts) (Perring et al., 2018b). Deposition history is also important: presurvey levels of N deposition determined subsequent diversity changes of vegetation. Environmental conditions such as light availability and density of large herbivores were also important. These different elements of context-dependency at specific locations are the reason for the range of the suggested $CL_{emp}N$ for ground vegetation in temperate European forests. There also needs to be enhanced awareness of how field-layer vegetation changes in response to N relate to alterations in other aspects of the forest ecosystem, also mediated by N deposition. This includes competition, herbivory, mycorrhizal infection, disease and species invasions (Gilliam, 2006).

From individual case studies to synthetic analyses

Previously, a large number of observations showed an increase in the abundance of nitrophilous species in forests, with N deposition typically ranging (historically) from 15 to more than 40 kg N ha^{-1} yr^{-1}. These species include *Galeopsis tetrahit*, *Rubus idaeus*, *Rubus fruticosum*, *Deschampsia flexuosa*, *Calamagrostis epigejos, Prunus serotina*, *Poa trivialis, Milium effusum, Molinia caerulea, Urtica dioica, Epilobium angustifolium, Frangula alnus, Arrhenaterum elatius, Impatiens parviflora, Galium aparine, Aegopodium podagraria, Sambucus* species, *Stellaria media, Stellaria holostea, Stellaria nemorum, Dryopteris filix mas, Dryopteris dilatata* and *Dryopteris cathusiana.* Such changes, with subsets of the species mentioned above, have been recorded in Dutch forests (Dirkse, 1993; Dirkse and Van Dobben, 1989), in German mixed fir and spruce forests (Kraft et al., 2000; Rodenkirchen, 1992), in young moraine forests in Germany (also with a comparison to N deposition experiments) (Hofmann, 1987; Hofmann et al., 1990; Anders et al., 2002;; Jenssen and Hofmann, 2005; Jenssen, 2009), in beech, oak and hornbeam forests in north east France (Bost, 1991), in 17 out of 18 forest sites in two regions in Switzerland (Kuhn et al., 1987), in the central Swiss plateau (Walther and Grundmann, 2001), in mixed deciduous Belgian forests (Lameire et al., 2000), in Atlantic old sessile oak woodlands (Wilkins and Aherne, 2016), and in woodland shelter belts of differing overstorey composition, along N deposition gradients from livestock farms in the UK (Pitcairn et al., 1998). As an example of an individual species, in Switzerland the cover of *Rubus fruticosus* agg. increased markedly in forest plots with a modelled N deposition rate of > 25 kg N ha^{-1} yr^{-1} (Flückiger and Braun, 2004, Figure 9.17). According to its Ellenberg N value, *Rubus fruticosus* would not be classified as a nitrophilous plant, but its shoot development seems to be highly stimulated by N. Gilliam et al. (2016) observed a significant N-mediated increase in *Rubus* spp. after the long-term application of 35 kg N $ha^{-1}yr^{-1}$ in the temperate forests of Fernow Experimental Forest (FEF), West Virginia. Interestingly, the degree of canopy openness determines whether light will be a limiting factor once N availability increases through deposition. Species such as *Oxalis acetosella* and *Mercurialis perennis* profit from high nutrient levels under closed canopies while open forest species such as *Melampyrum pratense* and *Tanacetum corymbosum* disappear (Heinrichs and Schmidt, 2017). Under open-forest conditions, the growth of light-demanding species such as *Anemone nemorosa* increases markedly at the expense of species such as *Primula elatior* and *Viola reichenbachiana* (Bernhardt-Römermann et al., 2010; Jantsch et al., 2013).

Figure 9.17. Percentage of cover by *Rubus fruticosus* agg. in Swiss forest observation plots, in relation to modelled N deposition.

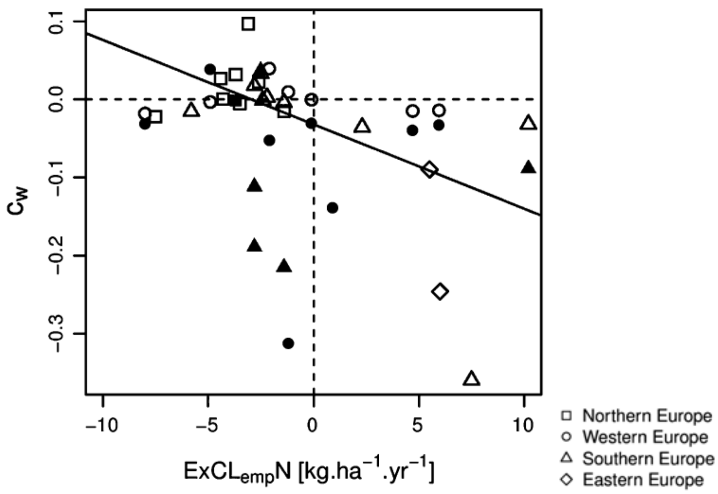


Source: Flückiger and Braun, 2004

By way of further examples, large-scale changes in vegetation have been observed in Scots pine forests in north-eastern Germany since the 1970s. N deposition of between 10 and 15 kg N ha^{-1} yr^{-1} (bulk deposition) over several decades led to N accumulation in oligotrophic Scots pine forests. This induced a shift in vegetation types over large areas. Mainly Scots pine forest types that were dominated by lichens, heather and bryophytes (C:N ratio 30-35) disappeared almost completely and were replaced by common forest types dominated by grasses (*Deschampsia flexuosa, Festuca ovina*). The N deposition gradient study by Wilkins and Aherne (2016), which involved applying TITAN to infer plant species changes in Atlantic oak woodland, resulted in a $CL_{emp}N$ of 13.2 kg N ha^{-1} y^{-1}.

In parallel to these findings of taxonomic shifts, the average N indicator number (according to Ellenberg, 1988) had been shown to rise using ICP Forests data (Van Dobben and De Vries, 2017). However, the authors also noted that the N deposition effect could not be clearly delimited from the overall signal (see also Verheyen et al., 2012). In contrast, the abundance of species with low N indicator values – comprising many endangered species (Ellenberg, 1985) – had been declining significantly in European forests with N deposition above the currently defined $CL_{emp}N$ (Dirnböck et al., 2014) (Figure 9.18). Combined taxonomic and indicator analyses from resurveys in north-east France confirmed a community increase in N indicator values, an increase in N demanding species, and an increased nutrient status as indicated by trophic level analyses (Thimonier et al., 1992). However, the clear attribution of vegetation changes to N deposition was not always straightforward, as seen for similar forests in north-east France (Thimonier et al., 1994). In Austria, N deposition effects on the forest floor vegetation of temperate spruce-fir-beech and pure Norway spruce forests were studied at the ICP Integrated Monitoring site Zöbelboden (northern Limestone Alps). Annual throughfall deposition of between 11 and 19 kg N ha^{-1} yr^{-1} (1994-2013) did cause homogenisation in the plant composition across the entire 90 ha study area (Hülber et al., 2008), as predicted by the N homogeneity hypothesis (Gilliam, 2006). However, wind and bark beetle disturbances hid any further eutrophication signal after 2005 (Helm et al., 2017).

Figure 9.18. Relationships between weighted averaged changes (cw) in the cover of nitrotrophic (Ellenberg N value 7-9: filled symbols) and oligotrophic plant species (Ellenberg N value 1-3, open symbols) and critical load exceedance (ExCLempN). Significant linear regression (P < 0.05, t-test) was found for oligotrophic species.

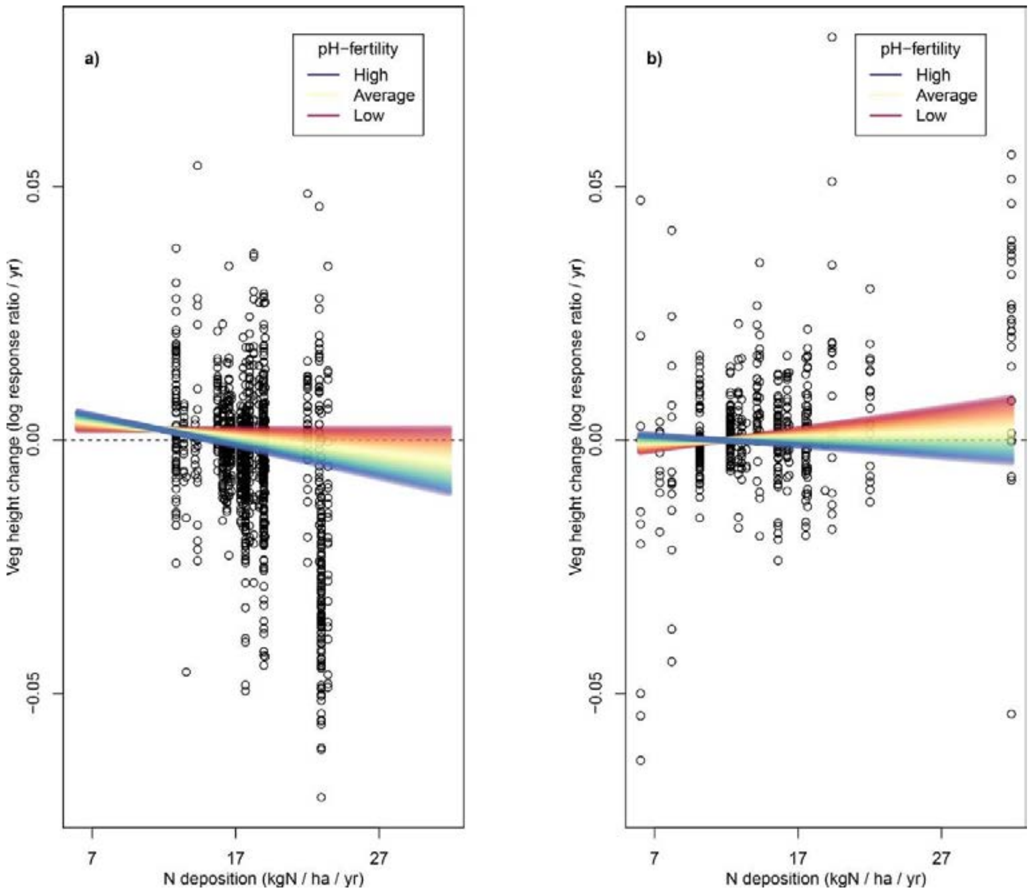


Source: Dirnböck et al., 2014

It is difficult to ascribe understorey vegetation changes to specific driving factors, specifically N (in the case of deriving critical loads), in single case or gradient studies with co-occurring environmental changes. This led to the awareness that combining studies from across sites and regions, coupled with careful analyses that account for study characteristics, can advance understanding within global change research (Verheyen et al., 2017). Such combination studies revealed the critical importance of overstorey dynamics in explaining an apparent eutrophication shift in European deciduous forests (Verheyen et al., 2012). Indeed, cumulative N deposition by the time of an initial survey had been shown to explain subsequent diversity changes that were related more to temporal changes in light availability and large herbivore density than to instantaneous N deposition rates (Bernhardt-Römermann et al., 2015). However, N deposition was shown to lead to declining herb cover in an observation resurvey dataset of forest understorey vegetation across Europe, but only in forests that have a land use history of continuous forest cover (Depauw et al., 2020). The importance of previous land use in dictating community responses to N deposition was also demonstrated in a functional trait framework for a greater number of plots, albeit with a less orthogonal design (Perring et al., 2018b). In Perring et al. (2018b) higher rates of N deposition actually led to greater species richness (and plant height) in forests managed less intensively in 1800 (i.e. as high forests), compared to decreases that were seen in forests managed more intensively through coppicing. Such results, across Europe, reinforce earlier findings from site-based studies. For instance, at forest sites with earlier litter raking in Germany, the average Ellenberg N indicator values had increased by 0.6 units while at forest sites without litter raking the increase was 0.3 units (Rodenkirchen, 1992). N deposition is the major factor driving the decline of specialised species with low distribution ranges in temperate forest understoreys (Staude et al., 2020). Hence, species compositional rather than richness changes are the predominant effects of N deposition in temperate forests.

Awareness that the impact of N deposition may be masked or may depend on other site factors, led to a review of forest understorey responses to this driving factor, and the development of a conceptual framework with the focus on critical load derivation (Perring et al., 2018b). They suggested that land and forest management, overstorey composition, climate change, forest continuity, landscape context, browsers and grazers can all affect forest understorey communities, with potential interactions with cumulative N deposition, and the forms and amounts of contemporary N deposition. They then focused on how light and soil pH value gradients influenced the community response to N deposition (while N deposition itself can influence these gradients). This was because soil pH can precondition subsequent community responses to the effects of increased N (and reduced sulphur deposition) (Smart et al., 2014; Stevens et al., 2011). In the case of light, as explained above, individual species (and plant communities) may respond to additional N in high-light environments but not when there are other resource limitations (De Frenne et al., 2015; Heinrichs and Schmidt, 2017).

Figure 9.19. **Plant community responses to N deposition between surveys across European deciduous temperate forests. Shown are predicted (a) graminoid (including sedges, grasses and rushes) and (b) forb responses to N deposition based on the best-fit model using 1814 plots across 40 forest regions. Changes in cover of these structural groups between surveys depend on the magnitude of N deposition and whether or not initial survey plots are shown to be covered by light demanding species (blue line) or shade tolerant species (red line). Black circles show the actual data.**



Source: Reproduced from Perring et al., 2018b

Using resurveys of 1814 plots from 40 forest regions across Europe, Perring et al. (2018b) confirmed the importance of context dependency in determining understorey community changes to N deposition. Plant community change, as measured by species richness, structural composition and functional traits, rarely depended on N deposition as the main effect (Perring et al., 2018b). Instead, and as shown by Figure 9.19a, cover change for graminoids is negative in response to N deposition in open environments, while it is positive in closed environments. What this means is that in sites initially characterised by high-light species assemblages (blue line), large amounts of N deposition can lead to decreased graminoid cover, while low amounts of N can lead to an increase. On the other hand, in communities initially characterised by plants with a low demand for light (red line), higher N deposition led to increased graminoid cover, and low N deposition decreased cover. In contrast, forbs (Figure 9.19b) show a quadratic response to N deposition, with different curvature depending on the light environment at the time of the initial survey. Community change, as measured by the change in community-weighted mean height, depended on N deposition, but only in a complex manner related to historical forest management (Perring et al., 2018a) and the position of sites on a pH-fertility gradient (Perring et al., 2018b).

These synthetic results, and others that show conditional vulnerability of plant communities to N deposition (Simkin et al., 2016), have implications for our understanding of $CL_{emp}N$. As well as awareness of how $CL_{emp}N$ can vary among species within communities and within species depending on environmental context (Henrys et al., 2011; Payne et al., 2013), the results described above mean that critical loads need to be set reflecting other environmental conditions, especially forest-management history, fine-resolution light availability and nutritional condition of the site. For instance, in forests with infrequent biomass removal in the past (i.e, high forests) critical loads could be less risk averse (i.e. higher) than for those forests with frequent biomass removal in the past (i.e. former coppices). $CL_{emp}N$ can be adjusted according to the species pool and fertility context. As explained in Perring et al. (2018b), fertile sites predisposed light-demanding species to decline in high forests exposed to elevated rates of N deposition. This decline was even more pronounced in former coppices. In a different context of lower fertility, shade-tolerant species increased in high forests while they held steady in coppices. Low $CL_{emp}N$ in one context are not apparent in another. Dynamic plant-soil modelling approaches to calculate $CL_{emp}N$ can explain context dependency, for instance by taking into account how historical management effects, such as litter raking, alter soil nutrient depletion and acidity. Expected future climate change can likewise be factored into these models to explore likely joint impacts together with N deposition trends (Dirnbock et al., 2017, 2018; Dirnböck et al., 2020). The main conclusions from these studies were that climate-induced tree growth enhancement may increase the competition for soil N, thereby relaxing eutrophication in the future. However, it is important to note that a significant degree of uncertainty is still associated with these kinds of models (De Vries et al., 2010).

Empirical critical loads can also be set by experiments where N is directly added, allowing the attribution of N effects, although the exact mechanism (i.e. responses due to eutrophication and/or acidification) would require careful interpretation. Until recently, there have been few experimental investigations of forest herb layer responses to N deposition, especially in temperate deciduous forests in Europe (Gilliam, 2006). However, as with observational datasets, the increase in research over the last years does allow synthetic investigation, such as meta-analyses, often incorporating responses in forests as well as other biome types. In such an analysis focusing on biomass and species richness, De Schrijver et al. (2011) showed no significant response of forest understorey vegetation to in situ instantaneous N addition. It also showed that, in general, species loss occurs faster at low levels of cumulative N input with slower species loss as cumulative N increases. These findings complement observational studies highlighting the importance of cumulative effects of N addition (e.g. Bernhardt-Römermann et al., 2015). In a more recent meta-analysis using experimental results, Midolo et al. (2019) discovered that no response metrics showed an interaction between ecosystem type and N application. Forests, therefore, exhibited similar declines as did other ecosystems regarding species richness, individual abundance, mean species abundance and geometric mean abundance with increasing N addition.

Critical load for forest field-layer vegetation

Taking into account all the new findings from predominantly gradient studies, we suggest that the $CL_{emp}N$ for temperate forest field-layer vegetation in Europe remains in the range of 10 to 15 kg N $ha^{-1}yr^{-1}$ and is 'reliable'. Since the last review of $CL_{emp}N$, substantial work has been carried out on the detailed responses of forest field-layer vegetation when considering the multitude of other drivers (land use change, management, climate change, etc.). This opens up the possibility that $CL_{emp}N$ can be adjusted according to the species pool and fertility context.

Soil fauna

The effects of increased anthropogenic N deposition on soil fauna are still an understudied area, with few long-term studies from which to identify conclusive impacts or critical loads for the vast range of forest soil fauna. Most research on the impacts of N on soil biota to date focussed on plant, fungi and microbial communities. However, the responses of organisms at higher trophic levels, such as mesofauna and earthworms, are less well understood and may be species-specific (Gan et al., 2013). Since these organisms are important for mediating key ecosystem functions such as litter decomposition and N mineralisation (Bardgett and Wardle, 2010), this is an area that merits more attention in future research. Most invertebrate and microarthropod groups generally show a negative short-term effect of N deposition on species richness and abundance, followed by varying long-term responses dependent upon the type, duration and dosage of N input (Gan et al., 2013; Lohm et al., 1977; Vilkamaa and Huhta, 1986; Xu et al., 2019).

A short-term (five years) application of N at a dose of 57 kg N ha^{-1} yr^{-1} showed little effect on the biomass of soil microarthropods from litterbags in a *Pinus sylvestris* stand at the low-background-deposition NITREX site Ysselsteyn in Sweden (Boxman et al., 1998). The dominance of Proturans increased in the fertiliser treatment. However, the abundance and species richness of *Oribatida* mites and soil mesofauna generally decreased in the N addition treatment compared with the control plot.

Conversely, repeated N deposition to forest systems appears to result in N saturation and chronic declines in both the abundance and diversity of soil invertebrates. Negative responses in *Collembola* were shown at a site in the Swiss Alps (NITREX site; background deposition of 12 kg N ha^{-1} yr^{-1}) by Xu et al. (2009) after 13 years of N addition (25 kg N ha^{-1} yr^{-1}). Total *Collembola* density and the density of *Isotomiella minor*, the most abundant species, decreased significantly in the upper soil layer (0-5 cm). In addition, the genera *Tomocerus*, *Arrhopalites*, *Sminthurus* and *Neanura* were completely absent from the N-treated plots, and the density group index of the community was negatively affected. Gan et al. (2013) observed a decline in abundance of both detritivores (*Oribatida* and *Collembola*) and predaceous mites (*Mesostigmata*) after 17 years of experimental N deposition (30 kg N ha^{-1} yr^{-1}) on four sugar maple (*Acer saccharum*) forest stands in North America. There was a shift in the community composition of *Oribatida* mites between N treatments, indicating species-specific responses to N deposition (Gan et al., 2013). In a state forest in Brittany, France (estimated background deposition 10-20 kg N ha^{-1} yr^{-1}), a single application of 100 kg N ha^{-1} yr^{-1} (in the form of NH_4NO_3) produced an effect on soil microorganisms that was still significant 23 years later (Deleporte and Tillier, 1999). There were also decreases in populations of the microarthropods *Acarina* (genera *Oribatida* and *Gamarida*), *Collembola*, *Symphyla* and *Pseudoscorpionida*. Seven years of N fertilisation (20 kg N ha^{-1} yr^{-1}) of a young beech stand (atmospheric deposition of 12 kg N ha^{-1} yr^{-1}) resulted in a significant decrease (66%) in the abundance of earthworms (Flückiger and Braun, 1999). This decline may also have been the result of soil acidification, as the pH of the upper soil layer (30 cm) decreased from 3.7 to 3.5, and earthworms are sensitive to changes in soil pH (Muys and Granval, 1997). In Sweden, a significant decrease in snails was observed over a period of 14 to 46 years in areas with N depositions of 15 to 25 kg N ha^{-1} yr^{-1}, while in areas with N depositions of 3 to 6 kg N ha^{-1} yr^{-1}, no significant changes were found (Gärdenfors et al., 1995). However, in this Swedish study, there was a sulphur deposition gradient (soil acidification) as well. Hence, no $CL_{emp}N$ could be defined on the basis of these data.

The observed short-term decreases in forest soil invertebrate populations after N fertiliser application were mostly attributed to acute ammonium toxicity and/or the 'salt effect' of increased osmotic potential in soil solutions (Lohm et al., 1977). Ammonium is very toxic to microarthropods, and hydrophilous soil organisms such as nematodes and earthworms are most likely to be negatively impacted by such osmotic stress and changes in forest soil pH (Lohm et al., 1977; Muys and Granval, 1997). Subsequent recoveries and increases in soil fauna populations after singular fertiliser applications were attributed to increases in the populations of the microorganisms upon which they feed, and increased litter input by the fertilised vegetation (Gan et al., 2013; Lohm et al., 1977). In contrast, the chronic declines in soil fauna populations observed after long-term N deposition are linked to decreasing pH and changes in the soil microbial community, particularly a reduction in soil fungi (Berg and Verhoef, 1998; Xu et al., 2009). Many species of *Collembola* and *Acarina* are fungivores and will thus experience population decline due to loss of food, which then leads to a population reduction in their predators, such as *Mesotigmata* mites (Gan et al., 2013; Xu et al., 2009). N deposition-induced changes in microarthropod community composition and microbial activity can significantly reduce litter decomposition and soil respiration. This leads to an increase in both C storage in organic matter and soil nutrient leaching (Gan et al., 2013; Xu et al., 2009; Zak et al., 2008). Owing to the lack of long-term research into soil fauna responses to N deposition, $CL_{emp}N$ have not yet been defined conclusively. Based on microarthropod community responses, Ochoa-Hueso et al. (2014) suggest a $CL_{emp}N$ of between 20 and 50 kg N ha^{-1} yr^{-1} for a semi-arid Mediterranean shrub system, similar to the critical threshold of 37 kg N ha^{-1} yr^{-1} previously reported for a Swiss subalpine forest (Xu et al., 2009). Research thus far indicates that the abundance, diversity and species richness of N-sensitive soil mesofauna such as *Collembola* (Figure 9.20) may be promising biological indicators of $CL_{emp}N$ and N saturation in forest systems (Jandl et al., 2003; Xu et al., 2009).

Figure 9.20. Springtails (*Collembola*) such as this *Tomocerus sp.* are sensitive to N deposition and may be useful bioindicators of forest soil N saturation.

Source: Frank Ashwood

9.3.6 Summary of $CL_{emp}N$ for coniferous and deciduous temperate forests

This section provides an overview of the impacts of N deposition on different components of general forest classes. The $CL_{emp}N$ ranges for the different classes are summarised in Table 9.1.

Table 9.1. $CL_{emp}N$ and effects of exceedances on different components of temperate forest classes. ## reliable; # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Component	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Soil processes			
Deciduous and Coniferous	10-15 10-15	# ##	increased N mineralisation, increased nitrification increased NO_3^- leaching
Trees			

Component	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Temperate forests	**10-15**[a]	(#)	Nutrient imbalances, increased N and decreased concentrations of P, K and Mg in foliage, increased susceptibility to pathogens and pests, change in fungistatic phenolics
Mycorrhiza			
Deciduous Coniferous	**13-16** **10-13**	## ##	Reduced sporocarp production, reduced belowground growth, reduced species richness and changed species composition
Ground vegetation			
Temperate forests	10-15	##	Changed species composition, increase in nitrophilous species, decrease in oligophilic species,
Lichens and algae			
Temperate forests	**3-5**	**#**	Decline of epiphytic lichens and N_2 fixation by cyanolichens, increase in free-living algae

a) Use the lower end under drought.

9.4 Effects of N deposition on spruce and pine taiga woodlands (T3F and T3G)

Together, spruce (T3F - dark taiga) and pine (T3G - light taiga) taiga woodlands constitute the westernmost part of the continuous Eurasian northern taiga belt (Chytrý et al., 2020). Spruce taiga woodlands include the boreal spruce and spruce-pine forests of Fennoscandia, north-eastern Poland, the Baltic States, Belarus and European Russia. Pine taiga woodlands include the boreal pine forests in the regions mentioned above. Broadleaved deciduous taiga woodlands include forests with broadleaved deciduous species, with extended cold winters and short mild summers. Tree height rarely exceeds 15 meters and may be as low as two meters in subarctic conditions. Lichens and mosses frequently dominate the ground layer.

Both spruce and pine taiga woodlands are mostly very poor in N. Any increase in N input may influence major soil processes as well as tree growth rates, changes in the species composition and diversity of forest understorey vegetation including bryophytes (mosses), epiphytic lichens on tree trunks and branches and the abundance of free-living algae and/or cyanobacteria living in association with bryophytes. Because soil responses to increased N input are likely to be similar in both spruce and pine taiga woodlands, the effects of N deposition on soil processes are presented together, whereas the impacts on biodiversity are treated separately.

9.4.1 Soil processes in spruce and pine taiga woodlands

Soil acidification

A long-term N fertilisation experiment in northern Sweden showed that 34 years of N additions (30 kg N ha^{-1} yr^{-1}, background deposition 2-3 kg N ha^{-1} yr^{-1}) led to increased N mineralisation rates in Scots pine forests (Chen and Högberg, 2006). Furthermore, N addition has been shown to result in soil acidification (Högberg et al., 2006; Solberg et al., 2004) which is reflected in a decrease in base cation saturation. This decrease in base cations was recorded only in the mineral soil, whereas for mor-layer base cation concentrations, no differences were detected between N treated plots and control plots (Högberg et al., 2006). In a long-term experiment (45 to 52 years with N additions every 5-10 years) carried out in Finland on 28 sites (spruce or pine), the added N varied from 10.5 to 37 kg N ha^{-1} yr^{-1}. During the experimental period, background N deposition decreased from 9 kg N ha^{-1} yr^{-1} to 4 kg N ha^{-1} yr^{-1}. Stand production, amounts of organic matter, carbon and most nutrients increased, but C:N ratio and pH decreased in surface soil layers. This was an indication of a slight soil acidification effect (Saarsalmi et al., 2014). A study of 204 sites with Norway spruce and Scots pine across mid and south-eastern Norway showed that N deposition (6-8.5 kg N ha^{-1} yr^{-1}) correlated negatively with base saturation in the humus layer, with soil pH and with Ca:Al ratio. It correlated positively with Al^{3+} concentration in the mineral soil layer (Solberg et al., 2004). However, Solberg et al. (2004) suggested that climate, geology and natural processes may also explain these relationships. Acidification of soil water at sites with low N depositions in northern Norway suggested other sources of acidification, such as plant and microbial uptake of base cations and NH_4^+ and increased production of organic acids (Kvaalen et al., 2002).

N leaching

N deposition may lead to increased N leaching from coniferous forests. Boreal forest ecosystems have large capacities to retain N (Petrone et al., 2007, Moldan et al., 2018). There are empirical studies that show a positive relationship between N leaching and N deposition, and others that show no such relationship (Akselsson et al., 2010; Futter et al., 2009; Gundersen et al., 1998b; Kaste et al., 2004; Nadelhoffer et al., 1999; Sjøeng et al., 2009). Under undisturbed conditions, boreal forests have a high N-retention capacity. However, forest disturbances such as clearcut harvests (Zanchi et al., 2014), storm fellings (Hellsten et al., 2015) and bark beetle attacks (Karlsson et al., 2018) have all been shown to initiate the leakage of NO_3^- to the soil water. Furthermore, it has been shown that the N leached to the soil water after clearcut harvests can cause increased NO_3^- concentrations in groundwater (Kubin, 1995; Norrström, 2002) and surface waters, too (Grip, 1982; Karlsson et al., 2021; Löfgren et al., 2009).

In south Sweden it has been demonstrated that increased N depositions elevated the risk of N leaching (Akselsson et al., 2004). Furthermore, studies in Finland indicated that timber harvesting has effects on N leaching, because the uneven distribution of logging residues (tree tops, foliage and branches) can lead to hot spots for N cycling and losses on clear cut sites. For example, studies in spruce, pine and birch forests demonstrated that logging residues increase the rates of net N mineralisation and net nitrification in the humus layer under the piles of logging residue (Adamczyk et al., 2016; Smolander et al., 2019; Törmänen et al., 2018). These effects can last for decades (Smolander et al., 2010).

Bahr et al. (2013) studied the growth of ectomycorrhizal fungi and their effect on N leakage on a broad N gradient in Norway spruce forests across Sweden. They concluded that mycorrhizal fungi are probably important for N retention capacity since high N leaching coincided with low fungal growth. But they could not differentiate between the effects of mycorrhizal fungal growth and the direct effect of N deposition on N leaching. Decreasing N deposition may have rapid positive effects on soil processes. In Finland, the long-term monitoring of deposition and run-off chemistry over a period of 12 to 25 years, showed decreased SO_4^{2-} and NO_3^- concentrations in bulk deposition since the 1980s. This had led to a rapid decrease in concentrations of these elements in run-off (Forsius et al., 2021; Moldan et al., 2001).

Effects on litter decomposition and soil C stocks

Boreal forest soils store a major proportion of global terrestrial carbon. Belowground inputs contribute as much as aboveground plant litter to the total C stored in the soil (Pan et al., 2011). Clemmensen et al. (2013, 2014) demonstrated that mycorrhizal fungi and root-associated fungi are important contributors to soil carbon stocks and that variation in belowground C and N sequestration is affected by mycorrhizal fungal type and growth. Recent studies have shed some light on the effects of N deposition on litter decomposition and soil C stocks. In boreal pine (*Pinus sylvestris* L.) and spruce (*Picea abies* Karst.) forests in northern Sweden with a low background deposition (2 kg N ha^{-1} yr^{-1}), Maaroufi et al. (2019) found that long-term N enrichment (ten years of 3, 6, 12 and 50 kg N ha^{-1} yr^{-1}) impeded mass loss of litter but not of humus, only slightly at low levels but significantly at the highest deposition level. They also found that saprophytic fungi and especially actinomycetes were more important for driving litter decomposition, whereas mycorrhizal fungi appeared more important for reducing N and P concentrations in humus and litter. In the same N experiment, Forsmark et al. (2020) were able to show that only at the highest N addition rate did litter accumulation increase by 46% and soil respiration per mass unit of soil C decrease by 31.2%, mainly by lowering autotrophic respiration. Thus, under the actual ambient low N deposition levels in boreal forest ecosystems, litter decomposition, soil respiration and soil carbon cycle seemed to be unaffected. From this study it was concluded that N deposition in temperate forest soils, where N is not limiting, impeded organic matter decomposition by reducing microbial growth, thereby stimulating carbon sequestration. The authors stated that the concomitant reduction in soil carbon emissions was substantial and equivalent in magnitude to the amount of carbon taken up by trees owing to N fertilisation. These findings seem to be in line with previous findings from a meta-analysis of Janssens et al. (2010).

Although the decomposition of high-quality litter, which is low in lignin, may be stimulated after increased N input, decomposition of litter with a high lignin concentration was found to be reduced by N (Knorr et al., 2005). Accordingly, the increase in soil C after N additions to coniferous forests was attributed not only to decreased decomposition rates of lignin-rich litter (Knorr et al., 2005) but also to N-induced increases in litter production (Franklin et al., 2003).

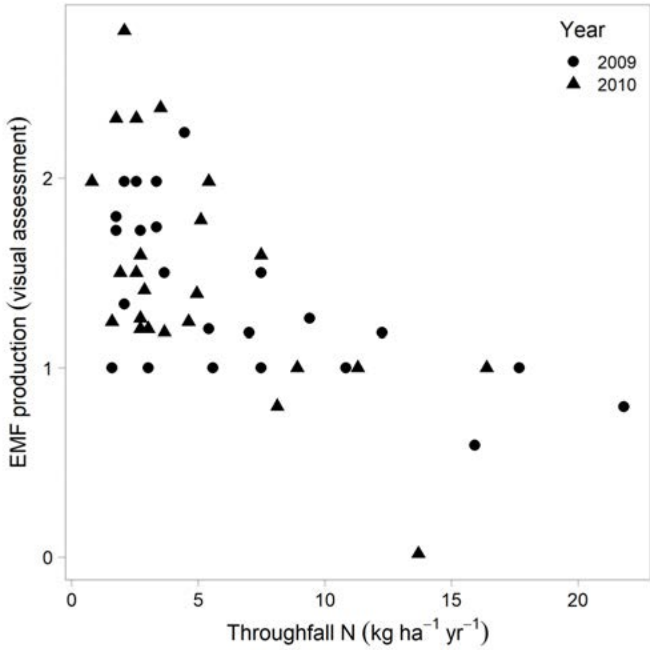
Soil microbial biomass

A review of N effects on soil microbial biomass by Treseder (2008) showed that N deposition reduced microbial biomass in boreal forests by up to 18%. Bacterial response ratios increased in boreal forests and decreased in tundra. After excluding very young forests and elevated CO_2 treatments from a meta-analysis by Janssens et al. (2010), the average reduction in microbial biomass due to N deposition was 16%. But the authors state that in N limited boreal forests or in open forests and very young plantations, bacterial biomass may even rise with increasing N deposition due to increasing rates of photosynthesis.

In a long-term N enrichment experiment (ten years of 0, 3, 6, 12, 50 kg N ha^{-1} yr^{-1}) in boreal conifer forests with very low background deposition (2 kg N ha^{-1} yr^{-1}), Maaroufi et al. (2019) found that microbial biomass (fungi, gram-positive and gram-negative bacteria) decreased significantly in the high N treatment compared to the control and low N treatments. The biomass of actinomycetes (bacteria) grew with increasing N addition. The results of this study also showed that the biomass of fungi and some bacteria was reduced in the 12 kg N ha^{-1} yr^{-1} treatment, suggesting a negative effect already at low N deposition levels commonly found in some boreal ecosystems.

In a N gradient study across conifer forests in Sweden, Bahr et al. (2013) reported a reduction in mycelial biomass of ectomycorrhizal fungi even at moderate levels of N deposition (throughfall N load < 10 kg N ha^{-1} yr^{-1}; Figure 9.21). This study included 29 thoroughly monitored Norway spruce stands from a large geographical region in Sweden and evaluated the importance of N deposition on growth of extramatrical mycelia, N leaching and C sequestration in a broader context.

Figure 9.21. Moderate levels of N deposition reduce the biomass of ectomycorrhizal fungi (EMF) in conifer forests across Sweden.



Source: Bahr et al., 2013

A different result was reported by Tahovská et al. (2020). They found that the soil microbial biomass was much larger in a spruce forest catchment in south-west Sweden that had received an N addition of 40 kg N ha^{-1} yr^{-1} over 24 years (cumulative N input of > 1200 kg N ha^{-1}), compared to a control catchment. The N addition did not change the fungi:bacteria ratio, but a larger share of the bacterial community was made up of copiotrophs. Furthermore, fungal community composition shifted to more nitrophilic ectomycorrhiza fungi. The restructured microbial community showed faster net N mineralisation and nitrification.

Effects on greenhouse gases

N addition also affects emissions of the important greenhouse gases, CH_4 and N_2O. It has been shown that boreal forest soils may be an important sink for atmospheric CH_4 as they contain CH_4-oxidising microbes. Long-term additions (27 years) of around 31 kg N ha^{-1} yr^{-1} to a Norway spruce stand in south-eastern Finland showed that the CH_4 uptake by soil was not negatively affected by N fertilisation (Saari et al., 2006) in contrast to results for a temperate system described in Chapter 9.3.1.

On organogenic forest soils (forests on former peatlands) in Sweden and Finland, a strong correlation between N_2O emissions and soil C:N ratio was demonstrated: the lower the C:N ratio, the higher the N_2O emissions (Klemedtsson et al., 2005). For other forests soils, not more than 0.5 to 1% of the N input evaporated as N_2O (Maljanen et al., (2006)): boreal soil, Papen and Butterbach-Bahl (1999): temperate soil (Germany)).

The information on the effect of N deposition on greenhouse gases is important for climate change models. There are not enough data to set a specific critical load.

9.4.2 Effects on mycorrhiza in spruce and pine taiga woodlands

Productivity in spruce- and pine-dominated taiga forests is primarily limited by a cold climate and low N availability (Tamm, 1991). Increased N input in boreal ecosystems alters growth and species composition of mycorrhizal fungi, as measured both in the production of fruit bodies (aboveground) and mycelia (belowground) (Lilleskov et al., 2001, 2002; Treseder, 2004); Lilleskov et al., 2011, 2019). In Norway spruce forests in southern Sweden aboveground fruit-body production in species of the genera *Cortinarius* and *Russula* decreased, while species such as *Paxillus involutus* and *Lactaria rufus* were less sensitive, or even responded positively to N additions of 35 kg N ha^{-1} yr^{-1}, for 4.5 years (background deposition 13 kg N ha^{-1} yr^{-1}) (Brandrud and Timmermann, 1998). Strengbom et al. (2003) found that the production of fruit-bodies in *Cortinarius* species in two conifer forests in northern Sweden was around 300 times higher on control plots than on plots receiving 34 kg N ha^{-1} yr^{-1} (background deposition 2-3 kg N ha^{-1} yr^{-1}); on the latter, production was very small. In Scots pine forests in northern Sweden ectomycorrhizal sporocarp production and richness were investigated in several N addition treatments (20, 35, 70 and 110 kg N ha^{-1} yr^{-1}, 2 kg N ha^{-1} yr^{-1} background deposition). The authors reported an elimination of sporocarps at the highest N treatment and a shift in community composition and dominant sporocarp taxa at 35 kg N ha^{-1} yr^{-1} toward a higher abundance of nitrophilic taxa, especially *Lactarius* (Hasselquist and Högberg, 2014). On a natural N gradient in boreal forests of northern America (1 to 20 kg N ha^{-1} yr^{-1}), sporocarp production and richness were found to be strongly reduced and community composition was found to change dramatically with increasing N availability (Lilleskov et al., 2001, 2002). Nitrophobic and nitrophilic taxa could be identified. Species of the genera *Cortinarius*, *Piloderma*, *Amphinema*, *Tomentella*, *Russula*, *Tricholoma*, *Lactarius* and *Hebeloma* declined in species richness or abundance with increasing organic horizon mineral N, while *Lactarius theiogalus*, *Laccaria, Paxillus involutus*, *Hygrophorus olivaceoalbus*, *Tylospora fibrillosa*, *Tomentella sublilacina* and *Thelephora terrestris* increased with rising N enrichment.

Mycorrhizal fungi on ericaceous dwarf-shrubs in boreal ecosystems appear to be less sensitive than ectomycorrhizal fungi associated with trees. Ishida and Nordin (2010) found that, although species composition of fungi on roots of *Vaccinium* species differed between spruce and pine forests, N addition had no negative effect on fungal communities (12.5 and 50 kg N ha^{-1} yr^{-1} for four years in pine and 12 years in spruce forests, with background depositions of 2 kg N ha^{-1} yr^{-1}). Maaroufi et al. (2019) found that N treatments (ten years of 0, 3, 6, 12, 50 kg N ha^{-1} yr^{-1}) in boreal conifer forests with very low background deposition (2 kg N ha^{-1} yr^{-1}) caused substantial changes in microbial community abundance and composition. Many of these effects occurred independently of trenching, and despite a minor decline in soil pH at the highest N application (0.22 pH units). Abundance was reduced for fungi and several bacterial groups, except for the bacterial group actinomycetes which increased in response to N enrichment. This also caused significant changes in both the mycorrhizal and saprotrophic fungal community composition, indicating that different species that are likely more N demanding became dominant as plots were enriched with N. While many species contributed to the compositional differences that emerged in response to N, the mycorrhizal fungi *Xerocomus ferrugineus* and *Tylospora asterophora* increased the most in response to N, whereas a variety of other operational taxonomic units decreased (e.g. *Tylospor*a. *Echinospora* and *Sebacinales*). Other studies also reported an increase in *Xerocomus* sp. in response to N addition (Almeida et al., 2019). The genus *Xerocomus* belongs to the long-distance exploration type (i.e. ectomycorrhizal fungi foraging strategy) that produces larger hyphal biomass contributing to fungal necromass with slow turnover compared to the short-distance exploration type.

Termination of N fertilisation experiments in conifer forests in Sweden led to residual fungal community effects even after 23 years (Choma et al., 2017) or 47 years (Strengbom et al., 2001). The former study revealed that the relative abundance of mycorrhizal fungal species shifted closer to, but did not reach, control levels. The latter study showed that sporocarp production did not fully recover and that species composition differed largely from the control plots.

However, Högberg et al. (2014) reported a recovery of many ectomycorrhizal fungal species (mainly *Atheliaceae* and *Russulaceae*, but not *Cortinariaceae* and *Thelephoraceae*) 14 years after termination of high-level N treatment (108 kg N ha^{-1} yr^{-1} for 19 years) in a Scots pine forest in northern Sweden. Meanwhile, bacterial species in this forest did not show any sign of recovery.

Bahr et al. (2013) showed that ectomycorrhizal fungi contributed substantially to carbon sequestration, but their growth was limited as nitrate leaching increased. This correlated positively with N deposition. They used a gradient of 29 plots with throughfall deposition between 0.95 and 24.6 kg N ha^{-1} yr^{-1}.

Along a soil fertility gradient in boreal forests in Sweden with NH_4^+ values between 1 and 53 µg N per g organic matter, Sterkenburg et al. (2015) observed changes in the community composition of ectomycorrhizal fungi. They also reported a reduction in litter and root associated ascomycetes as well as yeast and mould fungi while basidiomycetes increased in abundance in more fertile forests sites.

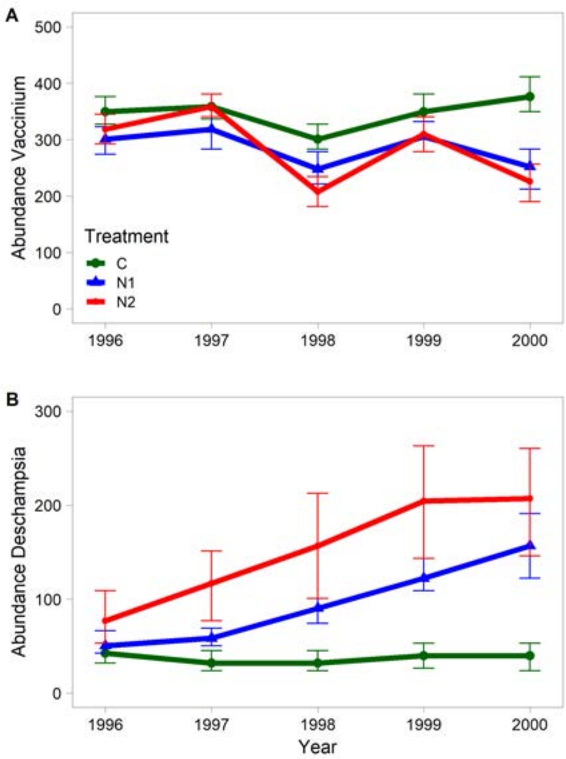
The N addition experiments did not permit any conclusions on a critical load as either the treatments were too high or an effect threshold was not indicated. However, the gradient study of Bahr et al. (2013) supports the results presented in the chapter on temperate forests.

9.4.3 Spruce taiga woodlands (T3F - dark taiga)

Field- and ground-layer vegetation

The responses in understorey vegetation in boreal spruce forests have been comprehensively studied. A common initial response after N addition in areas with low background depositions (2-6 kg N ha^{-1} yr^{-1}) is elevated tissue N concentration in plants, such as bryophytes, grasses and ericaceous dwarf-shrubs (Forsum et al., 2006; Mäkipää, 1998; Nordin et al., 1998, 2006; Strengbom et al., 2002). Several studies also reported that the composition of field-layer vegetation was sensitive to increased N input. Over a period of 20 years, Rosén et al. (1992) observed a significant positive correlation between *Deschampsia flexuosa*-dominated coniferous forests in Sweden and the pattern of N deposition, based on comparisons between field-layer vegetation surveys in the Swedish Forest Inventories of 1973 to 1977 and 1983 to 1987. *Deschampsia flexuosa* increased significantly during this period. These changes occurred above an N deposition of 7 to 11 kg N ha^{-1} yr^{-1}. Other studies reported altered abundance of commonly occurring dwarf shrubs such as *Vaccinium myrtillus* and *Vaccinium vitis-idaea* and the grass *Deschampsia flexuosa*. There have been reports of such changes already at low (5 kg N ha^{-1} yr^{-1}: Kellner and Redbo-Torstensson (1995), 12.5 kg N ha^{-1} yr^{-1}: Nordin et al. (2005)) or moderate levels (40 kg N ha^{-1} yr^{-1}: Manninen et al. (2009)). In northern Sweden, four years of additions of 6 kg N ha^{-1} yr^{-1} (background deposition of around 2 kg N ha^{-1} yr^{-1}) increased the abundance of *Deschampsia flexuosa* by around 50% (UNECE, 2007). Additions of 12.5 kg N ha^{-1} yr^{-1} over five years (background deposition of around 2 kg N ha^{-1} yr^{-1}), resulted in a 300% higher abundance of *Deschampsia flexuosa*, and a 34% lower abundance of *Vaccinium myrtillus* (Figure 9.22).

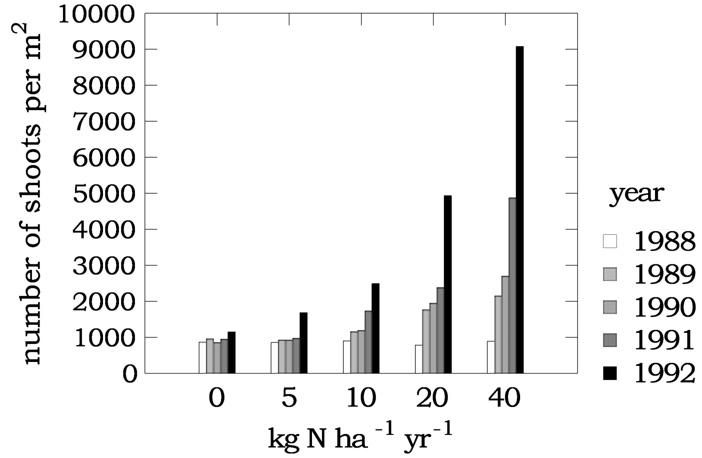
Figure 9.22. Response in the two dominant plant species of the ground vegetation (the dwarf shrub *Vaccinium myrtillus* (A) and the grass *Deschampsia flexuosa* (B)) in a Swedish boreal forest (T3F) that had been exposed to N additions corresponding to 12.5 (N1) and 50 kg N ha^{-1} yr^{-1} (N2) for five years. The fertilised plots were sized 1000 m^2 and each treatment was replicated six times (n = 6). Vertical bars show S.E.



Source: Nordin et al., 2005

In central Sweden (background deposition of 2-6 kg N ha^{-1} yr^{-1}), the shoot density of *Deschampsia flexuosa* increased by 70, 250, 430 and 780%, after four years of additions of 5, 10, 20, and 40 kg N ha^{-1} yr^{-1} (Figure 9.23). At the same site, the shoot density of *Trientalis europaea* showed significant increases when N additions exceeded 10 kg N ha^{-1} yr^{-1}.

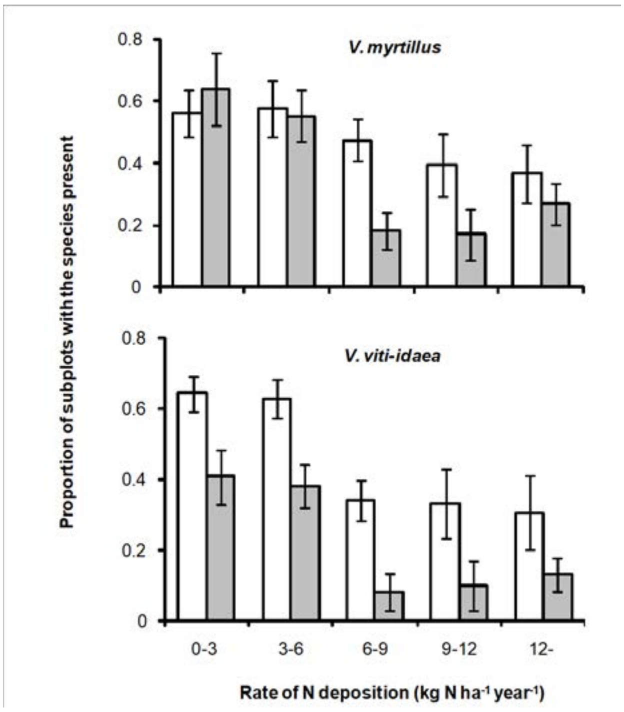
Figure 9.23. Number of shoots of *Deschampsia flexuosa* at different levels of N supply (kg N ha^{1} yr^{1}), between 1988 and 1992, in a boreal forest (T3F) located outside Söderhamn, in central Sweden.



Source: Kellner and Redbo-Torstensson, 1995

The above-listed effects after increased N inputs are supported by surveys along N deposition gradients and changes in understorey composition over time. A field survey in Sweden, examining the occurrence of understorey species at 557 sites (a few located outside T3F and T3G classes), showed that occurrences of *Vaccinium myrtillus* and *Vaccinium vitis-idea* (Figure 9.24) were less frequent in areas where N depositions had been greater than or equal to 6 kg N ha^{-1} yr^{-1} than in areas with lower N depositions (Strengbom et al., 2003). In Norway, the occurrence of *Deschampsia flexuosa* increased in spruce forests between 1988 and 1993 (background deposition (wet) of 7.9 kg N ha^{1} yr^{1}) (Økland, 1995). In addition, *Vaccinium myrtillus* proved to be more susceptible to the leaf pathogen *Valdensia heterodoxa* in areas with high levels of N deposition (Strengbom et al., 2003).

Figure 9.24. Proportion of subplots with *Vaccinium myrtillus* and *Vaccinium vitis-idaea* in forest stands with various N deposition rates. White bars represent Scots pine and dark bars represent stands dominated by Norway spruce. Vertical bars: mean and confidence intervals (95%).



Source: Strengbom et al., 2003

Results from experiments showed that understorey responses to high levels of N may depend on forest type. At experimental sites with high tree canopy cover, for example sites with high spruce domination, the effects of the input of high levels of N could be limited by reduced light availability as tree canopy cover increased. In comparison, light limitation for more open experimental sites, for instance with a higher proportion of pine trees, could be less severe and understorey responses to N additions more linear. In a study by Kellner and Redbo-Torstensson (1995), the density of *Deschampsia flexuosa* rose as N additions increased (Figure 9.23). Nordin et al. (2005) reported relatively small differences in effects on *Deschampsia flexuosa* and *Vaccinium myrtillus* resulting from five years of N additions of 12.5 and 50 kg N ha^{-1} yr^{-1} (Figure 9.24). Kellner and Redbo-Torstensson (1995) studied a mixed pine-spruce forest, while Nordin et al. (2005) studied a spruce-dominated forest, in which light limitation for understorey species may have limited N responses at high input rates. This illustrates that the responses to increased N deposition may differ depending on site conditions, and that complex biotic interactions, including altered shading from the tree canopy at high levels of N, may complicate the interpretation of the effects of N addition (Gilliam, 2006; Nordin et al., 2009).

Bryophytes depend directly on the wet and dry deposition of N and are, therefore, considered to be highly sensitive, even to small changes in supply. Additional N doses of between 12.5 and 50 kg N ha^{-1} yr^{-1} to coniferous forests in northern Sweden, led to an increase in arginine concentrations of *Pleurozium schreberi* and *Dicranum majus* in an irrigation experiment (Nordin et al., 1998). This was an indication that the mosses were unable to respond to N additions by increased growth and that, instead, N was accumulated in the form of arginine. High amino acid concentrations may be harmful to bryophytes, and correlated with reductions in the growth length of *Sphagnum* (Nordin and Gunnarsson, 2000). Additionally, it was found that N-induced decreases in the abundance of specific bryophytes may persist long after N input has been terminated (Nordin et al., 2005).

The concentration of total N in moss sampled in Sweden, Norway and Finland correlated linearly with the wet deposition of N in the range of 0 to 15 kg N ha^{-1} yr^{-1} (Harmens et al., 2011). A recent field study (Salemaa et al., 2020) showed accumulation of free NH_4^+ and total N concentrations of up to 2% in *Pleurozium schreberi* and *Hylocomium splendens* at throughfall N deposition < 3.5 kg N ha^{-1} yr^{-1} in background areas in Finnish boreal forests. Overall, the bryophyte total N concentrations were higher in Norway spruce than in Scots pine forests, and higher inside than outside forests, as the throughfall especially in southern Finnish spruce forests was enriched with DON leached from the canopy. The high total N concentration in bryophytes at such a low total N deposition level highlights the role of dry-deposited N and DON in areas with low precipitation (400-700 mm yr^{-1}) and low temperatures. In support of this, Pitcairn et al. (2006) concluded that bryophytes reached high N concentrations in areas with high precipitation only when rainfall constantly contained large concentrations of N. Pearce and Van der Wal (2008) demonstrated in experiments the importance of NO_3^- and NH_4^+ concentrations in wet deposition for the growth of a sensitive bryophyte species, for instance *Racomitrium lanuginosum*, irrespective of the N dose.

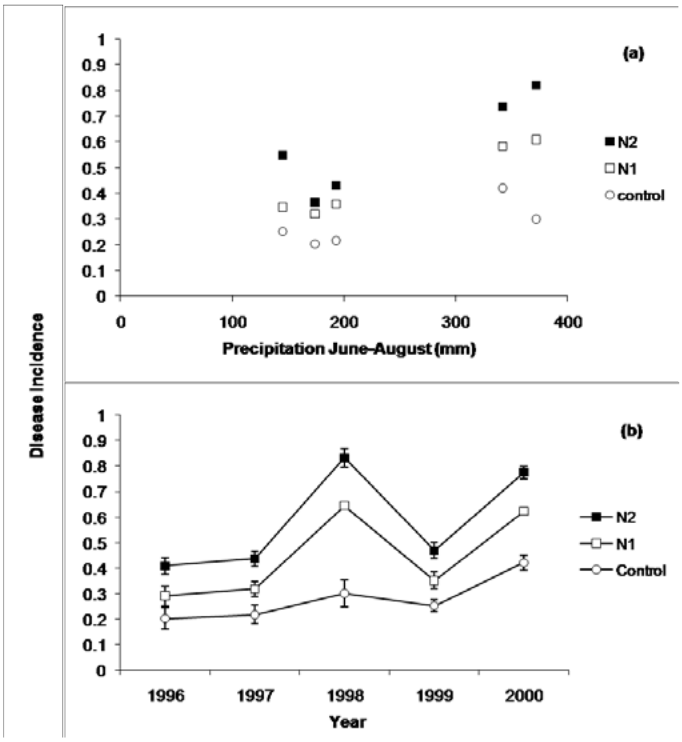
Biological N-fixation is a major driver of terrestrial ecosystem processes in natural ecosystems (Chapin et al., 1986). For instance, it was suggested that the symbiosis of feather mosses such as *Pleurozium schreberi* with cyanobacteria (*Nostoc*) was the main source of ecosystem N in natural boreal forest and tundra ecosystems (DeLuca et al., 2002). N deposition may affect biological N-fixation already at low levels. A $CL_{emp}N$ < 5 kg N ha^{-1} yr^{-1} is proposed based on biological N_2 fixation by cyanobacteria living in association with bryophytes (Gundale et al., 2011; Salemaa et al., 2019; Zackrisson et al., 2004). Zackrisson et al., (2004) found that a surface application of 4.5 kg N ha^{-1} yr^{-1} as NH_4NO_3 eliminated N_2 fixation in *Pleurozium schreberi* from northern Sweden (65-66°N). In their field experiment with NH_4NO_3 granules applied within one week of snow melt, the lowest N addition (3 kg N ha^{-1} yr^{-1}) resulted in a nearly 50% reduction in N_2 fixation per unit mass of *Pleurozium schreberi* relative to control (background deposition of 2 kg N ha^{-1} yr^{-1}) in a northern Swedish (64°N) Scots pine forest (Gundale et al., 2011). Salemaa et al. (2019) reported, in turn, that biological N_2 fixation was switched off in *Pleurozium schreberi* and *Dicranum* sp. in northern Finnish Scots pine and Norway spruce forests at a total N deposition in BD of 1-2 kg N ha^{-1} yr^{-1}, while *Hylocomium splendens* still showed very weak N_2 fixation in southern Finland at 3-4 kg N ha^{-1} yr^{-1} and a N concentration of 1.48% in bryophyte tissue. The amounts of N in TF were about 0.5-1.5 kg N ha^{-1} yr^{-1} in northern and 2-3 kg N ha^{-1} yr^{-1} in southern Finland. Inorganic N was a significant predictor of N_2 fixation in both BD and TF deposition models (Salemaa et al., 2019).

Increased sensitivity to pathogens and herbivores

Higher concentrations of N in plant tissue after increased N input may result in greater damage from pathogens and herbivores. Attacks by foliar pathogens on *Vaccinium myrtillus* and *Deschampsia fleuxosa* increased after N additions of 12.5 kg N ha^{-1} yr^{-1} for 5-10 years (background deposition of around 2 kg N ha^{-1} yr^{-1}; Nordin et al. (1998, 2005, 2006)). In areas with N depositions of over 6 kg N ha^{-1} yr^{-1}, *Vaccinium myrtillus* was more susceptible to leaf pathogens than in areas with lower levels of N deposition (Strengbom et al., 2003).

For the host plants, the effects of increased N availability on pathogens often depend on interactions with weather conditions. For example, the disease incidence of *Valdensia heterodoxa* infecting *Vaccinium myrtillus* correlates positively with summer precipitation. Higher precipitation levels increase the effect of added N (Figure 9.25). This means that the effect of increased N input may be small during dry years and large during wet years. In *Deschampsia flexuosa,* the two pathogens *Uromyces airae-flexuosae* and *Telimenella gangraena* showed opposite responses to drought stress. *Uromyces airae-flexuosae* decreased, while *Telimenella gangraena* increased in response to drought treatment (Nordin et al., 2006).

Figure 9.25. Disease incidence (proportion of diseased leaves) from the parasitic fungus *Valdensia heterodoxa* on *Vaccinium myrtillus* leaves, in relation to summer precipitation (a), and N addition (b). N corresponds to 0 (control), and to treatments of 12.5 (N1), and 50 kg N ha^{-1} yr^{-1} (N2). Each treatment was replicated six times (n = 6). Vertical bars show one SE.



Source: Strengbom et al., 2006

Plant damage caused by insect herbivores may increase after increased N input as insect population densities may be limited by low concentrations of N in plants. Nordin et al. (1998) demonstrated that damage caused by insect herbivores in *Vaccinium myrtillus* increased after N additions corresponding to 12.5 kg N ha^{-1} yr^{-1} (background deposition of around 2 kg N ha^{-1} yr^{-1}). Strengbom et al. (2005) demonstrated that *Operophtera brumata* larvae feeding on N-fertilised *Vaccinium myrtillus,* showed larger adult mass (indicating higher fecundity). Although N addition may have a positive effect on the population densities of *Operopthera* larvae (Nordin et al., 2009), increased predation or higher parasitoid load may limit such positive effects (Kytö et al., 1996; Strengbom et al., 2005), resulting in no or only minor effects on insect population densities.

Effects of different N forms

When assessing the effects of N deposition, it is important to bear in mind that plant species exhibit differences in their capacities to utilise N. Although the effect of N deposition on an ecosystem is mainly related to the quantity of N deposited, the qualitative aspect also needs to be acknowledged. Few studies have addressed the quantitative differences of N deposition on taiga habitats. Nordin et al. (2006) compared the effects of additions of 12.5 and 50 kg N ha^{-1} yr^{-1} in the form of NH_4NO_3, $(NH_4)_2SO_4$, or KNO_3, to a spruce forest in northern Sweden (background deposition of around 2 kg N ha^{-1} yr^{-1}). There was also a K_2SO_4 treatment to test the effect of the counter ions. *Deschampsia flexuosa* took up more NO_3^- than NH_4^+. Consequently, NO_3^- addition induced more grass growth than additions of NH_4^+. It was concluded that increased grass growth is less likely to occur when N is deposited as NH_4^+ than when it is deposited as NO_3.

Epiphytic lichens

Whole mature Norway spruces were fertilised during four vegetation seasons with an automated irrigation-fertilisation system administered daily as a diffuse spray across the entire canopy, with N in the form of NH_4NO_3 at five levels, equivalent to 0.6, 6, 12.5, 25 and 50 kg N ha^{-1} yr^{-1} at Vindeln Experimental Forests in northern Sweden (Johansson et al., 2012). The background deposition in TF was 2 kg N ha^{-1} in the area between late May and early October with 78% NH_4^+, 17% amino acid N and 5% NO_3^- (Forsum et al., 2006). The composition of the epiphytic lichen community was monitored on a yearly basis. N deposition reduced the species richness of the epiphytic lichen communities even at the lowest level of fertilisation application (6 kg N ha^{-1} yr^{-1}). The authors concluded that the applied wet deposition of NH_4NO_3 of 6 kg N ha^{-1} yr^{-1} (over the background deposition) was above the critical load of N deposition for boreal epiphytic lichen communities.

Esseen et al. (2016) studied the distribution of epiphytic hair lichens in the lower canopy of Norway spruce throughout Sweden from temperate to boreal and subalpine forests. They calculated the mean annual N deposition of N (dry + wet depositions) for 20 km x 20 km grid cells from data provided by the Swedish Meteorological and Hydrological Institute and then extracted the data for each study plot. The results suggest a $CL_{emp}N$ of 3.9 kg N ha^{-1} yr^{-1} for *Alectoria* and of 5.7 kg N ha^{-1} yr^{-1} for *Usnea*.

Most hair lichens have been classified as oligotrops in nutrient-poor sites (McCune and Geiser, 2009). The lower end of the total deposition range of 3.5-5 kg N ha^{-1} yr^{-1} by Esseen et al. (2016) corresponds to the moderate-risk critical load of 3.5 kg N ha^{-1} yr^{-1} resulting in a 50% decline in the diversity and abundance of forage lichens (including hair lichens) estimated by Geiser et al. (2019). A $CL_{emp}N$ of 3-5 kg N ha^{-1} yr^{-1} is proposed for taiga spruce forests (T3F), especially those with low precipitation but high concentrations of dissolved N, based on the European studies by Esseen et al. (2016) and Giordani et al. (2014).

Reversibility of N-induced effects

Although more than 47 years had passed since N addition (~100 kg N ha^{-1} yr^{-1}) to a spruce forest in northern Sweden had ceased, the production of fruit-bodies by N-sensitive mycorrhizal fungi such as *Russula* and *Cortinarius* species were still 39 to 85% lower than in unfertilised control plots (background deposition of around 2-3 kg N ha^{-1} yr^{-1} total N). In addition, the abundance of the bryophyte *Hylocomium splendens* was still 70% lower in plots formerly treated with N than in control plots, while N-favouring bryophytes such as *Brachytecium* and *Plagiothecium* species only occurred in plots previously treated with N (Strengbom et al., 2001). This is an indication that the reversibility of N-induced effects on this forest habitat is low, and that spruce taiga woodlands are very susceptible to high N depositions.

Summary

In Sweden, additions of 6 kg N ha^{-1} yr^{-1} over a four-year period (background deposition of around 2 kg N ha^{-1} yr^{-1}) were found to increase the abundance of *Deschampsia flexuosa* by around 50% (UNECE, 2007), whereas the abundance and occurrence of *Vaccinium myrtillus* was lower when N deposition exceeded this level (Strengbom et al., 2003). Additions of 5 kg N ha^{-1} yr^{-1} over four years resulted in 70% higher shoot densities of *Deschampsia flexuosa* in N-treated plots, compared to control plots (Kellner and Redbo-Torstensson, 1995). In addition, the increased occurrence of *Deschampsia flexuosa* in Norway, between 1988 and 1993, was also attributed to N deposition (Økland, 1995). Large effects on species composition and increased sensitivity to leaf pathogens have been reported from N additions of 12.5 kg N ha^{-1} yr^{-1} within a decade (Nordin et al., 2005, 2006; Strengbom et al., 2002). The low reversibility of N-induced effects (Strengbom et al., 2001) support the suggestion that spruce taiga woodlands is a habitat class that is sensitive to N deposition. The most sensitive organisms in terms of biodiversity are the epiphytic lichens and the critical loads intended to protect these organisms very much depend on the N concentration of the water on their surfaces. Based on the current European data on epiphytic lichens and bryophytes, a $CL_{emp}N$ of 3-5 kg N ha^{-1} yr^{-1} – the total deposition of inorganic N – is considered as a 'reliable' critical load for spruce taiga woodlands (T3F).

9.4.4 Pine taiga woodlands (T3G – light taiga)

Ground vegetation

Response to N deposition may partly depend on initial site productivity. In general, forests dominated by Scots pine, *Pinus sylvestris* (T3G), tend to be less productive than forests dominated by Norway spruce, *Picea abies* (T3F). There are indications that ground vascular plant species in forests dominated by Scots pine may be more resistant to increased N input than forests dominated by Norway spruce. Correlative data along the Swedish N deposition gradient show that in spruce forests, there is a significant drop in the occurrence of *Vaccinium myrtillus* when N deposition exceeds 6 kg N ha^{1} yr^{1}. However, in pine-dominated forests, this effect is seen not until N deposition exceeds 12 kg N ha^{1} yr^{1} (Figure 9.24; Strengbom et al., 2003). For *Vaccinium vitis-idaea* there appears to be less difference in response to N deposition between pine- and spruce-dominated forests. Effects on plant community compositions from increased N inputs often depend on how light availability is influenced by increased N availability (Hautier et al., 2009; Strengbom et al., 2004). This is especially important in forested systems where the response in understory vegetation is often determined by the overstorey response to increased N availability (Gilliam, 2006; Oberle et al., 2009). Compared to Norway spruce stands, Scots pine stands tend to have less canopy cover with more light reaching the forest floor, due to lower productivity and different plant architecture. The implication here is that such stands will be less sensitive to the reduced light availability resulting from N-induced increased tree growth. This may explain why *Vaccinium myrtillus* shows lower responsiveness to elevated N input in pine forests than in spruce forests (Strengbom et al., 2003). The lower productivity of pine forests may also explain why, in these habitats, *Vaccinium myrtillus* may initially be unresponsive or may even increase (at the expense of other dwarf shrubs such as *Vaccinium vitis-idaea*) after N addition (Kellner and Redbo-Torstensson, 1995). However, if N input is sufficiently high, or lasts long enough, the abundance of *Vaccinium myrtillus* will also be reduced in these pine-dominated stands (at the expense of grasses and herbs) (Strengbom et al., 2001). The pattern of gradual replacement of species according to their N strategy (e.g. nutrient-use efficiency) may also explain why the species richness of ground- and field-layer vegetation in low-productivity habitats such as boreal forests, may remain unchanged or may initially even increase in response to increased N deposition (Bobbink, 2004). Despite these indications of differences in N sensitivity between spruce- and pine-dominated forests, the available data on such differences are sparse, and more data are needed to elucidate these differences.

Ground-living lichens often make up most of the species' richness of the ground vegetation in pine forests. Several experiments identified this group of plants as sensitive and one of the first plant groups to disappear as a consequence of increased inputs of N (Dirkse and Martakis, 1992; Mäkipää, 1994, 1998; Skrindo and Økland, 2002; Strengbom et al., 2001). In Sweden, all ground-living lichens disappeared from plots receiving 30 to 60 kg N ha^{-1} yr^{-1} (background deposition of around 2-3 kg N ha^{-1} yr^{-1}) for 20 to 30 years (Strengbom et al., 2001; Van Dobben et al., 1999). In Norway the reduced occurrence of lichens was observed after N additions of 30 to 90 kg N ha^{-1} yr^{-1} (background deposition of 5-6 kg N ha^{-1} yr^{-1}) (Skrindo and Økland, 2002). The mechanism by which N deposition negatively influences the abundance of ground-living lichens is not clearly understood. Mäkipää and Heikkinen (2003) reported that the relative abundance of *Peltigera aphtosa*, which has cyanobacteria as its photobiont (and is expected to be N sensitive), decreased in Finland between 1951 and 1986, and again in 1995. It should be noted that the observed changes may not have been due solely to N deposition. Although several studies reported that increased N availability may disrupt physiological processes in lichens (Dahlmann et al., 2002; Kytöviita and Crittenden, 2007), the decreased abundance of lichens may also be an effect of increased competition from vascular plants that respond positively to N (Cornelissen et al., 2001).

Together with ground-living lichens, bryophytes make up the plant group that appears to be most sensitive to elevated N deposition. There are, however, major differences in responses between species, ranging from strong negative effects to positive effects. In pine forests, the decreased abundance of *Hylocomium splendens* and *Pleurozium schreberi,* and increased abundance of litter-dwelling species, such as *Brachythecium* and *Plagiothecium* species, are frequently reported after increased N input (Dirkse and Martakis, 1992; Mäkipää, 1994; Nordin et al., 2005; Skrindo and Økland, 2002; Strengbom et al., 2001; Van Dobben et al., 1999). Strengbom et al. (2001) reported a more than 70% reduction in the abundance of *Pleurozium schreberi* and the increased abundance of *Brachythecium* species, after 29 years of N additions of 34 kg N ha^{-1} yr^{-1} (background deposition of around 2-3 kg N ha^{-1} yr^{-1}). Skrindo and Økland (2002) reported that the occurrence of *Ceratodon purpureus*, *Dicranum fuscescens*, *Dicranum polysetum* and *Dicranum spurium* decreased with increasing N additions (30-90 kg N ha^{-1} yr^{-1}).

Optimal growth in relation to N inputs varies between species, and may partly explain why some species are more sensitive to increased N input than others (Salemaa et al., 2008). Moreover, the abundance of *Pleurozium schreberi* correlated negatively with tissue concentrations of amino acids (Nordin et al., 2005), indicating that excess N may be detrimental to this moss species. In addition, as for lichens, the reduced abundances of species such as *Hylocomium splendens* and *Pleurozium schreberi*, may also be partly explained by more intense competition from vascular plants under increased levels of N. The $CL_{emp}N$ of 3-5 kg N ha^{-1} yr^{-1} based on bryophyte responses in spruce taiga woodlands also applies to pine taiga woodlands (Salemaa et al., 2019, 2020).

Effects of different N forms

As far as we know, no data are available on the specific effects on pine forests of the different forms of N, except that of Salemaa et al. (2020). These authors found that the relationship of N concentrations for mosses in clearings was best with bulk deposition outside the forest and for mosses in the forest it was DON in throughfall. The responses reported for spruce forests are mostly thought to be valid for pine forests, too (see Chapter 9.4.3). However, the dry deposition of N is most likely higher in Norway spruce than in Scots pine forests (Lövblad et al., 1992), although dry deposition in northern coniferous forests is difficult to monitor (Karlsson et al., 2019).

Green algae and epiphytic lichens

Most of the recent studies on epiphytic lichens and N pollution, especially those done in Europe, focussed on assessing the critical levels of reactive gaseous N compounds for lichen diversity etc. instead of the critical loads. A Canadian study on epiphytic lichens in Jack pine (*Pinus banksiana* forests) in northern Alberta and Saskatchewan proposed a total deposited N threshold (TDN) of 1.5-3 kg N ha^{-1} yr^{-1} (Vadinther, 2019). The modelled N deposition data used by Vadinther (2019) accounted for 12 different forms of N. The TDN correlated highly with dry nitrogen oxide deposition (DNO) and dry nitrogen dioxide deposition (DNO_2), both of which were highly relevant drivers in the gradient Forest model which was used. Lichen community thresholds (based on epiphytic macrolichens such as *Vulpicida pinastri* and *Evernia mesomorpha* and a ground-living lichen *Cladina mitis*) across both DNO and DNO_2 gradients corresponded to a TDN threshold of 1.4-2.4 kg N ha^{-1} yr^{-1}. The results of Vadinther (2019) from a smaller scale study suggest, however, a biodiversity-based community $CL_{emp}N$ of 5.6 kg N ha^{-1} yr^{-1}. The latter $CL_{emp}N$ is based on several vascular plant species and bryophytes in addition to epiphytic lichens in Canadian pine taiga woodlands.

N concentrations in lichens can be used to estimate the throughfall deposition of inorganic N in areas for which no measured throughfall data are available (Root et al., 2013). Average N concentrations of 0.3-0.6% have been reported in *Platismatia glauca* and *Hypogymnia physodes* in background areas receiving 0.5-2.0 kg N ha^{-1} yr^{-1} as total or TF deposition (Dahlman et al., 2003; Johansson et al., 2010, 2011)), about 2.5 kg NO_3^--N ha^{-1} yr^{-1} as WD (Bruteig, 1993) or 0.02-0.1 mg NH_4^+-N l^{-1} in precipitation (Bruteig, 1993; Geiser and Neitlich, 2007). Based on the lichen N concentrations, atmospheric NO_2 and NH_3 concentrations, the frequencies of green algae + *Scoliociosporum chlorococcum* and selected acidophytic macrolichen indicator species on Scots pine trunks, Manninen (2018) proposed a $CL_{emp}N$ of 2-3 kg N ha^{-1} yr^{-1} as a total NO_2+NH_3 deposition for Scots pine forest in Finland. There, the precipitation is low and the concentration of dissolved N may thus be higher than in areas with similar atmospheric concentrations of gaseous N pollutants but high precipitation. In support of the importance of dry deposition and N concentration, Frahm (2013) related the N sensitivity of *Hypogymnia physodes* to its low conductivity (osmotic value) and low water uptake from salt solutions compared to the nitrophyte species such as *Phaeophyscia orbicularis* and *Physcia adscendens* with high osmotic tolerance of the salt effects on N compounds. Frahm (2013) also concluded that the nitrophilous lichen species were drought resistant thanks to their high osmotic values and were, therefore, more competitive in areas with low humidity than other lichen species.

The responses of epiphytic lichens highlight the need for measurements and more detailed data on dry deposition of N compounds to assess the role of dry deposition on the most sensitive organisms in taiga forests.

Reversibility of N-induced effects

Quist et al. (1999) reported swift recovery in soil N concentrations after the cessation of 20 years of N additions of 108 kg N ha^{-1} yr^{-1} (Norrliden site, background deposition of around 2 kg N ha^{-1} yr^{-1}). However, data on the reversibility of the N-induced effects on biodiversity at this site, suggest that reversibility is a slow process since the composition of understorey differed markedly from control. Strengbom et al. (2001) did not observe any recovery of plant species composition or fruit-body production nine years after N additions had ceased. And 14 years later, Chen and Högberg (2006) noted that the N mineralisation rates in Norrliden were still elevated for the plots that had been treated with N. In a gradient study, Weldon and Grandin (2021) also observed only slow recolonisation of N- and/or S-sensitive epiphytic lichen species in pine or spruce dominated forests over 20 years (1997-2016) in Sweden, despite the current suitable environmental conditions.

Summary

Compared to spruce taiga woodlands, there are indications that, for pine forests, negative effects on the biodiversity of vascular plant species from increased N input start to occur at higher N input rates. However, the data that support that pine forests are less sensitive to increased N input compared to spruce forests are not conclusive. Strengbom et al. (2003) found that, in pine forests, *Vaccinium vitis-idaea* had a significantly lower occurrence when N depositions were higher than 6 kg N ha^{-1} yr^{-1}. However, they also found that N depositions had to be over 12 kg N ha^{-1} yr^{-1} before *Vaccinium myrtillus* would occur at significantly lower rates. Experiments using higher N loads (over 20-30 kg N ha^{-1} yr^{-1}) often show major effects on the composition of understorey vegetation (Dirkse and Martakis, 1992; Strengbom et al., 2001; Van Dobben et al., 1999). The effects of N on plant community composition (Strengbom et al., 2001) and N mineralisation (Chen and Högberg, 2006) also appear to reverse only slowly when external N inputs are halted. In summary, these results suggest that pine taiga woodlands are sensitive to increased N deposition. With the support of gradient studies that revealed the effects at N loads of less than 10 kg N ha^{-1} yr^{-1}, it was recommended in the previous review version to set the $CL_{emp}N$ range for Pine taiga woodlands (T3G, formerly G3B) at 5 to 10 kg N ha^{-1} yr^{-1}. It was considered as 'quite reliable'. The study of Giordani et al. (2014) included both northern and southern European pine forests. A ´quite reliable´ $CL_{emp}N$ of 2-5 kg N ha^{-1} yr^{-1} is proposed for pine taiga woodland (T3G) based on epiphytic lichen responses, especially species' composition, to inorganic N deposition in TF reported in Europe, Canada and the USA, where there are still more pristine areas in terms of N deposition than in Europe. In comparison, the study by Geiser et al. (2021) indicated initial shifts from pollution-sensitive toward pollution-tolerant species already at 1.5 kg N ha^{-1} yr^{-1} and 2.7 kg S ha^{-1} yr^{-1}. Moreover, these $CL_{emp}N$ values are considered constant under any climate regime nationwide in the USA.

9.4.5 Summary T3F and T3G

Table 9.2 summarises the CLempN ranges on spruce and pine taiga woodlands (T3F and T3G)

Table 9.2. $CL_{emp}N$ and effects of exceedances on spruce and taiga woodlands. ## reliable; # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Dark taiga	T3F	5-10	##	**3-5**[a]	##	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation
Pinus sylvestris light taiga	T3G	5-10	#	**2-5**[a]	#	Changes in epiphytic lichen and ground-layer bryophyte communities, increase in

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
						free-living algae; decline in N-fixation

a) Mainly based on N deposition impacts on lichens and bryophytes.

9.5 Effects of N deposition on Mediterranean ecosystems

The Mediterranean biogeographical region is characterised by a climate with hot dry summers and rainy mild-cool winters. Spring and autumn are usually the growing seasons since the typical summer drought imposes limitations on biological activity. Besides the strong seasonal changes, the climate also presents high interannual variability. In Europe, this biogeographical region is located around the Mediterranean Basin. The climate variability acts together with a contrasting topography and is the driver behind a very rich biodiversity, particularly of plants. 25,000 flowering plants were identified that represent 10% of all known plants on earth. Half of the plants are also endemic. Mediterranean forests are also very diverse, with up to 100 different tree species. Almost half of the plants and animals listed in the Habitats Directive occur in the Mediterranean region. This high biodiversity makes the Mediterranean area one of the biodiversity hotspots for conservation priorities. The main threats to biodiversity in this region include land occupation for construction, forest fires, chronic water shortage linked to climate change, invasive species and an abandonment of traditional agriculture and pastoral regimes (Commission, 2009). Atmospheric N deposition has only recently been recognised as an additional pressure for Mediterranean biodiversity (Ochoa-Hueso et al., 2011; García-Gómez et al., 2020). High ozone concentrations are another particularity of the area. They are caused by the typical high air temperatures and solar radiation, together with the stability of air masses that constitute an additional threat to plant functioning and development (Ochoa-Hueso et al., 2017).

9.5.1 Deciduous broadleaved forest (T1)

Fagus forest on acid soils (T18)

The long-term variability of growth rates was analysed in four beech-dominated forests selected along a latitudinal transect across the Italian peninsula in the montane belt. The long-term median values of atmospheric N deposition, estimated with the WCRP-CMIP6 model, ranged from 4 to 9 kg N ha^{-1} yr^{-1} with maximum annual values up to 15 kg N ha^{-1} yr^{-1} (Gentilesca et al., 2018). The positive N deposition effect on annual height and volume growth N was more important than climate variation and this effect reached a threshold value of 10 kg N ha^{-1} yr^{-1}. On the basis of the results of this study and the joint consideration of the results of similar forests growing in temperate areas, a $CL_{emp}N$ of 10-15 kg N ha^{-1} yr^{-1} is proposed based on 'expert judgement'.

9.5.2 Broadleaved evergreen forest (T2)

Mediterranean evergreen *Quercus* forest (T21)

A $CL_{emp}N$ lower than 26 kg N ha^{-1} yr^{-1} was proposed for evergreen cork-oak (*Quercus suber*) woodlands in Portugal along a gradient from a point-source barn using epiphytic lichen functional groups diversity (Pinho et al., 2012). Increasing N deposition promoted a change from oligotrophic to nitrophytic communities. A long-term monitoring study in a dense forest dominated by holm-oak (*Quercus ilex*) and beech (*Fagus sylvatica*) in north-eastern Spain found that an average total atmospheric inorganic N deposition of 14.3 kg N ha^{-1} yr^{-1} was mostly retained in the catchments and only 2% was exported as DIN (Avila et al., 2020; Avila and Rodà, 2012). However, NO_3^- concentrations in the stream increased in recent years probably linked to forest maturation and climate warming. Another study comparing four holm-oak forests in Spain with a range of measured total inorganic N deposition of 9.4-28.9 kg N ha^{-1} yr^{-1} (Garcia-Gomez et al., 2018) found that the forest with the lowest deposition registered events of high concentrations of NO_3^- in the soil water (up to 28.15 mg NO_3-N l^{-1} at 20 cm depth) linked to ephemeral pulses of N deposition with the first rainfall events after the summer drought (García-Gomez et al., 2016). This phenomenon of temporal losses of N caused by a temporal asynchrony between N availability and plant N demand has been described as typical in Mediterranean ecosystems (Meixner and Fenn, 2004).

A fertilisation experiment of a holm-oak forest dominated by the evergreen tree *Quercus ilex* and the tall shrub *Phillyrea latifolia*, accompanied by other Mediterranean shrub species, with 60 kg N ha^{-1} yr^{-1} over background deposition, showed that after one year of N addition, the biomass of all microbial groups increased except for fungi. This increased the relative dominance of bacteria (Peguero et al., 2021). The higher bacterial biomass strongly favoured oribatid mites over springtails, which caused changes in the structure of the mesofauna arthropod community. This propagated the effects of N inputs throughout the soil food web. The effect of increasing N availability may be compensated by the strong constraint of drought on soil microbe activity and biomass.

With regard to forest ground vegetation, a $CL_{emp}N$ of 5-15 kg N ha^{-1} yr^{-1} is proposed for Mediterranean xeric grasslands (see Chapter 7.2.2), communities that typically constitute the understory vegetation of Dehesas (Spain) and Montados (Portugal), a traditional agroforestry system with open evergreen oak woodland and an herbaceous layer.

Based on the new information available, a $CL_{emp}N$ of 10-15 kg N ha^{-1} yr^{-1} is proposed for Mediterranean evergreen *Quercus* forests (T21), based on 'expert judgement'.

9.5.3 Coniferous forests (T3)

Mediterranean mountain *Abies* forest (T33)

N effects have been described in Mediterranean mountain *Abies* forests (T33, formerly G3.1) of the endemic fir *Abies pinsapo* along a gradient of atmospheric N deposition in the south of Spain. Three comparable fir forests were analysed in a bulk N deposition gradient ranging from 3.5 to 10.4 kg N ha^{-1} yr^{-1}. Chronic N deposition reduced fine root biomass, decreased photosynthetic nutrient use efficiency and shifted forests from N l to P limitation (Blanes et al., 2013a, b). The site with the highest N deposition showed changes in foliar N:P stoichiometry, smaller photosynthetic nitrogen use efficiency (PNUE), higher photosynthetic phosphorus use efficiency (PPUE) and indications of initial soil N saturation and N losses, compared to sites with up to 4.9 kg N ha^{-1} yr^{-1} of N deposition (Blanes et al., 2013a, b). The results indicated that the atmospheric N bulk deposition of 10.4 kg N ha^{-1} yr^{-1} had already exceeded the threshold where increasing N availability was beneficial for tree growth and physiology. Since dry deposition needs to be considered, a $CL_{emp}N$ of 10-15 kg N ha^{-1} yr^{-1} can be proposed for the protection of Mediterranean fir forests (T33), based on 'expert judgement'.

Mediterranean montane *Pinus sylvestris- Pinus nigra* forest (T37)

No specific fertilisation experiments or gradient studies are available for montane pine forests in the Mediterranean region. Sardans et al. (2016) described how N deposition was causing nutrient imbalances with increasingly limiting roles of P and other nutrients such as K in European *P. sylvestris* forests, including forests in the Mediterranean region. However, these effects were more evident in the central area of distribution of this species where higher levels of N deposition were observed. In the most recent revision of empirical critical loads, a value of 3 to 15 kg N ha^{-1} yr^{-1} was established for Mediterranean *Pinus* woodland based on 'expert judgement' of the results of Californian studies on N deposition effects in *Pinus ponderosa* (Fenn et al., 2008). A subsequent review established a $CL_{emp}N$ of 3 to 39 kg N ha^{-1} yr^{-1} for Mediterranean mixed-conifer forests (Pardo et al., 2011). The lowest $CL_{emp}N$ was based on lichen chemistry and community changes. For NO_3^- leaching and fine root biomass the critical load was 17 kg N ha^{-1} yr^{-1}. For soil acidification and understory biodiversity the critical load was 26 kg N ha^{-1} yr^{-1}. These values took into account throughfall N deposition. Since some N deposited on leaf surfaces could have been absorbed by plants or the phyllosphere, particularly in smaller deposition areas, a $CL_{emp}N$ of 5 to 17 kg N ha^{-1} yr^{-1} is proposed, based on 'expert judgement', for the protection of Mediterranean montane *Pinus* forests (T37).

Mediterranean lowland to submontane *Pinus* forest (*P. pinaster, P. halepensis, P. pinea*) (T3A)

The structure and functioning of microbial communities in biocrusts were studied in a N gradient study across semiarid Mediterranean ecosystems throughout Spain including shrubland, grassland and woodland sites (Ochoa-Hueso et al., 2013, 2016). This gradient included two *Pinus halepensis* woodlands. The authors found that the species abundance of both soil bacteria and fungi was reduced with N availabilities while green algae and cyanobacteria richness increased, thereby contributing to ecosystem eutrophication (Ochoa-Hueso et al., 2013, 2016). Since linear responses were found in most soil indicators along the 4.3-7.3 kg N ha^{-1} yr^{-1} gradient, a $CL_{emp}N$ of 4.3 kg N ha^{-1} yr^{-1} was proposed as the lowest end of the gradient (Ochoa-Hueso et al., 2013). In another N gradient in Spain including 28 sites of *Quercus coccifera* shrublands (n=18) and *P. halepensis* forests (n=10) covering a range of modelled inorganic N deposition between 4.4 and 8.1 kg N ha^{-1} yr^{-1}, soil acidification was detected in *Q. coccifera* but not *in P. halepensis* sites (Ochoa-Hueso et al., 2014). On the other hand, N deposition increased the C and N stored in the soils of Aleppo pine forests. A $CL_{emp}N$ of 5-10 kg N ha^{-1} yr^{-1} may be sufficient to protect Mediterranean *Pinus* forests (T3A), based on 'expert judgement'. This critical load is a good fit with the $CL_{emp}N$ of 5-15 kg N ha^{-1} yr^{-1} established as 'quite reliable', for Garrigue shrublands (S6) that frequently accompany these forests.

Effects on soil microbiota in Mediterranean ecosystems

The abundance of soil bacteria and fungi was studied in an N gradient study across semiarid Mediterranean ecosystems throughout Spain including sites in shrubland, grassland and woodland (Ochoa-Hueso, 2016). The author observed a reduction in the species abundance of both bacteria and fungi with N availabilities above 4-7 kg N ha^{-1} yr^{-1}. In an earlier study on AMF spore abundance in a coastal sage scrub vegetation in California, also representative for Mediterranean ecosystems, a shift in AM community composition along a gradient in N deposition from 10 to 35 kg N ha^{-1} yr^{-1} was observed (Egerton-Warburton and Allen, 2000).

9.5.4 Summary

Table 9.3 summarises the $CL_{emp}N$ ranges of the Mediterranean ecosystems.

Table 9.3. $CL_{emp}N$ and effects of exceedances on Mediterranean forest ecosystems (EUNIS class T). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Mediterranean *Fagus* forest on acid soils	**T18**	**10-15**	**(#)**	Annual height and volume tree growth; analogy to temperate *Fagus* forest
Mediterranean evergreen *Quercus* forest	**T21**	**10-15**	**(#)**	NO_3^- in soil water and streams
Mediterranean mountain *Abies* forest	**T33**	**10-15**	**(#)**	Tree foliar stoichiometry, tree physiology; soil N losses
Mediterranean montane *Pinus sylvestris-Pinus nigra* forest	**T37**	**5-17**	**(#)**	Lichen chemistry and community changes in Mediterranean mixed-conifer forests in USA
Mediterranean lowland to submontane *Pinus* forest (*P. pinaster, P. halepensis, P. pinea*)	**T3A**	**5-10**	**(#)**	C and N stored in soils; changes in soil microorganism communities

9.6 Overall summary for forests and other wooded land (EUNIS class T)

Table 9.4 gives an overview of the $CL_{emp}N$ ranges for forests and other wooded land.

Table 9.4. $CL_{emp}N$ and effects of exceedances on forests and other wooded land (T). ## reliable, # quite reliable and (#) expert judgement. Changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2011 reliability	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Broadleaved deciduous forest	T1	10-20	##	10-**15**	##	Changes in soil processes; nutrient imbalance; altered composition mycorrhiza and ground vegetation
Fagus forest on non-acid and acid soils	T17, T18	10-20	(#)	10-**15**	(#)	Changes in ground vegetation and mycorrhiza; nutrient imbalance, changes in soil fauna
Mediterranean *Fagus* forest on acid soils	T18			**10-15**	**(#)**	Annual height and volume tree growth; analogy to temperate Fagus forest
Acidophilous *Quercus* forest	T1B	10-15	(#)	10-15	(#)	Decrease in mycorrhiza; loss of epiphytic lichens and bryophytes; changes in ground vegetation

Ecosystem type	**EUNIS code**	**2011 kg N ha^{-1} yr^{-1}**	**2011 reliability**	**2022 kg N ha^{-1} yr^{-1}**	**2022 reliability**	**Indication of exceedance**
Carpinus and *Quercus* mesic deciduous forest	T1E	15-20	(#)	15-20	(#)	Changes in ground vegetation
Mediterranean evergreen *Quercus* forest	T21	10-20	(#)	10-**15**	(#)	NO_3 in soil water and streams
Coniferous forests	T3	5-15	##	**3**-15	##	Changes in soil processes, nutrient imbalance; altered composition mycorrhiza and ground vegetation; increase in mortality with drought
Temperate mountain *Picea* forest, Temperate mountain *Abies* forest	T31, T32	10-15	(#)	10-15	(#)	Decreased biomass of fine roots; nutrient imbalance; decrease in mycorrhiza; changed soil fauna
Mediterranean mountain *Abies* forest	T33			**10-15**	**(#)**	Tree foliar stoichiometry; tree physiology; soil N losses
Temperate continental *Pinus sylvestris* forest	T35	5-15	#	5-15	#	Changes in ground vegetation and mycorrhiza; nutrient imbalances; increased N_2O and NO emissions
Mediterranean montane *Pinus sylvestris-Pinus nigra* forest	T37			**5-17**	**(#)**	Lichen chemistry and community changes in Mediterranean mixed-conifer forests in USA
Mediterranean lowland to submontane *Pinus* forest	T3A	3-15	(#)	**5-10**	(#)	Reduction in fine-root biomass; shift in lichen community
Picea abies, dark taiga	T3F	5-10	##	**3-5** [a]	##	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation
Pinus sylvestris light taiga	T3G	5-10	#	**2-5** [a]	#	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation

a) Mainly based on N deposition impacts on lichens and bryophytes

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9.8 References

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M. (1989). Nitrogen saturation in northern forest ecosystems. *BioScience* **39**, 378-386.

Aber, J., McDowell, W. and Nadelhoffer, K. (1998). Nitrogen saturation in temperate forest ecosystems. *BioScience* **48**, 921-934.

Adamczyk, S., Kitunen, V., Lindroos, A.-J., Adamczyk, B. and Smolander, A. (2016). Soil carbon and nitrogen cycling processes and composition of terpenes five years after clear-cutting a Norway spruce stand: Effects of logging residues. *Forest Ecology and Management* **381**, 318-326.

Adriaenssens, S., Hansen, K., Staelens, J., Wuyts K., De Schrijver A., Baeten L., Boeckx P., Samson R. and Verheyen, K. (2012). Throughfall deposition and canopy exchange processes along a vertical gradient within the canopy of beech (*Fagus sylvatica* L.) and Norway spruce (*Picea abies* (L.) Karst). *Science of The Total Environment* **420**, 168-182.

Agren, G. I. (1983). Nitrogen productivity of some conifers. *Canadian Journal of Forest Research* **13**, 494-500.

Aguillaume, L., Avila, A., Pinho, P., Matos, P., Llop, E. and Branquinho, C. (2017). The critical levels of atmospheric ammonia in a Mediterranean holm-oak forest in north-eastern Spain. *Water Air and Soil Pollution* **228**, 13.

Ahrends, B., Schmitz, A., Prescher, A.-K., Wehberg, J., Geupel, M., Henning, A. and Meesenburg, H. (2020). Comparison of methods for the estimation of total inorganic nitrogen deposition to forests in Germany. *Frontiers in Forests and Global Change* **3**, 1-22.

Akselsson, C., Westling, O. and Örlander, G. (2004). Regional mapping of nitrogen leaching from clearcuts in southern Sweden. *Forest Ecology and Management* **202**, 235-243.

Akselsson, C., Belyazid, S., Hellsten, S., Klarqvist, M., Pihl-Karlsson, G., Karlsson, P.-E. and Lundin, L. (2010). Assessing the risk of N leaching from forest soils across a steep N deposition gradient in Sweden. *Environmental Pollution* **158**, 3588-3595.

Almeida, J.P., Rosenstock, N. P., Forsmark, B., Bergh, J. and Wallander, H. (2019). Ectomycorrhizal community composition and function in a spruce forest transitioning between nitrogen and phosphorus limitation. *Fungal Ecology* **40**, 20-31.

Ambus, P. and Robertson, G.P. (2006). The effect of increased N deposition on nitrous oxide, methane and carbon dioxide fluxes from unmanaged forest and grassland communities in Michigan. *Biogeochemistry* **79**, 315-337.

Anders, S., Beck, W., Bolte, A., Hofmann, G., Jenssen, M., Krakau, U. and Müller, J. (2002). *Ökologie und Vegetation der Wälder Nordostdeutschlands*, Dr. Kessel, Oberwinter.

Andrew, C., Halvorsen, R., Heegaard, E., Kuyper, T.W., Heilmann-Clausen, J., Krisai-Greilhuber, I., Bässler, C., Egli, S., Gange, A.C., Høiland, K., Kirk, P.M., Senn-Irlet, B., Boddy, L., Büntgen, U. and Kauserud, H. (2018). Continental-scale macrofungal assemblage patterns correlate with climate, soil carbon and nitrogen deposition. *Journal of Biogeography* **45**, 1942-1953.

Arnolds, E. (1985). *Veranderingen in de paddestoelenflora (mycoflora), Hoagwoud (NL)*, p. 101.

Arnolds, E. (1991). Decline of ectomycorrhizal fungi in Europe. *Agriculture, Ecosystems & Environment* **35**, 209-244.

Augustin, S., Bolte, A., Holzhausen, M. and Wolff, B. (2005). Exceedance of critical loads of nitrogen and sulphur and its relation to forest conditions. *European Journal of Forest Research* **124**, 289-300.

Avila, A. and Rodà, F. (2012). Changes in atmospheric deposition and streamwater chemistry over 25 years in undisturbed catchments in a Mediterranean mountain environment. *Science of The Total Environment* **434**, 18-27.

Avila, A., Aguillaume, L., Izquieta-Rojano, S., García-Gómez, H., Elustondo, D., Santamaría, J. and Alonso, R. (2017). Quantitative study on nitrogen deposition and canopy retention in Mediterranean evergreen forests. *Environmental Science and Pollution Research* **24**, 26213-26226.

Avila, A., Molowny-Horas, R. and Camarero, L. (2020). Stream chemistry response to changing nitrogen and sulfur deposition in two mountain areas in the Iberian Peninsula. *The Science of the Total Environment* **711**, 134697.

Avis, P.G., Mueller, G.M. and Lussenhop, J. (2008). Ectomycorrhizal fungal communities in two North American oak forests respond to nitrogen addition. *New Phytologist* **179**, 472-483.

Bahl-Butterbach, K., Gasche, R., Breuer, L. and Papen, H. (1997). Fluxes of NO and N_2O from temperate forest soils: impact of forest type, N deposition and of liming on the NO and N_2O emissions. *Nutrient Cycling in Agroecosystems* **48**, 79-90.

Bahr, A., Ellström, M., Akselsson, C., Ekblad, A., Mikusinska, A. and Wallander, H. (2013). Growth of ectomycorrhizal fungal mycelium along a Norway spruce forest nitrogen deposition gradient and its effect on nitrogen leakage. *Soil Biology and Biochemistry* **59**, 38-48.

Balsberg-Påhlsson, A. (1992). Influence of nitrogen fertilization on minerals, carbohydrates, amino acids and phenolic compounds in beech (*Fagus sylvatica*) leaves. *Tree Physiology* **10**, 93-100.

Bardgett, R. D. and Wardle, D. A. (2010). *Aboveground-belowground linkages: biotic interactions, ecosystem processes and global change*, Oxford University Press.

Berg, B. and Meentemeyer, V. (2002). Litter quality in a north European transect versus carbon storage potential. *Plant and Soil* **242**, 83-92.

Berg, M.P. and Verhoef, H. A. (1998). Ecological characteristics of a nitrogen-saturated coniferous forest in the Netherlands. *Biology and Fertility of Soils* **26**, 258-267.

Bernhardt-Römermann, M., Römermann, C., Pillar, V. D., Kudernatsch, T. and Fischer, A. (2010). High functional diversity is related to high nitrogen availability in a deciduous forest – evidence from a functional trait approach. *Folia Geobotanica* **45**, 111-124.

Bernhardt-Römermann, M., Baeten, L., Craven, D., De Frenne, P., Hédl, R., Lenoir, J., Bert, D., Brunet, J., Chudomelová, M., Decocq, G., Dierschke, H., Dirnböck, T., Dörfler, I., Heinken, T., Hermy, M., Hommel, P., Jaroszewicz, B., Keczyński, A., Kelly, D.L., Kirby, K.J., Kopecký, M., Macek, M., Máliš, F., Mirtl, M., Mitchell, F.J.G., Naaf, T., Newman, M., Peterken, G., Petřík, P., Schmidt, W., Standovár, T., Tóth, Z., Van Calster, H., Verstraeten, G., Vladovič, J., Vild, O., Wulf, M. and Verheyen, K. (2015). Drivers of temporal changes in temperate forest plant diversity vary across spatial scales. *Global Change Biology* **21**, 3726-3737.

Bigras, F. J., Ryyppö, A., Lindström, A. and Stattin, E. (2001). Cold acclimation and deacclimation of shoots and roots of conifer seedlings. In: Bigras, F.J., and Colombo, S.J. (eds.) *Conifer cold hardiness*. Kluwer Academic Publishers, Dordrecht, The Netherlands.

Blanes, C., Viñegla, B., Merino, J. and Carreira, J. A. (2013a). Nutritional status of Abies pinsapo forests along a nitrogen deposition gradient: do C/N/P stoichiometric shifts modify photosynthetic nutrient use efficiency? *Oecologia* **171**, 797-808.

Blanes, M. C., Viñegla, B., Salido, M. T. and Carreira, J. A. (2013b). Coupled soil-availability and tree-limitation nutritional shifts induced by N deposition: insights from N to P relationships in Abies pinsapo forests. *Plant and Soil* **366**, 67-81.

Blodgett, J. T., Kruger, E. L. and Stanosz, G. R. (1997). Effects of moderate water stress on disease development by *Sphaeropsis sapinea* on red pine. *Phytopathology* **87**, 422-434.

Blondeel, H., Perring, M. P., Depauw, L., De Lombaerde, E., Landuyt, D., De Frenne, P. and Verheyen, K. (2020). Light and warming drive forest understorey community development in different environments. *Global Change Biology*, **26**, 1681-1696.

Bobbink, R., Hornung, M. and Roelofs, J. G. M. (1996). *Empirical nitrogen critical loads for natural and semi-natural ecosystems*. In: Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded, UNECE Convention on Long-range Transboundary Air Pollution, Federal Environmental Agency, Berlin.

Bobbink, R. (2004). *Plant species richness and the exceedance of empirical nitrogen critical loads: an inventory*. Report Landscape Ecology, Utrecht University, Utrecht, The Netherlands.

Bobbink, R. and Hettelingh, J.-P. (2011). *Review and revision of empirical critical loads and dose-response relationships. Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010*, CCE.

Bolsinger, M. and Flückiger, W. (1989). Ambient air pollution induced changes in amino acid pattern of phloem sap in host plants—relevance to aphid infestation. *Environmental Pollution* **56**, 209-216.

Borken, W. and Matzner, E. (2004). Nitrate leaching in forest soils: an analysis of long-term monitoring sites in Germany. *Journal of Plant Nutrition and Soil Science* **167**, 277-283.

Bost, F. (1991). *Evolution de la flore herbacée forestière sur deux sites de la forêt de Haye entre 1972 et 1991*. Ecole Nationale Supérieure d'Agronomie et des Industries Alimentaires; INRA Seichamps, pp. 1-31.

Bowden, R. D., Davidson, E., Savage, K., Arabia, C. and Steudler, P. (2004). Chronic nitrogen additions reduce total soil respiration and microbial respiration in temperate forest soils at the Harvard Forest. *Forest Ecology and Management* **196**, 43-56.

Boxman, D., Van Dijk, H., Houdijk, A. and Roelofs, J. (1988). Critical loads for nitrogen, with special emphasis on ammonium. In: Nilsson J, Grennfelt P (eds.) *Critical loads for sulphur and nitrogen*. Skokloster, Sweden 19-24 March, 1988, pp 295-323.

Boxman, A. W., Krabbendam, H., Bellemakers, M. J. and Roelofs, J. G. (1991). Effects of ammonium and aluminium on the development and nutrition of *Pinus nigra* in hydroculture. *Environmental Pollution*, **73**, 119-136.

Boxman, A. W. and Van Dijk, H. (1994). Soil and vegetation responses to decreased atmospheric nitrogen and sulphur inputs into a Scots pine stand in The Netherlands. *Forest Ecology and Management* **68**, 39-45.

Boxman, A. W., Van Dam, D., Van Dijk, H. F. G., Hogervorst, R. F. and Koopmans, C. J. (1995). Ecosystem responses to reduced nitrogen and sulphur inputs into two coniferous forest stands in the Netherlands. *Forest Ecology and Management* **71**, 7-29.

Boxman, A. W., Van der Ven, P. J. M. and Roelofs, J. G. M. (1998). Ecosystem recovery after a decrease in nitrogen input to a Scots pine stand at Ysselsteyn, the Netherlands. *Forest Ecology and Management* **101**, 155-163.

Brandrud, T. E. and Timmermann, V. (1998). Ectomycorrhizal fungi in the NITREX site at Gardsjön, Sweden; below- and above-ground responses to experimentally-changed nitrogen inputs 1990-1995. *Forest Ecology and Management* **101***,* 207-214.

Branzanti, M. B., Rocca, E. and Pisi, A. (1999). Effect of ectomycorrhizal fungi on chestnut ink disease. *Mycorrhiza* **9**, 103-109.

Braun, S., Cantaluppi, L. and Flückiger, W. (2005). Fine roots in stands of *Fagus sylvatica* and *Picea abies* along a gradient of soil acidification. In *Environmental Pollution* **137**, 574-579.

Braun, S., Flückiger, W. and Braun, S. (2009). *Wie geht es unserem Wald?* Bericht 3, Schönenbuch: Institut für Angewandte Pflanzenbiologie.

Braun, S., Thomas, V. F. D., Quiring, R. and Flückiger, W. (2010). Does nitrogen deposition increase forest production? The role of phosphorus. *Environmental Pollution* **158**, 2043-2052.

Braun, S., Schindler, C. and Rihm, B. (2017). Growth trends of beech and Norway spruce in Switzerland: The role of nitrogen deposition, ozone, mineral nutrition and climate. *Science of the Total Environment* **599**-**600**, 637-646.

Braun, S., Hopf, S.-E. and De Witte, L.C. (2018). *Wie geht es unserem Wald? 34 Jahre Jahre Walddauerbeobachtung.*, Schönenbuch: Institut für angewandte Pflanzenbiologie. Retrieved from https://iap.ch/waldbericht.html

Braun, S., Schindler, C. and Rihm, B. (2020a). Foliar nutrient concentrations of European beech in Switzerland: relations with nitrogen deposition, ozone, climate and soil chemistry. *Frontiers in Forests and Global Change*, **3**, 1-15.

Braun, S., Tresch, S. and Augustin, S. (2020b). Soil solution in Swiss forest stands: A 20 year's time series. *PLOS ONE*, **15**, e0227530.

Braun, S., Ahrends, B., Alonso, R., Augustin, S., Garcia-Gomez, H., Hajjar, N., Karlsson, P.-E., Pihl-Karlsson, G., Schmitz, A. and Thimonier, A. (2022a). Nitrogen deposition in forests: throughfall and total deposition. In prep.

Braun, S., Rihm, B. and Schindler, C. (2022b). Epidemiological estimate of growth reduction by ozone in *Fagus sylvatica* L. and *Picea abies* Karst: sensitivity analysis and comparison with experimental results. *Plants* **11**, 777.

Brown, N., Vanguelova, E., Parnell, S., Broadmeadow, S. and Denman, S. (2018). Predisposition of forests to biotic disturbance: Predicting the distribution of Acute Oak Decline using environmental factors. *Forest Ecology and Management* **407**, 145-154.

Brunet, J., Diekmann, M. and Falkengren-Grerup, U. (1998). Effects of nitrogen deposition on field layer vegetation in south Swedish oak forests. *Environmental Pollution* **102**(1), 35-40.

Bruteig, I. E. (1993). The epiphytic lichen *Hypogymnia physodes* as a biomonitor of atmospheric nitrogen and sulphur deposition in Norway. *Environmental Monitoring and Assessment* **26**, 27-47.

Butterbach-Bahl, K., Breuer, L., Gasche, R., Willibald, G. and Papen, H. (2002). Exchange of trace gases between soils and the atmosphere in Scots pine forest ecosystems of the northeastern German lowlands. *Forest Ecology and Management* **167**, 123-134.

Cape, J.N., Freersmith, P.H., Paterson, I.S., Parkinson, J.A. and Wolfenden, J. (1990). The nutritional-status of *Picea abies* (L) Karst across Europe and implications for forest decline. *Trees-Structure and Function* **4**, 211-224.

Cape, J.N., Van der Eerden, L.J., Sheppard, L.J., Leith, I.D. and Sutton, M.A. (2009). Reassessment of critical levels for atmospheric ammonia. In: Sutton, M.A., Reis, S. and Baker, S.M. (eds.) *Atmospheric Ammonia*. Springer, Dordrecht, pp. 15-40.

Carreiro, M.M., Sinsabaugh, R.L., Repert, D.A. and Parkhurst, D.F. (2000). Microbial enzyme shifts explain litter decay responses to simulated nitrogen deposition. *Ecology* **81**, 2359-2365.

Chapin, F. S., Vitousek, P. M. and Van Cleve, K. (1986). The nature of nutrient Limitation in plant communities. *The American Naturalist* **127**, 48-58.

Chen, Y. and Högberg, P. (2006). Gross nitrogen mineralization rates still high 14 years after suspension of N input to a N-saturated forest. *Soil Biology and Biochemistry* **38**, 2001-2003.

Cheng, Y., Wang, J., Ge, Z., Zhang, J., Cai, Y., Chang, S.X., Cai, Z. and Chen, H.Y.H. (2020). Background nitrogen deposition controls the effects of experimental nitrogen addition on soil gross N transformations in forest ecosystems. *Biogeochemistry* **151**, 335-341.

Choma, M., Rappe-George, M. O., Bárta, J., Čapek, P., Kaštovská, E., Gärdenäs, A.I. and Šantrůčková, H. (2017). Recovery of the ectomycorrhizal community after termination of long-term nitrogen fertilisation of a boreal Norway spruce forest. *Fungal Ecology* **29**, 116-122.

Choma, M., Tahovská, K., Kaštovská, E., Bárta, J., Růžek, M. and Oulehle, F. (2020). Bacteria but not fungi respond to soil acidification rapidly and consistently in both a spruce and beech forest. *FEMS Microbiology Ecology* **96**, 1-13.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**(4), 648-675.

Clemmensen, K.E., Bahr, A., Ovaskainen, O., Dahlberg, A., Ekblad, A., Wallander, H., Stenlid, J., Finlay, R.D., Wardle, D.A. and Lindahl, B.D. (2013). Roots and associated fungi drive long-term carbon sequestration in boreal forest. *Science* **339**, 1615-1618.

Clemmensen, K. E., Finlay, R. D., Dahlberg, A., Stenlid, J., Wardle, D. and Lindahl, B. D. (2014). Carbon sequestration is related to mycorrhizal fungal community shifts during long term succession in boreal forests. *New Phytologist* **205**, 1525-1536.

CLRTAP (2017). *Mapping Critical Loads for Ecosytems. Chapter V of Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. Update 2017-09-10*, UNECE Convention on Long-range Transboundary Air Pollution. Retrieved from https://www.umweltbundesamt.de/sites/default/files/medien/4292/dokumente/ch5-mapman-2017-09-10.pdf

Commission, E. (2009). *Natura 2000 in the Meditarranean Region*.

Cornelissen, J.H.C., Callaghan, T.V and Alatalo, J.M. (2001). Global change and arctic ecosystems: is lichen decline a function of increases in vascular biomass? *Journal of Ecology* **89**, 984-994.

Cox, F., Barsoum, N., Bidartondo, M.I., Børja, I., Lilleskov, E., Nilsson, L.O., Rautio, P., Tubby, K. and Vesterdal, L. (2010a). A leap forward in geographic scale for forest ectomycorrhizal fungi. *Annals of Forest Science* **67**, 200.

Cox, F., Barsoum, N., Lilleskov, E.A. and Bidartondo, M.I. (2010b). Nitrogen availability is a primary determinant of conifer mycorrhizas across complex environmental gradients. *Ecology Letters* **13**, 1103-1113.

Currie, W. S., Aber, J. D., McDowell, W. H., Boone, R. D. and Magill, A. H. (1996). Vertical transport of dissolved organic C and N under long-term N amendments in pine and hardwood forests. *Biogeochemistry* **35**, 471-505.

Dahlmann, L., Näsholm, T. and Palmqvist, K. (2002). Growth, nitrogen uptake and resource allocation in the two tripartite lichens *Nephroma arcticum* and *Peltigera aphthosa* during nitrogen stress. *New Phytologist* **153**, 307-315.

Dahlman, L., Persson, J., Näsholm, T. and Palmqvist, K. (2003). Carbon and nitrogen distribution in the green algal lichens *Hypogymnia physodes* and *Platismatia glauca* in relation to nutrient supply. *Planta* **217**, 41-48.

Dahlman, L., Persson, J., Palmqvist, K. and Näsholm, T. (2004). Organic and inorganic nitrogen uptake in lichens. *Planta* **219**, 459-467.

Davidson, E. A. (1991). Fluxes of nitrous and nitric oxide from terrestrial ecosystems. In J.E. Rogers and W.B. Whitman (eds.) *Microbial production and consumption of greenhouse gases: methane, nitrogen oxides and Halomethanes*, Washington D.C.: American Society for Microbiology, pp. 219-235.

Davidson, E. A. and Kingerlee, W. (1997). A global inventory of nitric oxide emissions from soils. *Nutrient Cycling in Agroecosystems* **48**, 37-50.

De Frenne, P., Rodríguez-Sánchez, F., De Schrijver, A., Coomes, D.A., Hermy, M., Vangansbeke, P. and Verheyen, K. (2015). Light accelerates plant responses to warming. *Nature Plants* **1**, 1-3.

De Kam, M., Versteegen, C. M., Van den Burg, J. and Van der Werf, D. C. (1991). Effects of fertilization with ammonium sulphate and potassium sulphate on the development of *Sphaeropsis sapinea* in Corsican pine. *Netherlands Journal of Plant Pathology* **97**, 265-274.

De Schrijver, A., De Frenne, P., Ampoorter, E., Van Nevel, L., Demey, A., Wuyts, K. and Verheyen, K. (2011). Cumulative nitrogen input drives species loss in terrestrial ecosystems. *Global Ecology and Biogeography* **20**, 803-816.

De Vries, W., Reinds, G.J., Van der Salm, C., Draaijers, G.P.J., Bleeker, A., Erisman, J.W., Auée, J., Gundersen, P., Kristensen, H.L., Van Dobben, H.F., De Zwart, D., Derome, J., Voogd, J.C.H. and Vel, E.M. (2001). *Intensive Monitoring of Forest Ecosystems in Europe*. EC-UN/ECE, Brussels, Geneva, 177 pp.

De Vries, W., Reinds, G.J., Van der Salm, C., Van Dobben, H., Erisman, J.W., De Zwart, D., Bleeker, A., Draaijers, G.-PJ., Gundersen, P., Vel, E.M. and Haussmann, T. (2003). Results on nitrogen impacts in the EC and UN/ECE ICP Forests programme. *Environmental Documentation* **164**,199-208. Berne, BUWAL.

De Vries, W., Wamelink, G. W. W., Van Dobben, H., Kros, J., Reinds, G.J., Mol-Dijkstra, J.P., Smart, S.M., Evans, C.D., Rowe, E.C., Belyazid, S., Sverdrup, H.U., Van Hinsberg, A., Posch, M., Hettelingh, J.-P, Spranger, T. and Bobbink, R. (2010). Use of dynamic soil - Vegetation models to assess impacts of nitrogen deposition on plant species composition: An overview. *Ecological Applications* **20**, 60-79.

De Witte, L.C., Rosenstock, N.P., Van der Linde, S. and Braun, S. (2017). Nitrogen deposition changes ectomycorrhizal communities in Swiss beech forests. *Science of the Total Environment* **605**-**606**, 1083-1096.

DeForest, J.L., Zak, D.R., Pregitzer, K.S. and Burton, A.J. (2004). Atmospheric nitrate deposition and the microbial degradation of cellobiose and vanillin in a northern hardwood forest. *Soil Biology and Biochemistry* **36**, 965-971.

DeHayes, D.H., Ingle, M.A. and Wark, C.E. (1989). Nitrogen fertilization enhances cold tolerance of red spruce seedlings. *Canadian Journal of Forest Research* **19**, 1037-1043.

Deleporte, S. and Tillier, P. (1999). Long-term effects of mineral amendments on soil fauna and humus in an acid beech forest floor. *Forest Ecology and Management* **118**, 245-252.

DeLuca, T.H., Zackrisson, O., Nilsson, M.-C. and Sellstedt, A. (2002). Quantifying nitrogen-fixation in feather moss carpets of boreal forests. *Nature* **419**, 917-920.

Denman, S., Brown, N., Kirk, S., Jeger, M. and Webber, J. (2014). A description of the symptoms of Acute Oak Decline in Britain and a comparative review on causes of similar disorders on oak in Europe. *Forestry* **87**, 535-551.

Depauw, L., Perring, M. P., Landuyt, D., Maes, S.L. Blondeel, H., De Lombaerde, E., Brūmelis, G., Brunet, J., Closset-Kopp, D., Czerepko, J., Decocq, G., Den Ouden, J., Gawryś, R., Härdtle, W., Hédl, R., Heinken, T., Heinrichs, S., Jaroszewicz, B., Kopecký, M., Liepiņa, I., Macek, M., Máliš, F., Schmidt, W., Smart, S.M., Ujházy, K., Wulf, M. and Verheyen, K. (2020). Light availability and land-use history drive biodiversity and functional changes in forest herb layer communities. *Journal of Ecology* **108**, 1411-1425.

Desie, E., Vancampenhout, K., Nyssen, B., Van den Berg, L., Weijters, M., Van Duinen, G.-J., Den Ouden, J., Van Meerbeek, K. and Muys, B. (2020). Litter quality and the law of the most limiting: Opportunities for restoring nutrient cycles in acidified forest soils. *Science of the Total Environment* **699**, 134383.

Dickie, I. A., Xu, B. and Koide, R.T. (2002). Vertical niche differentiation of ectomycorrhizal hyphae in soil as shown by T-RFLP analysis. *New Phytologist* **156**, 527-535.

Diekmann, M., Brunet, J., Rühling, Å. and Falkengren-Grerup, U. (1999). Effects of nitrogen deposition: Results of a temporal-spatial analysis of deciduous forests in South Sweden. *Plant Biology* **1**, 471-481.

Dirkse, G.M. and Van Dobben, H.F. (1989). Het effect van bemesting op de samenstelling van de kruidlaag van dennenbossen. *Natura* **9**, 208-212.

Dirkse, G.M. and Martakis, G.F.P. (1992). Effects of fertilizer on bryophytes in Swedish experiments on forest fertilization. *Biological Conservation* **59**, 155-161.

Dirkse, G. M. (1993). Forest communities in the Netherlands (in Dutch), Stichting uitgeverij KNNV, Utrecht

Dirnböck, T., Grandin, U., Bernhardt-Römermann, M., Beudert, B., Canullo, R., Forsius, M., Grabner, M.-T., Holmberg, M., Kleemola, S., Lundin, L., Mirtl, M., Neumann, M., Pompei, E., Salemaa, M., Starlinger, F., Staszewski, T. and Uziębło, A.K. (2014). Forest floor vegetation response to nitrogen deposition in Europe. *Global Change Biology* **20**, 429-440.

Dirnböck , T., Djukic, I., Kitzler, B., Kobler, J., Mol-Dijkstra, J.P., Posch, M., Reinds, G.J., Schlutow, A., Starlinger, F. and Wamelink, W.G.W. (2017). Climate and air pollution impacts on habitat suitability of Austrian forest ecosystems. *Plos One* **12**, 16.

Dirnböck , T., Proll, G., Austnes, K., Beloica, J., Beudert, B., Canullo, R., De Marco, A., Fornasier, M.F., Futter, M., Goergen, K., Grandin, U., Holmberg, M., Lindroos, A.-J., Mirtl, M., Neirynck, J., Pecka, T., Nieminen, T.M., Nordbakken, J.-F., Posch, M., Reinds, G.-J., Rowe, E.C., Salemaa, M., Scheuschner, T., Starlinger, F., Uziębło, A.K., Valinia, S., Weldon, J., Wamelink, W.G.W. and Forsius, M. (2018). Currently legislated decreases in nitrogen deposition will yield only limited plant species recovery in European forests. *Environmental Research Letters* **13**, 11.

Dirnböck, T., Brielmann, H., Djukic, I., Geiger, S., Hartmann, A., Humer, F., Kobler, J., Kralik, M., Liu, Y., Mirtl, M. and Pröll, G. (2020). Long- and short-term inorganic nitrogen runoff from a karst catchment in Austria. *Forests* **11**, 1-20.

Dise, N. B., Rothwell, J. J., Gauci, V., Van der Salm, C. and De Vries, W. (2009). Predicting dissolved inorganic nitrogen leaching in European forests using two independent databases. *Science of The Total Environment* **407**, 1798-1808.

Draaijers, G. P. J., Erisman, J. W., Spranger, T. and Wyers, G. P. (1996). The application of throughfall measurements for atmospheric deposition monitoring. *Atmospheric Environment* **30**, 3349-3361.

Du, E., Van Doorn, M. and De Vries, W. (2021). Spatially divergent trends of nitrogen versus phosphorus limitation across European forests. *Science of The Total Environment* **771**, 145391.

Dunfield, P. F., Liesack, W., Henckel, T., Knowles, R. and Conrad, R. (1999). High-Affinity Methane Oxidation by a Soil Enrichment Culture Containing a Type II Methanotroph. *Applied and Environmental Microbiology* **65**, 1009-1014.

Duquesnay, A., Dupouey, J. L., Clement, A., Ulrich, E. and Le Tacon, F. (2000). Spatial and temporal variability of foliar mineral concentration in Beech (*Fagus sylvatica*) stands in northeastern France. *Tree Physiology* **20**, 13-22.

Edfast, A.B., Näsholm, T. and Ericsson, A. (1990). Free amino acid concentrations in needles of Norway spruce and Scots pine trees on different sites in areas with two levels of nitrogen deposition. *Canadian Journal of Forest Research* **20**, 1132-1136.

Edfast, A.B., Näsholm, T., Aronsson, A. and Ericsson, A. (1996). Applications of mineral nutrients to heavily N-fertilized Scots Pine trees: effects on arginine and mineral nutrient concentrations. *Plant and Soil* **184**, 57-65.

EFI. (2002). *Causes and consequences of increased forest growth in Europe*. Conference held in Copenhagen on Aug 28, 2002.

Egerton-Warburton, L.M. and Allen, E.B. (2000). Shifts in arbuscular mycorrhizal communities along an anthropogenic nitrogen deposition gradient. *Ecological Applications* **10**, 484-496.

Ellenberg, H. (1985). Veraenderungen der flora mitteleuropas unter dem Einfluss von Duengung und Immissionen. *Schweiz. z. Forstwes*, pp. 19-39.

Ellenberg, H. (1988). *Vegetation ecology of Central Europe*, Cambridge: Univ. Press.

Elling, W, Heber, U., Polle, A., Beese, F. (2007). *Schädigung von Waldökosystemen. Auswirkungen anthropogener Umweltveränderungen und Schutzmassnahmen*, München: Elsevier.

Emmet, B.A., Boxman, D., Bredemeier, M., Gundersen, P., Kjønas, O.J., Moldan, F., Schleppi, P., Tietema, A. and Wright, R.F. (1998). Predicting the effects of atmospheric nitrogen depositon in conifer stands: evidence from the NITREX ecosystem-scale experiments. *Ecosystems* **1**, 352-360.

Emmet, B.A. (1999). The impact of nitrogen on forest soils and feedbacks on tree growth. *Water, Air and Soil Pollution* **116**, 65-74.

Emmett, B. (2002). *The impact of nitrogen deposition in forest ecosystems: a review*, Bangor: Centre for Ecology and Hydrology.

Ericsson, A., Walheim, M., Nordén, L.-G., Näsholm, T., Norden, L. G. and Nasholm, T. (1995). Concentrations of mineral nutrients and arginine in needles of *Picea abies* trees from different areas in southern Sweden in relation to nitrogen deposition and humus form. *Effects of Acid Deposition and Tropospheric Ozone on Forest Ecosystems in Sweden* **44**, 147-157.

Erland, S. and Taylor, A. F. S. (2001). Diversity of ectomycorrhizal fungal communities in relation to the abiotic environment. (Van der Heijden, Ed.), Berlin: Springer, pp. 163-200.

Esseen, P.-A., Ekström, M., Westerlund, B., Palmqvist, K., Jonsson, B.G., Grafström, A. and Ståhl, G. (2016). Broad-scale distribution of epiphytic hair lichens correlates more with climate and nitrogen deposition than with forest structure. *Canadian Journal of Forest Research* **46**, 1348-1358.

Evans, C., Goodale, C., Caporn, S., Dise, N., Emmett, B., Fernandez, I., Field, C., Findlay, S., Lovett, G., Meesenburg, H., Moldan, F. and Sheppard, L. (2008). Does elevated nitrogen deposition or ecosystem recovery from acidification drive increased dissolved organic carbon loss from upland soil? A review of evidence from field nitrogen addition experiments. *Biogeochemistry* **91**,13-35.

Falkengren-Grerup, U. (1986). Falkengren-Grerup, U. (1986). Soil acidification and vegetation changes in deciduous forest in southern Sweden. *Oecologia* **70**, 339-347.

Falkengren-Grerup, U. (1995). Long-term changes in flora and vegetation in deciduous forests of southern Sweden. *Ecological Bulletins* **44**, 215-226.

Falkengren-Grerup, U., Brunet, J. and Diekmann, M. (1998). Nitrogen mineralisation in deciduous forest soils in south Sweden in gradients of soil acidity and deposition. *Environmental Pollution* **102**, 415-420.

Falkengren-Grerup, U. and Diekmann, M. (2003). Use of a gradient of N-deposition to calculate effect-related soil and vegetation measures in deciduous forests. *Forest Ecology and Management* **180**, 113-124.

Fenn, M.E., Poth, M.A. and Johnson, D.W. (1996). Evidence for nitrogen saturation in the San Bernardino Mountains in southern California. *Forest Ecology and Management* **82**, 211-230.

Fenn, M. E., Jovan, S., Yuan, F., Geiser, L., Meixner, T. and Gimeno, B. S. (2008). Empirical and simulated critical loads for nitrogen deposition in California mixed conifer forests. *Environmental Pollution* **155**, 492-511.

Fenn, M.E., Bytnerowicz, A., Schilling, S.L. and Ross, C.S. (2015). Atmospheric deposition of nitrogen, sulfur and base cations in jack pine stands in the Athabasca Oil Sands Region, Alberta, Canada. *Environmental Pollution (Barking, Essex: 1987)* **196**, 497-510.

Flechard, C.R., Nemitz, E., Smith, R.I., Fowler, D., Vermeulen, A.T., Bleeker, A., Erisman, J.W., Simpson, D., Zhang, L., Tang, Y.S. and Sutton, M.A. (2011). Dry deposition of reactive nitrogen to European ecosystems: A comparison of inferential models across the NitroEurope network. *Atmospheric Chemistry and Physics* **11**, 2703–2728.

Fleischer, K., Rebel, K.T., Van der Molen, M.K., Erisman, J.W., Wassen, M.J., Van Loon, E.E., Montagnani, L., Gough, C. M., Herbst, M., Janssens, I.A. Gianelle, D. and Dolman, A. J. (2013). The contribution of nitrogen deposition to the photosynthetic capacity of forests. *Global Biogeochemical Cycles* **27**, 187-199.

Flückiger, W., Braun, S., Flückiger-Keller, H., Leonardi, S., Asche, N., Bühler, U. and Lier, M. (1986). Untersuchungen über Waldschäden in festen Buchenbeobachtungsflächen der Kantone Basel-Landschaft, Basel-Stadt, Aargau, Solothurn, Bern, Zürich und Zug. Schweiz. *Zeitschrift für Forstwesen* **137**, 917-1010.

Flückiger, W. and Braun, S. (1998). Nitrogen deposition in Swiss forests and its possible relevance for leaf nutrient status, parasite attacks and soil acidification. *Environmental Pollution* **102**, 69-76.

Flückiger, W. and Braun, S. (1999). Nitrogen and its effect on growth, nutrient status and parasite attacks in beech and Norway spruce. *Water, Air and Soil Pollution* **116**, 99-110.

Flückiger, W. and Braun, S. (2004). *Wie geht es unserem Wald? Ergebnisse aus Dauerbeobachtungsflächen von 1984 bis 2004*, Bericht 2, Schönenbuch: Institut für Angewandte Pflanzenbiologie, 67 pp.

Flückiger, W. and Braun, S. (2011). *Auswirkung erhöhter Stickstoffbelastung auf die Stabilität des Waldes. Synthesebericht*, Schönenbuch: Institut für Angewandte Pflanzenbiologie. Retrieved from http://www.bafu.admin.ch/wald/01198/01206/index.html?lang=de

Forsius, M., Posch, M., Holmberg, M., Vuorenmaa, J., Kleemola, S., Augustaitis, A., Beudert, B., Bochenek, W., Clarke, N., De Wit, H.A., Dirnböck, T., Frey, J., Grandin, U., Hakola, H., Kobler, J., Krám, P., Lindroos, A.-J., Löfgren, S., Pecka, T., Rönnback, P., Skotak, K., Szpikowski, J., Ukonmaanaho, L., Valinia, S. and Váňa, M. (2021). Assessing critical load exceedances and ecosystem impacts of anthropogenic nitrogen and sulphur deposition at unmanaged forested catchments in Europe. *Science of The Total Environment* **753**, 141791.

Forsmark, B., Nordin, A., Maaroufi, N. I., Lundmark, T. and Gundale, M. J. (2020). Low and High Nitrogen Deposition Rates in Northern Coniferous Forests Have Different Impacts on Aboveground Litter Production, Soil Respiration and Soil Carbon Stocks. *Ecosystems* **23**, 1423-1436.

Forstner, S. J., Wechselberger, V., Müller, S., Keibinger, K.M., Díaz-Pinés, E., Wanek, W., Scheppi, P., Hagedorn, F., Gundersen, P., Tatzber, M., Gerzabek, M.H. and Zechmeister-Boltenstern, S. (2019). Vertical redistribution of soil organic carbon pools after twenty years of nitrogen addition in two temperate coniferous forests. *Ecosystems* **22**, 379-400.

Forsum, Å., Dahlman, L., Näsohlm, T. and Nordin, A. (2006). Nitrogen utilization by *Hylocomium splendens* in a boreal forest fertilization experiment. *Functional Ecology* **20**, 421-426.

Fowler, D., Cape, J. N. and Unsworth, M. H. (1989). Deposition of atmospheric pollutants on forests. *Philosophical Transactions of the Royal Society of London. B, Biological Sciences* **324**, 247-265.

Frahm, J. (2013). Contents of amino acids and osmotic values of epiphytic lichens as indicators for regional atmospheric nitrogen loads. *Archive for Lichenology* **9**, 1-11.

Franklin, O., Högberg, P., Ekblad, A. and Ågren, G. I. (2003). Pine forest floor carbon accumulation in response to N and PK additions: Bomb ^{14}C modelling and respiration studies. *Ecosystems* **6**, 644-658.

Frey, S. D., Knorr, M., Parrent, J.L. and Simpson, R.T. (2004). Chronic nitrogen enrichment affects the structure and function of the soil microbial community in temperate hardwood and pine forests. *Forest Ecology and Management* **196**, 159-171.

Frey, S. D., Ollinger, S., Nadelhoffer, K., Bowden, R., Brzostek, E., Burton, A., Caldwell, B.A., Crow, S., Goodale, C.L., Grandy, A.S., Finzi, A., Kramer, M.G., Lajtha, K., LeMoine, J., Martin, M., McDowell, W.H., Minocha, R., Sadowsky, J.J., Templer, P.H. and Wickings, K. (2014). Chronic nitrogen additions suppress decomposition and sequester soil carbon in temperate forests. *Biogeochemistry* **121**, 305-316.

Futter, M. N., Skeffington, R. A., Whitehead, P. G. and Moldan, F. (2009). Modelling stream and soil water nitrate dynamics during experimentally increased nitrogen deposition in a coniferous forest catchment at Gårdsjön, Sweden. *Hydrology Research* **40**, 187-197.

Gan, H., Zak, D. R. and Hunter, M. D. (2013). Chronic nitrogen deposition alters the structure and function of detrital food webs in a northern hardwood ecosystem. *Ecological Applications* **23**, 1311-1321.

García-Gomez, H., Izquieta-Rojano, S., Aguillaume, L., González-Fernández, I., Valiño, F., Elustondo, D., Santamaría, J.M., Àvila, A., Fenn, M.E. and Alonso, R. (2016). Atmospheric deposition of inorganic nitrogen in Spanish forests of *Quercus ilex* measured with ion-exchange resins and conventional collectors. *Environmental Pollution* **216**, 653-661.

Garcia-Gomez, H., Izquieta-Rojano, S., Aguillaume, L., González-Fernández, I., Valiño, F., Elustondo, D., Santamaría, J.M., Àvila, A., Bytnerowicz, A., Bermejo, V. and Alonso, R. (2018). Joining empirical and modelling approaches to estimate dry deposition of nitrogen in Mediterranean forests. *Environmental Pollution* **243**, 427-436.

García-Gómez, H., Calvete-Sogo, H., González-Fernández, I., Rábago, I., Bermejo, V., Valiño, F., Sanz, J., Elvira, S. and Alonso, R. (2020). Atmospheric nitrogen deposition in Spain: Emission and deposition trends, critical load exceedances and effects on terrestrial ecosystems. In: Sutton, M.A. (ed.) *Just Enough Nitrogen*, Springer, Cham, pp. 319-328.

Gärdenfors, U., Waldén, H. W. and Wäreborn, I. (1995). Effects of soil acidification on forest land snails. *Ecological Bulletins* **44**, 259-270.

Gasche, R. and Papen, H. (1999). A 3-year continuous record of nitrogen trace gas fluxes from untreated and limed soil of a N-saturated spruce and beech forest ecosystem in Germany. 2. NO and NO_2 fluxes. *Journal of Geophysical Research Atmospheres* **104**, 505-520.

Geiser, L. H. and Neitlich, P. N. (2007). Air pollution and climate gradients in western Oregon and Washington indicated by epiphytic macrolichens. *Environmental Pollution* **145**, 203-218.

Geiser, L. H., Nelson, P. R., Jovan, S. E., Root, H. T. and Clark, C. M. (2019). Assessing ecological risks from atmospheric deposition of nitrogen and sulfur to US forests using epiphytic macrolichens. *Diversity* **11**, 87.

Geiser, L.H., Root, H.T., Smith, R.J., Jovan, S.E., Clair, L., Dillmann, K.L., (2021). Lichen-based critical loads for deposition of nitrogen and sulfur in US forests. *Environmental Pollution* **291**, 118187.

Gentilesca, T., Rita, A., Brunetti, M., Giammarchi, F., Leonardi, S., Magnani, F., Van Noije, T., Tonon, G. And Borghetti, M. (2018). Nitrogen deposition outweighs climatic variability in driving annual growth rate of canopy beech trees: Evidence from long-term growth reconstruction across a geographic gradient. *Global Change Biology* **24**, 2898-2912.

Gillet, F., Peter, M., Ayer, F., Butler, R. and Egli, S. (2010). Long-term dynamics of aboveground fungal communities in a subalpine Norway spruce forest under elevated nitrogen input. *Oecologia* **164**, 499-510.

Gilliam, F. S. (2006). Response of the herbaceous layer of forest ecosystems to excess nitrogen deposition. *Journal of Ecology* **94**, 1176-1191.

Gilliam, F.S., Welch, N.T., Phillips, A.H., Billmyer, J.H., Peterjohn, W.T., Fowler, Z.K., Walter, C.A., Burnham, M.B., May, J.D. and Adams, M.B. (2016). Twenty-five-year response of the herbaceous layer of a temperate hardwood forest to elevated nitrogen deposition. *Ecosphere* **7**, e01250.

Giordani, P., Calatayud, V., Stofer, S., Seidling, W., Granke, O. and Fischer, R. (2014). Detecting the nitrogen critical loads on European forests by means of epiphytic lichens. A signal-to-noise evaluation. *Forest Ecology and Management* **311**, 29-40.

Göransson, A. (1990). *Alger, lavar och Garrupprättning hos unggranar längs en kvävegradient Sverige-Holland*. Report SNV, Solna.

Grennfelt, P. and Thörnelöf, E. (1992). Critical Loads for Nitrogen. Report from a workshop held at Lökeberg, Sweden, 6.-10. April 1992. Organized by the Nordic Council of Ministers in collaboration with The Convention on Long-range Transboundary Air Pollution (UNECE). In: Grennfelt, P. and Thörnelöf, E. (eds.), 1-428.

Grip, H. (1982). *Water chemistry and runoff in forest streams at Kloten*, Uppsala.

Grosse-Branckmann, H. and Grosse-Branckmann, G. (1978). Zur Pilzflora der Umgebung von Darmstadt vor 50 Jahren und heute (ein Vergleich der floristischen Befunde Franz Kellenbachs aus der Zeit von 1918 bis 1942 mit dem gegenwärtigen Vorkommen der Arten). *Z. Mykol.* **44**, 257-269.

Guerrieri, R., Vanguelova, E. I., Michalski, G., Heaton, T. H. E. and Mencuccini, M. (2015). Isotopic evidence for the occurrence of biological nitrification and nitrogen deposition processing in forest canopies. *Global Change Biology* **21**, 4613-4626.

Guerrieri, R., Lecha, L., Mattana, S., Cáliz, J., Casamayor, E.O., Barceló, A., Michalski, G., Peñuelas, J., Avila, A. and Mencuccini, M. (2020). Partitioning between atmospheric deposition and canopy microbial nitrification into throughfall nitrate fluxes in a Mediterranean forest. *Journal of Ecology* **108**, 626-640.

Gundale, M. J., Deluca, T. H. and Nordin, A. (2011). Bryophytes attenuate anthropogenic nitrogen inputs in boreal forests. *Global Change Biology* **17**, 2743-2753.

Gundersen, P. (1998). Effects of enhanced nitrogen deposition in a spruce forest at Klosterhede, Denmark, examined by moderate NH_4NO_3 addition. *Forest Ecology and Management* **101**, 251-268.

Gundersen, P., Callesen, I. and De Vries, W. (1998a). Nitrate leaching in forest ecosystems is related to forest floor C/N ratios. *Environmental Pollution* **102**, 403-407.

Gundersen, P., Emmett, B. A., Kjønas, O. J., Koopmans, C. J. and Tietema, A. (1998b). Impact of nitrogen deposition on nitrogen cycling in forests: a synthesis of the NITREX data. *Forest Ecology and Management* **101**, 37-55.

Gundersen, P., Schmidt, I. K. and Raulund-Rasmussen, K. (2006). Leaching of nitrate from temperate forests - effects of air pollution and forest management. *Environmental Reviews* **14**, 1-57.

Hällgren, J.E. and Näsholm, T. (1988). Critical loads for nitrogen. Effects on forest ecosystems. In: Nilsson, J., and Grennfelt, P. (eds.). *Critical Loads for Sulphur and Nitrogen*. Report from a Workshop held at Skokloster, Sweden, 19-24 March 1988. pp. 323-342.

Harmens, H., Norris, D.A., Cooper, D.M., Mills, G., Steinnes, E., Kubin, E., Thöni, L., Aboal, J.R., Alber, R., Carballeira, A., Coşkun, M., De Temmerman, L., Frolova, M., González-Miqueo, L., Jeran, Z., Leblond, S., Liiv, S., Maňkovská, B., Pesch, R., Poikolainen, J., Rühling, Å., Santamaria, J.M., Simonèiè, P., Schröder, W., Suchara, I., Yurukova, L. and Zechmeister, H.G. (2011). Nitrogen concentrations in mosses indicate the spatial distribution of atmospheric nitrogen deposition in Europe. *Environmental Pollution* **159**, 2852-2860.

Harrison, A. F., Schulze, E.-D., Gebauer, G. and Bruckner, G. (2000). Canopy uptake and utilization of atmospheric pollutant nitrogen. In: Schulze, E.D. (eds.) *Carbon and nitrogen cycling in European forest ecosystems*. Ecological Studies, vol 142. Springer, Berlin, Heidelberg, pp. 171-188.

Hasselquist, N.J. and Högberg, P. (2014). Dosage and duration effects of nitrogen additions on ectomycorrhizal sporocarp production and functioning: An example from two N-limited boreal forests. *Ecology and Evolution* **4**, 3015-3026.

Hautier, Y., Niklaus, P.A. and Hector, A. (2009). Competition for light causes plant biodiversity loss after eutrophication. *Science* **324**(5927), 636-638.

Hawksworth, D.L. and McManus, P.M. (1989). Lichen recolonization in London under conditions of rapidly falling sulphur dioxide levels and the concept of zone skipping. *Botanical Journal of the Linnean Society* **100**, 99-109.

Heinrichs, S. and Schmidt, W. (2017). Biotic homogenization of herb layer composition between two contrasting beech forest communities on limestone over 50 years. *Applied Vegetation Science* **20**, 271-281.

Heinsdorf, D. and Schulzke, D. (1969). Zur Feinwurzelverteilung junger Kiefern und Roteichen in einem humusarmen Sandboden nach Tiefenumbruch bzw. Pflugstreifenbearbeitung und mineralischer Düngung. *Arch.Forstw.* **18**, 731-745.

Hellsten, S., Stadmark, J., Karlsson, G.P., Karlsson, P.E. and Akselsson, C. (2015). Increased concentrations of nitrate in forest soil water after windthrow in southern Sweden. *Forest Ecology and Management* **356**, 234-242.

Helm, N., Essl, F., Mirtl, M. and Dirnböck, T. (2017). Multiple environmental changes drive forest floor vegetation in a temperate mountain forest. *Ecology and Evolution* **7**, 2155-2168.

Henrys, P.A., Stevens, C.J., Smart, S.M., Maskell, L.C., Walker, K.J., Preston, C.D., Crowe, A., Rowe, E.C., Gowing, D.J. and Emmett, B.A. (2011). Impacts of nitrogen deposition on vascular plants in Britain: an analysis of two national observation networks. *Biogeosciences* **8**, 3501-3518.

Hess, C., Niemeyer, T., Fichtner, A., Jansen, K., Kunz, M., Maneke, M., von Wehrden, H., Quante, M., Walmsley, D., von Oheimb, G. and Härdtle, W. (2018). Anthropogenic nitrogen deposition alters growth responses of European beech (*Fagus sylvativa* L.) to climate change. *Environmental Pollution* **233,** 92-98.

Hesse, C. N., Mueller, R. C., Vuyisich, M., Gallegos-Graves, V., Gleasner, C.D., Zak, D.R. and Kuske, C.R. (2015). Forest floor community metatranscriptomes identify fungal and bacterial responses to N deposition in two maple forests. *Frontiers in Microbiology* **6**, 1-15.

Hettelingh, J.-P., Stevens, C. J., Posch, M., Bobbink, R. and Vries, W. de. (2015). Assessing the impacts of nitrogen deposition on plant species richness in Europe. *Environmental Pollution* **25**, 573-586.

Heuck, C., Smolka, G., Whalen, E.D., Frey, S., Gundersen, P., Moldan, F., Fernandez, I.J., Spohn, M. (2018). Effects of long-term nitrogen addition on phosphorus cycling in organic soil horizons of temperate forests. *Biogeochemistry* **141**, 167-181.

Hofmann, G. (1987). Vegetationsänderungen in Kiefernbeständen durch Mineraldüngung. *Hercynia N.F. Leipzig* **24**, 271-278.

Hofmann, G., Heinsdorf, G. and Krauss, H.H. (1990). Wirkung atmogener Stickstoffeinträge auf Produktivität und Stabilität von Kiefern-Forstökosystemen. *Beiträge Für Die Forstwirtschaft* **24**, 59-73.

Högberg, P., Fan, H., Quist, M., Binkley, D. and Tamm, C. O. (2006). Tree growth and soil acidification in response to 30 years of experimental nitrogen loading on boreal forest. *Global Change Biology* **12**, 489-499.

Högberg, M. N., Yarwood, S. A. and Myrold, D. D. (2014). Fungal but not bacterial soil communities recover after termination of decadal nitrogen additions to boreal forest. *Soil Biology and Biochemistry* **72**, 35-43.

Hora, F. B. (1959). Quantitative experiments on foodstool production in woods. *Trans.Br.Myc.Soc.* **42**, 1-14.

Horn, K.J., Thomas, R.Q., Clark, C.M., Pardo, L.H., Fenn, M.E., Lawrence, G.B., Perakis, S.S., Smithwick, E.A.H., Baldwin, D., Braun, S., Nordin, A., Perry, C.H., Phelan, J.N., Schaberg, P.G., St. Clair, S.B., Warby, R. and Watmough, S. (2018). Growth and survival relationships of 71 tree species with nitrogen and sulfur deposition across the conterminous U.S. *PloS One* **13**, e0205296.

Hu, Y., Zhao, P., Zhu, L., Zhao, X., Ni, G., Ouyang, L., Schäfer, K.V.R. and Shen, W. (2019). Responses of sap flux and intrinsic water use efficiency to canopy and understory nitrogen addition in a temperate broadleaved deciduous forest. *Science of the Total Environment* **648**, 325-336.

Huber, D. M. (1980). The role of mineral nutrition in defence. In: Horsfall, J.G. and Cowling, E.B. (eds.). *Plant Disease*. Academic Press, pp. 381-406.

Huhn, G. and Schulz, H. (1996). Contents of free amino acids in Scots pine needles from field sites with different levels of nitrogen deposition. *New Phytologist* **134**, 95-101.

Hülber, K., Dirnböck, T., Kleinbauer, I., Willner, W., Dullinger, S., Karrer, G. and Mirtl, M. (2008). Long-term impacts of nitrogen and sulphur deposition on forest floor vegetation in the Northern limestone Alps, Austria. *Applied Vegetation Science* **11**, 395-404.

Hůnová, I., Kurfürst, P., Vlček, O., Stráník, V., Stoklasová, P., Schovánková, J. and Srbová, D. (2016). Towards a better spatial quantification of nitrogen deposition: A case study for Czech forests. *Environmental Pollution* **213**, 1028–1041.

Hyvönen, R., Persson, T. andersson, S., Olsson, B., Ågren, G. I. and Linder, S. (2008). Impact of long-term nitrogen addition on carbon stocks in trees and soils in northern Europe. *Biogeochemistry* **89**, 121-137.

ICP Forests (2001). *Intensive Monitoring of Forest Ecosystems in Europe. Technical Report 2001*, Brussels, Geneva: EC-UN/ECE.

Insarova, I.D., Insarov, G.E., Bråkenhielm, S., Hultengren, S., Martinsson, P.O. and Semenov, S.M. (1992). *Lichen sensitivity and air pollution - a review of literature data*. Swedish Environmental Protection Agency Report 4007.

Ishida, T. A. and Nordin, A. (2010). No evidence that nitrogen enrichment affect fungal communities of Vaccinium roots in two contrasting boreal forest types. *Soil Biology and Biochemistry* **42**, 234-243.

Jandl, R., Kopeszki, H., Bruckner, A. and Hager, H. (2003). Forest Soil Chemistry and Mesofauna 20 Years After an Amelioration Fertilization. *Restoration Ecology* **11**, 239-246.

Janssens, I. A., Dieleman, W., Luyssaert, S., Subke, J.-A. Reichstein, M., Ceulemans, R., Ciais, P., Dolman, A.J., Grace, J., Matteucci, G., Papale, D., Piao, S.L., Schulze, E.-D., Tang, J. and Law, B. E. (2010). Reduction of forest soil respiration in response to nitrogen deposition. *Nature Geoscience* **3**, 315-322.

Jantsch, M.C., Fischer, A., Fischer, H.S. and Winter, S. (2013). Shift in Plant Species Composition Reveals Environmental Changes During the Last Decades: A Long-Term Study in Beech (*Fagus sylvatica*) Forests in Bavaria, Germany. *Folia Geobotanica* **48**, 467-491.

Jarvis, S., Woodward, S., Alexander, I.J. and Taylor, A.F.S. (2013). Regional scale gradients of climate and nitrogen deposition drive variation in ectomycorrhizal fungal communities associated with native Scots pine. *Global Change Biology* **19**, 1688-1696.

Jenssen, M., Butterbach-Bahl, K., Hofmann, G. and Papen, H. (2002). Exchange of trace gases between soils and the atmosphere in Scots pine forest ecosystems of the northeastern German lowlands. 2. A novel approach to scale up N_2O- and NO-fluxes from forest soils by modelling their relationships to vegetation structure. *Forest Ecology and Management* **167**, 135-147.

Jenssen, M. and Hofmann, G. (2005). Einfluss atmogener Stickstoffeinträge auf die Vielfalt der Vegetation in Wäldern Nordostdeutschlands. *Beiträge Forstwirtschaft Und Landschaftsökologie* **39**, 132-141.

Jenssen, M. (2009). Assessment of the effects of top-soil changes on plant species diversity in forests, due to nitrogen deposition. In: Hettelingh, J.P., Posch M. and Slootweg, J. (eds). *Progress in the modelling of critical thresholds, impacts to plant species diversity and ecosystem services in Europe: CCE Status Report*, Coordination Centre for Effects, pp. 83-99.

Johansson, O., Nordin, A., Olofsson, J. and Palmqvist, K. (2010). Responses of epiphytic lichens to an experimental whole-tree nitrogen-deposition gradient. *New Phytologist* **188**, 1075-1084.

Johansson, O., Olofsson, J., Giesler, R. and Palmqvist, K. (2011). Lichen responses to nitrogen and phosphorus additions can be explained by the different symbiont responses. *New Phytologist* **191**, 795-805.

Johansson, O., Palmqvist, K. and Olofsson, J. (2012). Nitrogen deposition drives lichen community changes through differential species responses. *Global Change Biology* **18**, 2626-2635.

Johnson, D. W. and Taylor, G. E. (1989). Role of air pollution in forest decline in Eastern North America. *Water, Air and Soil Pollution* **48**, 21-43.

Jonard, M., Fürst, A., Verstraeten, A., Thimonier, A., Timmermann, V., Potočić, N., Waldner, P., Benham, S., Hansen, K., Merilä, P., Ponette, Q., De la Cruz, A.C., Roskams, P., Nicolas, M., Croisé, L., Ingerslev, M., Matteucci, G., Decinti, B., Bascietto, M. and Rautio, P. (2015). Tree mineral nutrition is deteriorating in Europe. *Global Change Biology* **21**, 418-430.

Jones, M.E., Paine, T.D., Fenn, M.E. and Poth, M.A. (2004). Influence of ozone and nitrogen deposition on bark beetle activity under drought conditions. *Forest Ecology and Management* **200**, 67-76.

Jönsson, A.M., Ingersten, M. and Rauland-Rasmussen, K. (2004a). Frost sensitivity and nutrient status in a fertilized Norway spruce stand in Denmark. *Forest Ecology and Management* **201**, 199-209.

Jönsson, A.M., Rosengren, U. and Nihlgård, B. (2004b). Excess nitrogen affects the frost sensitivity of the inner bark of Norway spruce. *Annals of Forest Science* **61**, 293-298.

Karlsson, P.E., Akselsson, C., Hellsten, S. and Pihl Karlsson, G. (2018). A bark beetle attack caused elevated nitrate concentrations and acidification of soil water in a Norway spruce stand. *Forest Ecology and Management* **422**, 338-344.

Karlsson, P.E., Pihl Karlsson, G., Hellsten, S., Akselsson, C., Ferm, M. and Hultberg, H. (2019). Total deposition of inorganic nitrogen to Norway spruce forests – Applying a surrogate surface method across a deposition gradient in Sweden. *Atmospheric Environment* **217**, 116964.

Karlsson, P.E., Karlsson, G.P., Hellsten, S. and Akselsson, C. (2021). *Transport of different compounds in runoff from a catchment with Norway spruce in west Sweden, before and after final cut harvest*. IVL Report 569 (in Swedish with English summary).

Kaste, Ø., Rankinen, K. and Lepistö, A. (2004). Modelling impacts of climate and deposition changes on nitrogen fluxes in northern catchments of Norway and Finland. *Hydrology and Earth System Sciences* **8**(4), 778-792.

Kellner, P.S. and Redbo-Torstensson, P. (1995). Effects of elevated nitrogen deposition on the field-layer vegetation in coniferous forests. *Ecological Bulletins* **44**, 227-237.

Kennedy, F. (2003). *How extensive are the impacts of nitrogen pollution in Great Britain's forests?* Forest Research Annual Report and Accounts 2002-2003.

Kiebacher, T., Keller, C., Scheidegger, C. and Bergamini, A. (2017). Epiphytes in wooded pastures: Isolation matters for lichen but not for bryophyte species richness. *PLOS ONE* **12**, e0182065.

King, G.M. and Schnell, S. (1998). Effects of ammonium and non-ammonium salt additions on methane oxidation by *Methylosinus trichosporium* OB3b and Maine Forest Soils. *Applied and Environmental Microbiology* **64**, 253-257.

Kint, V., Aertsen, W., Campioli, M., Vansteenkiste, D., Delcloo, A. and Muys, B. (2012). Radial growth change of temperate tree species in response to altered regional climate and air quality in the period 1901-2008. *Climatic Change* **115**, 343-363.

Kjøller, R., Nilsson, L.O., Hansen, K., Kappel Schmidt, I., Vesterdal, L. and Gundersen, P. (2012). Dramatic changes in ectomycorrhizal community composition, root tip abundance and mycelial production along a stand-scale nitrogen deposition gradient. *New Phytologist* **194**, 278-286.

Klein, R.M., Perkins, T.D. and Myers, H.L. (1989). Nutrient status and winter hardiness in red spruce foliage. *Canadian Journal of Forest Research* **19**, 754-758.

Klemedtsson, L., Von Arnold, K., Weslien, P. and Gundersen, P. (2005). Soil CN ratio as a scalar parameter to predict nitrous oxide emissions. *Global Change Biology* **11**, 1142-1147.

Knorr, M., Frey, S.D. and Curtis, P.S. (2005). Nitrogen additions and litter decomposition: A Meta-Analysis. *Ecology* **86**, 3252-3257.

Korhonen, J.F.J., Pihlatie, M., Pumpanen, J., Aaltonen, H., Hari, P., Levula, J., Kieloaho, A.J., Nikinmaa, E., Vesala, T. and Ilvesniemi, H. (2013). Nitrogen balance of a boreal Scots pine forest. *Biogeosciences* **10**, 1083–1095.

Kraft, M., Schreiner, M., Reif, A. and Aldinger, E. (2000). Veränderung von Bodenvegetation und Humusauflage im Nordschwarzwald. *AFZ* **55**, 222-224.

Kubin, E. (1995). The effect of clear cutting, waste wood collecting and site preparation on the nutrient leaching to groundwater. In: Nilsson, L.O., Hüttl, R.F., Johansson, U.T. (eds.) *Nutrient uptake and cycling in forest ecosystems*. Developments in Plant and Soil Sciences, vol 62. Springer, Dordrecht, pp. 661-670.

Kuhn, N., Amiet, R. and Hufschmid, N. (1987). Veranderungen in der Waldvegetation der Schweiz infolge Nahrstoffanreicherungen aus der Atmosphare. *Berichte - Eidgenossische Anstalt Fur Das Forstliche Versuchswesen* **158**, 77-84.

Kvaalen, H., Solberg, S., Clarke, N., Torp, T. and Aamlid, D. (2002). Time series study of concentrations of SO_4^{2-} and H^+ in precipitation and soil waters in Norway. *Environmental Pollution* **117**, 215-224.

Kwon, T., Shibata, H., Kepfer-Rojas, S. et al. (2021). Effects of climate and atmospheric nitrogen deposition on early to mid-term stage litter decomposition across biomes. *Frontiers in Forests and Global Change*, **4**, 1.

Kytö, M., Niemelä, P. and Annila, E. (1996). Vitality and bark beetle resistance of fertilized Norway spruce. *Forest Ecology and Management* **84**, 149-157.

Kytöviita, M.-M. and Crittenden, P.D. (2007). Growth and nitrogen relations in the mat-forming lichens *Stereocaulon paschale* and *Cladonia stellaris*. *Annals of Botany* **100**, 1537-1545.

L'Hirondelle, S.J., Jacobson, J.S. and Lassoie, J.P. (1992). Acidic mist and nitrogen fertilization effects on growth, nitrate reductase activity, gas exchange and frost hardiness of red spruce seedlings. *New Phytologist* **121**, 611-622.

Laiho, O. (1970). *Paxillus involutus* as a mycorrhizal symbiont of forest trees. *Acta Forestalia Fennica* **79**, 1-35.

Lameire, S., Hermy, M. and Honnay, O. (2000). Two decades of change in the ground vegetation of a mixed deciduous forest in an agricultural landscape. *Journal of Vegetation Science* **11**, 695-704.

Latte, N., Perin, J., Kint, V., Lebourgeois, F. and Claessens, H. (2016). Major changes in growth rate and growth variability of beech (*Fagus sylvatica* L.) related to soil alteration and climate change in Belgium. *Forests* **7**, 174.

Latty, E. F., Canham, C. D. and Marks, P. L. (2003). Beech bark disease in northern hardwood forests: The importance of nitrogen dynamics and forest history for disease severity. *Canadian Journal of Forest Research* **33**, 257-268.

Lehtonen, A., Linkosalo, T., Peltoniemi, M., Sievänen, R., Mäkipää, R., Tamminen, P., Salemaa, M., Nieminen, T., Ťupek, B., Heikkinen, J. and Komarov, A. (2016). Forest soil carbon stock estimates in a nationwide inventory: evaluating performance of the ROMULv and Yasso07 models in Finland. *Geoscientific Model Development* **9**, 4169-4183.

Liang, X., Zhang, T., Lu, X., Ellsworth, D.S., BassiriRad, H., You, C., Wang, D., He, P., Deng, Q., Liu, H., Mo, J. and Ye, Q. (2020). Global response patterns of plant photosynthesis to nitrogen addition: A meta-analysis. *Global Change Biology* **26**, 3585-3600.

Lilleskov, E.A., Fahey, T.J. and Lovett, G.M. (2001). Ectomycorrhizal Fungal Aboveground Community Change over an Atmospheric Nitrogen Deposition Gradient. *Ecological Applications* **11**, 397.

Lilleskov, E. A., Fahey, T. J., Horton, T. R. and Lovett, G. M. (2002). Belowground ectomycorrhizal fungal community change over a nitrogen deposition gradient in Alaska. *Ecology* **83**, 104-115.

Lilleskov, E.A., Hobbie, E.A. and Horton, T.R. (2011). Conservation of ectomycorrhizal fungi: exploring the linkages between functional and taxonomic responses to anthropogenic N deposition. *Fungal Ecology* **4**, 174-183.

Lilleskov, E.A., Kuyper, T.W., Bidartondo, M.I., and Hobbie, E.A. (2018). Atmospheric nitrogen deposition impacts on the structure and function of forest mycorrhizal communities: A review. *Environmental Pollution* **246**, 148-162.

Lilleskov, E.A., Kuyper, T.W., Bidartondo, M.I., Hobbie, E.A., (2019). Atmospheric nitrogen deposition impacts on the structure and function of forest mycorrhizal communities: A review. *Environmental Pollution* **246**, 148-162.

Liu, X.-Y., Koba, K., Makabe, A., Li, X.-D., Yoh, M. and Liu, C.-Q. (2013). Ammonium first: natural mosses prefer atmospheric ammonium but vary utilization of dissolved organic nitrogen depending on habitat and nitrogen deposition. *The New Phytologist* **199**, 407-419.

Lladó, S., López-Mondéjar, R. and Baldrian, P. (2017). Forest soil bacteria: diversity, involvement in ecosystem processes and response to global change. *Microbiology and Molecular Biology Reviews* **81**, 1-27.

Löfgren, S., Ring, E., von Brömssen, C., Sørensen, R. and Högbom, L. (2009). Short-term effects of clear-cutting on the water chemistry of two boreal streams in northern Sweden: A paired catchment study. *Ambio* **38**, 347-356.

Lohm, U., Lundqvist, H., Persson, T. and Wirén, A. (1977). *Effects of nitrogen fertilization on the abundance of enchytraeids and microarthropods in Scots pine forests*. Retrieved from http://urn.kb.se/resolve?urn=urn:nbn:se:slu:epsilon-9-58

Lövblad, G. andersen, B., Joffre, S., Reissell, A., Pedersen, U. and Hovmand, M. (1992). *Mapping deposition of sulphur, nitrogen and base cations in the Nordic countries*.

Lovett, G.M., Arthur, M.A., Weathers, K.C., Fitzhugh, R.D. and Templer, P.H. (2013). Nitrogen addition increases carbon storage in soils, but not in trees, in an eastern U.S. deciduous forest. *Ecosystems* **16**, 980-1001.

Lu, M., Zhou, X., Luo, Y., Yang, Y., Fang, C., Chen, J. and Li, B. (2011). Minor stimulation of soil carbon storage by nitrogen addition: A meta-analysis. *Agriculture, Ecosystems and Environment* **140**, 234-244.

Ma, S., Verheyen, K., Props, R., Wasof, S., Vanhellemont, M., Boeckx, P., Boon, N. and De Frenne, P. (2018). Plant and soil microbe responses to light, warming and nitrogen addition in a temperate forest. *Functional Ecology* **32**, 1293-1303.

Maaroufi, N.I., Nordin, A., Hasselquist, N.J., Bach, L.H., Palmqvist, K. and Gundale, M.J. (2015). Anthropogenic nitrogen deposition enhances carbon sequestration in boreal soils. *Global Change Biology* **21**, 3169-3180.

Maaroufi, N.I., Nordin, A., Palmqvist, K., Hasselquist, N.J., Forsmark, B., Rosenstock, N.P., Wallander, H. and Gundale, M.J. (2019). Anthropogenic nitrogen enrichment enhances soil carbon accumulation by impacting saprotrophs rather than ectomycorrhizal fungal activity. *Global Change Biology* **25**, 2900-2914.

Maes, S. L., Perring, M. P., Vanhellemont, M., Depauw, L., Van den Bulcke, J., Brūmelis, G., Brunet, J., Decocq, G., den Ouden, J., Härdtle, W., Zédl, R., Heinken, T., Heinrichs, S., Jaroszewicz, B., Kopecký, M., Máliš, F., Wulf, M. and Verheyen, K. (2019). Environmental drivers interactively affect individual tree growth across temperate European forests. *Global Change Biology* **25**, 201-217.

Magill, A. and Aber, J. D. (1998). Long-term effects of experimental nitrogen additions on foliar litter decay and humus formation in forest ecosystems. *Plant and Soil* **203**, 301-311.

Magill, A., Aber, J.D., Currie, W.S., Nadelhoffer, K.J., Martin, M.E., McDowell, W.H., Melillo, J.M., Steudler, P.S. (2004). Ecosystem response to 15 years of chronic nitrogen additions at the Harvard Forest LTER, Massachusetts, USA. *Forest Ecology and Management* **196** 7-28.

Mäkipää, R. (1994). Effects of nitrogen fertilization on the humus layer and ground vegetation under closed canopy in boreal coniferous stands. *Silva Fennica*, **28**, 81-94.

Mäkipää, R. (1998). Sensitivity of understorey vegetation to nitrogen and sulphur deposition in a spruce stand. *Ecological Engineering* **10**, 87-95.

Mäkipää, R. and Heikkinen, J. (2003). Large-scale changes in abundance of terricolous bryophytes and macrolichens in Finland. *Journal of Vegetation Science* **14**, 497-508.

Maljanen, M., Jokinen, H., Saari, A., Strommer, R. and Martikainen, P.J. (2006). Methane and nitrous oxide fluxes and carbon dioxide production in boreal forest soil fertilized with wood ash and nitrogen. *Soil Use and Management* **22**, 151-157.

Mannerkoski, H., Finér, L., Piirainen, S. and Starr, M. (2005). Effect of clear-cutting and site preparation on the level and quality of groundwater in some headwater catchments in eastern Finland. *Forest Ecology and Management* **220**, 107-117.

Manninen, O.H., Stark, S., Kytöviita, M.-M., Lampinen, L. and Tolvanen, A. (2009). Understorey plant and soil responses to disturbance and increased nitrogen in boreal forests. *Journal of Vegetation Science* **20**, 311-322.

Manninen, S. (2018). Deriving nitrogen critical levels and loads based on the responses of acidophytic lichen communities on boreal urban *Pinus sylvestris* trunks. *The Science of the Total Environment* **613**-**614**, 751-762.

Marx, D.H. (1969). The influence of ectotrophic mycorrhizal fungi on the resistance of pine roots to pathogenic infections. I Antagonism of mycorrhizal fungi to root pathogenic fungi and soil bacteria. *Phytopathology* **59**, 153-163.

Matzner, E. and Murach, D. (1995). Soil changes induced by air pollutant deposition and their implication for forests in Central Europe. *Water Air and Soil Pollution* **85**, 63-76.

McClure, M.S. (1980). Foliar nitrogen: a basis for host suitability for elongate hemlock scale, *Fiorinia externa* (Homoptera: Diaspididae). *Ecology* **61**, 72-79.

McCune, B., Geiser, L. (2009). *Macrolichens of the Pacific Northwest*, 3rd edn, Cornwallis, Ore, USA: Oregon State University.

Meixner, T. and Fenn, M. (2004). Biogeochemical budgets in a Mediterranean catchment with high rates of atmospheric N deposition – Importance of scale and temporal asynchrony. *Biogeochemistry* **70**, 331-356.

Meyer, M., Schröder, W., Nickel, S., Leblond, S., Lindroos, A.-J., Mohr, K., Poikolainen, J., Santamaria, J.M., Skudnik, M., Thöni, L., Beudert, B., Dieffenbach-Fries, H., Schulte-Bisping, H. and Zechmeister, H.G. (2015). Relevance of canopy drip for the accumulation of nitrogen in moss used as biomonitors for atmospheric nitrogen deposition in Europe. *Science of the Total Environment* **538**, 600-610.

Midolo, G., Alkemade, R., Schipper, A.M., Benítez-López, A., Perring, M.P. and De Vries, W. (2019). Impacts of nitrogen addition on plant species richness and abundance: A global meta-analysis. *Global Ecology and Biogeography* **28**, 398-413.

Mitchell, R.J., Truscott, A.M., Leith, I.D., Tang, Y.S., Van Dijk, N., Smith, R.I. and Sutton, M.A. (2003). *Impact of atmospheric nitrogen deposition on epiphytes in Atlantic oakwoods*. Environmental Documentation 164, 265-272. Berne, SAEFL.

Moldan, F., Wright, R.F., Löfgren, S., Forsius, M., Ruoho-Airola, T. and Skjelkvåle, B.L. (2001). Long-term changes in acidification and recovery at nine calibrated catchments in Norway, Sweden and Finland. *Hydrology and Earth System Sciences* **5**, 339-350.

Moldan, F., Jutterstrom, S., Hruska, J. and Wright, R.F. (2018). Experimental addition of nitrogen to a whole forest ecosystem at Gardsjon, Sweden (NITREX): Nitrate leaching during 26 years of treatment. *Environmental Pollution* **242**, 367-374.

Moraal, L. G. (1996). Bionomics of Haematoloma dorsatum (Hom., Cercopidae) in relation to needle damage in pine forests. *Anz.Schädlingskde., Pflanzenschutz, Umweltschutz* **69**, 114-118.

Morrison, E.W., Frey, S.D., Sadowsky, J.J., Van Diepen, L.T.A., Thomas, W.K. and Pringle, A. (2016). Chronic nitrogen additions fundamentally restructure the soil fungal community in a temperate forest. *Fungal Ecology* **23**, 48-57.

Murach, D. and Parth, A. (1999). Feinwurzelwachstum von Fichten beim Dach-Projekt im Solling. *AFZ/Der Wald* **54**, 58-60.

Mustajarvi, K., Merila, P., Derome, J., Lindroos, A-J., Helmisaari, H-S., Nöjd, P. and Ukonmaanaho, L. (2008). Fluxes of dissolved organic and inorganic nitrogen in relation to stand characteristics and latitude in Scots pine and Norway spruce stands in Finland. *Boreal Environment Research* **13**, 3-21.

Muys, B. and Granval, P. (1997). Earthworms as bio-indicators of forest site quality. *Soil Biology and Biochemistry* **29**, 323-328.

Nabuurs, G.J., Lindner, M., Verkerk, P.J., Gunia, K., Deda, P., Michalak, R. and Grassi, G. (2013). First signs of carbon sink saturation in European forest biomass. *Nature Climate Change* **3**, 792-796.

Nadelhoffer, K.J., Emmett, B.A., Gundersen, P., Kjønaas, O.J., Koopmans, C.J., Schleppi, P., Tietema, A. and Wright, R.F. (1999). Nitrogen deposition makes a minor contribution to carbon sequestration in temperate forests. *Nature* **398**, 145-148.

Näsholm, T. and Ericsson, A. (1990). Seasonal changes in amino acids, protein and total nitrogen in needles of fertilized Scots pine trees. *Tree Physiology* **6**, 267-281.

Näsholm, T., Nordin, A., Edfast, A.-B. and Högberg, P. (1997). Identification of Coniferous Forests with Incipient Nitrogen Saturation through Analysis of Arginine and Nitrogen-15 Abundance of Trees. *Journal of Environmental Quality* **26**, 302-309.

NEGTAP (2001). *Transboundary air pollution: acidification, eutrophication and ground level ozone in the UK*. Prepared by the Expert Group on Transboundary Air Pollution, Defra Contract EPG 1/3/153., London: Department for Environment, Food and Rural Affairs (DEFRA).

Neirynck, J., Kowalski, A.S., Carrara, A., Genouw, G., Berghmans, P. and Ceulemans, R. (2007). Fluxes of oxidised and reduced nitrogen above a mixed coniferous. *Environmental Pollution Volume* **149**, 31-43.

Nellemann, C. and Thomsen, M.G. (2001). Long-term changes in forest growth: Potential effects of nitrogen deposition and acidification. *Water, Air and Soil Pollution* **128**, 197-205.

Nilsen, P. (1995). Effect of nitrogen on drought strain and nutrient uptake in Norway spruce *Picea abies* (L.) Karst.) trees. *Plant and Soil* **172**, 73-85.

Nilsson, J. and Grennfelt, P. (1988). Critical loads for sulphur and nitrogen. Report from a workshop held at Skokloster, Sweden, 19-24 March, 1988, 418 pp.

Nilsson, L.O., Bååth, E., Falkengren-Grerup, U. and Wallander, H. (2007). Growth of ectomycorrhizal mycelia and composition of soil microbial communities in oak forest soils along a nitrogen deposition gradient. *Oecologia* **153**, 375-384.

Nordin, A., Näsholm, T. and Ericson, L. (1998). Effects of simulated N deposition on understorey vegetation of a boreal coniferous forest. *Functional Ecology* **12**, 691-699.

Nordin, A. and Gunnarsson, U. (2000). Amino acid accumulation and growth of *Sphagnum* under different levels of N deposition. *Ecoscience* **7**, 474-480.

Nordin, A., Strengbom, J., Witzell, J., Näsholm, T. and Ericson, L. (2005). Nitrogen deposition and the biodiversity of boreal forests: Implications for the nitrogen critical load. *AMBIO: A Journal of the Human Environment* **34**, 20-24.

Nordin, A., Strengbom, J. and Ericson, L. (2006). Responses to ammonium and nitrate additions by boreal plants and their natural enemies. *Environmental Pollution* **141**, 167-174.

Nordin, A., Strengbom, J., Forsum, Å. and Ericson, L. (2009). Complex biotic interactions drive long-term vegetation change in a nitrogen enriched boreal forest. *Ecosystems* **12**, 1204-1211.

Norrström, A.-C. (2002). *Även skogen läcker näring*, Linköping University Electronic Press.

Nybakken, L., Lie, M.H., Julkunen-Tiitto, R., Asplund, J. and Ohlson, M. (2018). Fertilization Changes Chemical Defense in Needles of Mature Norway Spruce (*Picea abies*). *Frontiers in Plant Science* **9**, 9.

Oberle, B., Grace, J.B. and Chase, J.M. (2009). Beneath the veil: plant growth form influences the strength of species richness-productivity relationships in forests. *Global Ecology and Biogeography* **18**), 416-425.

Ochoa-Hueso, R., Allen, E. B., Branquinho, C., Cruz, C., Dias, T., Fenn, M.E., Manrique, E., Pérez-Corona, M.E., Sheppard, L.J. and Stock, W.D. (2011). Nitrogen deposition effects on Mediterranean-type ecosystems: An ecological assessment. *Environmental Pollution* **159**, 2265-2279.

Ochoa-Hueso, R., Maestre, F. T., De Los Ríos, A., Valea, S., Theobald, M.R., Vivanco, M.G., Manrique, E. and Bowker, M.A. (2013). Nitrogen deposition alters nitrogen cycling and reduces soil carbon content in low-productivity semiarid Mediterranean ecosystems. *Environmental Pollution* **179**, 185-193.

Ochoa-Hueso, R., Arroniz-Crespo, M., Bowker, M.A., Maestre, F.T., Pérez-Corona, M.E., Theobald, M.R., Vivanco, M.G. and Manrique, E. (2014). Biogeochemical indicators of elevated nitrogen deposition in semiarid Mediterranean ecosystems. *Environmental Monitoring and Assessment* **186**, 5831-5842.

Ochoa-Hueso, R. (2016). Nonlinear disruption of ecological interactions in response to nitrogen deposition. *Ecology* **97**, 2802-2814.

Ochoa-Hueso, R., Delgado-Baquerizo, M., Gallardo, A., Bowker, M.A. and Maestre, F.T. (2016). Climatic conditions, soil fertility and atmospheric nitrogen deposition largely determine the structure and functioning of microbial communities in biocrust-dominated Mediterranean drylands. *Plant and Soil* **399**, 271-282.

Ochoa-Hueso, R., Munzi, S., Alonso, R., Arróniz-Crespo, M., Avila, A., Bermejo, V., Bobbink, R., Branquinho, C., Concostrina-Zubiri, L., Cruz, C., Cruz de Carvalho, R., De Marco, A., Dias, T., Elustondo, D., Elvira, S., Estébanez, B., Fusaro, L., Gerosa, G., Izquieta-Rojano, S., Lo Cascio, M., Marzuoli, R., Matos, P., Mereu, S., Merino, J., Morillas, L., Nunes, A., Paoletti, E., Paoli, L., Pinho, P., Rogers, I.B., Santos, A., Sicard, P., Stevens, C.J. and Theobald, M.R. (2017). Ecological impacts of atmospheric pollution and interactions with climate change in terrestrial ecosystems of the Mediterranean Basin: Current research and future directions. *Environmental Pollution* **227**, 194-206.

Ohenoja, E. (1988). Behaviour of mycorrhizal fungi in fertilized soils. *Karstenia* **28**, 27-30.

Økland, R. H. (1995). Changes in the occurrence and abundance of plant species in a Norwegian boreal coniferous forest, 1988-1993. *Nordic Journal of Botany* **15**, 415-438.

Ostonen, I., Truu, M., Helmisaari, H.-S., Lukac, M., Borken, W., Vanguelova, E., Godbold, D.L., Lõhmus, K., Zang, U., Tedersoo, L., Preem, J.-K., Rosenvald, K., Aosaar, J., Armolaitis, K., Frey, J., Kabral, N., Kukumägi, M., Leppälammi-Kujansuu, J., Lindroos, A.-J., Merilä, P., Napa, Ü., Nöjd, P., Parts, K., Uri, V., Varik, M. and Truu, J. (2017). Adaptive root foraging strategies along a boreal–temperate forest gradient. *New Phytologist* **215**, 977-991.

Oulehle, F., Goodale, C.L., Evans, C.D., Chuman, T., Hruška, J., Krám, P., Navrátil, T., Tesař, M., Ač, A., Urban, O. and Tahovská, K. (2021). Dissolved and gaseous nitrogen losses in forests controlled by soil nutrient stoichiometry. *Environmental Research Letters* **16**, 064025.

Påhlsson, A. M. B. (1992). Influence of nitrogen fertilization on minerals, carbohydrates, amino acids and phenolic compounds in beech (*Fagus sylvatica* L.) leaves. *Tree Physiology* **10**, 101-110.

Pan, Y., Birdsey, R. A., Fang, J., Houghton, R., Kauppi, P.E., Kurz, W.A., Phillips, O.L., Shvidenko, A., Lewis, S.L., Canadell, J.G., Ciais, P., Jackson, R.B., Pacala, S.W., McGuire, A.D., Piao, S., Rautiainen, A., Sitch, S. and Hayes, D. (2011). A large and persistent carbon sink in the world's forests. *Science* **333**, 988-993.

Papen, H. and Butterbach-Bahl, K. (1999). A 3-year continuous record of nitrogen trace gas fluxes from untreated and limed soil of a N-saturated spruce and beech forest ecosystem in Germany: 1. N_2O emissions. *Journal of Geophysical Research: Atmospheres* **104**, 18487-18503.

Pardo, L.H., Fenn, M.E., Goodale, C.L., Geiser, L.H., Driscoll, C.T., Allen, E.B., Baron, J.S., Bobbink, R., Bowman, W.D., Clark, C.M., Emmett, B., Gilliam, F.S., Greaver, T.L., Hall, S.J., Lilleskov, E.A., Liu, L., Lynch, J.A., Nadelhoffer, K.J., Perakis, S.S., Robin-Abbott, M.J., Stoddard, J.L., Weathers, K.C. and Dennis, R.L. (2011). Effects of nitrogen deposition and empirical nitrogen critical loads for ecoregions of the United States. *Ecological Applications* **21**, 3049-3082.

Park, J.-H. and Matzner, E. (2006). Detrital control on the release of dissolved organic nitrogen (DON) and dissolved inorganic nitrogen (DIN) from the forest floor under chronic N deposition. *Environmental Pollution* **143**, 178-185.

Payne R.J., Dise, N.B., Stevens, C.J., Gowing, D.J. and BEGIN Partners (2013). Impact of nitrogen deposition at the species level. *Proceedings of the National Academy of Sciences* **110**(3), 984-987.

Payne, R.J., Campbell, C., Britton, A.J., Mitchell, R.J., Pakeman, R.J., Jones, L., Ross, L.C., Stevens, C.J., Field, C., Caporn, S.J.M., Carroll, J., Edmondson, J.L., Carnell, E.J., Tomlinson, S., Dore, A.J., Dise, N. and Dragosits, U. (2019). What is the most ecologically-meaningful metric of nitrogen deposition? *Environmental Pollution* **247**, 319-331.

Pearce, I.S.K. and Van der Wal, R. (2008). Interpreting nitrogen pollution thresholds for sensitive habitats: The importance of concentration versus dose. *Environmental Pollution* **152**, 253-256.

Peguero, G., Folch, E., Liu, L., Ogaya, R. and Peñuelas, J. (2021). Divergent effects of drought and nitrogen deposition on microbial and arthropod soil communities in a Mediterranean forest. *European Journal of Soil Biology* **103**, 103275.

Peng, Y.F., Peng, Z.P., Zeng, X.T. and Houx, J.H. (2019). Effects of nitrogen-phosphorus imbalance on plant biomass production: a global perspective. *Plant and Soil* **436**, 245-252.

Perkins, T.D., Adams, G.T., Lawson, S.T., Schaberg, P.G. and McNulty, S.G. (1999). Long-term nitrogen fertilization increases winter injury in montane Red Spruce (*Picea rubens*) foliage. *Journal of Sustainable Forestry* **10**, 165-172.

Perring, M.P., Bernhardt-Römermann, M., Baeten, L., Midolo, G. Blondeel, H., Depauw, L., Landuyt, D. Maes, S.L., De Lombaerde, E., Carón, M.M., Vellend, M., Brunet, J., Chudomelová, M., Decocq, G., Diekmann, M., Dirnböck, T., Dörfler, I., Durak, T., De Frenne, P., Gilliam, F.S., Hédl, R., Heinken, T., Hommel, P., Jaroszewicz, B., Kirby, K.J., Kopecký, M., Lenoir, J., Li, D., Máliš, F., Mitchell, F.J.G., Naaf, T., Newman, M., Petřík, P., Reczyńska, K., Schmidt, W., Standovár, T., Świerkosz, K., Van Calster, H., O., Vild, Wagner, E.R., Wulf, M. and Verheyen, K. (2018a). Global environmental change effects on plant community composition trajectories depend upon management legacies. *Global Change Biology* **24**, 1722-1740.

Perring, M. P., Diekmann, M., Midolo, G., Schellenberger Costa, D., Bernhardt-Römermann, M., Otto, J.C.J., Gilliam, F.S., Hedwall, P.-O., Nordin, A., Dirnböck, T., Simkin, S.M., Málišk, F., Blondeel, H., Brunet, J., Chudomelová, M., Durak, T., De Frenne, P., Hédlm, R., Kopeckýp, M., Landuyt, D., Li, D., Manning, P., Petřík, P., Reczyńska, K., Schmidt, W., Standovár, T., Świerkosz, K., Vild, O., Waller, D.M., Verheyen, K. and Dirnböck, T. (2018b). Understanding context dependency in the response of forest understorey plant communities to nitrogen deposition. *Environmental Pollution* **242**, 1787-1799.

Persson, H. (1980). Fine-root dynamics in a Scots pine stand with and without near-optimum nutrient and water regimes. *Acta Phytogeographica Suecica* **68**, 101-110.

Persson, H. and Ahlström, K. (2002). Fine-root response to nitrogen supply in nitrogen manipulated Norway spruce catchment areas. *Forest Ecology and Management* **168**, 29-41.

Petrone, K., Buffam, I. and Laudon, H. (2007). Hydrologic and biotic control of nitrogen export during snowmelt: A combined conservative and reactive tracer approach. *Water Resources Research* **43**, W06420.

Pietilä, M., Lähdesmäki, P., Pietiläinen, P., Ferm, A., Hytönen, J. and Pätilä, A. (1991). High nitrogen deposition causes changes in amino acid concentrations and protein spectra in needles of the Scots pine (*Pinus sylvestris*). *Environmental Pollution* **72**, 103-115.

Pinho, P., Bergamini, A., Carvalho, P., Branquinho, C., Stofer, S., Scheidegger, C. and Maguas, C. (2012). Lichen functional groups as ecological indicators of the effects of land-use in Mediterranean ecosystems. *Ecological Indicators* **15**, 36-42.

Pitcairn, C.E.R., Leith, I., Sheppard, L., Sutton, M.A., Fowler, D., Munro, R.C., Tang, S. and Wilson, D. (1998). The relationship between nitrogen deposition, species composition and foliar nitrogen concentrations in woodland flora in the vicinity of livestock farms. *Environmental Pollution* **102**, 41-48.

Pitcairn, C., Fowler, D., Leith, I., Sheppard, L., Tang, S., Sutton, M. and Famulari, D. (2006). Diagnostic indicators of elevated nitrogen deposition. *Environmental Pollution* **144**, 941-950.

Pöyry, J., Carvalheiro, L.G., Heikkinen, R.K., Kühn, I., Kuussaari, M., Schweiger, O., Valtonen, A., Van Bodegom, P.M. and Franzén, M. (2017). The effects of soil eutrophication propagate to higher trophic levels. *Global Ecology and Biogeography* **26**, 18-30.

Preusser, S., Marhan, S., Poll, C. and Kandeler, E. (2017). Microbial community response to changes in substrate availability and habitat conditions in a reciprocal subsoil transfer experiment. *Soil Biology and Biochemistry* **105**, 138-152.

Quiring, R., Braun, S. and Flückiger, W. (1997). *N-deposition, N-nutrition and free amino acids in the foliage of Picea abies (L.) Karst and Fagus sylvatica (L.) from Swiss forest stands.* In: The Fourth International Symposium on Responses of Plant Metabolism to Air Pollution and Global Change, April 1-5, 1997. Egmond aan Zee, pp 56.

Quist, M.E., Näsholm, T., Lindeberg, J., Johannisson, C., Högbom, L. and Högberg, P. (1999). Responses of a nitrogen-saturated forest to a sharp decrease in nitrogen input. *Journal of Environmental Quality* **28**, 1970-1977.

Reynolds, B., Stevens, P.A., Hughes, S., Parkinson, J.A. and Weatherley, N.S. (1995). Stream chemistry impacts of conifer harvesting in Welsh catchments. *Water, Air and Soil Pollution* **79**, 147-170.

Ritter, G. (1990). Zur Wirkung van Stickstoffeinträgen auf Feinwurzelsystem und Mykorrhizabildung in Kieferbeständen. *Beitr.Forstwirtschaft* **24**, 100-104.

Rodenkirchen, H. (1992). Effects of acidic precipitation, fertilization and liming on the ground vegetation in coniferous forests of southern Germany. *Water, Air and Soil Pollution* **61**, 279-294.

Roelofs, J.G.M., Kempers, A.J., Houdijk, A.L. and Jansen, J. (1985). The effects of air-borne ammonium sulphate on *Pinus nigra* var. *maritima* in the Netherlands. *Plant and Soil* **84**, 45-56.

Root, H.T., Geiser, L.H., Fenn, M.E., Jovan, S., Hutten, M.A., Ahuja, S., Dillman, K., Schirokauer, D., Berryman, S. and McMurray, J.A. (2013). A simple tool for estimating throughfall nitrogen deposition in forests of western North America using lichens. *Forest Ecology and Management* **306**, 1-8.

Rosén, K., Gundersen, P., Tegnhammar, L., Johansson, M. and Frogner, T. (1992). Nitrogen enrichment of Nordic forest ecosystems: the concept of critical loads. *Ambio* **21**, 364-368.

Rosling, A., Lindahl, B.D. and Finlay, R.D. (2004). Carbon allocation to ectomycorrhizal roots and mycelium colonising different mineral substrates. *New Phytologist* **162**, 795-802.

RoTAP (2012). *Review of transboundary air pollution: acidification, eutrophication, ground-level ozone and heavy metals in the UK*. Contract report to Defra. Penicuik, Midlothian.

Roth, T., Kohli, L., Rihm, B., Meier, R. and Achermann, B. (2017). Using change-point models to estimate empirical critical loads for nitrogen in mountain ecosystems. *Environmental Pollution* **220**, 1480-1487.

Roth, T., Tresch, S., Kohli, L. and Braun, S. (2022). Bayesian change-point regression models including random effects to estimate empirical critical loads for nitrogen using brms and JA. *MethodX* submitted.

Rothe, A. and Mellert, K.H. (2004). Effect of forest management on nitrate concentrations in seepage water of forests in southern Bavaria, Germany. *Water Air and Soil Pollution* **156**, 337-355.

Rowe, E.C., Jones, L., Dise, N.B., Evans, C.D., Mills, G., Hall, J., Stevens, C.J., Mitchell, R.J., Field, C., Caporn, S.J.M., Helliwell, R.C., Britton, A.J., Sutton, M.A., Payne, R.J., Vieno, M., Dore, A.J. and Emmett, B.A. (2016). Metrics for evaluating the ecological benefits of decreased nitrogen deposition. *Biological Conservation* **212**, 1-10.

Rücker, T. and Peer, T. (1988). Die Pilzflora des Hellbrunner Berges - ein historischer Vergleich. *Ber. Nat. Med. Ver. Salzburg* **9**, 147-161.

Růžek, M., Tahovská, K., Guggenberger, G. and Oulehle, F. (2021). Litter decomposition in European coniferous and broadleaf forests under experimentally elevated acidity and nitrogen addition. *Plant and Soil* **463**, 471-485.

Saari, A., Smolander, A. and Martikainen, P.J. (2006). Methane consumption in a frequently nitrogen-fertilized and limed spruce forest soil after clear-cutting. *Soil Use and Management* **20**, 65-73.

Saarsalmi, A., Tamminen, P. and Kukkola, M. (2014). Effects of long-term fertilisation on soil properties in Scots pine and Norway spruce stands. *Silva Fennica* **48**, id 989.

Salemaa, M., Mäkipää, R. and Oksanen, J. (2008). Differences in the growth response of three bryophyte species to nitrogen. *Environmental Pollution* **152**, 82-91.

Salemaa, M., Lindroos, A.-J., Merilä, P., Mäkipää, R. and Smolander, A. (2019). N_2 fixation associated with the bryophyte layer is suppressed by low levels of nitrogen deposition in boreal forests. *Science of The Total Environment* **653**, 995-1004.

Salemaa, M., Kieloaho, A. J., Lindroos, A. J., Merilä, P., Poikolainen, J. and Manninen, S. (2020). Forest mosses sensitively indicate nitrogen deposition in boreal background areas. *Environmental Pollution* **261**, 114054.

Sandström, P., Cory, N., Svensson, J., Hedenås, H., Jougda, L. and Borchert, N. (2016). On the decline of ground lichen forests in the Swedish boreal landscape: Implications for reindeer husbandry and sustainable forest management. *Ambio* **45**, 415-429.

Sanz, M., Carratalá, A., Gimeno, C. and Millán, M. (2002). Atmospheric nitrogen deposition on the east coast of Spain: relevance of dry deposition in semi-arid Mediterranean regions. *Environmental Pollution* **118**, 259-272.

Sardans, J., Alonso, R., Janssens, I.A., Carnicer, J., Vereseglou, S., Rillig, M.C., Fernández-Martínez, M., Sanders, T.G.M. and Peñuelas, J. (2016). Foliar and soil concentrations and stoichiometry of nitrogen and phosphorous across European *Pinus sylvestris* forests: Relationships with climate, N deposition and tree growth. *Functional Ecology* **30**, 676-689.

Schlechte, G. (1986). Zur Mykorrhizapilzflora in geschädigten Forstbeständen. *Z.Mykol.* **52**, 225-232.

Schulze, E-D., Oren, R. and Lange, O.L. (1989). Nutrient relations of trees in healthy and declining Norway spruce stands. *Ecological Studies* 77**,** 392-417. Berlin, Springer-Verlag.

Simkin, S.M., Allen, E.B., Bowman, W.D., Clark, C.M., Belnap, J., Brooks, M.L., Cade, B., Collins, S.L., Geiser, L.H., Gilliam, F.S., Jovan, S.E., Pardo, L.H., Schulz, B.K., Stevens, C.J., Suding, K.N., Throop, H.L., Waller, D.M. and Waller, D.M. (2016). Conditional vulnerability of plant diversity to atmospheric nitrogen deposition across the United States. *Proceedings of the National Academy of Sciences* **113**, 4086-4091.

Sinsabaugh, R.L., Carreiro, M.M. and Repert, D.A. (2002). Allocation of extracellular enzymatic activity in relation to litter composition, N deposition and mass loss. *Biogeochemistry* **60**, 1-24.

Sjöberg, G., Bergkvist, B., Berggren, D. and Nilsson, S. I. (2003). Long-term N addition effects on the C mineralization and DOC production in mor humus under spruce. *Soil Biology and Biochemistry* **35**, 1305-1315.

Sjøeng, A.M.S., Kaste, Ø. and Wright, R.F. (2009). Modelling future NO_3 leaching from an upland headwater catchment in SW Norway using the MAGIC model: II. Simulation of future nitrate leaching given scenarios of climate change and nitrogen deposition. *Hydrology Research* **40**, 217-233.

Skrindo, A. and Økland, R.H. (2002). Effects of fertilization on understorey vegetation in a Norwegian *Pinus sylvestris* forest. *Applied Vegetation Science* **5**, 167-172.

Sleutel, S., Vandenbruwane, J., De Schrijver, A., Wuyts, K., Moeskops, B., Verheyen, K. and De Neve, S. (2009). Patterns of dissolved organic carbon and nitrogen fluxes in deciduous and coniferous forests under historic high nitrogen deposition. *Biogeosciences* **6**, 2743-2758.

Smart, S.M., Ellison, A.M., Bunce, R.G.H., Marrs, R.H., Kirby, K.J., Kimberley, A., Scott, A.W. and Foster, D.R. (2014). Quantifying the impact of an extreme climate event on species diversity in fragmented temperate forests: the effect of the October 1987 storm on British broadleaved woodlands. *Journal of Ecology* **102**, 1273-1287.

Smolander, A., Kitunen, V., Tamminen, P. and Kukkola, M. (2010). Removal of logging residue in Norway spruce thinning stands: Long-term changes in organic layer properties. *Soil Biology and Biochemistry* **42**, 1222-1228.

Smolander, A., Törmänen, T., Kitunen, V. and Lindroos, A.-J. (2019). Dynamics of soil nitrogen cycling and losses under Norway spruce logging residues on a clear-cut. *Forest Ecology and Management* **449**, 117444.

Solberg, S., Andreassen, K., Clarke, N., Torseth, K., Tveito, O.E., Strand, G.H. and Tomter, S. (2004). The possible influence of nitrogen and acid deposition on forest growth in Norway. *Forest Ecology and Management* **192**, 241-249.

Spiecker, H., Mielikäinen, R., Köhl, M. and Skorgsgaard, J. P. (1996). *Growth Trends in European Forests*. Springer-Verlag, New York, 372 pp.

Staude, I. R., Waller, D. M., Bernhardt-Römermann, M., Bjorkman, A.D., Brunet, J., De Frenne, P., Hédl, R., Jandt, U., Lenoir, J., Máliš, F., Verheyen, K., Wulf, M., Pereira, H.M., Vangansbeke, P., Ortmann-Ajkai, A., Pielech, R., Berki, I., Chudomelová, M., Decocq, G., Dirnböck, T., Durak, T., Heinken, T., Jaroszewicz, B., Kopecký, M., Macek, M., Malicki, M., Naaf, T., Nagel, T.A., Petřík, P., Reczyńska, K., Schei, F.H., Schmidt, W., Standovár, T., Świerkosz, K., Teleki, B., Van Calster, H., Vild, O. and Baeten, L. (2020). Replacements of small- by large-ranged species scale up to diversity loss in Europe's temperate forest biome. *Nature Ecology and Evolution* **4**, 802-808.

Sterkenburg, E., Bahr, A., Brandström Durling, M., Clemmensen, K. E. and Lindahl, B. D. (2015). Changes in fungal communities along a boreal forest soil fertility gradient. *New Phytologist* **207**, 1145-1158.

Stevens, C.J., Manning, P., Van den Berg, L.J.L., De Graaf, M.C.C., Wamelink, G.W.W., Boxman, A.W., Bleeker, A. Vergeer, P., Arroniz-Crespo, M., Limpens, J., Lamers, L.P.M., Bobbink, R. and Dorland, E. (2011). Ecosystem responses to reduced and oxidised nitrogen inputs in European terrestrial habitats. *Environmental Pollution* **159**, 665-676.

Strengbom, J., Nordin, A., Näsholm, T. and Ericson, L. (2001). Slow recovery of boreal forest ecosystem following decreased nitrogen input. *Functional Ecology* **15**, 451-457.

Strengbom, J., Nordin, A., Näsholm, T. and Ericson, L. (2002). Parasitic fungus mediates change in nitrogen-exposed boreal forest vegetation. *Journal of Ecology* **90**, 61-67.

Strengbom, J., Walheim, M., Näsholm, T. and Ericson, L. (2003). Regional differences in the occurrence of understorey species reflect nitrogen deposition in Swedish forests. *Ambio* **32**, 91-97.

Strengbom, J., Näsholm, T. and Ericson, L. (2004). Light, not nitrogen, limits growth of the grass *Deschampsia flexuosa* in boreal forests. *Canadian Journal of Botany* **82**, 430-435.

Strengbom, J., Witzell, J., Nordin, A. and Ericson, L. (2005). Do multitrophic interactions override N fertilization effects on *Operophtera* larvae? *Oecologia* **143**, 241-250.

Strengbom, J., Englund, G. and Ericson, L. (2006). Experimental scale and precipitation modify effects of nitrogen addition on a plant pathogen. *Journal of Ecology* **94**, 227-233.

Suz, L.M., Barsoum, N., Benham, S., Dietrich, H.-P., Fetzer, K.D., Fischer, R., García, P., Gehrman, J., Kristöfel, F., Manninger, M., Neagu, S., Nicolas, M., Oldenburger, J., Raspe, S., Sánchez, G., Schröck, H.W., Schubert, A., Verheyen, K., Verstraeten, A. and Bidartondo, M.I. (2014). Environmental drivers of ectomycorrhizal communities in Europe's temperate oak forests. *Molecular Ecology* **23**, 5628-5644.

Sverdrup, H. and Warfvinge, P. (1993). The effect of soil acidification on the growth of trees, grass and herbs as expressed by the (Ca+Mg+K)/Al ratio. Lund University, Department of Chemical Engineering II., *Reports in Ecology and Environmental Engineering* **2***,* 1-108.

Swanston, C., Homann, P.S., Caldwell, B.A., Myrold, D.D., Ganio, L. and Sollins, P. (2004). Long-term effects of elevated nitrogen on forest soil organic matter stability. *Biogeochemistry* **70**(2), 229-252.

Tahovská, K., Choma, M., Kaštovská, E., Oulehle, F., Bárta, J., Šantrůčková, H. and Moldan, F. (2020). Positive response of soil microbes to long-term nitrogen input in spruce forest: Results from Gårdsjön whole-catchment N-addition experiment. *Soil Biology and Biochemistry* **143**, 107732.

Tamm, C. O. (1991). *Nitrogen in Terrestrial Ecosystems*. Springer-Verlag, Berlin, pp. 1-115.

Termorshuizen, A. J. and Schaffers, A. P. (1987). Occurrence of carpophores of mycorrhizal fungi in selected stands of *Pinus sylvestris* in the Netherlands in relation to stand vitality and air pollution. *Plant and Soil* **104**, 209-217.

Termorshuizen, A. J. (1990). *Decline of carpophores of mycorrhizal fungi in stands of Pinus sylvestris*, Agricultural University Wageningen, The Netherlands.

Thelin, G., Rosengren-Brinck, U., Nihlgård, B. and Barkman, A. (1998). Trends in needle and soil chemistry of Norway spruce and Scots pine stands in South Sweden 1985-1994. *Environmental Pollution* **99**, 149-158.

Thimonier, A., Dupouey, J. and Timbal, T. (1992). Floristic changes in the herb-layer vegetation of a deciduous forest in the Lorrain plain under the influence of atmospheric deposition. *Forest Ecology and Management* **55**, 149-167.

Thimonier, A., Dupouey, J.L., Bost, F. and Becker, M. (1994). Simultaneous eutrophication and acidification of a forest ecosystem in North-East France. *New Phytologist* **126**, 533-539.

Thimonier, A., Kosonen, Z., Braun, S., Rihm, B., Schleppi, P., Schmitt, M., Seitler, E., Waldner, P. and Thöni, L. (2019). Total deposition of nitrogen in Swiss forests: Comparison of assessment methods and evaluation of changes over two decades. *Atmospheric Environment* **198**, 335-350.

Thorpe, E. (2011). *Impact of nitrogen deposition on methane- and ammonia-oxidising microbial communities in forest soils*, University of Essex.

Throop, H. L. and Lerdau, M. T. (2004). Effects of nitrogen deposition on insect herbivory: Implications for community and ecosystem processes. *Ecosystems* **7**, 109-133.

Tietema, A., Boxman, A.W., Emmet, B.A., Moldan, F., Gundersen, P., Schleppi, P. and Wright, R.F. (1998). Nitrogen saturation experiments (NITREX) in coniferous forest ecosystems in Europe: a summary of results. *Environmental Pollution* **102**, 433-437.

Tomova, L., Braun, S. and Flückiger, W. (2005). The effect of nitrogen fertilization on fungistatic phenolic compounds in roots of beech (*Fagus sylvatica*) and Norway spruce (*Picea abies*). *Forest Pathology* **35**, 262-276.

Törmänen, T., Kitunen, V., Lindroos, A.-J., Heikkinen, J. and Smolander, A. (2018). How do logging residues of different tree species affect soil N cycling after final felling? *Forest Ecology and Management* **427**, 182-189.

Tresch, S., Rihm, B., Braun, S. and Schindler, C. (2022). Does Norway spruce have a future in Switzerland? Insights from increased tree mortality due to climate change indicators and N depositions. In prep.

Treseder, K.K. (2004). A meta-analysis of mycorrhizal responses to nitrogen, phosphorus and atmospheric CO_2 in field studies. *New Phytologist* **164**, 347–355.

Treseder, K.K. (2008). Nitrogen additions and microbial biomass: a meta-analysis of ecosystem studies. *Ecology Letters* **11**, 1111–1120.

Tyler, G. (1987). Probable effects of soil acidification and nitrogen deposition on the floristic composition of oak (*Quercus robus* L.) forest. *Flora* **179**, 165-170.

UNECE (2005). *Forest Condition in Europe. 2005 Technical Report. Convention on Long-Range Transboundary Air Pollution*. International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests 1-99. Geneva and Brussels, UNECE, pp. 1-99.

UNECE (2007). *Recent results and updating of scientific and technical knowledge. Workshop on effects of low-level nitrogen deposition.* UN report ECE/EB.AIR/WG.1/2007/15.

UNECE (2015). *Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads and Levels and Air Pollution Effects, Risks and Trends*. With updated chapters: http://www.icpmapping.org/Mapping_Manual, United Nations Economic Commission for Europe, Convention on Long-range Transboundary Air Pollution, Working Group on Effects, Task Force on Modelling and Mapping Air Pollution Effects, Risks and Trends.

UNECE Mapping Manual (2004). Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. Convention on Long-range Transboundary Air Pollution.

Vadinther, N. (2019). *The influence of nitrogen deposition on community composition in Pinus banksiana forests across nortwestern Canada.* MSc thesis, Trent University, Peterborough, Ontario Canada.

Valliere, J.M., Irvine, I.C., Santiago, L. and Allen, E.B. (2017). High N, dry: Experimental nitrogen deposition exacerbates native shrub loss and nonnative plant invasion during extreme drought. *Global Change Biology* **23**, 4333-4345.

Van der Linde, S., Suz, L.M., Orme, C.D.L., Cox, F., Andreae, H., Asi, E., Atkinson, B., Benham, S., Carroll, C., Cools, N., De Vos, B., Dietrich, H.-P., Eichhorn, J., Gehrmann, J., Grebenc, T., Gweon, H.S., Hansen, K., Jacob, F., Kristöfel, F., Lech, P., Manninger, M., Martin, J., Meesenburg, H., Merilä, P., Nicolas, M., Pavlenda, P., Rautio, P., Schaub, M., Schröck, H.-W., Seidling, W., Šrámek, V., Thimonier, A., Thomsen, I.M., Titeux, H., Vanguelova, E., Verstraeten, A., Vesterdal, L., Waldner, P., Wijk, S., Zhang, Y., Žlindra, D., Bidartondo, M.I. (2018). Environment and host as large-scale controls of ectomycorrhizal fungi. *Nature* **558**, 243-248.

Van Diepen, L.T.A., Lilleskov, E.A., Pregitzer, K.S. and Miller, R.M. (2007). Decline of arbuscular mycorrhizal fungi in northern hardwood forests exposed to chronic nitrogen additions. *The New Phytologist* **176**, 175-83.

Van Diepen, L.T.A., Lilleskov, E.A. and Pregitzer, K.S. (2011). Simulated nitrogen deposition affects community structure of arbuscular mycorrhizal fungi in northern hardwood forests. *Molecular Ecology* **20**, 799-811.

Van Dijk, H.F.G. and Roelofs, J.G.M. (1988). Effects of excessive ammonium deposition on the nutritional status and condition of pine needles. *Physiologia Plantarum* **73**, 494-501.

Van Dijk, H., Van der Gaag, M., Perik, P.S.M. and Roelofs, J.G.M. (1992). Nutrient availability in Corsican pine stands in the Netherlands and the occurrence of *Sphaeropsis sapinea*: a field study. *Canadian Joernal of Botany* **70**, 870-875.

Van Dobben, H.F., Terbraak, C.J.F. and Dirkse, G.M. (1999). Undergrowth as a biomonitor for deposition of nitrogen and acidity in pine forest. *Forest Ecology and Management* **114**, 83-95.

Van Dobben, H.F. and De Vries, W. (2017). The contribution of nitrogen deposition to the eutrophication signal in understorey plant communities of European forests. *Ecology and Evolution* **7**, 214-227.

Van Strien, A.J., Boomsluiter, M., Noordeloos, M.E., Verweij, R.J.T. and Kuyper, T.W. (2018). Woodland ectomycorrhizal fungi benefit from large-scale reduction in nitrogen deposition in the Netherlands. *Journal of Applied Ecology* **55**, 290-298.

Vanguelova, E. I., Benham, S., Pitman, R., Moffat, A.J., Broadmeadow, M., Nisbet, T., Durrant, D., Barsoum, N., Wilkinson, M., Bochereau, F., Hutchings, T., Broadmeadow, S., Crow, P., Taylor, P. and Houston, T.D. (2010). Chemical fluxes in time through forest ecosystems in the UK - Soil response to pollution recovery. *Environmental Pollution* **158**, 1857-1869.

Vanguelova, E., Reynolds, B., Nisbet, T. and Godbold, D. (2011). The cycling of pollutants in nonurban forested environments, Springer, Dordrecht, pp. 679-710.

Vanguelova, E.I., Nisbet, T.R., Moffat, A.J., Broadmeadow, S., Sanders, T.G.M. and Morison, J.I.L. (2013). A new evaluation of carbon stocks in British forest soils. *Soil Use and Management* **29**, 169-181.

Vanguelova, E., Pitman, R. and Benham, S. (2018). Long term trends and effects of nitrogen input on forests and soils in the UK. In *Proceedings of the International Conference of forest soils*.

Verheyen, K., Baeten, L., De Frenne, P., Bernhardt-Römermann, M., Brunet, J., Cornelis, J., Decocq, G., Dierschke, H., Eriksson, O., Hédl, R., Heinken, T., Hermy, M., Hommel, P., Kirby, K., Naaf, T., Peterken, G., Petřík, P., Pfadenhauer, J., Van Calster, H., Walther, G.-R., Wulf, M. and Verstraeten, G. (2012). Driving factors behind the eutrophication signal in understorey plant communities of deciduous temperate forests. *Journal of Ecology* **100**, 352-365.

Verheyen, K., De Frenne, P., Baeten, L., Waller, D.M., Hédl, R., Perring, M.P., Blondeel, H., Brunet, J., Chudomelová, M., Decocq, G., De Lombaerde, E., Depauw, L., Dirnböck, T., Durak, T., Eriksson, O., Gilliam, F.S., Heinken, T., Heinrichs, S., Hermy, M., Jaroszewicz, B., Jenkins, M.A., Johnson, S.E., Kirby, K.J., Kopecký, M., Landuyt, D., Lenoir, J., Li, D., Macek, M., Maes, S.L., Máliš, F., Mitchell, F.J.G., Naaf, T., Peterken, G., Petřík, P., Reczyńska, K., Rogers, D.A., Schei, F.H. Schmidt, W., Standovár, T., Świerkosz, K., Ujházy, K., Van Calster, H., Vellend, M., Vild, O., Woods, K., Wulf, M. and Bernhardt-Römermann, M. (2017). Combining biodiversity resurveys across regions to advance global change research. *BioScience* **67**, 73-83.

Verstraeten, A., Neirynck, J., Cools, N., Roskams, P., Louette, G., De Neve, S. and Sleutel, S. (2017). Multiple nitrogen saturation indicators yield contradicting conclusions on improving nitrogen status of temperate forests. *Ecological Indicators* **82**, 451-462.

Vigdis, V. (2001). *Emission data reported to UNECE/EMEP: Evaluation of the spatial distributions of emissions.* EMEP/MSC-W Note 1/01, July 2001.

Vilkamaa, P. and Huhta, V. (1986). Effect of fertilization and pH on communities of Collembola in pine forest soil. *Annales Zoologici Fennici* **23**, 167-174.

Villada, A., Vanguelova, E.I., Shaw, L. and Verhoef, A. (2013). *Evaluation of tree species and soil type interactions for their potential for long term C sequestration*, PhD thesis, University of Reading, UK.

Vuorenmaa, J., Augustaitis, A., Beudert, B., Clarke, N., De Wit, H.A., Dirnböck, T., Frey, J., Forsius, M., Indriksone, I., Kleemola, S., Kobler, J., Krám, P., Lindroos, A.-J., Lundin, L., Ruoho-Airola, T., Ukonmaanaho, L. and Váňa, M. (2017). Long-term sulphate and inorganic nitrogen mass balance budgets in European ICP Integrated Monitoring catchments (1990-2012). *Ecological Indicators* **76**, 15-29.

Wallenda, T. and Kottke, I. (1998). Nitrogen deposition and ectomycorrhizas. *New Phytologist* **139**, 169-187.

Walther, G. R. and Grundmann, A. (2001). Trends of vegetation change in colline and submontane climax forests in Switzerland. *Bulletin of the Geobotanical Institute ETH* **67**, 3-12.

Wang, R., Goll, D., Balkanski, Y., Hauglustaine, D., Boucher, O., Ciais, P., Janssens, I., Penuelas, J., Guenet, B., Sardans, J., Bopp, L., Vuichard, N., Zhou, F., Li, B., Piao, S., Peng, S., Huang, Y. and Tao, S. (2017). Global forest carbon uptake due to nitrogen and phosphorus deposition from 1850 to 2100. *Global Change Biology* **23**(11), 4854-4872.

Weil, R.R. and Brady, N.C. (2017). *The nature and properties of soils*, 15th edition, Pearson.

Weldon, J. and Grandin, U. (2021). Weak recovery of epiphytic lichen communities in Sweden over 20 years of rapid air pollution decline. *The Lichenologist* **53**, 203-213.

Westling, O., Hallgren, L. and Sjöblad, K. (1992). *Deposition och effekten av luftföronreninger i södra och mellersta Sverige*. IVL Rapport B 0283-877x1079.

Wilkins, K. and Aherne, J. (2016). Vegetation community change in Atlantic oak woodlands along a nitrogen deposition gradient. *Environmental Pollution* **216**, 115-124.

Wilkins, K., Aherne, J. and Bleasdale, A. (2016). Vegetation community change points suggest that critical loads of nutrient nitrogen may be too high. *Atmospheric Environment*, **146**(Supplement C), 324-331.

Williams, M.W., Hood, E. and Caine, N. (2001). Role of organic nitrogen in the nitrogen cycle of a high-elevation catchment, Colorado Front Range. *Water Resources Research* **37**, 2569-2581.

Williams, M.W., Clow, D., Blett, T., Williams, M.W., Clow, D. and Blett, T. (2004). A novel indicator of ecosystem N status: DIN to DON ratio in riverine waters. In: *AGUFM*, Vol. 2004, pp. H53F-05.

Woods, C., Hunt, S., Morris, D. and Gordon, A. (2012). Epiphytes influence the transformation of nitrogen in coniferous forest canopies. *Boreal Environment Research* **17**, 411-424.

Xu, G.-L., Schleppi, P., Li, M.-H. and Fu, S.-L. (2009). Negative responses of Collembola in a forest soil (Alptal, Switzerland) under experimentally increased N deposition. *Environmental Pollution* **157**, 2030-2036.

Xu, X.N., Yan, L.M. and Xia, J.Y. (2019). A threefold difference in plant growth response to nitrogen addition between the laboratory and field experiments. *Ecosphere* **10**, 14.

Yan, L.M., Xu, X.N. and Xia, J.Y. (2019). Different impacts of external ammonium and nitrate addition on plant growth in terrestrial ecosystems: A meta-analysis. *Science of the Total Environment* **686**, 1010-1018.

Zackrisson, O., DeLuca, T.H., Nilsson, M.-C., Sellstedt, A. and Berglund, L.M. (2004). Nitrogen fixation increases with successional age in boreal forests. *Ecology* **85**, 3327-3334.

Zak, D.R., Holmes, W.E., Burton, A. J., Pregitzer, K.S. and Talhelm, A.F. (2008). Simulated atmospheric NO_3^- deposition increses soil organic matter by slowing decomposition. *Ecological Applications* **18**, 2016-2027.

Zanchi, G., Belyazid, S., Akselsson, C. and Yu, L. (2014). Modelling the effects of management intensification on multiple forest services: a Swedish case study. *Ecological Modelling* **284**, 48-59.

Zechmeister-Boltenstern, S., Michel, K. and Pfeffer, M. (2011). Soil microbial community structure in European forests in relation to forest type and atmospheric nitrogen deposition. *Plant and Soil* **343**, 37-50.

Zimmermann, F., Plessow, K., Queck, R., Bernhofer, C. and Matschullat, J. (2006). Atmospheric N- and S-fluxes to a spruce forest - Comparison of inferential modelling and the throughfall method. *Atmospheric Environment* **40**, 4782-4796.

10 Use of empirical critical loads of nitrogen ($CL_{emp}N$) in risk assessment and nature protection

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10.1 Introduction

In Europe, empirical critical loads of nitrogen ($CL_{emp}N$) are mainly used by the Parties to the Geneva Air Convention to quantify risks to ecosystems from air pollution, and to set emission reduction targets. The levels of implementation range from more common national broad-scale use in national risk assessments to specific regional and local applications in several countries to inform decisions on new and existing emission sources near protected habitats.

$CL_{emp}N$ for natural and semi-natural ecosystems were first presented in a background document at the 1992 workshop on critical loads staged during the UNECE Convention in Lökeborg (Sweden). Since then, they have been continuously developed on the basis of the latest scientific findings over the past 30 years. They pursue a harmonised, scientific approach developed with international experts. Based on field evidence, $CL_{emp}N$ are derived from observed N addition studies and N gradient studies which identify dose-effect relationships. The original rationale behind developing the $CL_{emp}N$ was to include the growing empirical evidence of negative nitrogen effects with a view to increasing the credibility of nitrogen CLs in policy support and regulation. This has proved very successful. $CL_{emp}N$ have come to play an important role used alone or in combination with computed critical loads. $CL_{emp}N$ constitute a powerful tool for setting (interim) targets (e.g. reducing the negative impacts of nitrogen deposition in the UK (Rowe et al., 2020)) for the protection of ecosystems or habitats.

The aim of this chapter is to provide examples of how $CL_{emp}N$ can be used on different scales and in different European countries. This is not a complete list of currently available applications. It is, rather, a selection in order to provide guidance for practitioners and policy makers on how $CL_{emp}N$ can be used in practice. The values are valid for all habitats regardless of the legal protection status of the ecosystem. Furthermore, this chapter considers how $CL_{emp}N$ are used in nature conservation practice. It also illustrates the extent to which they are a suitable metric to assess the risks for biodiversity and the diversity of ecosystems from atmospheric nitrogen inputs.

10.2 Examples of applications of $CL_{emp}N$ in risk assessments in Europe

In the Call for Data 2019-2021, the Parties to the Convention were asked if and how $CL_{emp}N$ are used at national or local level (CCE, 2019). In summary, countries use $CL_{emp}N$ at national level to determine eutrophication status, and to estimate nitrogen exceedances in all relevant natural and semi-natural ecosystems (e.g. Switzerland (FOEN, 2016) and Germany (UBA-DzU, 2021)), or only for terrestrial non-forest habitats (e.g. Austria, Ireland). In the UK, $CL_{emp}N$ are used to assess both the eutrophication of terrestrial habitats (except for managed woodland) and the acidification of soils in impacts assessments. Some specific examples of use are presented in the following section.

10.2.1 Critical load for the eutrophication ($CL_{eut}N$) approach

Since 2005, $CL_{emp}N$ have been part of the call for data published by the CCE. In 2015, the CCE published a combined dataset entitled the critical load for eutrophication ($CL_{eut}N$). The $CL_{eut}N$ of an ecosystem is defined as either the empirical ($CL_{emp}N$) or the modelled ($CL_{nut}N$) critical load of nitrogen or – if a site has been assigned both values – the lower of the two. Since 2017, $CL_{eut}N$ have been reported by several NFCs to the CCE.

10.2.2 Parameter to verify modelled critical loads

$CL_{emp}N$ corroborate modelled critical loads from an effect-oriented perspective. Some countries use $CL_{emp}N$ to adjust the results for modelled simple mass balance (SMB) critical loads thereby increasing their reliability (e.g. Austria, Switzerland, UK). For example, based on $CL_{emp}N$, the minimum modelled CL for productive forests in Switzerland is set at 10 kg N ha^{-1} yr^{-1}.

10.2.3 NEC Directive reporting (2016/2284/EU) and other National Emissions Ceilings Regulations

Critical loads and exceedances are not listed in the required parameters specified under the reporting on effects in Article 9 of the NEC Directive. The reporting is, however, required to include sites under pressure from N deposition. Furthermore, critical load exceedances have been reported by several countries as a detail for the sites (EEA, 2020).

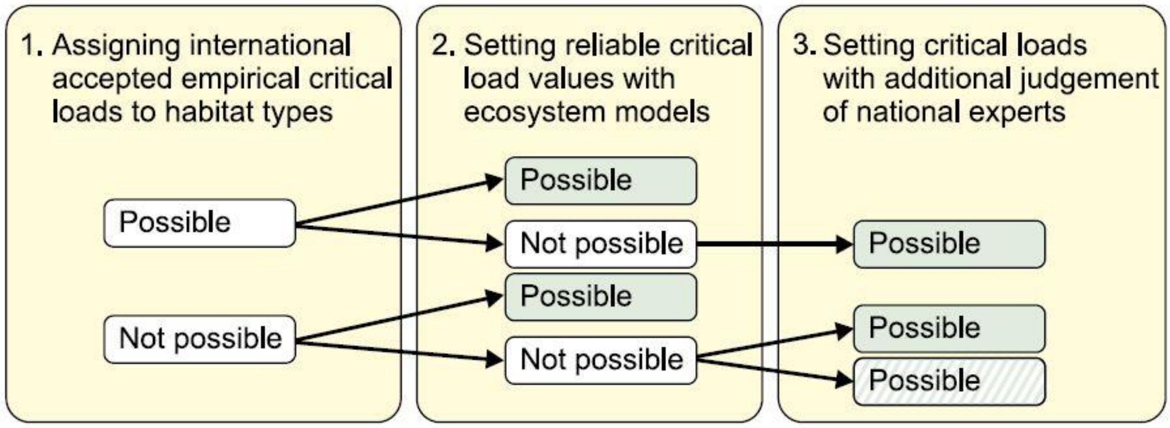
The UK measures and reports on the impacts of air pollution on ecosystems in line with Part 5 of the National Emissions Ceilings (NEC) Regulations 2018. The obligation set out in Article 9 of the NEC Directive 2016 was transposed to the national regulations prior to the exit of the UK from the EU. $CL_{emp}N$ values feature in supplemental monitoring data and statistics that compare the ecological risk from air pollution to habitats at country level. Data are collated from the UK APIENs monitoring network. The reporting template mentioned in Article 9 is used as a starting point but both it and the monitoring network are being reviewed following the first round of Article 9 reporting (WER, 2019).

10.2.4 Use of empirical critical loads in the integrated approach in the Netherlands

Critical loads for Natura 2000 habitats have been identified using an integrated method of the empirical and the modelling approach (Van Dobben et al., 2014). The method for setting a unique critical load value per habitat is as follows: for each Dutch habitat type it was determined whether a $CL_{emp}N$ was available (Bobbink and Hettelingh, 2011). If so, the critical load was further specified with results from the simulation models for the combined effect of eutrophication and acidification (Van Dobben et al., 2006). If no $CL_{emp}N$ was available, the critical load value was based on the mean value of the results from the national simulation model. Furthermore, if no result was available from the simulation models either, the critical load value was based on expert judgement. The EUNIS type for which the critical load range was set, had to be clearly related to the habitat type. For an overview of the procedure, see Figure 10.1.

The procedure yielded N critical loads for most Annex I habitat types in the Netherlands. In most cases, empirical ranges and/or reliable model estimates were available, and critical loads could be set based on published information. In about 70 per cent of all the habitats, the models yielded critical load values within the given empirical range. In those cases where simulated critical loads were outside the empirical range, the critical load was set to the nearest extreme of the $CL_{emp}N$ values. The difference between the middle of the $CL_{emp}N$ range and the modelled values per habitat type was on average less than 1 kg ha^{-1} yr^{-1}. 60 out of the 75 habitat types found in The Netherlands were sensitive to N deposition (CLN < 34 kg N ha^{-1} yr^{-1}). These critical loads for N deposition (Van Dobben et al., 2014) are the given standard in The Netherlands, and are also accepted by the government and by the judges of the highest administrative court. And because these critical loads have unique values instead of ranges, it is clear whether or not the critical load of a specific habitat is exceeded. The habitat-specific critical loads are used together with detailed maps of habitat occurrences and detailed deposition maps to reveal exceedances across Natura 2000 nature reserves.

Figure 10.1. Procedure used in the Netherlands for setting critical loads for Annex I habitat types. In most cases, reliable critical loads could be set after steps 1 and 2 (green boxes). In some cases, additional judgement from national experts was used for setting reliable or relatively reliable critical loads (green boxes), or even to give a best possible estimate (striped box).



Source: Van Hinsberg and Van Dobben, 2011

10.2.5 Air Pollution Information System in the UK

At local level, the UK publishes habitat and site-specific $CL_{emp}N$ values on the Air Pollution Information System (APIS) along with other information about current and historic concentrations and deposition (CEH, 2016). APIS is a publicly available resource for risk assessment used in decision-making by regulators, local authorities, and land managers to understand air pollution effects on ecosystems. The use of site-specific evidence to ascertain which part of the $CL_{emp}N$ range (i.e. lower or higher value) should be utilised has been harmonised for UK practitioners (see APIS risk assessment guidance) in most cases (APIS Steering Group, 2013). Where $CL_{emp}N$ are less certain or require additional information prior to their application, users are advised to consult the relevant national nature conservation body. The way in which that critical load then feeds into risk assessment and decision-making processes currently varies across UK countries and decision-making bodies. For example, current approaches compare the predicted change in concentration or deposition that is expected to result from a licensing proposal with a percentage of the critical level or load in two stages: screening for likely significant effects and determination of whether adverse effects can be ruled out.

$CL_{emp}N$ also form an important part of the UK characterisation of risks to ecosystems from air pollution and promote understanding of where emission reduction measures can be targeted to maximise the benefits to those habitats. Although the form of these interventions and the locations where they have the most impact may vary between UK countries, $CL_{emp}N$ account for a large part of this activity. The national and international reporting approaches are harmonised for UK level reporting by habitat type, and are similar for the UK countries and protected sites in Scotland, Wales, Northern Ireland and England (Rowe et al., 2021). This is demonstrated both in the annual reporting for CLRTAP and in the biodiversity indicator for air pollution pressure reported for international biodiversity goals and targets (Defra, 2020). As increasing attention is paid to air pollution effects on ecosystems, the UK is investing in new evidence to support the use of critical loads and their underpinning data in risk assessment. The aims here are to streamline their use and make the meaning of critical loads clearer for policy makers, proposers, and decision-makers.

10.2.6 Guidelines for immission control approval

In several countries, $CL_{emp}N$ are used as part of national standardised guidelines for the determination and assessment of the impact of nitrogen deposition on nearby ecosystems caused by installations requiring immission control approval. In Germany for example, the assessments are carried out for facilities that are to be newly licensed and that reach a certain size according to the Federal Immission Control Act, i.e. exceed a certain number of animals in the case of livestock facilities or a certain firing rate in the case of incineration plants. For all biotopes and ecosystems located in the vicinity of such a new facility, nitrogen sensitivity must be defined with the help of empirical critical loads (LAI N-Dep, 2012). The guideline is intended to contribute to a greater degree of legal certainty in the authorisation of installations and, by extension, to simplify and accelerate enforcement. In Denmark, $CL_{emp}N$ have served as a basis for defining classes of nitrogen sensitivity for different ecosystem types used in ammonia regulation and agricultural approval (Bak, 2014).

However, the interpretation of the ecosystem damage caused by nitrogen and the evaluation of the impact in any licensing process should also consider other non-atmospheric sources of nitrogen. This is particularly relevant for wetland habitats and aquatic systems where there are additional pathways of N input into sites, for instance, from surface water or groundwater (Rhymes et al., 2015) or from seawater for coastal habitats. There are no easy solutions to this problem, but some studies are starting to address this issue, such as trial-based approaches which calculate combined fluxes from atmospheric and other sources (e.g. Farr et al., 2019).

10.3 Empirical critical loads in nature conservation practice

Nitrogen deposition poses a serious threat to sensitive ecosystems throughout Europe. This has been confirmed by the latest international or national assessments that compare critical loads to modelled nitrogen deposition estimates (EMEP, 2020; Schaap et al., 2018; Rowe et al., 2021), and also by national survey data (Jones et al., 2016; Britton and Ross, 2018; Heinze et al., 2019). In large parts of sensitive ecosystems and habitats, the exceedance of critical loads may lead to significant changes in site conditions and biodiversity. This threatens many ecosystems, regardless of their protection status under both national and EU nature conservation law. Exceedances also have highly adverse effects on restoration and management efforts in oligotrophic and mesotrophic ecosystems and EU protected habitats, and result in far higher costs of the measures to compensate for the high nitrogen input.

Despite the evidence that nitrogen deposition is a major threat to European biodiversity, $CL_{emp}N$ have not been widely used up to now in European nature conservation practice and monitoring: For example, in the reporting guidelines for the obligatory, periodical reporting on the conservation status of the protected habitat types (reporting under Article 17 of the Habitats Directive 92/43/EEC), issued by the European Environment Agency (EEA) (DG Environment, 2017), the reporting of anthropogenic “eutrophication” is spread over the different sources of pollution in various subcategories of the main pressures and threats such as agriculture, forestry or air pollution, etc. Furthermore, only a limited number of main pressures can be reported using a ranking system and not all threats are reported in full. Anthropogenic nitrogen deposition is not a predefined threat category and the assessment with critical loads is not foreseen. Consequently, it is difficult to obtain a complete picture of eutrophication as a choice in the threat characterisation tables.

10.3.1 Critical loads for quantitative assessment of conservation status

To date, there is no guidance or recommendation on the application of critical loads to quantitatively assess conservation status and the threat of nitrogen deposition. This topic was also discussed at a workshop organised in 2013 by the Joint Nature Conservation Committee (JNCC) (Whitfield and McIntosh, 2014). The participants in the workshop already at that time recommended "*to establish a mechanism to bring together experts on the impacts of nitrogen deposition and conservation status to define common desired outcomes and develop integrated actions for consideration, how air pollution impacts relate to conservation status, and how critical load exceedance relates to achieving favourable conservation status."* The suggestion was also made to further develop *"appropriate indicators or metrics of air pollution impacts … for specific habitats, to support evaluation of air pollution policy".* The recommendations of the workshop were partly taken over into national activities.

Both JNCC and NatureScot (formerly known as Scottish Natural Heritage (SNH)) have explored decision frameworks for assessing whether nitrogen deposition is a threat leading to adverse habitat condition on protected sites (JNCC 579, 2016; SNH 958, 2018). Both sets of guidelines give practical advice on how to make use of and apply $CL_{emp}N$. However, in both cases, limitations of the methodology were reported, for instance, the required $CL_{emp}N$ are not available for all of the covered biotopes or the robustness of critical load data was too low for some of the habitats. While NatureScot found that the approach for determining conservation status, for the aforementioned reasons, required further development, the JNCC approach is being considered in the UK by the other national nature conservation agencies. For example, Natural England has been developing the Nitrogen Decision Framework (NDF) to refine the assessment of exceedances of nitrogen deposition on designated sites. The NDF will help to support the assessment of the degree of risk or threat arising from a proposal as a consequence of emissions to air, based on national exceedance data and habitat quality information where it is available. The intention is to produce an automated method by which Natural England can apply the NDF to each "Site of Special Scientific Interest" unit (SSSI) in England. The application of this method will be discussed internally and more widely with the nature conservation bodies.

10.3.2 Recommendations for nature conservation practice

In Germany, $CL_{emp}N$ were used to assess the relevance of various threat factors for endangered habitat types on the Red List of Threatened Habitat Types (Heinze, 2019). Given the availability of $CL_{emp}N$ data, a nitrogen sensitivity value could be attributed to 47% of the habitats. However, in contrast, this meant that no sensitivity value could be attributed to 53% of the open-land biotopes to date. Nevertheless, the assessment suggests that nitrogen deposition is one of the predominant threats to protected habitats in Germany.

Furthermore, the latest German Interpretation Manual of Natura 2000 habitats (Ssymank et al., 2021) paves the way for the use of $CL_{emp}N$ in a) appropriate assessments according to Article 6 (3-4) of the Habitats Directive, b) site management and c) the assessment of the conservation status with respect to atmospheric nitrogen deposition as a threat. In this manual an indicative value of nitrogen sensitivity is attributed as an expert judgement to all habitat types in order to fill the gaps where $CL_{emp}N$ are still missing and need to be developed in future.

In summary, it is possible and beneficial to use $CL_{emp}N$ for both air pollution and biodiversity assessments at national and international levels. Technically, crosswise usage in both areas of application is possible. Transfer matrices of the $CL_{emp}N$ systematics and the Natura 2000 systematics have been available since the publication of the report of Bobbink and Hettelingh

(2011), and have been updated in this report (see Appendix 1). Furthermore, it is possible to narrow down the range values to concrete values with the help of limiting factors. This is already being done successfully for local approval procedures, for instance in Germany or The Netherlands.

Therefore, the recommendation is to include supplementary information on $CL_{emp}N$ also in the EU guidelines and manuals used for reporting on the conservation status of European habitat types. It is expected that a concurrent application of $CL_{emp}N$ in the area-wide assessments of risks from nitrogen deposition (EMEP, 2020; Rowe et al., 2021; Defra, 2020), as well as in national licensing practice and nature conservation, will further increase the visibility of the values, and help to harmonise risk assessment approaches at different application sites and for different polluters.

10.4 A metric to assess risks to ecosystem biodiversity from atmospheric N inputs

Threats to ecosystems from nutrient imbalances, high nitrogen saturation, and biodiversity loss have become an increasingly important issue under the Geneva Air Convention. The Long-term Strategy 2020-2030, which was adopted in 2018, specifically identifies the protection of biodiversity and the prevention of biodiversity loss as a goal. Indeed, this strategy states that the "disruption of global and regional nitrogen cycles is one of the most important environmental challenges (…). Current and future exceedances of critical loads of nitrogen as an indicator of biodiversity loss over large areas are dominated by ammonia emissions from agriculture (…) (Long-Term Strategy Decision 2018/5: p. 7)".

In general, biological diversity is the term used to depict the variety of life on Earth. It encompasses all organisms, species, and populations, the genetic variation among them and their complex assemblages of communities and ecosystems (UNEP, 2017). Each species within an ecosystem has its own niches and requirements. Changes in the ability to access these requirements – in this report through changes in nitrogen availability – affect the population size of a species resulting in changes to the composition of biological communities and ecosystems (CSS, 2020).

The results of the present review demonstrate and confirm that excessive atmospheric nitrogen deposition negatively affects species assemblages, and thus poses a serious risk to biodiversity. In many cases, it has already caused a decline. The scientific evidence available for the various existing dose-response relationships between nitrogen deposition and the risks for ecosystems and habitat quality of European ecosystems is summarised in Chapters 3 to 9 of this report. These chapters present many different scientific studies from around the world which provide evidence that exceedances of $CL_{emp}N$ can clearly be linked to reduced plant species richness in some major European ecosystem types.

Therefore, the revised $CL_{emp}N$ (Chapter 11, Table 11.1) are an appropriate and relevant metric to quantitatively assess risks to biodiversity (risks to ecosystem diversity) from nitrogen deposition.

It should, however, be noted, that the $CL_{emp}N$ approach also has some limitations. At first, it is sometimes difficult to establish the lower end of $CL_{emp}N$ ranges because current background depositions are above $CL_{emp}N$ in many places, and data has sometimes been collected in a situation where the system was not in a state of equilibrium. Secondly, the published studies draw on quite different impact indicators, which may not have included all relevant effects on the most sensitive species, and often do not include effects occurring over longer timescales. Thirdly, the type of habitat is not the only factor influencing $CL_{emp}N$ because climate conditions or soil type impact sensitivity as well. Therefore, $CL_{emp}N$ ranges tend to be quite broad (Bobbink and Hettelingh, 2011). Last but not least, for large-scale application in risk assessment, the accurate mapping of ecosystems and, by extension, the spatial attribution of thresholds still reveals uncertainties. As a consequence, it will often be necessary or preferential to base risk assessment on a mixture of methods, for instance $CL_{emp}N$ combined, where appropriate, with static or dynamic soil geochemistry models, plant competition, and plant occurrence models.

10.5 Conclusions

The various uses of $CL_{emp}N$ range from broad-scale, national risk assessments of negative impacts of area-wide atmospheric nitrogen deposition to very local appraisals of a single protected ecosystem affected by nitrogen emissions from a single source. $CL_{emp}N$ provide a way to include consideration of effects on ecosystem function in relation to the pressure from nitrogen deposition, and to quantify the tolerance of ecosystems. A major strength of the $CL_{emp}N$ is that they are based on observations in the field. Values for different habitats were first established more than 30 years ago and are continuously being updated with due consideration of the latest scientific findings. Existing values apply to the scientific and natural entity of an ecosystem or habitat regardless of the legal protection status of that system. It is important to emphasise that $CL_{emp}N$ are not only applicable to the risk assessment of Natura 2000 habitats but also to the same ecosystems with other classifications such as EUNIS and without protection under Natura 2000 legislation.

Nevertheless, continuous experimental and survey work is essential to further improve the robustness of $CL_{emp}N$, and to establish $CL_{emp}N$ for habitats that have not yet been studied, for instance coastal shingle, temperate and Mediterranean-montane scrub. The different methods for modelling critical loads should be further developed concurrently and their integration into risk assessment is an important task for the future. Also, the Europe-wide, closer linking of environmental monitoring and air pollution measurements could help to effectively close knowledge gaps. Joint efforts and a more in-depth exchange between experts in air pollution control and biodiversity conservation are necessary to further increase knowledge. Last but not least, for large-scale assessments it is important to have an accurate mapping procedure that offers robust information on the location of ecosystems and the attribution of critical loads.

$CL_{emp}N$ are a suitable indicator to identify risks and damage to biodiversity at the ecosystem level, which can be linked to policy-relevant biodiversity targets (e.g. the EU Biodiversity Strategy for 2030 which aims to ensure no net loss of biodiversity and ecosystem services), and to the Convention on Biodiversity (CBD).

10.6 References

APIS Steering Group (2013). *APIS indicative critical loads values: Recommended values within nutrient critical load ranges for use in air pollution impact assessments.* Available here: http://www.apis.ac.uk/indicative-critical-load-values

Bak, J.L. (2014). Critical Loads for Nitrogen Based on Criteria for Biodiversity Conservation. *Water, Air & Soil Pollution* **225**, 2180.

Bobbink, R. and Hettelingh J.-P. (eds.) (2011). *Review and revision of empirical critical Loads and dose-response relationships,* Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM).

Britton, A.J. and Ross, L.C. (2018). *Towards the development of a Nitrogen Deposition Decision Framework for vegetation assessment in Scotland.* Scottish Natural Heritage Research Report No. 958.

CCE (2019). Call for Data 2019-2021. Available here: https://www.umweltbundesamt.de/sites/default/files/medien/4292/dokumente/cce_call_for_data_2019_2021.pdf

CEH (2016). APIS – Air Pollution Information System. Information available here: http://www.apis.ac.uk/

CSS - Center for Sustainable Systems, University of Michigan (2020). *Biodiversity Factsheet.* Pub. No. CSS09-08. Available here: https://css.umich.edu/factsheets/biodiversity-factsheet

Defra (2020). *UK Biodiversity Indicators 2020. Indicator B5a – Air pollution 2020*. Available here: https://hub.jncc.gov.uk/assets/9be943eb-3066-4e8e-82d3-b1348d7ce9b9

DG Environment (2017). *Reporting under Article 17 of the Habitats Directive: Explanatory notes and guidelines for the period 2013-2018.* Brussels. Pp 188. Available here: https://www.bfn.de/fileadmin/BfN/natura2000/Dokumente/Reporting_guidelines_Article_17_final_May_2017.pdf

EEA (2020). *Analysis of Ecosystem Monitoring Data under Article 9 of Directive (EU) 2016/2284. Comprehensive Assessment*. Available here: http://cdr.eionet.europa.eu/help/habitats_art17

EMEP (2020). *Transboundary particulate matter, photo-oxidants, acidifying and eutrophying components.* Status report 1/2020, Norwegian Meteorological Institute.

FOEN - Federal Office for the Environment Switzerland (2016). *Critical Loads of Nitrogen and their Exceedances.* Swiss contribution to the effects-oriented work under the Convention on Long-range Transboundary Air Pollution (UNECE). Bern, Switzerland.

Gareth, F., Hall, J., Jones, L., Whiteman, M., Haslam, A., Phillips, N., Tang, S., Williams, H., Davison, P. and Lapworth, D. (2019). *Atmospheric deposition at groundwater dependent wetlands phase 2: nutrient source apportionment case studies from England and Wales.* Keyworth, Nottingham. *British Geological Survey*, 93pp. (OR/17/021)

Habitats Directive 92/43/EEC. Available here: http://eur-lex.europa.eu/legal-content/EN/ALL/?uri=CELEX:01992L0043-20070101

Heinze, S., Finck, P., Raths, U., Riecken, U. and Ssymanck, A. (2019). *Analyse der Gefährdungsursachen von Biotoptypen in Deutschland.*

Jones L., Hall, J., Strachan, I., Field, C., Rowe, E., Stevens, C.J., Caporn, S.J.M., Mitchell, R., Britton, A., Smith, R., Bealey, B., Masante, D., Hewison, R., Hicks, K., Whitfield, C. and Mountford, E. (2016). *A decision framework to attribute atmospheric nitrogen deposition as a threat to or cause of unfavourable habitat condition on protected sites.* JNCC Report No. 579. JNCC, Peterborough.

LAI N-Dep (2012). *Leitfaden zur Ermittlung und Bewertung von Stickstoffeinträgen* (Guidance on the identification and assessment of nitrogen inputs), Langfassung, Bund/Länder-Arbeitsgemeinschaft für Immissionsschutz. Available here: https://lkclp.de/uploads/files/oekosysteme_lai_n-leitfaden_01_03_12.pdf

Long-term strategy Decision 2018/5. Available here: https://unece.org/DAM/env/documents/2018/Air/EB/correct_numbering_Decision_2018_5.pdf

Rhymes J., Jones, L., Lapworth, D.J., White, D., Fenner, N., McDonald, J.E. and Perkins, T.L. (2015). Using chemical, microbial and fluorescence techniques to understand contaminant sources and pathways to wetlands in a conservation site. *Science of the Total Environment* **511,** 703-711.

Rowe, E.C., Sawicka, K., Tomlinson, S., Levy, P., Banin, L.F., Martín Hernandez, C., Fitch, A. and Jones, L. (2021). *Trends Report 2021: Trends in critical load and critical level exceedances in the UK.* Report to Defra under contract AQ0849, UKCEH Project 07617. Available here: https://uk-air.defra.gov.uk/library/reports?report_id=1020

Schaap, M., Hendriks, C., Kranenburg, R., Kuenen, J., Segers, A., Banzhaf, S., Schlutow, A., Nagel, H.-D. and Ritter, A. (2018). *Modellierung atmosphärischer Stoffeinträge von 2000 bis 2015 zur Bewertung der ökosystem-spezifischen Gefährdung von Biodiversität durch Luftschadstoffe in Deutschland*; eds. Geupel, M., Umweltbundesamt, Dessau-Roßlau.

Ssymank, A., Ellwanger, G., Ersfeld, M., Ferner, J., Lehrke, G., Müller, C., Raths, U., Röhling, M. and Vischer-Leopold, M. unter Mitarbeit von Balzer, S., Bernhardt, N., Fuchs, D., Sachteleben, J., Schubert, E. and Tschiche, J. (2021). *Das europäische Schutzgebietssystem Natura 2000. BfN-Handbuch zur Umsetzung der Fauna-Flora-Habitat-Richtlinie (92/43/EWG) und der Vogelschutzrichtlinie (2009/147/EG). Band 2.1: Lebensraumtypen der Meere und Küsten, der Binnengewässer sowie der Heiden und Gebüsche.* - 2. A, Naturschutz und Biologische Vielfalt Heft 172 (2.1), 1-795.

UNEP – UN Environment Programme (2017). *Biodiversity Factsheet*. Available here: https://www.unep.org/resources/factsheet/biodiversity-factsheet

Van Dobben, H.F., Van Hinsberg, A., Schouwenberg, E.P.A.G., Jansen, M., Mol-Dijkstra, J.P. Wiegers, H.J.J., Kros, J. and De Vries, W. (2006). Simulation of Critical Loads for Nitrogen for Terrestrial Plant Communities in The Netherlands. *Ecosystems* **9,** 32-45.

Van Dobben, H., Bobbink, R., Bal, D. and Van Hinsberg, A (2014). *Overview of critical loads for nitrogen deposition for Natura 2000 habitat (sub)types occurring in The Netherlands.* Wageningen, Alterra Wageningen UR (University and Research Center), Alterra report 2488.

Van Hinsberg, A. and Van Dobben, H.F. (2011). Setting critical loads for Dutch Natura 2000 sites using empirical information and dynamic modelling. In: W.K. Hicks, C.P. Whitfield, W.J. Bealey and M.A. Sutton (eds.) *Nitrogen deposition and Natura 2000: Science & practice in determining environmental impacts*. COST729/Nine/ESF/CCW/JNCC/SEI Workshop Proceedings, published by COST, pp. 101-104.

Wageningen Environmental Research – WER (2019). *First analysis of ecosystem monitoring as required under Art 9 of the NECD.* Ref. 3417/B2017/EEA Wageningen Environmental Research, Netherlands. Available here: https://ec.europa.eu/environment/air/pdf/reduction_reports/Final%20report%20ecosystem%20monitoring%20network%20Nov2019.pdf

Whitfield, C. and McIntosh, N. (2014). *Nitrogen Deposition and the Nature Directives Impacts and responses: Our shared Experiences.* Report of the Workshop held 2–4 December 2013, JNCC Peterborough. JNCC Report No. 521

UBA-DzU (2021). *Überschreitung der Belastungsgrenzen für Eutrophierung.* Available here: https://www.umweltbundesamt.de/daten/flaeche-boden-land-oekosysteme/land-oekosysteme/ueberschreitung-der-belastungsgrenzen-fuer-0

11 Conclusions and gaps in knowledge

11.1 Conclusions

Within the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP Convention), procedures have been developed to model and map critical loads for airborne N deposition in support of effect-based European policies for the abatement of air pollution (Bull et al., 2001; Hettelingh et al., 2001; 2007). Both the steady-state mass balance method and the empirical approach are used to scientifically support European policies aiming at effective emission reductions of air pollutants (ICP M&M, 2017).

Based on observed changes in the structure and function of ecosystems, empirical critical loads of N ($CL_{emp}N$) have been evaluated for specific receptor groups of natural and semi-natural ecosystems, reported in a range of publications (Bobbink et al., 1992; 1996; 2003). A synthesis of the knowledge for use of $CL_{emp}N$ under the LRTAP Convention was published by Achermann and Bobbink (2003) and by Bobbink and Hettelingh (2011). $CL_{emp}N$ were also included in the second edition of the Air Quality Guidelines for Europe of the World Health Organization Regional Office for Europe (WHO, 2000). Available new insights and data on the impacts of N deposition on natural and semi-natural ecosystems vegetation since the last publication of Bobbink and Hettelingh (2011) have justified a review and revision of $CL_{emp}N$ of which the results have materialised in this report.

The review and revision was conducted using a similar '*empirical approach*' (see Chapter 2 for more details) as described in Bobbink and Hettelingh (2011). For this purpose, the authors have reviewed all relevant European publications – as comprehensively as possible – on the effects of N on natural and semi-natural ecosystems for the period from 2010 to mid-2021. The authors used peer-reviewed publications, book chapters, nationally published papers and 'grey' reports by institutes or organisations, when available on request. For the classification of ecosystems the EUNIS habitat classification for Europe was used (Chytrý et al., 2020). To improve incorporation of the $CL_{emp}N$ in the EU habitat type classification, an appendix has been added, coupling the EUNIS system with the EU habitat classification (Appendix 1). Chapters 3 to 9 present evaluations of the effects of N enrichment per EUNIS class and $CL_{emp}N$ updated with newly published evidence, when available. $CL_{emp}N$ have been revised, if necessary, and summarised in separate tables, per chapter. In Chapter 10 the authors provide examples of how $CL_{emp}N$ can be used on different scales and in different European countries. In preparation of the CCE workshop (Berne, Switzerland, 26-28 October 2021), the draft of the background document was reviewed by a number of European experts and subsequently sent to all participants, after the suggested amendments had been processed.

11.2 Overview of the revised empirical N critical loads 2022

The text of this background document was intensively discussed and evaluated during the CCE workshop in Berne, held under the auspices of the Convention on Long-range Transboundary Air Pollution (CLRTAP Convention). At the end of this meeting, consensus was reached on the updated and revised list of empirical critical loads of N for natural and semi-natural ecosystems. Table 11.1 presents the new, revised empirical N critical loads.

Table 11.1. Overview of empirical N critical loads (kg N ha^{-1} yr^{-1}) to natural and semi-natural ecosystems (column 1), classified according to EUNIS (column 2), as established in 2011 (column 3), and as revised in 2022 (column 4). The reliability is indicated by ## reliable; # quite reliable and (#) expert judgement (column 5). Column 6 provides a selection of effects that may occur when critical loads are exceeded. Finally, changes with respect to 2011 are indicated as values in bold.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Marine habitats (MA)					
Atlantic upper-mid salt marshes	MA223	20-30	**10-20**	(#)	Increase in dominance of graminoids; decline positive indicator species
Atlantic mid-low salt marshes	MA224	20-30	**10-20**	(#)	Increase in late successional species; decline positive indicator species
Atlantic pioneer salt marshes	MA225	20-30	20-30	(#)	Increase in late successional species; increase in productivity species
Coastal habitat (N)					
Shifting coastal dunes	N13, N14	10-20	10-20	#	Biomass increase; increased N leaching; reduced root biomass
Coastal dune grasslands (grey dunes)	N15	8-15	**5**-15	##	Increased biomass and cover of graminoids and mesophilic forbs; decrease in oligotrophic species including lichens; increased tissue N; increased N leaching; soil acidification
Coastal dune heaths	N18, N19	10-20	10-**15**	#	Increased plant production; increased N leaching; accelerated succession; typical lichen C:N decrease; increased yearly increment *Calluna*
Moist and wet dune slacks	N1H	10-20	**5-15**	#	Increased cover of graminoids and mesophilic forbs; decrease in oligotrophic species; increased Ellenberg N
Dune-slack pools (freshwater aquatic communities of permanent Atlantic and Baltic or Mediterranean and Black Sea dune-slack water bodies)	N1H1, N1J1	10-20	10-20	(#)	Increased biomass and rate of succession

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Inland surface water habitats (C) [a]					
Permanent oligotrophic lakes, ponds and pools (including soft-water lakes)	C1.1	3-10	**2**-10 [b]	##	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P; shifts in macrophyte community
Alpine and sub-Arctic clear water lakes	C1.1		**2-4**	**##**	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Boreal clear water lakes	C1.1		**3-6**	**##**	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Atlantic soft water bodies	C1.1, elements C1.2	3-10	**5**-10	##	Change in species composition of macrophyte communities
Permanent dystrophic lakes, ponds and pools	C1.4	3-10	**5**-10 [c]	(#)	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P
Mire, bog and fen habitats (Q)					
Raised and blanket bogs	Q1	5-10	5-10	##	Increase in vascular plants; decrease in bryophytes; altered growth and species composition of bryophytes; increased N in peat and peat water
Valley mires, poor fens and transition mires	Q2	10-15	**5**-15	**##**	Increase in sedges and vascular plants; negative effects on bryophytes
Palsa and polygon mires	Q3		**3-10**	**(#)**	Increase in graminoids, tissue N concentrations and decomposition rate
Rich fens	Q41-Q44	15-30	15-**25**	**#**	Increase in tall vascular plants (especially graminoids); decrease in bryophytes
Arctic-alpine rich fens	Q45	15-25	15-25	(#)	Increase in vascular plants; decrease in bryophytes
Grasslands and tall forb habitats (R)					
Semi-dry Perennial calcareous grassland (basic meadow steppe)	R1A	15-25	**10-20**	##	Increase in tall grasses; decline in diversity; change in species composition; increased

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
					mineralisation; N leaching; surface acidification
Mediterranean closely grazed dry grasslands or Mediterranean tall perennial dry grassland or Mediterranean annual-rich dry grassland	R1D or R1E or R1F	15-25	**5-15**	(#)	Increased production; dominance by graminoids; changes to soil crusts; changes to soil nutrient cycling
Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta*	R1M	10-15	**6-10**	##	Increase in graminoids; decline of typical species; decrease in total species richness
Oceanic to subcontinental inland sand grassland on dry acid and neutral soils or Inland sanddrift and dune with siliceous grassland	R1P or R1Q	8-15	**5**-15	(#)	Decrease in lichens; increase in biomass
Low and medium altitude hay meadows	R22	20-30	**10-20**	(#)	Increase in tall grasses; decrease in diversity; decline of typical species
Mountain hay meadows	R23	10-20	10-**15**	**#**	Increase in nitrophilous graminoids; changes in diversity; decline of typical species
Moist or wet mesotrophic to eutrophic hay meadow	R35	15-25	15-25	(#)	Increase in tall graminoids; decreased diversity; decrease in bryophytes
Temperate and boreal moist and wet oligotrophic grasslands	R37	10-20	10-20	#	Increase in tall graminoids; decreased diversity; decrease in bryophytes
▶ Moss and lichen dominated	(Earlier E4.2)	5-10	5-10	#	Change in species composition; effects on bryophytes or lichens

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
mountain summits					
▶ Temperate acidophilous alpine grasslands	R43	5-10	5-10	#	Changes in species composition; increase in plant production
Arctic-alpine calcareous grassland	R44	5-10	5-10	#	Changes in species composition; increase in plant production
Heathland, scrub and tundra habitats (S)					
Tundra	S1	3-5	3-5 [d]	#	Changes in biomass; physiological effects; changes in bryophyte species composition; decrease in lichen species richness
Arctic, alpine and subalpine scrub habitats	S2	5-15	5-**10** [d]	#	Decline in lichens; bryophytes and evergreen shrubs
Lowland to montane temperate and submediterranean *Juniperus* scrub	S31		**5-15**	**(#)**	Shift in vegetation community composition; reduced seed viability
Northern wet heath	S411				
▶ 'U' *Calluna*-dominated wet heath (upland)	S411	10-20	**5–15** [e]	**##**	Decreased heather dominance; decline in lichens and mosses; increased N leaching
▶ 'L' *Erica tetralix*-dominated wet heath (lowland)	S411	10-20	**5-15** [e]	**##**	Transition from heather to grass dominance; decrease in heather cover; shift in vegetation community composition
Dry heaths	S42	10-20	**5-15** [e]	##	Transition from heather to grass dominance; decline in lichens; changes in plant biochemistry; increased sensitivity to abiotic stress
Maquis, arborescent matorral and thermo-	S5	20-30	**5-15**	(#)	Change in plant species richness and community composition; nitrate leaching; acidification of soil.

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Mediterranean scrub					
Garrigue	S6		**5-15**	**#**	Changes in species composition; decline in shrub cover; increased invasion of annual herbs
Forest habitats (T)					
Broadleaved deciduous forest	T1	10-20	10-**15**	##	Changes in soil processes; nutrient imbalance; altered composition mycorrhiza and ground vegetation
Fagus forest on non-acid and acid soils	T17, T18	10-20	10-**15**	(#)	Changes in ground vegetation and mycorrhiza; nutrient imbalance; changes in soil fauna
Mediterranean *Fagus* forest on acid soils	T18		**10-15**	**(#)**	Annual height and volume tree growth; analogy to temperate *Fagus* forest
Acidophilous *Quercus* forest	T1B	10-15	10-15	(#)	Decrease in mycorrhiza; loss of epiphytic lichens and bryophytes; changes in ground vegetation
Carpinus and *Quercus* mesic deciduous forest	T1E	15-20	15-20	(#)	Changes in ground vegetation
Mediterranean evergreen *Quercus* forest	T21	10-20	10-**15**	(#)	NO_3 in soil water and streams
Coniferous forests	T3	5-15	**3**-15	##	Changes in soil processes; nutrient imbalance; altered composition mycorrhiza and ground vegetation; increase in mortality with drought
Temperate mountain *Picea* forest, Temperate mountain *Abies* forest	T31, T32	10-15	10-15	(#)	Decreased biomass of fine roots; nutrient imbalance; decrease in mycorrhiza; changed soil fauna
Mediterranean mountain *Abies* forest	T33		**10-15**	**(#)**	Tree foliar stoichiometry; tree physiology; soil N losses
Temperate continental *Pinus sylvestris* forest	T35	5-15	5-15	#	Changes in ground vegetation and mycorrhiza; nutrient imbalances; increased N_2O and NO emissions

Ecosystem type	EUNIS code	2011 kg N ha^{-1} yr^{-1}	2022 kg N ha^{-1} yr^{-1}	2022 reliability	Indication of exceedance
Mediterranean montane *Pinus sylvestris-Pinus nigra* forest	T37		**5-17**	**(#)**	Lichen chemistry and community changes in Mediterranean mixed-conifer forests in USA
Mediterranean lowland to submontane *Pinus* forest	T3A	3-15	**5-10**	(#)	Reduction in fine-root biomass; shift in lichen community
Dark taiga	T3F	5-10	**3-5** [f]	##	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation
Pinus sylvestris light taiga	T3G	5-10	**2-5** [f]	#	Changes in epiphytic lichen and ground-layer bryophyte communities; increase in free-living algae; decline in N-fixation

a) The lower part of the $CL_{emp}N$ range should be applied for lakes in small catchments (with high lake to catchment ratios), because these are most exposed to atmospheric deposition, given that a relatively high fraction of their N inputs is deposited directly on the lakes and is not retained in the catchments. Similarly, the lower part of the range should be applied for lakes in catchments with thin soils, sparse vegetation and/or with a high proportion of bare rock.

b) This $CL_{emp}N$ should only be applied to oligotrophic waters with low alkalinity and with no significant agricultural or other human inputs. Apply the lower end of the range to clear-water sub-Arctic and alpine lakes, the middle range to boreal lakes and the higher end of the range to Atlantic soft waters.

c) This $CL_{emp}N$ should only be applied to waters with low alkalinity and with no significant agricultural or other direct human inputs. Apply the lower end of the range to boreal dystrophic lakes.

d) Use towards high end of range if phosphorus limited, and towards lower end if phosphorus is not limiting.

e) Use towards high end of range with high intensity management, and use towards lower end of range with low intensity management.

f) Mainly based on N deposition impacts on lichens and bryophytes.

Modifying factors

The modifying factors, i.e. general relationships between abiotic factors and empirical N critical loads, presented in Table 11.2, were defined as part of the earlier revision by Bobbink and Hettelingh (2011). This table was discussed at the $CL_{emp}N$ workshop in Noordwijkerhout in 2010. Each modifying factor was addressed by the workshop participants for each individual EUNIS category, with the aim of providing as specific instructions as possible for application in the different EUNIS classes (for more detailed information, see Bobbink and Hettelingh (2011), Appendices 5 to 7).

Table 11.2. Suggestions to apply lower, middle or upper parts of the set critical load ranges for terrestrial ecosystems (excluding wetlands), if national data are insufficient.

	Temperature/ frost period	Soil wetness	Base cation availability	Management intensity
Action				
Move to lower part	COLD/LONG	DRY	LOW	LOW
Use middle part	INTERMED	NORMAL	INTERMED	USUAL
Move to higher part	HOT/NONE	WET	HIGH	HIGH

As there is no consensus on how to quantify the modifying factors for broad regional scale assessments, it is proposed to use the minimum value of the ranges of $CL_{emp}N$ in each EUNIS class. This will enable comparison of their exceedances between different air pollution abatement scenarios. Furthermore, countries are advised to identify the highly sensitive receptor ecosystems within the EUNIS classification relating to their individual interest.

Several new national approaches have been developed in the UK and in the Netherlands to estimate or calculate critical loads for specific ecosystems that are part of a larger EUNIS class with an agreed range of $CL_{emp}N$ (Hall and Wadsworth, 2010; Van Dobben et al., 2014). Although this is an important development to provide a more complete list of critical loads for the large number of ecosystem types, it should be extended to a more European-based approach.

11.3 Gaps in knowledge and research needs

Most of the Earth's biodiversity is present in semi-natural and natural ecosystems. It is therefore crucial to control atmospheric N pollution to prevent negative impacts on these ecosystems. Effort should be directed to produce fine-resolution maps of sensitive ecosystems of high conservation value for each country in order to map critical loads of N for these systems. It is advisable to use both the mass balance and empirically derived N critical loads to improve the robustness of impact-based assessments of natural areas at risk from nitrogen deposition scenarios on a broad regional scale (see Hettelingh et al., 2007).

More information is needed on the relative effects of oxidised and reduced N deposition. At recent UNECE expert meetings, it was emphasised that there is increasing evidence that NH_x has a greater impact than NO_y. In particular, lichens and sometimes bryophytes in different ecosystems, as well as weakly buffered ecosystems, are likely to be more sensitive to deposition of reduced N. However, at present it is not possible to set critical loads for both forms of N, separately. In addition, the critical levels of NH_3 are in a process of revision in 2022 to 2023.

Serious gaps in knowledge exist on the effects of increased N deposition on semi-natural and natural ecosystems, although considerable progress has been made for several habitat groups, between 2010 and 2021. The following gaps in knowledge have been recognised as most important:

- more research and data are required to establish a critical load for the following ecosystems: several grasslands and hay meadows; all Mediterranean vegetation types; wet swamp forests; many mires and fens and several coastal habitats;
- more research is needed for all distinguished EUNIS habitat types that have an 'expert' judgement rating or for which only few studies are available;

- impacts of N enrichment in (sensitive) freshwater and shallow marine ecosystems (including coastal waters) need further research;
- additional efforts are needed to assign the observed N effects to the corresponding EUNIS forest habitat subtypes (level 3);
- in the last decade an increasing number of gradient studies on atmospheric N deposition in several EUNIS habitat types have been published and proved to be useful for evaluation of the ClempN. More gradient studies with both low and high N regions are needed, especially in EUNIS habitat types that are hardly investigated;
- more research is needed on the differential effects of the deposited N forms (NO_x or NH_y) in order to be able to determine the critical loads for oxidised and reduced nitrogen separately in the future;
- in order to refine the current critical loads, long-term (10-20 years) N addition experiments with a high frequency of N treatments between 5 and 50 kg N ha^{-1} yr^{-1} in regions with low background depositions are useful. This would increase the reliability of the derived critical loads if the lowest treatment level does not exceed the critical load;
- climate change and nitrogen deposition are likely to have strong interactive effects on ecosystem functioning and climate change may alter ecosystem responses to nitrogen deposition and vice versa. More experimental studies are needed to investigate these interactions, and also more gradient studies that explicitly examine the impacts of nitrogen deposition in combination with climatic gradients;
- at present, there are only a few studies that have looked at both the impact of and recovery from nitrogen deposition. More studies on this topic are needed to better understand the reversibility of nitrogen deposition impacts and the long-term projections for N-polluted ecosystems.

In conclusion, it is crucial to understand the long-term effects of increased N deposition on ecosystem processes in a representative range of ecosystems. Therefore, it is important to quantify the effects of N deposition through the manipulation of N inputs in long-term ecosystem studies in both pristine and affected areas. These data in combination with gradient studies are essential to validate critical loads and develop robust dynamic ecosystem models and/or multiple correlative species models that are reliable enough to use in the calculation of $CL_{emp}N$ for natural and semi-natural ecosystems, and to predict natural recovery rates for N-affected systems.

11.4 References

Achermann, B. and Bobbink, R. (eds.) (2003). *Empirical critical loads for nitrogen.* Environmental Documentation No.164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Bobbink, R., Boxman, D., Fremstad, E., Heil, G., Houdijk, A. and Roelofs, J. (1992). *Critical loads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna.* In: Grennfelt, P. and Thörnelöf, E. (eds.), Critical loads for nitrogen, Nord 41, Nordic Council of Ministers, Copenhagen. pp. 111.

Bobbink, R., Hornung, M. and Roelofs, J.G M. (1996). *Empirical nitrogen critical loads for natural and semi-natural ecosystems.* In: Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded, Texte 71/96, III-1-54. Federal Environmental Agency, Berlin.

Bobbink, R. and Hettelingh, J.P. (eds.) (2011). *Review and revision of empirical critical loads and dose-response relationships.* Coordination Centre for Effects, National Institute for Public Health and the Environment (RIVM).

Bobbink, R., Ashmore, M., Braun, S., Fluckiger, W. and Van den Wyngaert, I.J.J. (2003). *Empirical nitrogen critical loads for natural and semi-natural ecosystems: 2002 update.* Environmental Documentation No. 164 Air, pp. 43-170. Swiss Agency for Environment, Forest and Landscape SAEFL, Berne.

Bull K.R., Achermann B., Bashkin V., Chrast R., Fenech G., Forsius M., Gregor H-D., Guardans R., Haußmann T., Hayes F., Hettelingh J.-P., Johannessen T., Krzyzanowski M., Kucera V., Kvaeven B., Lorenz M., Lundin L., Mills G., Posch M., Skjelkvåle B.L., Ulstein M.J. (2001). Coordinated effects monitoring and modelling for developing and supporting international air pollution control agreements. *Water, Air and Soil Pollution* **130,** 119-130.

Chytrý, M., Tichý, L., Hennekens, S.M., Knollová, I., Janssen, J.A., Rodwell, J.S., Peterka, T., Marcenò, C., Landucci, F. and Danihelka, J. (2020). EUNIS Habitat Classification: Expert system, characteristic species combinations and distribution maps of European habitats. *Applied Vegetation Science* **23**, 648-675.

Hall, J. and Wadsworth, R. (2010). Estimating the effect of abiotic factors on modifying the sensitivity of vegetation to nitrogen deposition: an application of endorsement theory. *Water, Air and Soil Pollution* **212**: 441-459.

Hettelingh J.-P., Posch M. and De Smet P.A.M. (2001). Multi-effect critical loads used in multi-pollutant reduction agreements in Europe. *Water, Air and Soil Pollution* **130,** 1133-1138.

Hettelingh J.-P., Posch M., Slootweg J., Reinds G.J., Spranger T. and Tarrason L. (2007). Critical loads and dynamic modelling to assess European areas at risk of acidification and eutrophication. *Water, Air and Soil Pollution: Focus* **7**, 379-384.

ICP M&M (2017). *Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends.* Chapter 5: Mapping Critical Loads for Ecosystems. Available here: https://www.umweltbundesamt.de/en/manual-for-modelling-mapping-critical-loads-levels?parent=68093, last accessed 11.03.2022.

Van Dobben, H., Bobbink, R., Bal, D. and Van Hinsberg, A (2014). *Overview of critical loads for nitrogen deposition for Natura 2000 habitat (sub)types occurring in The Netherlands.* Wageningen, Alterra Wageningen UR (University and Research Center), Alterra report 2488.

WHO (2000). *Air quality guidelines for Europe, second edition.* WHO regional publications, European series, No. 91. World Health Organisation, Regional office for Europe, Copenhagen.

Glossary

Glossary Item	Description
Acidification (referring to soil)	Acidification is defined by the loss of acid neutralizing capacity (ANC) (soil buffering capacity). It is a gradual process leading to leaching of base cations (calcium, magnesium, potassium) from the topsoil, through leaching, and replacement by acidic elements (especially aluminium).
Aerosols	Solid or liquid particles suspended in the air. This includes dust, soot and sea-salt crystals, size 1 nm to 100 µm.
Al	Aluminium
Ammonia	NH_3 is a colourless gas with a pungent odour in high concentration Ammonia is product of 'biological nitrogen fixation' (see entry), it can also be synthesised using the 'Haber–Bosch' process (see entry). Livestock farming largely contributes to NH_3 emissions.
Ammonium	Is a monovalent cation (NH_4^+), closely linked to production and solution of ammonia in water. Is a form of reactive nitrogen. Constituent of many synthetic fertilisers, such as ammonium nitrate.
ANC	Acid Neutralizing Capacity
Anthropogenic	Effects which relate specifically to human activities, i.e. anthropogenic reactive nitrogen production, through the 'Haber–Bosch' process.
Anthroposphere	All parts of the planetary system which are affected by human activities.
Arborescent strata	Layer of trees in a plant community.
Arginine	N-rich amino acid, the arginine concentration in plant material often increased with higher N deposition.
Atmospheric deposition	Removal of suspended material from the atmosphere, this can be classified as either 'wet' or 'dry'. Wet deposition occurs when material is removed from the atmosphere by precipitation. In dry deposition, the material is removed from the atmosphere by interference with surfaces.
Autotroph	Organism (primary producer) that converts energy (sun light) into organic compounds which can be used by other organisms (see Heterotroph).
Biodiversity	Biodiversity is the variability of life on earth. It is a measure of variation at the genetic, species, and ecosystem level. The value of biodiversity is multifold, from preserving the integrity of the biosphere as a whole, to providing fodder, food and medicine, to spiritual and aesthetic well-being. Biodiversity and plant diversity are often used as synonyms.
Biological Nitrogen Fixation (BNF)	"Fixing" of unreactive di-nitrogen (N_2) to reactive nitrogen species by microorganisms (Rhizobium, Frankia, free-living etc.).

Glossary Item	Description
Bulk deposition	Deposition collected in the open field with a funnel, measuring wet deposition with some dry deposition (15-30%).
Bryophytes	Earliest land plants on earth, consisting of mosses, liverworts and hornworts. They are non- vascular plants.
Carbon sequestration	The capture and removal of carbon dioxide from the atmosphere and storing it in a reservoir, storage in soil organic matter is one of the most efficient ways.
CLRTAP	Convention on Long-range Transboundary Air Pollution, under the United Nations Economic Commission for Europe (UNECE).
C:N ratio	Ratio of carbon (C) to nitrogen (N) in soil and of litter. C:N < 25 mineralisation dominates, C:N > 25 immobilisation dominates.
Chlorophyll a	Photosynthetic pigment. In aquatic ecosystem, the chlorophyll a is used as a proxy for algal biomass production.
CH_4	Methane, is a colourless, odourless, flammable gas that is the simplest hydrocarbon and is the major constituent of natural gas.
CO_2	Carbon dioxide, is a chemical compound composed of two oxygen atoms and one carbon atom.
Critical level	Concentration of an atmospheric pollutants above which direct harmful effects on sensitive vegetation elements may occur according to present knowledge.
Critical loads	A quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge.
Cryptogam	A cryptogam (scientific name Cryptogamae) is a plant (in the wide sense of the word) or a plant-like organism that reproduces by spores, without flowers or seeds.
Denitrification	A microbially facilitated process (mainly by heterotrophic bacteria, but also by fungi) where nitrate is reduced and di-nitrogen (N_2) is ultimately produced through series of intermediate or nitrous oxide products, including the greenhouse gas N_2O.
DCA	Detrended Correspondence Analysis
DIN	Dissolved Inorganic Nitrogen
DOM	Dissolved Organic Matter
DON	Dissolved Organic Nitrogen
Di-nitrogen	N_2, a colourless, odourless, and inert gas which makes up approximately 78% of the atmosphere. Di-nitrogen is the thermodynamically stable state (‘unreactive nitrogen’) in contrast to many different reactive nitrogen forms. Also called nitrogen gas.

Glossary Item	Description
Dystrophic	Description for water bodies (lakes), rich in dissolved organic carbon (humic acids), low in productivity (see oligo- and eutrophic).
Ellenberg N indicator values	Ellenberg indicator values (EIVs) are ordinal estimates of species ecological optima along seven main ecological gradients - light, temperature, continentality, moisture, nutrients, soil reaction and salinity. Here for nutrients.
EMEP	European Monitoring and Evaluation Programme. Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe.
EUNIS	Stands for European Nature Information System. The EUNIS habitat classification is a comprehensive pan-European system for habitat identification. The classification is hierarchical and covers all types of habitats from natural to artificial, from terrestrial to aquatic. The habitat types are identified by specific codes (first revision with code changes started in 2015).
Eutrophication	The enrichment of the nutrient load in ecosystems (terrestrial and aquatic), especially by nitrogen and/or phosphorus compounds. This disturbs the balance of organisms in the ecosystem, affecting terrestrial and aquatic biodiversity and water quality.
Exceedance	The concentration or amount of pollution above a 'critical level' or 'critical load'.
Food webs	Terrestrial, aquatic food webs, interconnected food chains on "what/who" eats "what/who".
Forb	An herbaceous flowering plant that is not a graminoid (grass, sedge, or rush). The term is used in biology and in vegetation ecology, especially in relation to grasslands and understory.
Garrigue	Mediterranean shrub habitat, consisting of sclerophyllous shrubs, more open than maquis.
Gradient	Variable atmospheric N deposition in space or time. In the present revision, spatial gradients are included to assess critical N loads.
Greenhouse Gas	GHG includes carbon dioxide (CO_2), nitrous oxide (N_2O), methane (CH_4), ozone (O_3), water vapour and various other gases.
Haber–Bosch process	The high-pressure, chemical process which synthesises reactive nitrogen as ammonia (NH_3) from the reaction of N_2 and H_2. Fritz Haber discovered the process (1908) and Carl Bosch developed the technique at an industrial scale.
Heathland	Various types of plant communities, commonly with a dominance of dwarf shrubs.
Herbaceous	A plant whose stem does not become woody and persistent (as in a tree or shrub) but remains soft and succulent, and dies (completely or down to the root) after flowering.
Herbivores	Organisms (insects, mammals) that feed on plants.

Glossary Item	Description
Heterotroph	Organism that cannot produce its own food.
Isotopes	Same chemical element (non-radioactive) with same number of electrons in the nuclei, but different number of neutrons in the nuclei (stable isotopes).
Immobilisation (referring to soil)	The incorporation of reactive nitrogen compounds into soil microbial biomass.
Leaching	The washing out of soluble ions (mainly of mobile nitrate ions) and compounds by water draining through soil.
Legumes	Herbs that fix di-nitrogen from the atmosphere, by means of symbiotic microorganisms in the plant roots.
Lichen	Lichen is a symbiosis between fungi and algae. The algae photosynthesise (in some cases they also fix N_2) and the fungi provide nutrients from the environment. Terricolous lichens grow on soil, epiphytic lichens on branches or bark of trees. Most lichens can dehydrate completely without damage.
Litter	Dead, detached plant material. Litter decomposition (mineralisation) is an important part of the nutrient cycle.
Littoral	Part of a sea, lake, or river that is close to the shore, includes the intertidal zone.
Natura 2000	Protected areas in European Union, implanted by the EU Habitats Directive (1992).
Mineralisation	Decomposition process where organic compounds are transformed to inorganic compounds (N and C mineralisation).
Maquis	Mediterranean shrub habitat, consisting of densely growing evergreen shrubs (see Garrigue).
Macrophytes	Aquatic plants, term to distinguish them from algae and other microphytes.
N	Nitrogen
NDVI	Normalised Difference Vegetation Index = Cover of green biomass
NEP	Net Ecosystem Productivity
Nitrification	The biological oxidation of ammonia via nitrite to nitrate (aerobic process).
Nitrophilous	Translates to 'nitrogen-loving'. A species which may be nitrogen limited and then benefits from increased nitrogen availability in the environment.
N_2O	Laughing gas, produced through denitrification, strong greenhouse gas. Not accounted for the total atmospheric N deposition.
NPK fertiliser	Artificial fertiliser containing the three major plant nutrients (nitrogen, phosphorus and potassium).

Glossary Item	Description
Oligotrophic	'Poor in nutrient'. The opposite of eutrophic (see Eutrophication).
Retention	Retention of N in the soil, mainly in the soil organic matter (SOM). Expected N losses due to high N deposition are lower than expected.
Phosphorus limitation	Plant phosphorus (P) limitation is common, primarily because soil P forms often have a low plant availability. Chronically elevated N deposition often affects plant P availability (so called N-induced P limitation, increased foliar N:P ratios).
Photooxidation	The light-dependent generation of active oxygen species is termed photooxidative stress. This can occur in two ways: 1) the donation of energy or electrons directly to oxygen as a result of photosynthetic activity; 2) exposure of tissues to ultraviolet irradiation. Photooxidation is responsible for the accumulation of harmful reactive oxygen species in plant tissues.
PME:NR ratio	Phosphomonoesterase:nitrate reductase ratio
Pristine	State or area which is not affected by atmospheric N deposition or other pollutants.
Relevés	A vegetation relevé is a recording of a sample of a plant community. For each plant species, the cover, abundance, frequency, vitality, fertility and/or the phenological condition in a specific test area and of the spatial vegetation structure are recorded.
Sclerophyllous shrub	A type of shrub that is adapted to long periods of dryness and heat. The plants feature hard leaves, short internodes (the distance between leaves along the stem) and leaf orientation which is parallel or oblique to direct sunlight. The word comes from the Greek sklēros (hard) and phyllon (leaf).
Synthetic fertiliser	Industrially produced fertiliser, using the 'Haber–Bosch' process.
Simple mass balance	Calculation method based on inputs and outputs of the variable of concern (nitrogen, acidity, base cations).
Throughfall	Rainwater (precipitation) falling through the canopy (foliage) of a forest (or crop). Throughfall N is commonly lower than total atmospheric N.
TITAN	Stands for Threshold IndicatorTaxa ANalysis (TITAN). Analytical method to detect changes in taxa distributions along an environmental gradient (N gradient) over space or time.
TP	Total Phosphorus
Vascular plant	A vascular plant is any one of a number of plants with specialised vascular tissue. The two types of vascular tissue, xylem and phloem, are responsible for moving water, minerals, and the products of photosynthesis throughout the plant.

Appendix 1: Classification of habitats according to EUNIS and EU Habitats Directive (Natura 2000, Annex I)

This appendix gives the correspondence between ecosystems, classified according to EUNIS (European Nature Information System) (Chytrý et al., 2020) and the EU Habitats according to Directive Annex I. Only the EUNIS classes for which $Cl_{emp}N$ has been set are included in the table below. Empirical critical loads of N, as set in this background document, can be assigned to EU habitats with the same reliability as for the corresponding EUNIS classifications (see Table 11.1).

Ecosystem type	EUNIS code	EU habitats[a]
Marine habitats (MA)		
Atlantic upper-mid salt marshes	MA223	1330
Atlantic mid-low salt marshes	MA224	1330
Atlantic pioneer salt marshes	MA225	1310, 1320, 1330
Coastal habitat (N)		
Shifting coastal dunes	N13, N14	2120, *2110*
Coastal dune grasslands (grey dunes)	N15	2130, 21A0
Coastal dune heaths	N18, N19	2140, 2150
Moist and wet dune slacks	N1H	2190, 21A0
Dune-slack pools (freshwater aquatic communities of permanent Atlantic and Baltic or Mediterranean and Black Sea dune-slack water bodies)	N1H1, N1J1	x
Inland surface water habitats (C)		
Permanent oligotrophic lakes, ponds and pools (including soft-water lakes)	C1.1	3110, 3130
Alpine and sub-Arctic clear water lakes	C1.1	3110, 3130
Boreal clear water lakes	C1.1	3110, 3130
Atlantic soft water bodies	C1.1, elements C1.2	3110, 3130
Permanent dystrophic lakes, ponds and pools	C1.4	3160
Mire, bog and fen habitats (Q)		
Raised and blanket bogs	Q1	7110, 7130
Valley mires, poor fens and transition mires	Q2	7140
Palsa and polygon mires	Q3	7320
Rich fens	Q41-Q44	7230
Arctic-alpine rich fens	Q45	7240
Grasslands and tall forb habitats (R)		

Ecosystem type	EUNIS code	EU habitats[a]
Semi-dry perennial calcareous grassland (and basic meadow steppe) or Continental dry grassland (true steppe)	R1A or R1B	6210, 6280, *62A0, 6240, 6250, 6270, 62C0* or 6240, 6250, 62C0
Mediterranean closely grazed dry grasslands or Mediterranean tall perennial dry grassland or Mediterranean annual-rich dry grassland	R1D or R1E or R1F	6220
Lowland to montane, dry to mesic grassland usually dominated by *Nardus stricta* or Boreal and arctic acidophilous alpine grassland	R1M or R42	6230 or 6150
Oceanic to subcontinental inland sand grassland on dry acid and neutral soils or Inland sanddrift and dune with siliceous grassland	R1P or R1Q	6120, 2330, *6270* or 2330, 2340
Low- and medium altitude hay meadows	R22	6270, 6510
Mountain hay meadows	R23	6520
Moist or wet mesotrophic to eutrophic hay meadow	R355	x
Temperate and boreal moist and wet oligotrophic grasslands	R372	x
▶ Moss and lichen dominated mountain summits	(Earlier E4.2)	x
▶ Temperate acidophilous alpine grasslands	R43	6140, 6150, 62D0, *6230*
Arctic-alpine calcareous grassland	R44	6170
Heathland, scrub and tundra habitats (S)		
Tundra	S1	x
Arctic, alpine and subalpine scrub habitats	S2	4060, 4080
Lowland to montane temperate and submediterranean *Juniperus* scrub	S31	5130
Northern wet heath	S411	4010
▶ ‘U’ *Calluna*-dominated wet heath (upland)	S411	4010
▶ ‘L’ *Erica tetralix*-dominated wet heath (lowland)	S411	4010
Dry heaths	S42	2310, 2320, 4030, 4040, *5140*
Maquis, arborescent matorral and thermo-Mediterranean scrub	S5	x
Garrigue	S6	x

Ecosystem type	EUNIS code	EU habitats[a]
Forest habitats (T)		
Broadleaved deciduous forest	T1	x
Fagus forest on non-acid and acid soils	T17, T18	9110, 9120, 9130, 9140, 9150, 91W0, 9210, 9220, 9270, 9280, 91K0, 91S0, 91V0, 91W0, 91X0, *9260*
Mediterranean *Fagus* forest on acid soils	T18	9110, 9120, 91W0, *9260*
Acidophilous *Quercus* forest	T1B	9190, 9260, 91A0
Carpinus and *Quercus* mesic deciduous forest	T1E	9020, 9160, 9170, 91G0, 91L0, 91Y0, *9260*
Mediterranean evergreen *Quercus* forest	T21	9330, 9340, 9390, 93A0
Coniferous forests	T3	x
Temperate mountain *Picea* forest, Temperate mountain *Abies* forest	T31, T32	9410, 91BA, 91P0, *9110, 9120, 9140, 9270, 9410*
Mediterranean mountain *Abies* forest	T33	9510, 9520, *9270*
Temperate continental *Pinus sylvestris* forest	T35	91C0, 91T0, 91U0, *9060*
Mediterranean montane *Pinus sylvestris-Pinus nigra* forest	T37	9530
Mediterranean lowland to submontane *Pinus* forest	T3A	9540
Dark taiga	T3F	9050, *9010*
Pinus sylvestris light taiga	T3G	9010, 9060

a) Relationships concern revised EUNIS level 3 only. When the revised EUNIS habitat has no relationship to Annex I habitat types this is indicated with x in field 'EU habitats'. *Italics* indicate relationships of low importance, in the sense that only a very small part of the Annex I habitat is crosslinked with the EUNIS habitat. In many cases low importance indicates that the relationship exist only under a specific interpretation of the Annex I habitat.

Appendix 2: Agenda of the expert workshop on empirical critical loads of nitrogen

Tuesday 26th October 2021		
8:30	*Welcome coffee / Registration*	
Welcome and introduction *Chair: Reto Meier*		
9:00	Welcome Address and Organizational Information	Reto Meier
9:15	Objectives of the workshop	Alice James Casas, Christin Loran
Presentation of scientific background information *Chair: Roland Bobbink*		
9:30	Review and revision procedure	Roland Bobbink
10:00	Marine habitats (EUNIS class MA)	Roland Bobbink
10:20	Coastal habitats (EUNIS class N)	Laurence Jones
10:40	*Coffee Break*	
11:00	Inland surface waters (EUNIS class C)	Christin Loran, Heleen De Wit
11:20	Discussion (EUNIS class MA, N, C)	
11:40	Mires, bogs and fens (EUNIS class Q)	Chris Field
12:00	*Coffee Break*	
13:30	Grasslands and lands dominated by forbs, mosses or lichens (EUNIS class R)	Vegar Bakkestuen
13:50	Heathland, scrub and tundra (EUNIS class S)	Leon Van den Berg
14:10	Discussion (EUNIS class Q, R, S)	
14:30	*Coffee Break*	
15:00	Woodland, forest and other wooded land (EUNIS class T)	Sabine Braun
15:20	Discussion (EUNIS class T)	
15:40	General Discussion	

Wednesday 27th October 2021		
Application of empirical Critical Loads *Chair: Reto Meier*		
9:00	Aspects of application	Markus Geupel
10:30	*Coffee Break*	Alice James Casas, Christin Loran

Wednesday 27th October 2021		
Working group specific discussions *Discussions chaired by lead authors*		
11:00	Working group 1 - EUNIS class MA, N, C	
	Working group 2 - EUNIS Q, R	
	Working group 3 - EUNIS S, T	
12:30	*Lunch Break*	
14:00	Working group 1 - EUNIS class MA, N, C	
	Working group 2 - EUNIS Q, R	
	Working group 3 - EUNIS S, T	
19:00	*Light show "Rendez-vous Bundesplatz"*	
20:00	*Dinner at Restaurant Altes Tramdepot*	

Thursday 28th October 2021		
Concluding session *Chair: Christin Loran*		
09:00	Reporting of working groups to plenary	
10:40	*Coffee Break*	
11:00	Discussion	
12:00	Conclusion	

Appendix 3: List of participants of the expert workshop on empirical critical loads of nitrogen

Surname	Name	Institute
Aazem	Khalid	Joint Nature Conservation Committee
Aherne	Julian	Trent University
Alonso	Rocío	CIEMAT
Augustin	Sabine	BAFU
Bakkestuen	Vegar	NINA
Bobbink	Roland	B-WARE
Braun	Sabine	IAP
Britton	Andrea	James Hutton Institute
Caporn	Simon	Manchester Metropolitan University
De Wit	Heleen	NIVA
Field	Chris	Manchester Metropolitan University
Garcia Gómez	Héctor	CIEMAT
Geupel	Markus	UBA
Greaver	Tara	US EPA
Hayes	Felicity	CEH
Hicks	Kevin	University of York
Hiltbrunner	Erika	University of Basel
James Casas	Alice	INERIS
Jones	Laurence	CEH
Karlsson	Per Erik	IVL
Kohli	Lukas	Hintermann & Weber
Loran	Christin	UBA
Manninen	Sirkku	University of Helsinki
Meier	Reto	BAFU
Perring	Mike	CEH
Posch	Max	IIASA/CIAM
Prescher	Anne-Katrin	Thuenen Institute of Forest Ecosystems
Roth	Tobias	Hintermann & Weber
Rowe	Ed	CEH
Scheuschner	Thomas	UBA

Surname	Name	Institute
Tomassen	Hilde	B-WARE
Tresch	Simon	IAP
Ukonmaanaho	Liisa	Luke
Van den Berg	Leon	Bosgroep Zuid Nederland
Vanguelova	Elena	Forest Research UK
Velle	Liv Guri	Møreforsking
Zappala	Susan	Joint Nature Conservation Committee

A.2 Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations - NEA001

FOR INTERNAL USE ONLY

Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations

Version: June 2018

Green-winged orchids *Anacamptis morio* on a roadside verge (by kind permission of Mark Meijrink @ http://markmeijrink.wordpress.com)

Contents

Natural England Internal Note on Ways of Working:

Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations

1. Introduction

1.1 This internal operational Guidance Note describes how Natural England advises competent authorities and others on the assessment of plans and projects (as required by the Conservation of Habitats and Species Regulations 2017 ('the Habitats Regulations')) likely to generate road traffic emissions to air which are capable of affecting European Sites[1].

The terms used throughout this note are referred to with regard to the Habitats Regulations assessment (HRA) procedure. The meaning of these terms is separate and distinct from the meaning of similar terms associated with Environmental Impact Assessment (EIA) procedures[2]. HRA and EIA can be compared as follows:

Framework	Relevance step	Detailed assessment step
Habitats Regulations Assessment	Likely Significant Effect Test	Adverse Effect Test
Environmental Impact Assessment	Screening	Significance Test

Natural England's Role as Advisor under the Habitats Regulations

1.2 Natural England plays several roles in the implementation of the Habitats Regulations, acting as an advisory 'nature conservation body' under Regulation 5 and as a 'competent authority' as defined under Regulation 7. As a competent authority, Natural England must formally assess new plans or projects which are (a) subject to the section 28 SSSI notice and consent procedures under Regulation 24 and (b) any plans or projects we are planning to undertake ourselves or give our authorisation or permission to under regulation 63.

[1] The term 'European Site' applies here to the following Protected Sites occurring in England; Special Areas of Conservation (SACs), candidate SACs, Special Protection Areas (SPAs), Sites of Community Importance (SCIs), potential SPAs, possible SACs, listed or proposed Ramsar sites and sites identified, or required, as compensatory measures for adverse effects on these European sites (see also page 28 of the National Planning Policy Framework 2012 and regulation 8 of the Habitats Regulations 2017.

[2] The EIA of certain projects under the EU Directive (2014/52/EU) on the assessment of the effects of certain public and private projects on the environment as transposed by the UK into various EIA Regulations covering town and country planning, infrastructure planning, forestry, agriculture and marine works (for an overview see https://www.gov.uk/guidance/environmental-impact-assessment)

1.3 This guidance is concerned with Natural England's other role as advisor to other competent authorities, acting as a '*nature conservation body*' according to regulation 5, also referred to in the Regulations as '*the appropriate nature conservation body*'. This definition also includes our sister agencies the Natural Resources Wales and Scottish Natural Heritage.

1.4 It is a statutory requirement under regulation 64(3) for competent authorities to consult Natural England for its views when they are carrying out an Appropriate Assessment (AA) and to '*have regard*' to any representations that we may make. Although there is no statutory requirement at the earlier step of determining 'likely significant effect', we are also likely to be consulted by other competent authorities for a 'screening opinion' or for further advice on the scope of an appropriate assessment, particularly where they do not have access to ecological expertise. This advice is increasingly delivered through Natural England's Discretionary Advice Service.

Who is this Guidance Note for?

1.5 This is internal guidance designed to assist Natural England staff when giving practical and proportionate advice to competent authorities and others about their assessment of the potential impacts from road traffic emissions on the qualifying features of European Sites. This Guidance Note has been prompted by the High Court judgment in Wealden v SSCLG [2017] ('the Wealden Judgment 2017').

1.6 It is worth noting the Dutch courts request for a preliminary ruling from the Court of Justice of the European Union ('CJEU') in C-294/17 on a series of questions relating to the implementation of the Dutch State's national nitrogen strategy[3] in light of the Habitats Directive. Any ruling subsequently provided by the CJEU is also likely to be of interest to the UK and may affect the contents of this guidance.

1.7 This Guidance Note has been drafted to reflect Natural England's current operational approach to advising competent authorities on air quality matters affecting European Sites. External stakeholders should be mindful that this note may be subject to review in light of operational feedback, new authoritative decisions and any subsequent reform of or changes to Natural England's general approach to giving its advice.

[3] See http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:62017CN0294

Why has this guidance note been made public?

1.8 This internal guidance has been made public for general information purposes to explain Natural England's approach to assessing the effects of road traffic emissions on European Sites particularly in light of the Wealden Judgment 2017. This version of Natural England's internal guidance note has been modified to remove references to Natural England internal information sources so that it is clear to an external audience.

1.9 Natural England has provided this general guidance to its staff on the factors to consider when advising a competent authority on the HRA of plans and projects generating road traffic and air pollution effects. It cannot cater for all situations and where local factors or information indicate that it would be inappropriate to rely on this guidance, it advises staff to seek further internal advice and/or advise that the plan or project should progress to appropriate assessment.

1.10 Publication of this internal guidance does not replace the need for competent authorities to consult Natural England where appropriate. Competent authorities and other third parties seeking Natural England's advice in relation to specific plans or projects should continue to consult Natural England in the usual way.

1.11 In addition to this guidance note, competent authorities and other third parties may also wish to seek the expert advice of other relevant statutory bodies as appropriate, such as the Environment Agency, and refer to other technical guidance on air quality matters or the Habitats Regulations Assessment process.

This internal Guidance Note includes Natural England's own interpretation of the law as it applies to air quality matters affecting European Sites. It does not constitute legal or professional advice to competent authorities or to any other third party. No warranty is given nor liability accepted for the contents of this internal Guidance Note. Competent authorities and other parties should seek their own legal advice.

What's covered by this Internal Guidance Note

1.12 This guidance outlines Natural England's approach to advising competent authorities on air quality assessment and identifies data sources to:

- allow competent authorities to have regard to these matters when they undertake their statutory duties and reach their conclusions on Habitats Regulations Assessments

- identify when Natural England is likely to advise no further assessment is required

- identify when Natural England is likely to advise detailed assessment and bespoke advice may be required, and,
- assist Natural England staff when drafting advice on potential impacts from air pollution.

1.13 **This guidance is applicable when Natural England gives its advice on plans and projects involving the following;**

- Emissions from road traffic likely to be generated by new development projects including residential, mixed use and industrial/commercial developments
- Emissions from road traffic likely to result from allocations in strategic Local Plans
- Emissions from proposed road schemes

What's not covered by this Internal Guidance Note

1.14 This guidance focusses on ecological receptors and does not cover human health.

1.15 This guidance is limited to plans or projects with road traffic emissions. It does not apply where the subject plan or project relates to non-road point sources or Environmental Permitting of intensive livestock units.

1.16 This guidance does not specifically cover nationally significant sites such as Sites of Special Scientific Interest (SSSIs), which are covered by a different regulatory framework. However, the general principles for air quality assessment outlined here for European Sites are likely to be equally relevant for this and other designations.

1.17 This guidance does not cover the further stages of the HRA process (tests for alternative solutions, imperative reasons of overriding public interest and compensation measures (stages 3 and 4 in Figure 1) which will be based on more bespoke advice and should be led by the competent authority responsible for the HRA.

2. Overview - how might European sites be adversely affected by air pollution?

2.1 Air pollution that typically affects habitat will include dust and particulate matter (PM), nitrogen oxides (NOx), ammonia (NH_3) and sulphur dioxide (SO_2). Each proposal type will have emissions typically associated with its specific activity. For example, ammonia is typically associated with farming or waste management. Combustion sources such as industry or traffic are more likely to be associated with nitrogen oxides and particulate matter.

2.2 Generally speaking, the risks to qualifying features from air pollution (in simple terms) most frequently arise from:

a) The direct effects which arise when a pollutant which is dispersed in the air is taken up by vegetation (through pores on the surface called stomata). Pollutants taken up by vegetation can cause adverse impacts to plant health and viability. The relevant assessment benchmark for pollutant concentrations 'in the air' is referred to as a **critical level** expressed in units of $\mu g/m^3$ (micrograms per cubic metre).

b) There are indirect effects which arise when the pollutant settles onto the ground (referred to as 'deposition') causing nutrient enrichment of the soil ('eutrophication') or changes to the soil pH ('acidification'). These effects can decrease the ability of a plant to compete with other plants and can hinder the inherent capacity for self-repair and self-renewal under natural conditions. In other words, nitrogen acts as a fertiliser for plants that can thrive on high nitrogen levels and can dominate plant communities. The speed with which a given pollutant settles (or deposits) after it is released into the atmosphere is different for each pollutant, and is influenced by how dense (or heavy) the particles are. Some pollutants travel a long distance before deposition occurs whilst others will settle much closer to their source. Wind speed and direction will also have an influence on deposition properties.

   The relevant assessment benchmark for pollutant levels which settle from the air onto a surface (or deposit) is referred to as a **critical load** expressed in units of kilograms of nitrogen per hectare per year (Kg N/ha/yr) for nitrogen deposition or kilo-equivalents per hectare per year (Keq/ha/yr) for acid deposition**.**

2.3 The UK's Air Pollution Information System (APIS; http://www.apis.ac.uk/) provides an overview of deposition, air pollution effects on habitat and typical emissions arising from different proposal types in the APIS Starter's Guide to Air Pollution Sources. Further description of critical loads (deposition benchmarks) and critical levels (air concentration benchmarks) can be found on APIS Guide to Critical Loads and Levels. These topics are covered in more detail in subsequent sections of the guidance. All assessment stages rely on sufficient information to make a determination.

2.4 Road traffic is a source of NOx emissions, meaning that increases in traffic can represent a risk with regard to the potential effects associated with the exceedance of critical levels for sensitive vegetation. Traffic emissions can also be a short range contributor to nitrogen deposition.

3. Overview – an approach to the HRA of plans or projects with road traffic emissions

3.1 There are four stages to assessment for European Sites (see Figure 1). This guidance relates primarily to Stage 1 of the process and the scoping of a stage 2 appropriate assessment (as illustrated in Figure 1 below).

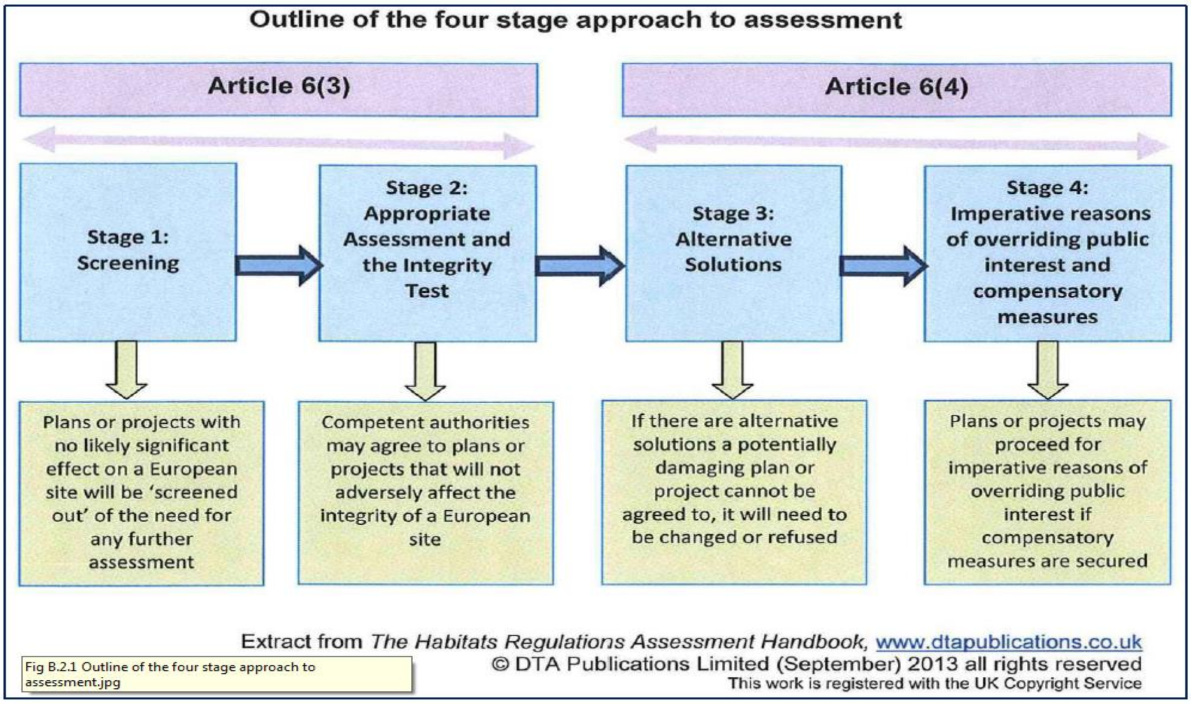


Figure 1: Overview of the Habitats Regulations Assessment procedures

3.2 Under the Habitats Regulations, it is the competent authority[4] who must carry out an appropriate assessment of any plan or project which is either not directly connected with or necessary to the management of a European Site and which is likely to have a significant effect on a European site. A competent authority should therefore decide for itself as to the likelihood of a significant effect on a site (stage 1 of Figure 1 above), but it is often the case that it may seek advice on this from Natural England (see section 4 below).

3.3 Furthermore, there is a statutory requirement for a competent authority to formally consult Natural England for the purposes of an appropriate assessment (Stage 2 in Figure 1 above). This is the only statutory input required from Natural England during the HRA process under the Habitats Regulations.

[4] The Habitats Regulations define a 'competent authority' as including any Minister of the Crown, government department, statutory undertaker, public body of any description or persons holding public office, or any person exercising those functions (regulation 7(1)).

Staff should be aware that, in accordance with Government's guidance on competent authority co-ordination when applying the Habitats Regulations, it is generally permissible for a competent authority to adopt, if it can, the assessment, reasoning and conclusions of another competent authority relating to the same plan or project, thus avoiding unnecessary duplication of effort. Staff are therefore encouraged to advise competent authorities to first check, at an early stage, the extent to which this might apply in relation to assessing road traffic emissions from an individual proposal. For example, the likely effects of a development proposal might have already been considered by a HRA of a Local Plan made by the same or another competent authority.

3.4 When specifically **advising** a competent authority at this screening stage of HRA as to whether the road traffic emissions associated with a plan or project are likely to have a significant effect on a European site, Natural England suggests a sequential approach can be taken to quickly filter out those proposals posing no credible risk.

3.5 Firstly it considers the evidence about emission types and distance that emissions are likely to travel to identify whether a plan or project might pose a risk to a European site (***step 1***).If a proposal gives rise to emissions that are likely to reach a designated site, the screening assessment should, secondly, consider the sensitivity of the qualifying feature(s) at the designated site (***step 2***). Next, if the necessary information is available, establish the feature's location and its likely exposure to emissions (***step 3***) to confirm the presence or absence of a credible risk.

3.6 Where there is the potential for interaction between a sensitive feature and emissions, ascertain either the predicted increase in flow of road traffic associated with the plan or project ('AADT flow') or the predicted process-contribution as a % of the pollution benchmark to act as a screening threshold alone (***step 4a***) and, where the threshold is not exceeded alone, in-combination (***step 4b & 4c***). These steps inform a decision as to whether a more detailed 'appropriate assessment' is required. The requirement to specifically consider the risks of 'in-combination' effects is explained further starting at paragraph 4.31. Together, these steps represent the "likely significant effect" or "screening" stage. If a proposal alone is above the likely significant effect thresholds, there is no need to also look for the risk of in-combination effects before proceeding to the appropriate assessment stage.

3.7 If the likelihood for significant effect cannot be ruled out, Natural England should advise the competent authority that an appropriate assessment is needed (***step 5***). Appropriate assessment is intended to be proportionate to the risk from a plan or project and does not always require detailed modelling or large amounts of reporting. The appropriate assessment should focus on assessing more precisely the ecological impacts of the emissions on the site in view of its qualifying features and conservation objectives. It should take into account any detailed modelling that is or becomes available, the best available evidence as to ecological impacts, background levels and likelihood for future reductions. Natural England will be consulted by the competent authority for the purposes of the assessment and asked for its advice (***step 6***).

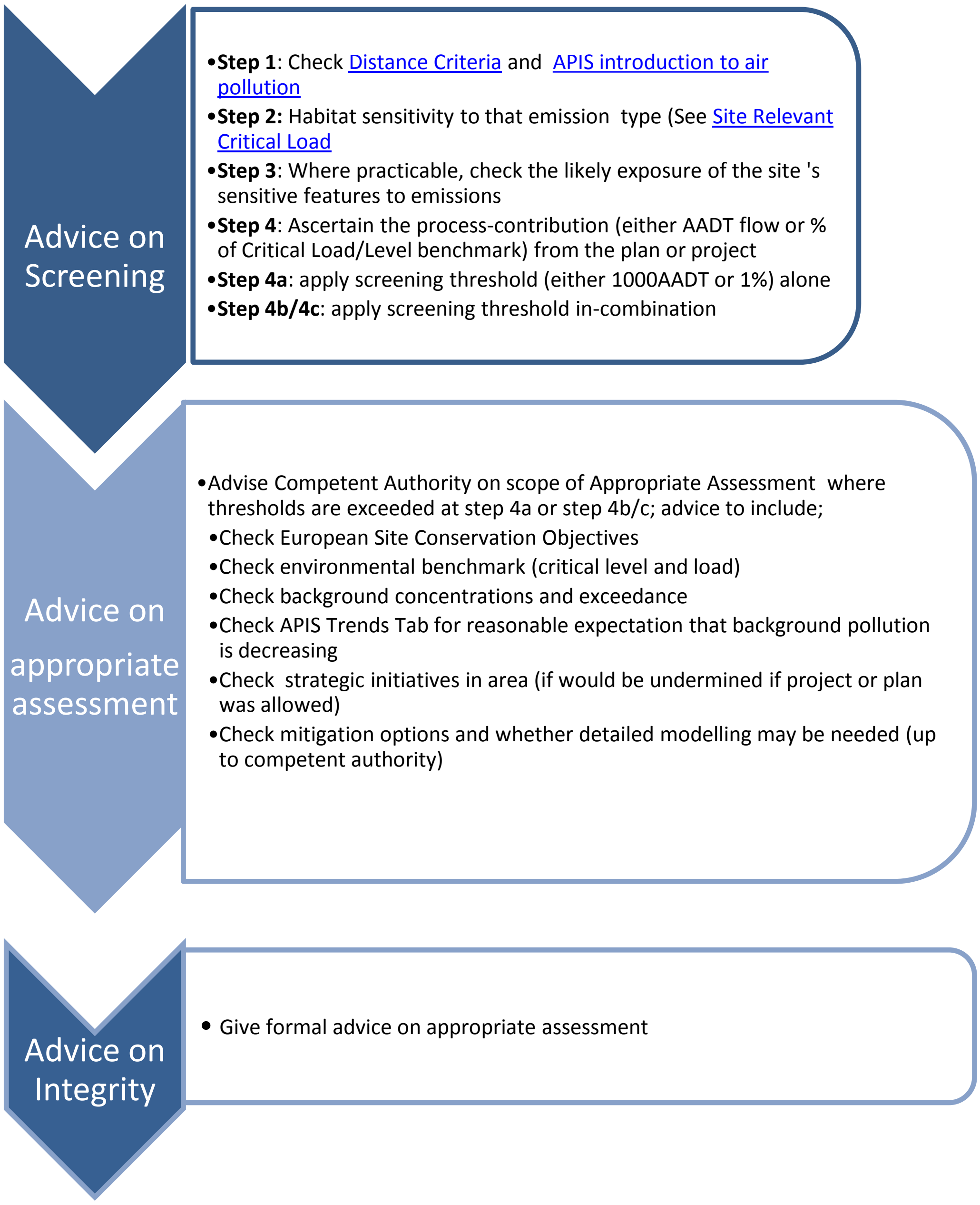
3.8 Natural England can direct competent authorities to further information they will find useful for undertaking an appropriate assessment and further guidance to inform the scope of an appropriate assessment is given in Section 5. It is at this stage that we would also detail why a likely significant effect could not be ruled out either because of the risk to a European site from the plan or project 'alone' or due to a risk of 'in-combination' effects.

3.9 A summary flowchart has been produced in *Appendix A* to this guidance, which is linked to the screening steps described in more detail below. It can help to guide staff in coming to a view as to the advice to be given on the assessment of plans or projects.

3.10 Staff should note that this document and the flowchart only provides general guidance on the factors to consider when advising a competent authority on the HRA of those plans and projects generating road traffic and air pollution effects. It cannot cater for all situations. Where there is information available that indicates it would be inappropriate to rely on this guidance (for example, there is uncertainty in the evidence base, there are development clusters that need to be accounted for or specific local evidence is available which undermine the application of this guidance), it will be necessary to consider whether further internal advice is needed and/or whether we should advise that the plan or project should progress to appropriate assessment. This adapted advice will need to be explained on a case by case basis.

Figure 2: Overview of stages and steps when advising a competent authority on the HRA of a road traffic project or plan

For road traffic emissions the distance criteria applied is 200m. Distance criteria applied to other emission sources is available on request and under review;



4. Advice on Screening for Likely Significant Effects

4.1 The purpose of the screening stage of the HRA process is to initially identify the risk or the possibility of significant adverse effects on a European site which could undermine the achievement of a site's conservation objectives and which therefore require further detailed examination through an appropriate assessment (see also paragraph 4.3 below). If risks which might undermine a site's conservation objectives can clearly be ruled out (based on the consideration of objective information), a proposal will have no likely significant effect and no appropriate assessment will be needed.

4.2 The Habitats Regulations place the responsibility for the screening decision as to whether appropriate assessment is required on the competent authority (see, for example, the text of regulations 63 and 105). There is no statutory requirement for a competent authority to seek or to rely on Natural England's screening opinion – it can come to its own view on likely significant effect. However, a competent authority, and/or the promoters or proposers of a plan/project, may request Natural England's advice on screening at formal consultation or at pre-application stages (under our Discretionary Advice Service). This section is intended to cover such circumstances.

4.3 In undertaking an assessment of 'likely significant effects' under the Habitats Regulations, authoritative case law has established that:

- An effect is likely if it '*cannot be excluded on the basis of objective information'*[5]
- An effect is significant if it '*is likely to undermine the conservation objectives'*[6]
- In undertaking a screening assessment for likely significant effects '*it is not that significant effects are probable, a risk is sufficient'….* but there must be credible evidence that there is '*a real, rather than a hypothetical, risk''*[7].

4.4 The Advocate General's opinion in *Sweetman* also offers some simple guidance that the screening step '*operates merely as a trigger'* which asks *'should we bother to check?''*[8].

4.5 As such, when determining whether air pollution from a plan or project has a 'likely significant effect' upon a given qualifying feature under the Habitats Regulations, the extent to which there are risks of air pollution that might undermine the conservation objectives for the site is central.

4.6 It is recommended that Natural England staff follow the sequential steps 1 – 5 outlined below to apply this screening procedure when Natural England is asked

[5] Case C127-02 *Waddenzee* (refer para 45)
[6] Case C127-02 *Waddenzee* (refer para 48)
[7] Boggis v Natural England and Waveney DC [2009] EWCA Civ 1061 (refer paras 36-37)
[8] Case C 258/11 *Sweetman* Advocate General Opinion (refer paras 49-50)

to advise competent authorities on the risks of air quality impacts within the framework of a HRA.

Step 1: Does the proposal give rise to emissions which are likely to reach a European site?

4.7 Any emissions from road traffic associated with a specific proposal and the proximity to European sites should be considered in the consultation documents. If they are not, further information should be requested from the competent authority consulting Natural England.

4.8 A key factor to consider at this initial screening step for air pollution assessment is the distance between an emission source and the receptor (in this case a European site). Emissions to air may have effects over both long and short ranges depending on the size, source and nature of the emission.

4.9 Distance-based criteria have been established for several sectors to identify consultations requiring consideration for potential effects from air pollution. These are listed on Natural England's Technical Information Exchange (TIE) air pollution pages (Distance Criteria) and currently under review[9].

4.10 With regard to potential risks from road traffic emissions, Natural England and Highways England are in agreement that protected sites falling within 200 metres of the edge of a road affected by a plan or project need to be considered further. This is based on evidence presented in ENRR580 (Bignal *et al.* 2004[10]) and is consistent with more current literature (Ricardo-AEA, 2016[11]). However, where (unusually) there is a credible risk that air quality impacts might extend beyond 200 metres from a road, Natural England may advise that additional sites should also be scoped into the HRA.

4.11 The distance between roads where increased traffic levels are predicted and specific designated sites can be checked using Magic.

4.12 If the consultation does not fall within the distance criterion for designated sites (i.e. 200m for road traffic proposals), no further steps of the assessment are necessary. Such proposals are likely to have no effect on sites at all and so do not need to be subject to assessment in-combination with other plans and projects. A screening conclusion of no likely significant effect on the site can be advised with regard to the risk of road traffic emissions affecting air quality.

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[9] Available upon request
[10] BIGNAL, K., ASHMORE, M. & POWER, S. 2004. *The ecological effects of diffuse air pollution from road transport*. English Nature Research Report No. 580, Peterborough.
[11] RICARDO-AEA, 2016. *The ecological effects of air pollution from road transport: an updated review*. Natural England Commissioned Report no.199.

Step 2: Are the qualifying features of sites within 200m of a road sensitive to air pollution?

4.13 The qualifying features of European Sites can be identified by reference to Natural England's formal advice on their Conservation Objectives, which include a definitive list of legally-qualifying features. These objectives are available here. Alternatively a list of qualifying features can also be found by searching for the European Site on Designated Sites View.

4.14 There are several ways to establish whether a qualifying feature is sensitive to the type of air emissions expected from a proposal. These range from broad, internationally agreed pollution benchmarks (critical loads and levels) to site specific information such as survey data.

4.15 APIS provides key information about feature sensitivity to specific pollutants:

- by broad category (habitat, ecosystem and species) and,
- by qualifying feature on each designated European site (Site Relevant Critical Loads Search Tool).

4.16 Where none of a site's qualifying features are considered to be sensitive to a pollutant, then no further assessment is required for that pollutant. For example a chalk river will not typically be sensitive to acid deposition because of its natural buffering capacity. In these circumstances a screening conclusion of no likely significant effect on the site can be reached with regard to air quality.

Where at least one of a site's features is known to be sensitive, further screening is advised at step 3 (where information is available) or at step 4. Where there is uncertainty over the sensitivity of the feature in close proximity to a road affected by the plan or project, then a precautionary approach should be taken with an assumption made that the feature may be sensitive.

Step 3: Could the sensitive qualifying features of the site be exposed to emissions?

4.17 Usually, only those European sites present within 200m of the edge of a road on which a plan or project will generate traffic will need to be considered when checking for the likelihood of significant effects from road traffic emissions (but see also paragraph 4.10).

4.18 Many sites are designated for several different qualifying features. Not all features are present within a given location within the site. In some cases, a road surface and its adjacent verges may be included within a designated site boundary. This does not necessarily mean that it, and its associated verges, will be of nature conservation interest and form part of a qualifying feature. The inclusion of the hard surface of a road and/or its adjacent verges might simply have been unavoidable when denoting a boundary and included simply for convenience. These areas will therefore constitute 'site-fabric'[12], being of no special nature conservation interest. Conversely, at some sites, roadside verges may have been deliberately included within a site boundary and be an integral part of a designated habitat. Therefore, a site's conservation objectives are unlikely to apply equally to all parts of a site and a competent authority may need to be made aware of this as necessary.

4.19 An early understanding of the spatial distribution of features within a site can help to decide whether or not appropriate assessment will be required. This is particularly relevant as contributions to air pollution from a road will typically decrease with distance away from that road (e.g. Ricardo-AEA, 2016[13]). Where the applicant has provided reliable and precise information that models the likely deposition of road-based pollutants in relation to the distribution of a site's features and any sensitive qualifying features are not present within the area to be affected by emissions (and Natural England's advice is that there is no conservation objective to restore the features to that area), it will be relatively straightforward to ascertain that the plan or project poses no credible air quality risk to it.

Where no information is provided that is able to sufficiently predict the deposition of pollutants in relation to the site's sensitive features, further screening is advised at step 4.

4.20 Information about the precise location of features within sites may be available from a variety of sources. Preferably, up to date ecological information will have been provided by the applicant to the competent authority as part of the submitted proposal being consulted upon. This may include further survey and spatial information about the location of Protected Sites, the distribution of sensitive features and their sensitivity to emissions from a road that, subject to our checks and validation, could be relied upon to inform this step.

Information is held in Natural England's Designated Sites System Viewer about the spatial location of individual features. Each feature is assigned to an underpinning monitoring 'unit' for condition reporting purposes. If a sensitive feature is not assigned to a unit (or intended to be restored to the unit) within the distance criterion then effects can be screened out. (Note that the current

[12] 'Site-fabric' is a general term used by Natural England to describe land and/or permanent structures present within a designated site boundary which are not, and never have been, part of the special interest of a site, nor do they contribute towards supporting a special interest feature of a site in any way, but which have been unavoidably included within a boundary for convenience or practical reasons. Areas of site-fabric will be deliberately excluded from condition assessment and will not be expected to make a contribution to the achievement of conservation objectives.
[13] Ricardo-AEA, 2016. The ecological effects of air pollution from road transport: an updated review' (NECR199).

reportable condition of a feature, based on latest condition assessment information, should not be used to justify screening out effects on a feature.)

4.21 If none of the site's sensitive qualifying features known to be present within 200m are considered to be at risk due to their distance from the road, there is no credible risk of a significant effect which might undermine a site's conservation objectives. The screening thresholds adopted in step 4 below need not be applied and no further assessment is required. In these circumstances, a screening conclusion of no likely significant effect on the site can be advised with regard to air quality.

4.22 If, at this stage, there is uncertainty over the presence or absence of the feature in close proximity to a road affected by the plan or project, then a precautionary approach should be taken with an assumption made that the feature may be present and step 4 undertaken.

Step 4: Application of screening thresholds

4.23 If a proposal has not been screened out by steps 1-3, the next step is to consider the risk from the road traffic emissions associated with the plan or project. Depending on the information available, this could be expressed in terms of either the predicted average annual daily traffic flow ('AADT' as proxy for emissions) or the predicted emissions themselves (the actual process-contribution). Each of these parameters have guideline thresholds to check whether the predicted change is likely to be significant (e.g.1000 AADT for traffic numbers or 1% of critical load or level for emissions). This information should have been provided to the competent authority by the applicant.

4.24 The use of the AADT screening threshold is advocated by Highways England in their Design Manual for Roads and Bridges[14] (DMRB) to check whether more detailed assessment of the impact of emissions from road traffic is required. This non-statutory or guideline threshold is based on a predicted change of daily traffic flows of 1,000 AADT or more (or heavy duty vehicle flows on motorways (HDV) change by 200 AADT or more).

4.25 The AADT thresholds do not themselves imply any intrinsic environmental effects and are used solely as a trigger for further investigation. Widely accepted Environmental Benchmarks for imperceptible impacts are set at 1% of the critical load or level, which is considered to be roughly equivalent to the DMRB thresholds for changes in traffic flow of 1000AADT and for HDV 200AADT. This has been confirmed by modelling using the DMRB Screening Tool that used average traffic flow and speed figures from Department of Transport data to calculate whether the NOx outputs could result in a change of > 1% of

[14] HIGHWAYS ENGLAND. Design Manual for Roads and Bridges Volume 11 Section 3, Part 1 - Air Quality

critical/load level on different road types. A change of >1000 AADT on a road was found to equate to a change in traffic flow which might increase emissions by 1% of the Critical Load or Level and might consequentially result in an environmental effect nearby (e.g. within 10 metres of roadside).

As a result, the AADT thresholds and 1% of critical load/level are considered by Natural England's air quality specialists (and by industry, regulators and other statutory nature conservation bodies) to be suitably precautionary, as any emissions below this level are widely considered to be imperceptible and, in the case of AADT, undetectable through the DMRB model. There can therefore be a high degree of confidence in its application to screen for risks of an effect.

If there is already detailed, locally-based modelling available about the plan or project that shows the 1% of the environmental benchmark is *not* exceeded, even if 1000 AADT is, then this level of precision is sufficient to override the use of the very generic 1000 AADT guideline threshold above.

Remember that 1000 AADT has been adopted here to simply help trigger when to look further where traffic projection data is the sole means of assessment - it does not immediately mean there *will* be an effect.

Considering the effect of avoidance and mitigation measures already incorporated into the plan/project

4.26 In a recent authoritative decision in C-323/17 *People Over Wind*, the CJEU concluded that it is not appropriate, at the screening stage of a HRA, to take account of measures intended to avoid or reduce the harmful effects of the plan or project on a European Site. This overrules previously established UK case law in *Hart*[15] which concluded that incorporated measures could be taken into account at this screening stage when judging the risk of a significant effect. These matters can now only be taken into account as part of the appropriate assessment stage of a HRA.

4.27 Where Natural England's advice is requested at the screening stage, it should ensure that the competent authority and/or the promoters or proposers of a plan or project have clearly identified the nature of the plan or project under review and whether there are avoidance and/or mitigation measures that are to be excluded from the screening assessment. Where Natural England considers there is doubt in these matters, the precautionary principle should be applied and these matters should *not* be taken into account when Natural England is advising

[15] Hart District Council v Secretary of State for Communities and Local Government, Luckmore Ltd and Barratt Homes Limited and Taylor Wimpey Developments Limited and Natural England [2008] EWHC 1204(Admin))

on applying the thresholds below to judge likely significant effect. Natural England should explain the reasoning for its advice, however the competent authority, as the decision maker, is entitled to disagree with this advice and reach its own reasoned and cogent decision.

Step 4a: apply the threshold alone

4.28 First consider the effects of the plan or project ‘alone’ against the screening threshold. Where a proposal is considered to have a likely significant effect because it breaches the screening threshold alone it should go through to an appropriate assessment ‘alone’ (at least initially). There is no need to consider the potential for in-combination effects (at steps 4b/c below) at this screening step as an appropriate assessment is needed in any event.

4.29 If the predicted change in traffic flow is less than 1000AADT (or the level of emissions is <1% of the critical load/level), the associated emissions are not likely to have a significant effect alone but the risk of in-combination effects should be considered further (go to step 4b/c).

4.30 At this stage, this is irrespective of the current background levels and whether critical load or level values are currently being exceeded or not. This is because 1% of the environmental benchmark or 1000AADT is considered to be so small that anything less than this will be, in any event, not likely to be perceptible and significant. We would advise that current background levels are considered later should appropriate assessment be needed.

Step 4b: apply the threshold in-combination with emissions from other road traffic plans and projects

4.31 Where a proposal is *below* the screening threshold *alone* at step 4a above (i.e. <1000 AADT or <1% depending on information available), step 4b must be considered to apply the same screening threshold ‘in-combination’. This step is explicitly included here to reflect the requirements of the Habitats Regulations and in response to the recent clarification provided in the Wealden Judgment 2017.

4.32 This is also because projects and plans that increase road traffic flow have a high likelihood of acting together, or in-combination, with other plans or projects that would also increase traffic on the same roads. Vehicles generated by different plans or projects can end up on the exact same road(s) (forming a line source of emissions) within or close to the same site. In these cases, it is difficult to justify use of a threshold alone for determining likelihood for significant effect by applying it solely to the project being assessed. The threshold should be applied in-combination.

4.33 An in-combination effect is one which does not represent a likely significant effect ‘alone’ but, when added to similar effects from other live plans and projects, becomes significant.

4.34 The Wealden Judgment 2017 found that the use of the 1000 AADT guidelines (the proxy for 1% (on road) of the critical level/load (for the receiving habitat)) as the sole means of catering for in-combination effects lacked coherence, particularly where other figures are known which, when added together, would cause that threshold to be exceeded. From that, the Court concluded that where the likely effect of an individual plan or project does not itself exceed the threshold of 1000 AADT (or 1%), its effect must still be considered alongside the similar effects of other ‘live’ plans and projects (see paragraph 4.44 below) to check whether their added or combined effect on a site could be significant. The threshold itself was not questioned.

4.35 Natural England recognises that at both the screening and appropriate assessment stages of a HRA, the likely effects of a plan or project need to be thought about individually and in combination with other relevant plans or projects. This is a legal requirement of the Habitats Regulations and it helps to ensure that European sites are not inadvertently damaged by the additive effects of multiple plans or projects.

4.36 It may be very obvious that there are no other plans or projects which are ‘live’ at the time of the assessment (see 4.44 below) whose effects could act together with the subject proposal. A competent authority should clearly record this in their assessment in such cases. Natural England’s advice is that where evidence concerning other live plans and projects is available, such as increases in road traffic from other plans or projects that will affect the same roads being assessed, the 1000 AADT threshold should also be applied to their combined value to screen for in-combination effects.

4.37 Natural England staff may be asked by a competent authority to advise on the scope of an in-combination screening step and how far they should look for other road traffic plans and projects which may be relevant to their risk assessment. In Natural England’s view, staff in a competent authority can apply their professional judgment when considering this. An exhaustive search for relevant plans and projects by a competent authority is normally required to comply with the Habitats Regulations. However, a pragmatic approach to identifying the most pertinent ones may need to be taken where there is a large number of proposals. It might be reasonable to *initially* limit a search to those plans and projects which are of most direct relevance to the subject plan or project under HRA. This may be those which are simply the closest to the site or within a certain distance from it, or the most influential in nature).

4.38 Once screening thresholds have been exceeded to indicate that there is a risk of a significant combined effect from the subject proposal and other plans or projects and an appropriate assessment is warranted, the search for other live plans/projects may stop. This may mean that more minor plans or projects can be excluded from the in-combination assessment being undertaken.

4.39 This search should not be limited to other plans or projects being proposed within the jurisdiction of that competent authority; other relevant proposals affecting the same European Site(s) may occur within adjoining local planning authority areas for example.

4.40 Where the in-combination effect of the subject plan or project with more than one plan or project is greater than the 1000 AADT (when using traffic flow data) or 1% (when using emissions data) threshold, appropriate assessment is advised.

Step 4c: apply the threshold in-combination with emissions from other non-road plans and projects

4.41 When considering the potential for in-combination effects, a competent authority should also recognise that different proposal types ('sectors') and different pollutants (e.g. ammonia (NH_3), nitrogen oxides (NO_x and NO_2)) can combine together to have the same or similar effect on a given area of habitat. By way of example, nitrogen deposition on a site can result from both the emissions of ammonia from a farm source and also from emissions of nitrogen oxides from a traffic source, with both having an eutrophication effect.

4.42 Where the in-combination effect of the subject plan or project with other road traffic plans or projects has not exceeded the relevant 1000 AADT (or 1%) threshold, we should advise the competent authority to look further for any other insignificant effects of live 'non-road' plans/projects to check that the 1% threshold is not exceeded in this way.

4.43 Where the in-combination effect of the subject plan or project with one or more plan or project is greater than the 1% threshold, appropriate assessment is advised.

4.44 It is generally well-established that the scope of an in-combination assessment is restricted to plans and projects which are 'live' at the same time as the assessment being undertaken. These can potentially include:

- The incomplete or non-implemented parts of plans or projects that have already commenced;
- Plans or projects given consent or given effect but not yet started.
- Plans or projects currently subject to an application for consent or proposed to be given effect;

- Projects that are the subject of an outstanding appeal;
- Ongoing plans or projects that are the subject of regular review and renewal
- Any draft plans being prepared by any public body
- Any proposed plans or projects that are reasonably foreseeable and/or published for consultation prior to application

As stated above, when considering this scope, competent authorities can be mindful of the assessment, reasoning and conclusions included in any previous HRAs for these plans or projects.

What 'plans and projects' are already included in the nationally modelled background?

APIS provides information about background pollution concentrations for each European site through the Site Relevant Critical Load Tool (on the Concentrations/Deposition tab). Projects and plans operational **on or before** dates included in background pollution data on APIS are typically considered as an integral part of the background. These should **not** be included as projects or plans for in-combination assessment as this would effectively be double-counting the emission sources.

4.45 It is the role of the competent authority, not Natural England, to acquire sufficient knowledge and information on other plans and projects that are included within an in-combination assessment to enable it to make a fair and reasonable assessment of the likelihood of a significant combined effect. This may mean the plan or project proposer may be asked by the competent authority to provide or compile this.

4.46 Sources of information that project proposers or competent authorities can use to identify plans or projects that might act in-combination include:

- Planning Portals to locate applications awaiting permissions
- Environmental Permits Register of Applications and Register of Issued Permits
- Local plans (including brownfield registers with permission in principle) and any allocations not yet permitted.

4.47 In general terms, it is important for a competent authority to remember that the subject plan or project remains the focus of any in-combination assessment. Therefore, it is Natural England's view that care should be taken to avoid unnecessarily combining the *insignificant* effects of the subject plan or project with the effects of other plans or projects which can be considered *significant* in their own right. The latter should always be dealt with by its own individual HRA alone. In other words, it is only the appreciable effects of those other plans and projects that are not themselves significant alone which are added into an in-combination assessment with the subject proposal (i.e. 'don't combine individual biscuits (=insignificant) with full packs (=significant)').

4.48 As stated above, an exhaustive search for relevant non-road plans and projects is normally required to comply with the Habitats Regulations. Where there is likely to be a large number of other live plans or projects which could all potentially fall within the scope of an in-combination assessment, it is Natural England's view that staff in a competent authority can apply their professional judgment when considering this. It might be that a pragmatic approach to identifying the most pertinent ones may be required from the competent authority. It might be reasonable to initially limit a search to those plans and projects which are of most direct relevance to the subject plan or project under HRA (i.e. the likelihood of that plan or project's effects impacting upon the same site in-combination with the proposed plan or project). This may be those which are simply the closest to the site or within a certain distance from it, or the most influential in nature.

4.49 As above, should screening thresholds be exceeded to indicate that there is a risk of a significant effect, this may mean that more minor plans or projects become immaterial to the in-combination assessment and can be discounted.

Similarly, this search should not be limited to other plans or projects being proposed within the jurisdiction or administrative boundaries of that competent authority; other relevant proposals affecting the same European Site(s) may occur within adjoining local authority areas for example.

Step 5: Advise on the need for Appropriate Assessment where thresholds are exceeded, either alone or in-combination

4.50 This can be summarised below:

Traffic Proxy or Process Contribution from a plan or project alone	Advice on screening for likely significant effect	Is Appropriate Assessment required by the competent authority?
More than 1000 AADT (or >1% of critical level/load)	There is a risk of a significant effect on air quality alone	Yes
Less than 1000 AADT (or <1% of critical level or load)	There is a risk of an appreciable effect on air quality but is unlikely to be significant alone and screen for in-combination effect	<u>Either</u> No – advise that appropriate assessment is not required if: • no other plans/projects can be identified that would act in-combination, or • **together** they add up to **less than** 1000 AADT (or 1% of critical level/load) <u>Or</u> Yes – advise that appropriate assessment is required if: • other plans/projects can be identified that would act in-combination, and • **together** they add up to **more than** 1000 AADT (or 1% of critical level/load)

5. Advising competent authorities on the scope and content of an Appropriate Assessment

About this section

5.1 This section aims to provide Area Team staff with further assistance when giving their advice to a competent authority on the scope and content of an appropriate assessment examining the likely effects of road traffic emissions.

5.2 This is not intended to provide a definitive or exhaustive checklist of factors to consider. A competent authority is entitled to make use of additional information and to seek the additional advice of others.

5.3 At this stage of HRA, it is a statutory requirement for competent authorities to formally consult Natural England '*for the purposes of'* an Appropriate Assessment (AA) and to '*have regard*' to any representations that Natural England may make. This consultation may include advice about further information that may be required from the applicant and advice as to whether the scope of the appropriate assessment fully addresses the likely risks to the site(s).

5.4 Typically, Natural England's expert advice is given significant weight; however a competent authority, as the decision maker, is also entitled to disagree with Natural England's advice and reach its own reasoned and cogent conclusion at Appropriate Assessment.

5.5 This section highlights a number of factors, in no particular order, that we could usefully advise a competent authority as being relevant for consideration in an assessment. It does **not** recommend sequential steps or provide definitive guidance about how or to what degree these factors should inform an assessment, which will depend on the facts and circumstances of each case.

Introduction

5.6 Having previously identified a risk or a possibility of a significant effect from a plan or project (either alone or in-combination), the purpose of the appropriate assessment stage is to more precisely assess the likely effects identified and to inform a conclusion as to whether an adverse effect on site integrity can be ruled out.

5.7 The 'integrity' of a site should be taken to mean the coherence of its ecological structure and function, across its whole area that enables it to sustain the habitat, complex of habitats and/or the levels of populations of the species for which it was, or will be, designated or classified. A site can also be described as having a high degree of integrity where '*the inherent potential for meeting site conservation objectives is realised, the capacity for self-repair and self-renewal* *under dynamic conditions is maintained and a minimum of external management is required*' (European Commission, 2000[16]).

5.8 Whilst the assessment should be an objective one which is contiguous with but more detailed than the previous screening stage, it should always be 'appropriate' in terms of its scope, content, length and complexity to the plan or project under assessment. This was recently reiterated by the Supreme Court decision in the case of *Champion*[17] which clarified:

'Appropriate' is not a technical term. It indicates no more than that the assessment should be appropriate to the task in hand: that task being to satisfy the authority that the project will not adversely affect the integrity of the site concerned'.

5.9 It should not be assumed that appropriate assessment will necessarily involve detailed and complex monitoring or modelling work. Whilst complex work *might* be necessary in fully understanding what will happen to a site if the plan or project goes ahead, and asking whether that would be consistent with maintaining or restoring a site's integrity, it is equally possible that a fairly concise and straightforward assessment might be entirely 'appropriate'.

5.10 This section provides some information on additional factors which may be relevant to the scope of an appropriate assessment that seeks to assess the impacts from air pollution in a more detailed manner to ascertain whether there will be an adverse effect on site integrity. The impacts resulting from a change in the atmospheric concentration or deposition of pollutants as a result of the plan or project might include:

- Changes in the species composition of a designated or supporting habitat, especially in nutrient poor ecosystems, with an (unnatural) shift towards species associated with higher nitrogen availability (e.g. leading to the dominance of tall grasses);
- Reduction in the species richness of designated habitat
- Damage or loss of sensitive lichens and bryophytes (which may be strongly typical of a designated habitat) which receive their nutrients largely from the atmosphere
- Increases in nitrate leaching and changes in soil nutrient status which may affect the structure and function of a designated or supporting habitat

[16] EUROPEAN COMMISSION, 2000. Managing Natura 2000 Sites (section 4.6.3).
[17] Champion v North Norfolk DC [2015] UKSC 52 (refer para 41)

5.11 Further technical guidance about the ecological impacts from road transport can also be found in the Natural England research report 'The ecological effects of air pollution from road transport: an updated review' (NECR199[18]).

5.12 The competent authority is therefore likely to require both ecological and air quality advice in order to undertake their appropriate assessment.

The use of thresholds at the appropriate assessment stage

5.13 At the previous screening stage, Natural England has advised that a threshold equivalent to 1% of the critical load/level can be applied as a guideline to initially check which road traffic plans and projects might require appropriate assessment. At appropriate assessment stage, Natural England recommends that this same 1% threshold is *not* used as a means of determining whether there is an adverse effect on site integrity from a road traffic project. Other factors are relevant which may mean that a plan or project that exceeds the 1% screening threshold can still demonstrate no adverse effect on site integrity through an appropriate assessment.

Issues recommended for further consideration by an appropriate assessment:

Consider whether the sensitive qualifying features of the site would be exposed to emissions

5.14 Where no information was available at the screening stage to consider the emissions from road traffic and the distance to sensitive qualifying features of sites within 200m of the road, this should be investigated further as part of the appropriate assessment.

5.15 This may require the applicant to provide further information about the actual predicted emissions at the behest of the competent authority to inform this assessment.

5.16 This is particularly relevant to this stage as contributions to air pollution from a road will typically decrease with distance away from that road (e.g. Ricardo-AEA, 2016[19]). Therefore, if, upon closer examination, the qualifying feature which is considered to be sensitive is shown not to be present within the area predicted to be affected by emissions (and Natural England's advice is that there is no

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[18] RICARDO-AEA, 2016. *The ecological effects of air pollution from road transport: an updated review*. Natural England Commissioned Report no.199.
[19] Ricardo-AEA, 2016. The ecological effects of air pollution from road transport: an updated review' (NECR199).

conservation objective to restore the feature to that area), it will be relatively straightforward to ascertain that the plan or project poses no credible risk to it and there is unlikely to be an adverse effect on the site's integrity.

5.17 Similarly, it may be possible at this stage to demonstrate that, despite their proximity, the sensitive features will actually only be exposed to emissions that are <1% of the Critical Load/Levels (both alone and in-combination) due to their distance from the affected road(s).

Consider the European Site's Conservation Objectives

5.18 The Habitats Regulations state that appropriate assessments of plans and projects must be undertaken '*in view of that site's conservation objectives*'. **The 'key question' for the appropriate assessment is, in view of these objectives, can it be ascertained that, should the plan or project go ahead, there will be no adverse effect from it on the site's integrity so that the site's conservation objectives will not be undermined.**

5.19 In England, Natural England provides formal advice on European Site Conservation Objectives, their purpose being in part to enable their effective use in HRAs and to expedite decision-making by competent authorities[20]. This advice is made publically available for all European terrestrial sites and European marine sites. This advice complements, but is broader than and different to, the narrower range of attributes and targets as set out in our SSSI 'Favourable Condition Tables' which are used for our own monitoring purposes to report on 'condition' status.

5.20 For Special Areas of Conservation, with reference to '*the key question'* above, the conservation objectives are to '*ensure that the integrity of the site is maintained or restored as appropriate, and ensure that the site contributes to achieving the Favourable Conservation Status of its Qualifying Features, by maintaining or restoring…'.*

The conservation objectives for any given site then go on to list a series of core attributes which form part of a site's integrity to be 'maintained' *or* ' restored'. When considering the risks associated with air pollution to a SAC, the attribute most likely to be undermined is '*the structure and function (including typical species) of qualifying natural habitats'*. These structural and functional changes might *in turn*, lead to changes to other attributes but most impacts from air pollution follow as a consequence of the structural and functional changes which are therefore of primary importance.

[20] Defra, 2012. Report of the Habitats and Wild Birds Directives Implementation Review. Pages 26-27.

5.21 Special Protection Areas (SPA) are different; the qualifying features are the bird populations for which the site has been classified. The conservation objectives are to 'e*nsure that the integrity of the site is maintained or restored as appropriate, and ensure that the site contributes to achieving the aims of the Wild Birds Directive, by maintaining or restoring…'.*

As with SACs, the conservation objectives then go on to list a series of core attributes which form part of that site's integrity to be 'maintained' or 'restored'. When considering the risks associated with air pollution to a SPA, the attribute most likely to be 'undermined' is '*the structure and function of the habitats of the qualifying species'* (N.B. there is not reference to typical species in the case of SPA supporting habitat).

Where a Natural England Area Team has provided further **Supplementary Advice about a European Site's Conservation Objectives**, air quality will, where appropriate, be highlighted as a specific attribute of a site's structure and function with regard to any air quality sensitive features.

The conservation objective for the air quality attribute will typically be to ensure that, over the long-term, air pollutants are either maintained below or restored to below the site-relevant Critical Loads and Levels given on APIS. The inclusion of this objective in this advice on conservation objectives reflects the condition threat that exceedance poses. The objective will be tailored to distinguish where air quality should be maintained or restored dependent on whether these air quality benchmarks are currently being exceeded or not. Over time, this advice should be updated accordingly by Area Teams in light of best available information.

These objectives do not affect our existing condition assessments of these sites as air quality benchmarks do not currently inform condition reporting directly; the effects of exceedance might, over time, show up when measuring specific attributes of a habitat's structure e.g. the dominance of nitrogen-tolerant species or a decline in the extent of bare ground.

The trajectory of deposition and concentration trends illustrated on APIS is perhaps a better measure of whether the air quality objectives for a site are likely to be met or not.

NOTE OF CAUTION

When considering the sensitivity of SPA qualifying features, the extent to which changes to the structure and function of the *supporting* habitats might represent a risk to the integrity of an SPA will vary significantly, depending on the ecological role that the structure and function of a supporting habitat plays in maintaining the population for which the site has been classified. The site relevant critical load pages on APIS provide information on the sensitivity of each SPA feature.

5.22 When considering the ‘key question’ above in view of the conservation objectives, it follows that a decision as to whether a proposal ‘undermines’ the conservation objectives (or not) should also be informed by whether the conservation objectives are to ‘maintain’ or to ‘restore’.

Where background levels show the site is not currently exceeding relevant air quality benchmarks and the conservation objectives are to maintain the concentrations and deposition of air pollutants either at current levels or below the relevant benchmarks

5.23 Where there is currently no exceedance of relevant benchmarks (such as Critical Loads and Levels – see also para 5.31) the site’s conservation objectives are to ‘maintain the concentrations and deposition of air pollutants at current levels or below the relevant benchmarks’ to protect the site’s integrity in respect of air pollution. As such, a new plan or project could undermine the conservation objectives of such a site where it leads to a deterioration in air quality that is significant in the context of the site, even where that site is below a critical load or level. The evidence presented by Caporn *et al.* (2016)[21] in NECR 210 shows that small contributions of nitrogen deposition from the air have the potential to lead to *more* significant changes in vegetation composition where a site is below but near to the Critical Load, compared to a site which significantly exceeds a critical load. The appropriate assessment will need to examine such risks, and likely effects, in more detail.

5.24 Even where an additional contribution is small (e.g. <1% of critical load/level but >1% of the critical load/level in-combination), a competent authority should undertake a more considered assessment with regard to sites that are currently meeting their conservation objectives (which is considered to be appropriate to the specific circumstances).

Where the background levels show the site is already exceeding relevant air quality benchmarks and the conservation objectives are to ‘restore the concentrations and deposition of air pollutants to within benchmarks’.

5.25 Where the conservation objectives are to ‘restore the concentrations and deposition of air pollutants to within benchmarks’ (i.e. where the relevant benchmarks such as Critical Loads/Levels are *already* exceeded) they will be *undermined* by any proposals for which there is credible evidence that further emissions will compromise the ability of other national or local measures and initiatives to reduce background levels.

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[21] CAPORN, S., FIELD, C., PAYNE, R., DISE, N., BRITTON, A., EMMETT, B., JONES, L., PHOENIX, G., S POWER, S., SHEPPARD, L. & STEVENS, C. 2016. *Assessing the effects of small increments of atmospheric nitrogen deposition (above the critical load) on semi-natural habitats of conservation importance*. Natural England Commissioned Reports, Number 210.

5.26 An exceedance alone is insufficient to determine the acceptability (or otherwise) of a project**.** Exceedance will represent a threat to the condition and integrity of the site. Hypothetically, it could be argued that any increase above a currently exceeded state compromises the extent to which improvements from other initiatives will deliver the restoration aims of the conservation objectives as any additional pollution could slow the rate at which progress is made towards meeting the relevant air quality benchmarks.

5.27 In terms of whether an 'adverse' effect can be ruled out, the Advocate General's Opinion in *Sweetman*[22] indicated that, in her view, a plan or project involving *'some strictly temporary loss of amenity which is capable of being fully undone'* would not be an adverse effect on integrity. By comparison, the *'lasting and irreparable loss'* of part of the SAC feature in *Sweetman*[23] was ruled to be an adverse effect on integrity.

5.28 In practice, where a site is already exceeding a relevant benchmark, the extent to which additional increments from plans and projects would undermine a conservation objective to 'restore' will involve further consideration of whether there is credible evidence that the emissions represent a real risk that the ability of other national or local measures and initiatives to otherwise reduce background levels will be compromised in a meaningful manner. This is a judgement to be taken by the competent authority which should be informed by, amongst others, the extent to which any declining national trends in air pollution or strategic work to tackle emissions affecting the site more locally might otherwise lead to improvements, the rate at which such improvement are anticipated to be delivered, any credible evidence on the extent of the impacts of a plan or project and whether those impacts can properly be considered temporary and reversible.

Consider background pollution

5.29 European sites are unlikely to be pristine in terms of air quality effects, and our advice will therefore be mindful of the current condition of the site's features and the site's long-term conservation objectives. Factors already affecting the site which are not related to the plan/project being assessed count as the current prevailing or background conditions. These factors may be having an adverse effect independent of the proposal being assessed (and should be addressed separately) but nevertheless may be currently undermining the site's resilience to new and additional pressures.

5.30 The background condition of the site will provide some further context to judging the risk of an adverse effect on integrity. This section explores where to obtain

[22]Advocate General Opinion in Case C-258/11 *Sweetman* (refer paras 58-61)
[23] Case C258-11 *Sweetman* (refer para 56)

background concentrations of air pollution to take into account as prevailing conditions.

(a) Review the Environmental Benchmarks ('critical loads and levels') and feature sensitivity to nitrogen

5.31 Habitats have varying sensitivity to air pollution effects. APIS provides environmental benchmarks for habitat either through the Site Relevant Critical Load Tool or the Habitat/pollutant impacts Tab on the home screen. These benchmarks are called critical loads or levels.

5.32 Critical levels and loads are set to take account of very long term contributions of pollution (20 – 30 year timeframe). Critical loads in particular are expressed as a range because they cover the situation across Europe for each nitrogen sensitive habitat. This range has to account for the variation in topography and precipitation/climate across Europe. In the UK, APIS outlines the part of the critical load range that is most appropriate based on available evidence (UK Indicative Critical Load Values).

5.33 Check whether the habitat being assessed has an environmental benchmark to assist with the assessment. If there is no benchmark on APIS that could mean there is lack of data. Absence of a benchmark is not assurance that a specific feature is insensitive to air pollution.

5.34 In addition, check and consider a feature's sensitivity to nitrogen more precisely. Some features and sites are much more sensitive to nitrogen than others; NECR 200 identifies three categories of sensitivity for traffic emissions; high (5-10 CL range), medium (10-20 CL range) and low (20-30 CL range).

5.35 Whilst the main impact mechanism of concern is through acid and nutrient nitrogen deposition (covered below), many assessments consider direct toxicity to vegetation from NOx. In this case the first relevant question to ask is the extent to which the relevant critical level might be exceeded as a result of the plan/project (either alone or in-combination with other plans and projects).

5.36 Ricardo-AEA (2016) in NECR200[24] found that background concentrations of NO_x in rural areas away from roads are typically in the range 15 – 20µg/m^3 i.e. some way off exceeding the critical level of 30µg/m^3.

Note that APIS provides background NOx values which are averaged over a 5km grid square. This means that higher levels along the roadside (but within a European site boundary) can be missed.

[24] RICARDO-AEA, 2016. *Potential risk of impacts of nitrogen oxides from road traffic on designated nature conservation sites*. Natural England Commissioned Report no. 200.

5.37 NECR200 measured designated site exposure to NOx from road traffic taking account of other background sources of NOx for 2011 and predicted 2020 data. High (>30µg/m^3), medium (> 25µg/m^3) or low (<25µg/m^3) categories of exposure to NOx from road traffic are identified based on a combination of road traffic NOx and background levels. Whilst this is a national snapshot in time (based on modelled data available at the time of the study in 2014), it could provide useful contextual data to supplement site specific data from APIS. Further information is provided here at NECR200.

5.38 When considering the impacts of a plan or project in relation to critical levels, it is important to understand the distance from the road that the critical level is exceeded and whether this represents a credible risk to qualifying features. We may wish to advise for example on how site boundaries have been defined and how the conservation objectives should be interpreted and applied to roads and road verges within a site boundary (see also step 3 in the screening stage above).

(b) Check for exceedance of Environmental Benchmarks

5.39 Exceedance of the benchmarks is determined by comparing the CBED (the 'Concentration Based Estimated Deposition' model) results (at 5km or 1km grid resolution) with critical levels or loads. Through this very direct approach for determining exceedance, more than 80% of the area of sensitive European Sites is currently in this exceedance state. This approach does not account for variability within the 5km grid square.

5.40 National maps to demonstrate where habitat sensitive to air pollution is predicted to be above its environmental benchmarks are available on Defra's UK AIR website[25].

5.41 Whilst most sensitive European Sites will be in this exceedance state, it does *not* automatically mean that further plans or projects affecting them would have an adverse effect on site integrity. Rather, it provides another piece of information to consider when determining whether a proposal might have a benign impact on site integrity and be acceptable or whether a conclusion of no adverse effect on site integrity cannot be reached by the assessment.

(c) Consider trends and whether there is evidence to indicate that background levels are decreasing

5.42 Acquiring information on whether local background pollution levels are declining or not can provide useful context to an appropriate assessment.

[25] 2013-2016 exceedances are in Defra AQ0826

5.43 This is available on the APIS Site Relevant Critical Load Tool and background concentrations are displayed under the "Trends" tab. This trend data currently covers the deposition and concentration trends over at least the last 8 years of national modelling. It is updated annually, though background trends are a 3-year average to account for weather variation (e.g. year 2005 is the average of years 2004, 2005 & 2006). The trend data is provided for maximum and minimum air concentrations (NOx, SOx, ammonia) as well as deposition (nutrient nitrogen and acid). A precautionary approach is to use the maximum value.

5.44 For deposition there are 3 sets of maximum and minimum values related to 3 rates of deposition:

- Moorland (or knee-high vegetation)
- Forest (or anything taller than knee high)
- Grid Average (average deposition for 5km grid square across habitat types)

5.45 Which value you use will depend on what type of habitat you are looking at. Figure 3 shows an example of nitrogen deposition trends at Breckland SAC. Nationally predicted declines in nitrogen deposition on heathland at Breckland SAC from 27 kg N/ha/year in 2005 to 24 kg N/ha/year in 2014 could mean that some increases in nitrogen from a plan or project (alone and in combination) may not impede this downward trend. Taking into account all relevant factors and information, it may be possible to consider some increases as temporary and reversible, which would be unlikely to undermine site objectives. In other words, we can still expect - even with the plan/project – the overall environmental loading will return to below critical level and loads within an appropriate timeframe.

5.46 While this may be a useful factor to consider in some cases, it should not be applied blindly. A range of matters will remain relevant, including whether any local survey evidence indicates that it is unsafe to rely on national modelling or where there are development clusters which would mean that any headroom that may be available should be more closely monitored or cannot be confidently relied on.

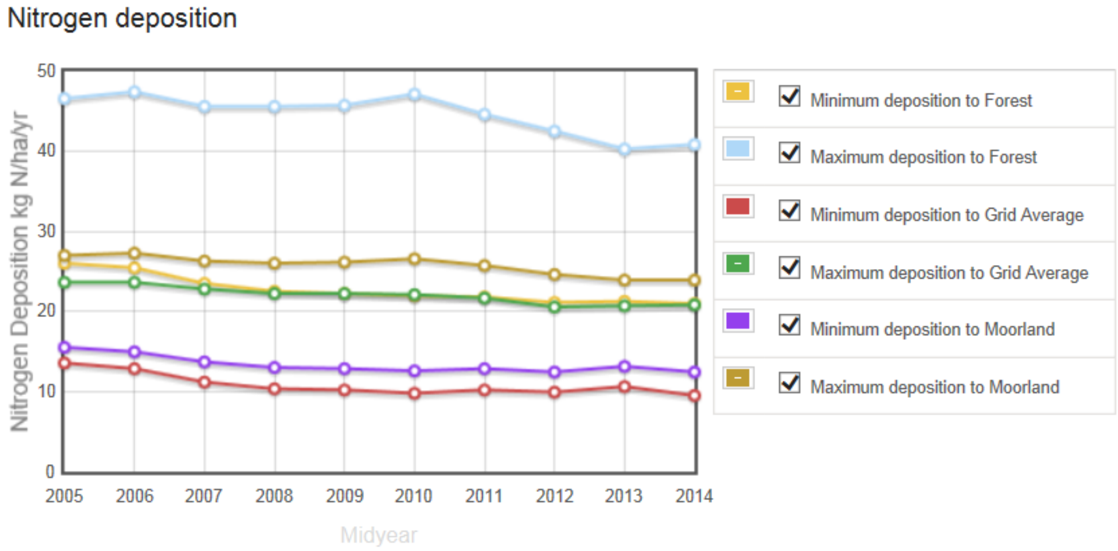


Figure 3: APIS Trends Tab for Breckland SAC Nitrogen Deposition

Consider the designated site in its national context

5.47 NECR200 provides contextual information to help inform relative risk within a wider national context. It provides an analysis of SAC and SSSI exposure to NOx from road traffic (taking into account other background sources of NOx), for 2011 and 2020 (based on 2014 modelling data).

5.48 It provides a relative categorisation of SSSI and SAC site exposure to road traffic NO_x in a national context and a relative risk categorisation of SACs based on exposure and site sensitivity. Whilst the data is a snapshot in time based on 2011 data and modelled 2020 data, it does provide a national context for local decision makers when assessing local plans and local development in relation to road traffic impacts on designated sites.

Consider the best available evidence on small incremental impacts from nitrogen deposition

5.49 When assessing likely adverse effects on site integrity, the Natural England Commissioned Report 210: *Assessing the effects of small increments of atmospheric nitrogen deposition (above the critical load) on semi-natural habitats of conservation importance* (referred to above) may be of relevance.

5.50 This research shows that habitats that have already been subject to high background nitrogen deposition can develop an effective tolerance to the effects of further deposition. However, this evidence is not appropriate for use to justify further exceedance on designated sites alone, without also considering all available factors and information and where this would undermine the conservation objectives to reverse this and restore pollutant levels to within an acceptable level.

5.51 The objective of this report was to examine recent vegetation survey data to understand the relationships that exist between species (composition and richness) and nitrogen deposition, and to determine the effect of incremental increases in nitrogen. Vegetation data were analysed from 226 sites, collected over 8 surveys of 5 UK priority habitats for conservation (sand dune, bog, lowland heath, upland heath, acid grassland). Further evidence was gained from published survey data and the network of UK nitrogen addition experiments.

5.52 This report provides detail about how much additional nitrogen might lead to a loss of one species on the following habitats (although in the case of bogs and sand dunes there was either insufficient information to develop a dose-response curve or the measure of effect (loss of one species) was too coarse to make a determination):

- Upland heath
- Lowland heath
- Bog (non-curvilinear response)
- Acid grassland
- Sand dunes

For certain habitats this information can inform a more precise assessment of the likely effect. The implications of any such predicted effects on overall species-richness should then be further evaluated in light of the site's Conservation Objectives to inform the conclusion of the appropriate assessment.

Consider the spatial scale and duration of the predicted impact and the ecological functionality of the affected area

5.53 The likely duration of any emission-impact(s) and the potential for recovery/reversibility of that impact are important factors to consider further when determining whether it is possible to demonstrate no adverse effect on integrity. For example, a conclusion of no adverse effect on integrity may be able to be reached in the case of a short-lived effect from which the site/feature can quickly recover (e.g. a peak caused by construction traffic).

5.54 The anticipated duration of any potential air quality impact, the ability for the affected feature to absorb or recover from that impact and the likely timescale of any anticipated recovery may be an important consideration in the assessment. The longer or more uncertain the feature's likely recovery time from an impact, the more difficult it may be to demonstrate no adverse effect on integrity.

5.55 A Natural England research report (NECR205) on how small scale effects[26] on European Sites have been considered in decision-making is of relevance here. Where the spatial extent of the affected area is small then the risk to the integrity of the site needs to be approached in a reasonable and proportionate manner. The Research Report concluded that:

'In the case of small scale effects on a qualifying Annex 1 habitat type for which a SAC had been designated, the decisions reviewed suggest that it is the relative importance of the area affected in terms of the rarity, location, distribution, vulnerability to change and ecological structure which is most influential. The contribution the affected area made to the overall integrity of the site (and hence that site's contribution to the conservation status of that habitat type at a member state level) exerted a stronger influence over decision makers than the spatial extent of the effect.

In the case of small scale effects on a supporting habitat for a species (whether a designated SAC species or a classified SPA species), the decisions reviewed suggest it is the ecological functioning of that supporting habitat which is most influential: that is, what ecological function the affected area was performing, or could perform, and it's importance to the population of the species for which the site had been designated / classified.'

Consider site survey information

5.56 Information available from site surveys will be relevant to an appropriate assessment. In particular any information which might indicate evidence of existing impacts from air pollution from similar sources which might introduce reasonable scientific doubt as to the absence of such adverse effects should the plan or project in question be permitted.

5.57 Such information which is available at the stage of the HRA could also enable a more detailed review of the likely exposure of sensitive features to emissions.

Consider national, regional and local initiatives or measures which can be relied upon to reduce background levels at the site

5.58 Where an existing national, regional or local initiative can be relied upon to lead to the reduction in background levels of pollution at a site, the competent authority should assess the implications of a plan or project against an improving background trend.

[26] CHAPMAN, C. & TYLDESLEY, D. 2016. *Small-scale effects: How the scale of effects has been considered in respect of plans and projects affecting European sites - a review of authoritative decisions*. Natural England Commissioned Reports, Number 205.

5.59 In order to rely on the fact that national, regional or local initiatives will positively affect the environmental context within which a decision is taken on a plan or project (at appropriate assessment), a high degree of certainty is required in order to satisfy the precautionary nature of the legislation. Competent authorities should consider in their assessment the full details of the national, regional or local initiatives that they intend to rely on in an HRA and ensure that they are confident that such schemes will be implemented and achieve the results predicted within the relevant timescales.

5.60 An appropriate assessment would need to consider whether the additional contribution against a reliably predicted declining background level would adversely affect the integrity of the site in question. This question would be informed by a judgement by the competent authority over any delay that the new plan or project might introduce to the timeframe within which the benchmark might have otherwise been achieved (had the plan or project not been consented) and whether it considers any delay would be acceptable or not (having regard to Natural England's advice).

5.61 Examples of strategic work could include:

- Measures to implement Shared Nitrogen Action Plans (SNAPs) that are measured and demonstrated as a certainty, not simply an aspirational plan of potential measures. See Improvement Programme for England's Natura 2000 Sites Atmospheric Nitrogen Theme Plan IPENSTP013.
- National projections given in reports on NE Evidence Catalogue (NECR200 roads report)
- National Policy resulting in emission reductions (e.g. Clean Air Zones, Ultra-low emission zone actions) – these would need to have measureable outcomes for emissions that are certain; again they cannot be aspirational only.
- Evidence of uptake of emission-reduction measures in local agri-environment schemes (whilst recognising the timeframe of any commitments)

Note the request of the Dutch courts for a preliminary ruling from the CJEU in C-294/17 on the Dutch national nitrogen programme (see earlier paragraph 1.6).

Consider measures to avoid or reduce the harmful effects of the plan or project on site integrity

5.62 In a recent decision in C-323/17 *People Over Wind*, the CJEU concluded that any measures intended to avoid or reduce the harmful effects of the plan or project on a European Site should be taken into account at the appropriate assessment stage, rather than the preceding screening stage.

5.63 A submitted proposal subject to appropriate assessment by a competent authority may already contain such measures that have already been voluntarily proposed by the applicant. Further 'additional' mitigation measures can also be imposed by that competent authority on the proposal by way of formal conditions or restrictions subject to which a permission or authorisation may be given. These may be different to or go further than any mitigation measures already proposed by the applicant.

5.64 However, it is relevant to consider these matters at the appropriate assessment stage and Natural England may wish to advise a competent authority on such measures.

5.65 Avoidance and mitigation measures must be capable of preventing adverse effects on site integrity over the full lifetime of the plan or project. To be viable, such measures should be considered to be effective, reliable, timely, guaranteed and of sufficient duration.

5.66 As a result, the inclusion of these measures should be supported by evidence and confidence that they will be effective and that they can be adequately secured and legally enforced to ensure they are strictly implemented by the plan/project proposer.

5.67 Examples of plan/project specific measures to mitigate air quality effects might include;

- Traffic management measures which reduce emissions at source e.g. road speed reduction measures aimed at reducing impacts on sensitive sites/features
- Planting of wooded shelterbelts or other types of green barriers such as trees, green walls and hedges to intercept and limit the dispersal of traffic emissions to sensitive sites/features.

Consider any likely in-combination effects with other live plans and projects from other sectors

5.68 Where a plan or project has been screened in for appropriate assessment based on the likelihood of it having a significant effect <u>alone</u>, it should initially be subject to appropriate assessment on this basis.

5.69 If, after considering and applying any further mitigation measures to the plan or project, the competent authority considers that the risk of residual effects remain which are appreciable (i.e. not inconsequential) but no longer adverse in their own right, then a further in-combination assessment of these residual effects would be required at this stage to check for a combined adverse effect (see principles included in step 4b/c).

5.70 Other plans or projects that could add to the road traffic effects of the subject plan or project and have a cumulative effect on a particular site could originate from other sectors (e.g. applications for intensive livestock permits or industrial installations).

6. Giving Natural England's advice to the competent authority for the purposes of the appropriate assessment

6.1 The competent authority must have regard to any representations that Natural England makes about its assessment and can give its views considerable weight in coming to its decision[27]. However, Natural England's advice on an appropriate assessment is not binding and it does not have to be given such weight if cogent reasons can be given by a competent authority for departing from it[28].

6.2 Competent authorities may consult Natural England on their final appropriate assessment and the conclusions that have been reached Natural England's response will represent its formal opinion, as the appropriate nature conservation body, on the effects of the proposals on the integrity of the European Site(s) in accordance with the Habitats Regulations.

6.3 Natural England should advise on the competent authority's conclusion reached by its appropriate assessment. Where we do not agree with the conclusions of the assessment, we should explain why not with clear and credible reasoning. We may wish to advise on further modifications/conditions/restrictions that could, in our view, enable the competent authority to conclude no adverse effect on the integrity of the site, for instance.

6.4 Where an adverse effect on a European site's integrity cannot be ruled out by a competent authority, despite the application of additional mitigation, it does not necessarily follow that the plan or project will not be permitted. In accordance with the Habitats Regulations, the competent authority (in conjunction with the project proposers and the relevant Government department) could then consider whether the proposal can satisfy stages 3 and 4 of the Habitats Regulations Assessment (consideration of alternative solutions and imperative reasons of overriding public interest) subject to securing the necessary compensatory measures. In these circumstances, the competent authority should initially be referred to current Government guidance on applying these stages of HRA.

[27] See (Ashdown Forest Economic Development LLP v SSCLG, Wealden District Council [2014] EWHC 406 (Admin) at paragraph 110)

[28] See R (Akester) v. DEFRA [2010] EWHC 232 (Admin) at paragraph 112; Wealden DC v. SSCLG [2016] EWHC 247 (Admin) at paragraphs 91 and 95; DLA Delivery v. Lewes District Council [2015] EWHC 2311 at paragraph 32; Mynydd y Gwynt at paragraph 20.

6.5 Natural England staff should act in accordance with Part 7 of Natural England's Non-Financial Schedule of Delegations when giving its advice to competent authorities on the appropriate assessment of certain plans and projects.

For further information about the content of this guidance note, please contact Natural England Planning Consultations Team at consultations@naturalengland.org.uk.

Appendix A: Summary Flowchart – advising on steps for HRA of plans/projects with road traffic emissions

Stage	Flowchart step		Supplemental evidence/ basis for judgment
Initial screening for credible risk of an effect	**1**	Check Distance criteria - could significant emissions reach a protected site? *Yes* = move to Step 2 *No* = no further HRA required	Industry standards based on likely distance for modelled emissions (scoping model); often related back to significance threshold Distance Criteria – 200m for roads and available upon request; note this is currently under review APIS Introduction to Air Pollution
	2	Check the sensitivity of qualifying habitats or supporting habitat of qualifying species. Are habitats in proximity sensitive to the emission type? *Yes* = move to Step 3 *No* = no further HRA required	APIS Site relevant Critical Loads and Levels (based on literature and professional judgement) http://www.apis.ac.uk/srcl
Detailed screening for determining whether screening thresholds are appropriate	**3**	Check habitat likelihood to be exposed to emissions Are the sensitive habitats where emissions are predicted to be? *Yes or Unsure* = move to Step 4a *No* = no further HRA required	Use application documents to understand predicted emissions (magnitude and location if available). If not available, assume emissions reach entire site in proximity. Investigate location of habitats determined as sensitive in Step 2. Use MAGIC priority habitat layers (internal staff: if necessary contact Site responsible Officer for advice to understand if sensitive habitats are present).
Applying screening thresholds	**4a**	Apply Screening Threshold Alone *If below threshold alone* = move to step 4b. *If above* = move straight to step 5.	Ascertain the Process Contribution (PC) or proxy increase in traffic from the plan or project (emissions and predicted deposition or AADT flow). This can be determined through application document, screening model results, detailed model results and information from APIS. Apply Screening threshold (1% of critical level or load or 1000AADT) alone.

Stage	Flowchart step		Supplemental evidence/ basis for judgment
	4b	Apply Screening Threshold In-combination with other traffic/roads *If below threshold in-combination* = move to step 4c. *If above* = move straight to step 5.	Use information from competent authority to determine if there are plans or projects in the pipeline (not in background pollution) that should be considered in-combination for emission from roads/ increase in traffic. For instance, add traffic increases/ emissions & deposition from other Local Plans together and apply 1000 AADT/ 1% to that sum.
	4c	Apply screening threshold in-combination across sectors *If below threshold in-combination*= no likely significant effect can be advised and no further assessment is required. *If above* = move to step 5.	Use information from other competent authorities (Planning Portal or Environmental Permitting register) to determine if there are nearby permissions that would have an in-combination effect with the roads being assessed. When all relevant proposals together (in-combination) fall below the 1% or 1000 AADT level of change, there is reasonable rationale to consider the proposal unlikely to have a significant effect.
Advise Appropriate Assessment is required and contribute scoping advice	**5**	Provide supporting evidence to Competent Authority (scoped as appropriate) Proceed to Step 6 when requested by competent authority and sufficient information is available to provide advice	• Check distance of sensitive habitats from emissions • Check European Site Conservation Objectives • Check environmental benchmark (critical level and load) • Check background concentrations and exceedance • Check APIS Trends Tab for reasonable expectation that background pollution is decreasing • Assess likely scale and duration of impacts on habitats from emissions • Check strategic initiatives in area (if would be undermined if project or plan was allowed) • Check mitigation options and whether detailed modelling may be needed (up to competent authority) • Consider any residual effects (after mitigation where practicable) and check for in-combination effects with other plans/projects
Advice on the appropriate assessment	**6**	Competent Authority has provided an Appropriate Assessment conclusion When requested by competent authority and information is available to provide advice	Give formal advice on appropriate assessment – provide reasoning for our advice

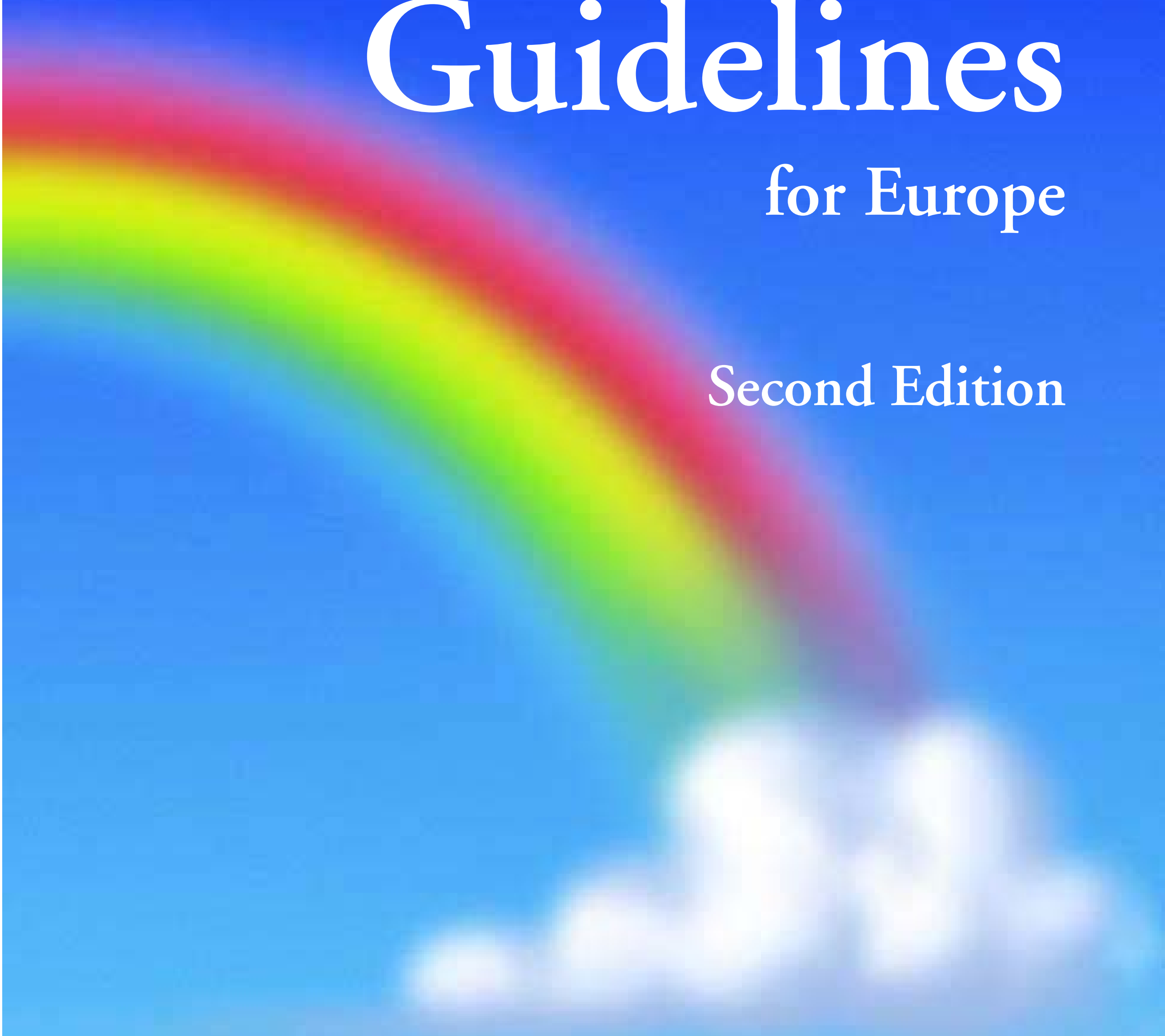
A.3 World Health Organization. 2000. Air Quality Guidelines for Europe. WHO Regional Publications, European Series, No. 91. Second Edition

World Health Organization
Regional Office for Europe
Copenhagen

Air Quality Guidelines

for Europe

Second Edition



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The World Health Organization was established in 1948 as a specialized agency of the United Nations serving as the directing and coordinating authority for international health matters and public health. One of WHO's constitutional functions is to provide objective and reliable information and advice in the field of human health, a responsibility that it fulfils in part through its publications programmes. Through its publications, the Organization seeks to support national health strategies and address the most pressing public health concerns.

The WHO Regional Office for Europe is one of six regional offices throughout the world, each with its own programme geared to the particular health problems of the countries it serves. The European Region embraces some 870 million people living in an area stretching from Greenland in the north and the Mediterranean in the south to the Pacific shores of the Russian Federation. The European programme of WHO therefore concentrates both on the problems associated with industrial and post-industrial society and on those faced by the emerging democracies of central and eastern Europe and the former USSR.

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Air Quality Guidelines
for Europe

Second Edition

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Contents

PART III. EVALUATION OF ECOTOXIC EFFECTS

Foreword

Clean air is considered to be a basic requirement for human health and wellbeing. In spite of the introduction of cleaner technologies in industry, energy production and transport, air pollution remains a major health risk. Recent epidemiological studies have provided evidence that in Europe hundreds of thousands of premature deaths are attributed to air pollution. The World Health Organization has been concerned with air pollution and its impact on human health for more than 40 years. In 1987 these activities culminated in the publication of the first edition of Air quality guidelines for Europe. *It was the aim of the guidelines to provide a basis for protecting public health from adverse effects of air pollutants, to eliminate or reduce exposure to hazardous air pollutants, and to guide national and local authorities in their risk management decisions. The guidelines were received with great enthusiasm and found wide application in environmental decision-making in the European Region as well as in other parts of the world.*

Since the publication of the first edition, new scientific data in the field of air pollution toxicology and epidemiology have emerged and new developments in risk assessment methodology have taken place. It was therefore necessary to update and revise the existing guidelines. Starting in 1993, the Bilthoven Division of the WHO European Centre for Environment and Health undertook this process in close cooperation with WHO headquarters and the European Commission. More than 100 experts contributed to the preparation of the background documents or participated in the scientific discussions that led to the derivation of guideline values for a great number of air pollutants. WHO is most grateful for their contribution and expert advice. Financial support received from the European Commission, the Swedish Environmental Protection Agency and the Government of the Netherlands during the preparation of the second edition of the guidelines made this effort possible and is warmly acknowledged.

The guidelines are a contribution to HEALTH21, the health for all policy framework for the WHO European Region. This states that, by the year 2015, people in the Region should live in a safer physical environment, with exposure to contaminants hazardous to health at levels not exceeding internationally agreed standards. WHO is therefore pleased to see that the revised air quality *guidelines are being used as a starting point for the derivation of legally binding limit values in the framework of the EU Air Quality Directive. Also, the global guidelines for air quality, recently issued by WHO headquarters, are based on the revised guidelines for Europe.*

Thus, the work and efforts of everybody who contributed to the revision of the guidelines has already had an important impact. It is expected that the publication of this second edition will provide the Member States with a sound basis for improving human health by ensuring adequate air quality for all. I should like to warmly thank all the WHO staff who made this important endeavour possible.

Marc A. Danzon
WHO Regional Director for Europe

Preface

The first edition of the WHO *Air quality guidelines for Europe* was published in 1987. Since then new data have emerged and new developments in risk assessment methodology have taken place, necessitating the updating and revision of the existing guidelines. The Bilthoven Division of the WHO European Centre for Environment and Health has undertaken this process in close cooperation with the International Programme on Chemical Safety (IPCS) and the European Commission.

At the start of the process, the methods to be used in the risk assessment process, the use of the threshold concept, the application of uncertainty factors, and the quantitative risk assessment of carcinogens were discussed, and the approach to be used was agreed on. In setting priorities for the compounds to be reviewed, a number of criteria were established: (*a*) the compound (or mixture) posed a widespread problem in terms of exposure sources; (*b*) the potential for personal exposure was large; (*c*) new data on health or environmental impact had emerged; (*d*) monitoring had become feasible since the previous evaluation; and (*e*) a positive trend in ambient air concentrations was evident. Application of these criteria has resulted in the selection of the air pollutants addressed in the review process.

It is the aim of the guidelines to provide a basis for protecting public health from adverse effects of air pollutants and to eliminate or reduce exposure to those pollutants that are known or likely to be hazardous to human health or wellbeing. The guidelines are intended to provide background information and guidance to (inter)national and local authorities in making risk assessment and risk management decisions. In establishing pollutant levels below which exposure – for life or for a given period of time – does not constitute a significant public health risk, the guidelines provide a basis for setting standards or limit values for air pollutants.

Although the guidelines are considered to be protective to human health they are by no means a "green light" for pollution, and it should be stressed that attempts should be made to keep air pollution levels as low as practically achievable. In addition, it should be noted that in general the guidelines do not differentiate between indoor and outdoor air exposure because, although the site of exposure determines the composition of the air and the concentration of the various pollutants, it does not directly affect the exposure–response relationship.

In general, the guidelines address single pollutants, whereas in real life exposure to mixtures of chemicals occurs, with additive, synergistic or antagonistic effects. In dealing with practical situations or standard-setting procedures, therefore, consideration should be given to the interrelationships between the various air pollutants. It should be emphasized, however, that the guidelines are health-based or based on environmental effects, and are not standards *per se*. In setting legally binding standards, considerations such as prevailing exposure levels, technical feasibility, source control measures, abatement strategies, and social, economic and cultural conditions should be taken into account.

It is a policy issue to decide which specific groups at risk should be protected by the standards and what degree of risk is considered to be acceptable. These decisions are influenced by differences in risk perception among the general population and the various stakeholders in the process, but also by differences in social situations in different countries, and by the way the risks associated with air pollution are compared with risks from other environmental exposures or human activities. National standards may therefore differ from country to country and may be above or below the respective WHO guideline value.

This publication includes an introduction on the nature of the guidelines and the methodology used to establish guideline values for a number of air pollutants. In addition, it describes the various aspects that need to be considered by national or local authorities when guidelines are transformed into legally binding standards. For the pollutants addressed, the sections on "Health risk evaluation" and "Guidelines" describe the most relevant considerations that have led to the recommended guideline values. For detailed information on exposure and on the potential health effects of the reviewed pollutants, the reader is referred to the Regional Office's web site, where the background documents on the individual air pollutants can be accessed.

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PART I

GENERAL

Introduction

Human beings need a regular supply of food and water and an essentially continuous supply of air. The requirements for air and water are relatively constant (10–20 m^3 and 1–2 litres per day, respectively). That all people should have free access to air and water of acceptable quality is a fundamental human right. Recognizing the need of humans for clean air, in 1987 the WHO Regional Office for Europe published *Air quality guidelines for Europe (1)*, containing health risk assessments of 28 chemical air contaminants.

These guidelines can be seen as a contribution to target 10 of HEALTH21, the health for all policy framework for the WHO European Region as formulated in 1999 *(2)*. This target states that by the year 2015, people in the Region should live in a safer physical environment, with exposure to contaminants hazardous to health at levels not exceeding internationally agreed standards. The achievement of this target will require the introduction of effective legislative, administrative and technical measures for the surveillance and control of both outdoor and indoor air pollution, in order to comply with criteria to safeguard human health. Unfortunately, this ambitious objective is not likely to be met in the next few years in many areas of Europe. Improvement in epidemiological research over the 1990s and greater sensitivity of the present studies have revealed that people's health may be affected by exposures to much lower levels of some common air pollutants than believed even a few years ago. While the no-risk situation is not likely to be achieved, a minimization of the risk should be the objective of air quality management, and this is probably a major conceptual development of the last few years.

Various chemicals are emitted into the air from both natural and man-made (anthropogenic) sources. The quantities may range from hundreds to millions of tonnes annually. Natural air pollution stems from various biotic and abiotic sources such as plants, radiological decomposition, forest fires, volcanoes and other geothermal sources, and emissions from land and water. These result in a natural background concentration that varies according to local sources or specific weather conditions. Anthropogenic air pollution has existed at least since people learned to use fire, but it has increased rapidly since industrialization began. The increase in air pollution resulting from the expanding use of fossil energy sources and the growth in the manufacture and use of chemicals has been accompanied by mounting public awareness of and concern about its detrimental effects on health and the environment. Moreover, knowledge of the nature, quantity, physico-chemical behaviour and effects of air pollutants has greatly increased in recent years. Nevertheless, more needs to be known. Certain aspects of the health effects of air pollutants require further assessment; these include newer scientific areas such as developmental toxicity. The proposed guideline values will undoubtedly be changed as future studies lead to new information.

The impact of air pollution is broad. In humans, the pulmonary deposition and absorption of inhaled chemicals can have direct consequences for health. Nevertheless, public health can also be indirectly affected by deposition of air pollutants in environmental media and uptake by plants and animals, resulting in chemicals entering the food chain or being present in drinking-water and thereby constituting additional sources of human exposure. Furthermore, the direct effects of air pollutants on plants, animals and soil can influence the structure and function of ecosystems, including their self-regulation ability, thereby affecting the quality of life.

In recent decades, major efforts have been made to reduce air pollution in the European Region. The emission of the main air pollutants has declined significantly. The most pronounced effect is observed for sulfur dioxide: its total emission was reduced by about 50% in the period 1980–1995. Reduction of emission of nitrogen oxides was smaller and was observed only after 1990: total emission declined by about 15% in the period from 1990 to 1995 *(3)*. The reduction of sulfur dioxide emission is reflected by declining concentrations in ambient air in urban areas. Trends in concentrations of other pollutants in urban air, such as nitrogen dioxide or particulate matter, are less clear and it is envisaged that these pollutants still constitute a risk to human health *(4)*.

Many countries of the European Region encounter similar air pollution problems, partly because pollution sources are similar, and in any case air pollution does not respect national frontiers. The subject of the transboundary long-range transport of air pollution has received increasing attention in Europe over the last decade. International efforts to combat emissions are undertaken, for instance within the framework of the Convention on Long-range Transboundary Air Pollution established by the United Nations Economic Commission for Europe *(5, 6)*.

The task of reducing levels of exposure to air pollutants is a complex one. It begins with an analysis to determine which chemicals are present in the air, at what levels, and whether likely levels of exposure are hazardous to human health and the environment. It must then be decided whether an unacceptable risk is present. When a problem is identified, mitigation strategies should be developed and implemented so as to prevent excessive risk to public health in the most efficient and cost-effective way.

Analyses of air pollution problems are exceedingly complicated. Some are national in scope (such as the definition of actual levels of exposure of the population, the determination of acceptable risk, and the identification of the most efficient control strategies), while others are of a more basic character and are applicable in all countries (such as analysis of the relationships between chemical exposure levels, and doses and their effects). The latter form the basis of these guidelines.

The most direct and important source of air pollution affecting the health of many people is tobacco smoke. Even those who do not smoke may inhale the smoke produced by others ("passive smoking"). Indoor pollution in general and occupational exposure in particular also contribute substantially to overall human exposure: indoor concentrations of nitrogen dioxide, carbon monoxide, respirable particles, formaldehyde and radon are often higher than outdoor concentrations *(7)*.

Outdoor air pollution can originate from a single point source, which may affect only a relatively small area. More often, outdoor air pollution is caused by a mixture of pollutants from a variety of diffuse sources, such as traffic and heating, and from point sources. Finally, in addition to those emitted by local sources, pollutants transported over medium and long distances contribute further to the overall level of air pollution.

The relative contribution of emission sources to human exposure to air pollution may vary according to regional and lifestyle factors. Although, as far as some pollutants are concerned, indoor air pollution will be of greater importance than outdoor pollution, this does not diminish the importance of outdoor pollution. In terms of the amounts of substances released, the latter is far more important and may have deleterious effects on animals, plants and materials as well as adverse effects on human health. Pollutants produced outdoors may penetrate into the indoor environment and may affect human health by exposure both indoors and outdoors.

NATURE OF THE GUIDELINES

The primary aim of these guidelines is to provide a basis for protecting public health from adverse effects of air pollution and for eliminating, or reducing to a minimum, those contaminants of air that are known or likely to be hazardous to human health and wellbeing. In the present context, guidelines are not restricted to a numerical value below which exposure for a given period of time does not constitute a significant health risk; they also include any kind of recommendation or guidance in the relevant field.

The guidelines are intended to provide background information and guidance to governments in making risk management decisions, particularly in setting standards, but their use is not restricted to this. They also provide information for all who deal with air pollution. The guidelines may be used in planning processes and various kinds of management decisions at community or regional level.

When guideline values are indicated, this does not necessarily mean that they should be used as the starting point for producing general countrywide standards, monitored by a comprehensive network of control stations. In the case of some pollutants, guideline values may be of use mainly for carrying out local control measures around point sources. To aid in this process, information on major sources of pollutants has been provided.

It should be emphasized that when numerical air quality guideline values are given, these values are not standards in themselves. Before transforming them into legally binding standards, the guideline values must be considered in the context of prevailing exposure levels, technical feasibility, source control measures, abatement strategies, and social, economic and cultural conditions (see Chapter 4). In certain circumstances there may be valid reasons to pursue policies that will result in pollutant concentrations above or below the guideline values.

Although these guidelines are considered to protect human health, they are by no means a "green light" for pollution. It should be stressed that attempts should be made to keep air pollution levels as low as practically achievable.

Ambient air pollutants can cause a range of significant effects that require attention: irritation, odour annoyance, and acute and long-term toxic effects. Numerical air quality guidelines either indicate levels combined with exposure times at which no adverse effect is expected in terms of noncarcinogenic endpoints, or they provide an estimate of lifetime cancer risk arising from those substances that are proven human carcinogens or carcinogens with at least limited evidence of human carcinogenicity. It should be noted that the risk estimates for carcinogens do not indicate a safe level, but they are presented so that the carcinogenic potencies of different carcinogens can be compared and an assessment of overall risk made.

It is believed that inhalation of an air pollutant in concentrations and for exposure times below a guideline value will not have adverse effects on health and, in the case of odorous compounds, will not create a nuisance of indirect health significance. This is in line with the definition of health: a state of complete physical, mental and social wellbeing and not merely the absence of disease or infirmity *(8)*. Nevertheless, compliance with recommendations regarding guideline values does not guarantee the absolute exclusion of effects at levels below such values. For example, highly sensitive groups such as those impaired by concurrent disease or other physiological limitations may be affected at or near concentrations referred to in the guideline values. Health effects at or below guideline values may also result from combined exposure to various chemicals or from exposure to the same chemical by multiple routes.

It is important to note that guidelines have been established for single chemicals. Mixtures of chemicals can have additive, synergistic or antagonistic effects. In general, our knowledge of these interactions is rudimentary. One exception can be found in a WHO publication on summer and winter smog *(9)*, which deals with commonly recurring mixtures of air pollutants.

In preparing this second edition of the guidelines, emphasis has been placed on providing data on the exposure–response relationships of the pollutants considered. It is expected that this will provide a basis for estimating the risk to health posed by monitored concentrations of these pollutants.

Although health effects were the major consideration in establishing the guidelines, evidence of the effects of pollutants on terrestrial vegetation was also considered and guideline values were recommended for a few substances (see Part III). These ecological guidelines have been established because, in the long term, only a healthy total environment can guarantee human health and wellbeing. Ecological effects on life-forms other than humans and plants have not been discussed since they are outside the scope of this book.

The guidelines do not differentiate between indoor and outdoor exposure (with the exception of exposure to mercury) because, although the sites of exposure influence the type and concentration of air pollutants, they do not directly affect the basic exposure–effect relationships. Occupational exposure has been considered in the evaluation process, but it was not a main focus of attention as these guidelines relate to the general population. However, it should be noted that occupational exposure may add to the effects of environmental exposure. The guidelines do not apply to very high short-term concentrations that may result from accidents or natural disasters.

The health effects of tobacco smoking have not been assessed here, the carcinogenic effects of smoking having already been evaluated by IARC in 1986 *(10)*. Neither have the effects of air pollutants on climate been considered, since too many uncertainties remain to allow a satisfactory evaluation of possible adverse health and environmental effects. Possible changes of climate, however, should be investigated very seriously by the appropriate bodies because their overall consequences, for example the "greenhouse effect", may go beyond direct adverse effects on human health or ecosystems.

PROCEDURES USED IN THE UPDATING AND REVISION PROCESS

The first step in the process of updating and revising the guidelines was the selection of pollutants. Air pollutants of special environmental and health significance to countries of the European Region were identified and selected by a WHO planning group in 1993 *(11)* on the basis of the following criteria:

(*a*) whether substances or mixtures posed a widespread problem in terms of sources;
(*b*) the ubiquity and abundance of the pollutants where the potential for exposure was large, taking account of both outdoor and indoor exposure;
(*c*) whether significant new information on health effects had become available since the publication of the first edition of the guidelines;
(*d*) the feasibility of monitoring;
(*e*) whether significant non-health (e.g. ecotoxic) effects could occur; and
(*f*) whether a positive trend in ambient levels was evident.

During the deliberations of the planning group, compounds that had not been dealt with in the first edition of the guidelines were also considered, including butadiene, fluoride, compounds associated with global warming and with alterations in global air pollution (and possibly with secondary health effects), and compounds associated with the development of alternative fuels and new fuel additives. Other factors affecting selection included the timetable of the project, and the fact that only those substances for which sufficient documentation was available could be considered.

The existence of relevant WHO Environmental Health Criteria documents was of great value in this respect. On the basis of these considerations, the following 35 pollutants were selected to be included in this second edition of the guidelines:

Organic air pollutants
Acrylonitrile[1]
Benzene
Butadiene
Carbon disulfide[1]
Carbon monoxide
1,2-Dichloroethane[1]
Dichloromethane
Formaldehyde
Polycyclic aromatic hydrocarbons (PAHs)
Polychlorinated biphenyls (PCBs)
Polychlorinated dibenzodioxins and dibenzofurans (PCDDs/PCDFs)
Styrene
Tetrachloroethylene
Toluene
Trichloroethylene
Vinyl chloride[1]

Inorganic air pollutants
Arsenic
Asbestos[1]
Cadmium
Chromium
Fluoride
Hydrogen sulfide[1]
Lead
Manganese
Mercury
Nickel
Platinum
Vanadium[1]

Classical air pollutants
Nitrogen dioxide
Ozone and other photochemical oxidants
Particulate matter
Sulfur dioxide

Indoor air pollutants
Environmental tobacco smoke
Man-made vitreous fibres
Radon

[1] 1987 evaluation retained, not re-evaluated.

In addition to the 35 pollutants listed above, this second edition expands on the ecological effects presented in the first edition in an enlarged section examining the ecotoxic effects of sulfur dioxide (including sulfur and total acid deposition), nitrogen dioxide (and other nitrogen compounds, including ammonia) and ozone.

To carry out the evaluation process, the planning group established a number of working groups on:

- methodology and format
- ecotoxic effects
- classical air pollutants
- inorganic air pollutants
- certain indoor air pollutants
- polychlorinated biphenyls, dioxins and furans
- volatile organic pollutants.

The dates of the meetings of these working groups and the membership are listed in Annex I.

Before the meeting of each working group, scientific background documents providing in-depth reviews of each pollutant were prepared as a basis for discussion. Guidelines were established on the basis of these discussions. After each meeting, a text on each pollutant or pollutant group was drafted on the basis of the amended background documents, incorporating the working group's conclusions and recommendations. The draft report of the working group was then circulated to all participants for their comments and corrections. A final consultation group was then convened to critically review the documents for clarity of presentation, adequacy of description of the rationale supporting each guideline and consistency in the application of criteria, and with a view to possibly considering newly emerged information. The process concluded with a review of the recommendations and conclusions of all the working groups.

It was appreciated, during preparation of this second edition, that the expanded range of pollutants being considered and the considerably expanded database available for some pollutants would lead to a significant lengthening of the text. It was therefore decided to publish in this volume summaries of the data on which the guidelines are based. The full background evaluation will become progressively available on the Regional Office's web site.

As in the first edition, detailed referencing of the relevant literature has been provided with indications of the periods covered by the reviews of individual pollutants. Every effort has been made to ensure that the material provided is as up-to-date as possible, although the extended period of preparation of this second edition has inevitably meant that some sections refer to more recently published material than others.

During the preparation of the second edition, the Directorate-General for Environment, Nuclear Safety and Civil Protection (DGXI) of the European Commission developed a Framework Directive and a number of daughter directives dealing with individual pollutants. It was agreed with the Commission that the final drafts of the revised WHO guideline documents would provide a starting point for discussions by the Commission's working groups aiming at setting legally binding limit values for air quality in the European Union.

REFERENCES

1. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
2. *HEALTH21. The health for all policy framework for the WHO European Region.* Copenhagen, WHO Regional Office for Europe, 1999 (European Health for All Series, No. 6).
3. *Europe's environment: the second assessment*. Copenhagen, European Environment Agency, 1998.
4. *Overview of the environment and health in Europe in the 1990s: Third Ministerial Conference on Environment and Health, London, 16–18 June 1999*. Copenhagen, WHO Regional Office for Europe, 1998 (document EUR/ICP/EHCO 02 02 05/6).
5. *Convention on Long-range Transboundary Air Pollution. Strategies and policies for air pollution abatement. 1994 major review.* New York, United Nations, 1995 (ECE/EB.AIR/44).
6. *Convention on Long-range Transboundary Air Pollution. Major review of strategies and policies for air pollution abatement.* New York, United Nations, 1998 (EB.AIR/1998/3, Add.1).
7. WHO EUROPEAN CENTRE FOR ENVIRONMENT AND HEALTH. *Concern for Europe's tomorrow. Health and the environment in the WHO European Region.* Stuttgart, Wissenschaftliche Verlagsgesellschaft, 1995.
8. *Constitution of the World Health Organization.* Geneva, World Health Organization, 1985.

9. *Acute effects on health of smog episodes.* Copenhagen, WHO Regional Office for Europe, 1992 (WHO Regional Publications, European Series, No. 43).
10. *Tobacco smoking*. Lyons, International Agency for Research on Cancer, 1986 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 38).
11. *Update and revision of the air quality guidelines for Europe. Report of a WHO planning meeting*. Copenhagen, WHO Regional Office for Europe, 1994 (document EUR/ICP/CEH 230).

Criteria used in establishing guideline values

Relevant information on the pollutants was carefully considered during the process of establishing guideline values. Ideally, guideline values should represent concentrations of chemical compounds in air that would not pose any hazard to the human population. Realistic assessment of human health hazards, however, necessitates a distinction between absolute safety and acceptable risk. To produce a guideline with a high probability of offering absolute safety, one would need a detailed knowledge of dose–response relationships in individuals in relation to all sources of exposure, the types of toxic effect elicited by specific pollutants or their mixtures, the existence or nonexistence of "thresholds" for specified toxic effects, the significance of interactions, and the variation in sensitivity and exposure levels within the human population. Such comprehensive and conclusive data on environmental contaminants are generally unavailable. Very often the relevant data are scarce and the quantitative relationships uncertain. Scientific judgement and consensus therefore play an important role in establishing guidance that can be used to indicate acceptable levels of population exposure. Value judgements are needed and the use of subjective terms such as "adverse effects" and "sufficient evidence" is unavoidable.

Although it may be accepted that a certain risk can be tolerated, the risks to individuals within a population may not be equally distributed: there may be subpopulations that are at considerably increased risk. Therefore, groups at special risk in the general population must be taken specifically into account in the risk management process. Even if knowledge about groups with specific sensitivity is available, unknown factors may exist that change the risk in an unpredictable manner. During the preparation of this second edition of the guidelines, attention has been paid to defining specific sensitive subgroups in the population.

INFORMATION COMMON TO CARCINOGENS AND NONCARCINOGENS

Sources, levels and routes of exposure

Available data are provided on the current levels of human exposure to pollutants from all sources, including the air. Special attention is given to atmospheric concentrations in urban and unpolluted rural areas and in the indoor environment. Where appropriate, concentrations in the workplace are also indicated for comparison with environmental levels. To provide information on the contribution from air in relation to all other sources, data on uptake by inhalation, ingestion from water and food, and dermal contact are given where relevant. For most chemicals, however, data on total human exposure are incomplete.

Toxicokinetics

Available data on toxicokinetics (absorption, distribution, metabolism and excretion) of air pollutants in humans and experimental animals are provided for comparison between test species and humans and for interspecies and intraspecies extrapolation, especially to assess the magnitude of body burden from long-term, low-level exposures and to characterize better the mode of toxic action. Data concerning the distribution of a compound in the body are important in determining the molecular or tissue dose to target organs. It has been appreciated that high-to-low-dose and interspecies extrapolations are more easily carried out using equivalent tissue doses. Metabolites are mentioned, particularly if they are known or believed to exert a greater toxic potential than the parent compound. Additional data of interest in determining the fate of a compound in a living organism include the rate of excretion and the biological half-life. These toxicokinetic parameters should be compared between test species and humans for derivation of interspecies factors where this is possible.

Terminology

The following terms and definitions are taken largely from Environmental Health Criteria No. 170, 1994 *(1)*.

Adverse effect Change in morphology, physiology, growth, development or life span of an organism which results in impairment of functional capacity or impairment of capacity to compensate for additional stress or increase in susceptibility to the harmful effects of other environmental influences.

Benchmark dose (BMD) The lower confidence limit of the dose calculated to be associated with a given incidence (e.g. 5% or 10% incidence) of effect estimated from all toxicity data on that effect within that study *(2)*.

Critical effect(s) The adverse effect(s) judged to be most appropriate for the health risk evaluation.

Lowest-observed-adverse-effect level (LOAEL) Lowest concentration or amount of a substance, found by experiment or observation, which causes an adverse alteration of morphology, functional capacity, growth, development or life span of the target organism distinguishable from normal (control) organisms of the same species and strain under the same defined conditions of exposure.

No-observed-adverse-effect level (NOAEL) Greatest concentration or amount of a substance, found by experiment or observation, which causes no detectable adverse alteration of morphology, functional capacity, growth, development or life span of the target organism under defined conditions of exposure. Alterations of morphology, functional capacity, growth, development or life span of the target may be detected which are judged not to be adverse.

Toxicodynamics The process of interaction of chemical substances with target sites and the subsequent reactions leading to adverse effects.

Toxicokinetics The process of the uptake of potentially toxic substances by the body, the biotransformation they undergo, the distribution of the substances and their metabolites in the tissues, and the elimination of the substances and their metabolites from the body. Both the amounts and the concentrations of the substances and their metabolites are studied. The term has essentially the same meaning as pharmacokinetics, but the latter term should be restricted to the study of pharmaceutical substances.

Uncertainty factor (UF) A product of several single factors by which the NOAEL or LOAEL of the critical effect is divided to derive a tolerable intake. These factors account for adequacy of the pivotal study, interspecies extrapolation, inter-individual variability in humans, adequacy of the overall database, and nature of toxicity. The choice of UF should be based on the available scientific evidence.

CRITERIA FOR ENDPOINTS OTHER THAN CARCINOGENICITY

Criteria for selection of NOAEL/LOAEL

For those compounds reportedly without direct carcinogenic effects, determination of the highest concentration at which no adverse effects are observed, or the lowest concentration at which adverse effects are observed in humans, animals or plants is the first step in the derivation of the guideline value. This requires a thorough evaluation of available data on toxicity. The decision as to whether the LOAEL or the NOAEL should be used as a starting point for deriving a guideline value is mainly a matter of availability of data. If a series of data fixes both the LOAEL and the NOAEL, then either might be used. The gap between the lowest-observed-effect level and the no-observed-effect level is among the factors included in judgements concerning the appropriate uncertainty factor. Nevertheless, one needs to consider that in studies in experimental animals, the value of the NOAEL (or LOAEL) is an observed value that is dependent on the protocol and design of the study from which it was derived. There are several factors that influence the magnitude of the value observed, such as the species, sex, age, strain and developmental status of the animals studied; the group size; the sensitivity of the methods applied; and the selection of dose levels. Dose levels are frequently widely spaced, so that the observed NOAEL can be in some cases considerably less than the true no-adverse-effect level, and the observed LOAEL considerably higher than the true lowest-adverse-effect level *(1)*.

A single, free-standing no-observed-effect level that is not defined in reference to a lowest-observed-effect level or a LOAEL is not helpful. It is important to understand that, to be useful in setting guidelines, the NOAEL must be the highest level of exposure at which no adverse effects are detected. It is difficult to be sure that this has been identified unless the level of exposure at which adverse effects begin to appear has also been defined. Opinions on this subject differ, but the working consensus was that the level of exposure of concern in terms of human health is more easily related to the LOAEL, and this level was therefore used whenever possible. In the case of irritant and sensory effects on humans, it is desirable where possible to determine the no-observed-effect level. These effects are discussed in more detail below.

On the basis of the evidence concerning adverse effects, judgements about the uncertainty factors needed to minimize health risks were made. Averaging times were included in the specification of the guidelines, as the duration of exposure is often critical in determining toxicity. Criteria applied to each of these key factors are described below.

Criteria for selection of adverse effect

Definition of a distinction between adverse and non-adverse effects poses considerable difficulties. Any observable biological change might be considered an adverse effect under certain circumstances. An adverse effect has been defined as "any effect resulting in functional impairment and/or pathological lesions that may affect the performance of the whole organism or which contributes to a reduced ability to respond to an additional challenge" *(3)*. Even with such a definition, a significant degree of subjectivity and uncertainty remains. Ambient levels of major air pollutants frequently cause subtle effects that are typically detected only by sensitive methods. This makes it exceedingly difficult, if not impossible, to achieve a broad consensus as to which effects are adverse. To resolve this difficulty, it was agreed that the evidence should be ranked in three categories.

1. The first category comprises observations, even of potential health concern, that are single findings not verified by other groups. Because of the lack of verification by other investigators, such data could not readily be used as a basis for deriving a guideline value. They do, however, indicate the need for further research and may be considered in deriving an appropriate uncertainty factor based on the severity of the observed effects.

2. The second category is a lowest-observed-effect level (or no-observed-effect level) that is supported by other scientific information. When the results are in a direction that might result in pathological changes, there is a higher degree of health concern. Scientific judgement based on all available health information is used to determine how effects in this category can be used in determining the pollutant level that should be avoided so that excessive risk can be prevented.

3. The third category comprises levels of exposure at which there is clear evidence for substantial pathological changes; these findings have had a major influence on the derivation of the guidelines.

Benchmark approach

The benchmark dose (BMD) is the lower confidence limit of the dose that produces a given increase (e.g. 5% or 10%) in the level of an effect to which an uncertainty factor can be applied to develop a tolerable intake. It has a number of advantages over the NOAEL/LOAEL approach *(2)*. First, the BMD is derived on the basis of the entire dose–response curve for the critical, adverse effect rather than that from a single dose group as in the case of the NOAEL/LOAEL. Second, it can be calculated from data sets in which a NOAEL was not determined, eliminating the need for an additional uncertainty factor to be applied to the LOAEL. Third, definition of the BMD as a lower confidence limit accounts for the statistical power and quality of the data; that is, the confidence intervals around the dose–response curve for studies with small numbers of animals or of poor quality and thus lower statistical power would be wide, reflecting the greater uncertainty of the database. On the other hand, better studies would result in narrow confidence limits, and thus in higher BMDs.

Although there is no consensus on the incidence of effect to be used as basis for the BMD, it is generally agreed that the BMD should be comparable with a level of effect typically associated with the NOAEL or LOAEL. Allen et al. *(4, 5)* have estimated that a BMD calculated from the lower confidence limit at 5% is, on average, comparable to the NOAEL, whereas choosing a BMD at 10% is more representative of a LOAEL *(6)*. Choosing a BMD that is comparable to the NOAEL has two advantages: (*a*) it is within the experimental dose-range, eliminating the need to interpolate the dose–response curve to low levels; and (*b*) it justifies the application of similar uncertainty factors as are currently applied to the NOAEL for interspecies and intraspecies variation. It should be noted, however, that the main disadvantage of the benchmark approach is that it is not applicable for discrete toxicity data, such as histopathological or teratogenicity data.

Criteria for selection of uncertainty factors

In previous evaluations by WHO, uncertainty factors (sometimes called safety factors) have been applied to derive guidelines from evidence that conforms to accepted criteria for adverse effects on health *(7–9)*. Traditionally, the uncertainty (safety) factor has been used to allow for uncertainties in extrapolation from animals to humans and from a small group of individuals to a large population, including possibly undetected effects on particularly sensitive members of the population. In addition, uncertainty factors also account for possible synergistic effects of multiple exposures, the seriousness of the observed effects and the adequacy of existing data *(1)*. It is important to understand that the application of such factors does not indicate that it is known that humans are more sensitive than animal species but, rather, that the sensitivity of humans relative to that of other species is usually unknown. It is possible that humans are less sensitive than animals to some chemicals.

In this second edition of the air quality guidelines, the terms “safety factor” and “protection factor” have been replaced by the term “uncertainty factor”. It is felt that this better explains the derivation and implications of such factors. Of course, such a factor is designed to provide an adequate level of protection and an adequate margin of safety, because these factors are applied in the derivation of guidelines for the protection of human health. They are not applied in the derivation of ecological guidelines because these already include a kind of uncertainty factor with regard to the variety of species covered.

A wide range of uncertainty factors are used in this second edition, based on scientific judgements concerning the interplay of various effects. The decision process for developing uncertainty factors has been complex, involving the transformation of mainly non-quantitative information into a single number expressing the judgement of a group of scientists.

Some of the factors taken into account in deciding the margin of protection can be grouped under the heading of scientific uncertainty. Uncertainty occurs because of limitations in the extent or quality of the database. One can confidently set a lower margin of protection (that is, use a smaller uncertainty factor) when a large number of high-quality, mutually supportive scientific experiments in different laboratories using different approaches clearly demonstrate the dose–response, including a lowest-observed-effect level and a no-observed-effect level. In reality, difficulties inherent in studying air pollutants, and the failure to perform much-needed and very specific research, means that often a large uncertainty factor has to be applied.

Where an uncertainty factor was adopted in the derivation of air quality guidelines, the reasoning behind the choice of this factor is given in the scientific background information. As previously mentioned, exceeding a guideline value with an incorporated uncertainty factor does not necessarily mean that adverse effects will result. Nevertheless, the risk to public health will increase, particularly in situations where the most sensitive population group is exposed to several pollutants simultaneously.

Individuals and groups within a population show marked differences in sensitivity to given pollutants. Individuals with pre-existing lung disease, for instance, may be at higher risk from exposure to air pollutants than healthy people. Differences in response can be due to factors other than pre-existing health, including age, sex, level of exercise taken and other unknown factors. Thus, the population must be considered heterogeneous in respect of response to air pollutants. This perhaps wide distribution of sensitivity combined with a distribution of exposure makes the establishment of population-based thresholds of effect very difficult. This problem is taken up in the section on particulate matter (page 186). Existing information tends not to allow adequate assessment of the proportion of the population that is likely to show an enhanced response. Nevertheless, an estimate of even a few percent of the total population entails a large number of people at increased risk.

Deriving a guideline from studies of effects on laboratory animals in the absence of human studies generally requires the application of an increased uncertainty factor, because humans may be more susceptible than laboratory animal species. Negative data from human studies will tend to reduce the magnitude of this uncertainty factor. Also of importance are the nature and reversibility of the reported effect. Deriving a guideline from data that show that a given level of exposure produces only slight alterations in physiological parameters requires a smaller uncertainty factor than when data showing a clearly adverse effect are used. Scientific judgement about uncertainty factors should also take into account the biochemical toxicology of pollutants, including the types of metabolite formed, the variability in metabolism or response in humans suggesting the existence of hypersusceptible groups, and the likelihood that the compound or its metabolites will accumulate in the body.

It is obvious, therefore, that diverse factors must be taken into account in proposing a margin of protection. The uncertainty factor cannot be assigned by a simple mathematical formula; it requires experience, wisdom and judgement.

Feasibility of adopting a standard approach

In preparing this second edition of the guidelines, the feasibility of developing a standard methodology for setting guidelines was discussed. It was agreed that Environmental Health Criteria No. 170 *(1)* was a valuable source of information. On the other hand, it was recognized that large variation in the data available for different compounds made the use of a standard approach impossible. Much of the difficulty concerns the adequacy of the database, and this has played a large part in controlling the methods of assessment adopted. This is illustrated in Table 1.

Table 1. Size and completeness of database in relation to assessment method

Examples	Completeness/ size of database	Uncertainties	Feasibility of expert judgement	Need for standardized approach
Nitrogen dioxide, ozone, lead	+++	+	+++	+
Manganese, nickel	++	++	++	++
Volatile organic compounds	+	+++	+	+++

It will be seen that when the database is strong (that is, when a good deal is known about the human toxicology of the compound) it is suggested that expert judgement can be used to set a guideline. In such circumstances the level of uncertainty is low. If, on the other hand, the database is weak, then a larger level of uncertainty will exist and there is much to be said for using a standardized approach, probably involving the application of a substantial uncertainty factor. The dangers of replacing expert judgement and the application of common sense with advanced, complex and sometimes not intuitively obvious statistical methods for deriving guidelines was discussed. It was agreed that a cautious approach should be adopted.

Criteria for selection of averaging times

The development of toxicity is a complex function of the interaction between concentration of a pollutant and duration of exposure. A chemical may cause acute, damaging effects after peak exposure for a short period and irreversible or incapacitating effects after prolonged exposure to lower concentrations. Our knowledge is usually insufficient to define accurately the relationship between effects on the one hand and concentration and time on the other. Expert judgement must be applied, therefore, based on the weight of the evidence available *(10)*.

Generally, when short-term exposures lead to adverse effects, short-term averaging times are recommended. The use of a long-term average under such conditions would be misleading, since the typical pattern of repeated peak exposures is lost during the averaging process and the risk manager would have difficulties in deciding on effective strategies. In other cases, knowledge of the exposure–response relationship may be sufficient to allow recommendation of a long averaging period. This is frequently the case for chemicals that accumulate in the body and thereby produce adverse effects. In such cases, the integral of concentration over a long period can have more impact than the pattern of peak exposure.

It should be noted that the specified averaging times are based on effects on health. Therefore, if the guidelines are used as a basis for regulation, the regulator needs to select the most appropriately and practically defined standards in relation to the guidelines, without necessarily adopting the guidelines directly. It was appreciated that monitoring techniques for some pollutants would not allow reporting of data in terms of the averaging times recommended in the guidelines. Under such circumstances, a compromise between the averaging time specified in the guidelines and that obtainable in practice has to be reached in setting an air quality standard.

A similar situation occurs for effects on vegetation. Plants are generally damaged by short-term exposures to high concentration as well as by long-term exposures to low concentration. Therefore, both short- and long-term guidelines to protect plants are proposed.

Criteria for consideration of sensory effects

Some of the substances selected for evaluation have malodorous properties at concentrations far below those at which toxic effects occur. Although odour annoyance cannot be regarded as an adverse health effect in a strict sense, it does affect the quality of life. Therefore, odour threshold levels have been indicated where relevant and used as a basis for separate guideline values.

For practical purposes, the following characteristics and respective levels were considered in the evaluation of sensory effects:

- intensity, where the *detection threshold level* is defined as the lower limit of the perceived intensity range (by convention the lowest concentration that can be detected in 50% of the cases in which it is present);

- quality, where the *recognition threshold level* is defined as the lowest concentration at which the sensory effect, such as odour, can be recognized correctly in 50% of the cases; and

- acceptability and annoyance, where the *nuisance threshold level* is defined as the concentration at which not more than a small proportion of the population (less than 5%) experiences annoyance for a small part of the time (less than 2%); since annoyance will be influenced by a number of psychological and socioeconomic factors, a nuisance threshold level cannot be defined on the basis of concentration alone.

During revision of the guidelines, the problems of irritation (for example, of the skin) and headache were also considered as possible problems of annoyance. It was agreed that headache should be regarded as a health endpoint and not merely as a matter of annoyance.

CRITERIA FOR CARCINOGENIC ENDPOINT

Cancer risk assessment is basically a two-step procedure, involving a qualitative assessment of how likely it is that an agent is a human carcinogen, and a quantitative assessment of the cancer risk that is likely to occur at given levels and duration of exposure (*11*).

Qualitative assessment of carcinogenicity

The decision to consider a substance as a carcinogen is based on the qualitative evaluation of all available information on carcinogenicity, ensuring that the association is unlikely to be due to chance alone. Here the classification criteria of the International Agency for Research on Cancer (IARC) have been applied (Box 1). In dealing with carcinogens, a "general rule" and exceptions from this were defined. The "general rule" states that for compounds in IARC Groups 1 and 2A (proven human carcinogens, and carcinogens with at least limited evidence of human carcinogenicity), guideline values are derived with the use of quantitative risk assessment with low-dose risk extrapolation. For compounds in Groups 2B (inadequate evidence in humans but sufficient evidence in animals), 3 (unclassifiable as to carcinogenicity in humans) and 4 (noncarcinogenic), guideline values are derived with the use of a threshold (uncertainty factor) method. For compounds in Group 2B, this may incorporate a separate factor for the possibility of a carcinogenic effect in humans.

In case of sufficient scientific evidence, one may be justified in deviating from the "general rule". First, a compound classified in Group 1 or 2A may be assessed with the use of the uncertainty factor methodology, provided that there is strong evidence that it is not genotoxic as judged from a battery of short-term test systems for gene mutation, DNA damage, etc. In such cases it can be established with certainty that an increase in exposure to the compound is associated with an increase in cancer incidence only above a certain level of exposure. It was considered that this required a level of understanding of the mechanisms of action not presently available for the compounds classified as Group 1 or 2A on the current list. Second, a compound in Group 2B may be assessed with the use of quantitative risk assessment methods instead of the uncertainty factor approach. This may be considered appropriate where the mechanism of carcinogenesis in animals is likely to be a non-threshold phenomenon as indicated, for example, by the genotoxic activity of the compound in different short-term test systems.

Quantitative assessment of carcinogenic potency

The aim of quantitative risk assessment is to use information available from very specific study situations to predict the risk to the general population posed by exposure to ambient levels of carcinogens. In general, therefore, quantitative risk assessment includes the extrapolation of risk from relatively high dose levels (characteristic of animal experiments or occupational exposures), where cancer responses can be measured, to relatively low dose levels, which are of concern in environmental protection and where such

Box 1. Classification criteria of the International Agency for Research on Cancer

Group 1 – the agent (mixture) is carcinogenic to humans.
The exposure circumstance entails exposures that are carcinogenic to humans.
This category is used when there is *sufficient evidence* of carcinogenicity in humans. Exceptionally, an agent (mixture) may be placed in this category when evidence in humans is less than sufficient but there is *sufficient evidence* of carcinogenicity in experimental animals and strong evidence in exposed humans that the agent (mixture) acts through a relevant mechanism of carcinogenicity.

Group 2
This category includes agents, mixtures and exposure circumstances for which, at one extreme, the degree of evidence of carcinogenicity in humans is almost sufficient, as well as those for which, at the other extreme, there are no human data but for which there is evidence of carcinogenicity in experimental animals. Agents, mixtures and exposure circumstances are assigned to either group 2A (probably carcinogenic to humans) or group 2B (possibly carcinogenic to humans) on the basis of epidemiological and experimental evidence of carcinogenicity and other relevant data.

Group 2A – the agent (mixture) is probably carcinogenic to humans.
The exposure circumstance entails exposures that are probably carcinogenic to humans.
This category is used when there is *limited evidence* of carcinogenicity in humans and sufficient evidence of carcinogenicity in experimental animals. In some cases, an agent (mixture) may be classified in this category when there is inadequate evidence of carcinogenicity in humans and *sufficient evidence* of carcinogenicity in experimental animals and strong evidence that the carcinogenesis is mediated by a mechanism that also operates in humans. Exceptionally, an agent, mixture or exposure circumstance may be classified in this category solely on the basis of limited evidence of carcinogenicity in humans.

Group 2B – the agent (mixture) is possibly carcinogenic to humans.
The exposure circumstance entails exposures that are possibly carcinogenic to humans.
This category is used for agents, mixtures and exposure circumstances for which there is *limited evidence* of carcinogenicity in humans and less than *sufficient evidence of* carcinogenicity in experimental animals. It may also be used when there is *inadequate evidence* of carcinogenicity in humans but there is *sufficient evidence* of carcinogenicity in experimental animals. In some instances, an agent, mixture or exposure circumstance for which there is *inadequate evidence* of carcinogenicity in humans but *limited evidence* of carcinogenicity in experimental animals together with supporting evidence from other relevant data may be placed in this group.

Box 1. (contd)

Group 3 – The agent (mixture or exposure circumstance) is not classifiable as to its carcinogenicity to humans.
This category is used most commonly for agents, mixtures and exposure circumstances for which the evidence of carcinogenicity is inadequate in humans and inadequate or limited in experimental animals. Exceptionally, agents (mixtures) for which the evidence of carcinogenicity is inadequate in humans but sufficient in experimental animals may be placed in this category when there is strong evidence that the mechanism of carcinogenicity in experimental animals does not operate in humans. Agents, mixtures and exposure circumstances that do not fall into any other group are also placed in this category.

Group 4 – The agent (mixture) is probably not carcinogenic to humans.
This category is used for agents or mixtures for which there is *evidence suggesting lack of carcinogenicity* in humans and in experimental animals. In some instances, agents or mixtures for which there is *inadequate evidence* of carcinogenicity in humans but *evidence suggesting lack of carcinogenicity* in experimental animals, consistently and strongly supported by a broad range of other relevant data, may be classified in this group.

Source: IARC *(12)*.

risks are too small to be measured directly, either by animal studies or by epidemiological studies.

The choice of the extrapolation model depends on the current understanding of the mechanisms of carcinogenesis *(13)*, and *no* single mathematical procedure can be regarded as fully appropriate for low-dose extrapolation. Methods based on a linear, non-threshold assumption have been used at the national and international level more frequently than models that assume a safe or virtually safe threshold.

In these guidelines, the risk associated with lifetime exposure to a certain concentration of a carcinogen in the air has been estimated by linear extrapolation and the carcinogenic potency expressed as the incremental unit risk estimate. The incremental unit risk estimate for an air pollutant is defined as "the additional lifetime cancer risk occurring in a hypothetical population in which all individuals are exposed continuously from birth throughout their lifetimes to a concentration of 1 $\mu g/m^3$ of the agent in the air they breathe" *(14)*.

The results of calculations expressed in unit risk estimates provide the opportunity to compare the carcinogenic potency of different compounds and can help to set priorities in pollution control, taking into account current levels of exposure. By using unit risk estimates, any reference to the "acceptability" of risk is avoided. The decision on the acceptability of a risk should be made by national authorities within the framework of risk management. To support authorities in the decision-making process, the guideline sections for carcinogenic pollutants provide the concentrations in air associated with an excess cancer risk of 1 in a population of 10 000, 1 in 100 000 or 1 in 1 000 000, respectively, calculated from the unit risk.

For those substances for which appropriate human studies are available, the method known as the "average relative risk model" has been used, and is therefore described in more detail below.

Several methods have been used to estimate the incremental risks based on data from animal studies. Two general approaches have been proposed. A strictly linearized estimate has generally been used by the US Environmental Protection Agency (EPA) *(14)*. Nonlinear relations have been proposed for use when the data derived from animal studies indicate a nonlinear dose–response relationship or when there is evidence that the capacity to metabolize the polluting chemical to a carcinogenic form is of limited capacity.

Quantitative assessment of carcinogenicity based on human data

Quantitative assessment using the average relative risk model comprises four steps: (*a*) selection of studies; (*b*) standardized description of study results in terms of relative risk, exposure level and duration of exposure; (*c*) extrapolation towards zero dose; and (*d*) application to a general (hypothetical) population.

First, a reliable human study must be identified, where the exposure of the study population can be estimated with acceptable confidence and the excess cancer incidence is statistically significant. If several studies exist, the best representative study should be selected or several risk estimates evaluated.

Once a study is identified, the relative risk as a measure of response is calculated. It is important to note that the 95% confidence limits around the central estimate of the relative risk can be wide and should be specifically stated and evaluated. The relative risk is then used to calculate the excess lifetime cancer risk expressed as unit risk (UR) associated with a lifetime exposure to 1 μg/m^3, as follows:

$$UR = \frac{P_0(RR - 1)}{X}$$

where: P_0 = background lifetime risk; this is taken from age/cause-specific death or incidence rates found in national vital statistics tables using the life table methodology, or it is available from a matched control population

RR = relative risk, being the ratio between the observed (O) and expected (E) number of cancer cases in the exposed population; the relative risk is sometimes expressed as the standardized mortality ratio SMR = (O/E) × 100

X = lifetime average exposure (standardized lifetime exposure for the study population on a lifetime continuous exposure basis); in the case of occupational studies, X represents a conversion from the occupational 8-hour, 240-day exposure over a specific number of working years and can be calculated as X = 8-hour TWA × 8/24 × 240/365 × (average exposure duration [in years])/(life expectancy [70 years]), where TWA is the time-weighted average (μg/m^3).

It should be noted that the unit lifetime risk depends on P_0 (background lifetime risk), which is determined from national age-specific cancer incidence or mortality rates. Since these rates are also determined by exposures other than the one of interest and may vary from country to country, it follows that the UR may also vary from one country to another.

Necessary assumptions for average relative risk method

Before any attempt is made to assess the risk in the general population, numerous assumptions are needed at each phase of the risk assessment process to fill in various gaps in the underlying scientific database. As a first step in any given risk assessment, therefore, an attempt should be made to identify the major assumptions that have to be made, indicating their probable consequences. These assumptions are as follows.

1. *The response (measured as relative risk) is some function of cumulative dose or exposure.*

2. *There is no threshold dose for carcinogens.*
Many stages in the basic mechanism of carcinogenesis are not yet known or are only partly understood. Taking available scientific findings into consideration,

however, several scientific bodies *(8, 15–17)* have concluded that there is no scientific basis for assuming a threshold or no-effect level for chemical carcinogens. This view is based on the fact that most agents that cause cancer also cause irreversible damage to deoxyribonucleic acid (DNA). The assumption applies for all non-threshold models.

3. *The linear extrapolation of the dose–response curve towards zero gives an upper-bound conservative estimate of the true risk function if the unknown (true) dose–response curve has a sigmoidal shape.*
The scientific justification for the use of a linear non-threshold extrapolation model stems from several sources: the similarity between carcinogenesis and mutagenesis as processes that both have DNA as target molecules; the strong evidence of the linearity of dose–response relationships for mutagenesis; the evidence for the linearity of the DNA binding of chemical carcinogens in the liver and skin; the evidence for the linearity in the dose–response relationship in the initiation stage of the mouse 2-stage tumorigenesis model; and the rough consistency with the linearity of the dose–response relationships for several epidemiological studies. This assumption applies for all linear models.

4. *There is constancy of the relative risk in the specific study situation.*
In a strict sense, constancy of the relative risk means that the background age/cause-specific rate at any time is increased by a constant factor. The advantage of the average relative risk method is that this needs to be true only for the average.

Advantages of the method

The average relative risk method was selected in preference to many other more sophisticated extrapolation models because it has several advantages, the main one being that it seems to be appropriate for a fairly large class of different carcinogens, as well as for different human studies. This is possible because averaging doses, that is, averaging done over concentration and duration of exposure, gives a reasonable measure of exposure when dose rates are not constant in time. This may be illustrated by the fact that the use of more sophisticated models *(14, 18, 19)* results in risk estimates very similar to those obtained by the average relative risk method.

Another advantage of the method is that the carcinogenic potency can be calculated when estimates of the average level and duration of exposure are the only known parameters besides the relative risk. Furthermore, the method has the advantage of being simple to apply, allowing non-experts in the field of risk models to calculate a lifetime risk from exposure to the carcinogens.

Limitations of the method

As pointed out earlier, the average relative risk method is based on several assumptions that appear to be valid in a wide variety of situations. There are specific situations, however, in which the method cannot be recommended, mainly because the assumptions do not hold true.

The cumulative dose concept, for instance, is inappropriate when the mechanism of the carcinogen suggests that it cannot produce cancer throughout all stages of the cancer development process. Also, specific toxicokinetic properties, such as a higher excretion rate of a carcinogen at higher doses or a relatively lower production rate of carcinogenic metabolites at lower doses, may diminish the usefulness of the method in estimating cancer risk. Furthermore, supralinearity of the dose–response curve or irregular variations in the relative risk over time that cannot be eliminated would reduce the value of the model. Nevertheless, evidence concerning these limitations either does not exist or is still too preliminary to make the average relative risk method inappropriate for carcinogens evaluated here.

A factor of uncertainty, rather than of methodological limitation, is that data on past exposure are nearly always incomplete. Although it is generally assumed that in the majority of studies the historical dose rate can be determined within an order of magnitude, there are possibly greater uncertainties, even of more than two orders of magnitude, in some studies. In the risk assessment process it is of crucial importance that this degree of uncertainty be clearly stated. This is often done simply by citing upper and lower limits of risk estimates. Duration of exposure and the age- and time-dependence of cancer caused by a particular substance are less uncertain parameters, although the mechanisms of relationship are not so well understood *(11)*.

Risk estimates from animal cancer bioassays

Animal bioassays of chemicals provide important information on the human risk of cancer from exposure to chemicals. These data enhance our confidence in assessing human cancer risks on the basis of epidemiological data.

There is little doubt of the importance of animal bioassay data in reaching an informed decision on the carcinogenic potential of a chemical. The collection and use of data such as those on saturation mechanisms, absorption, distribution and metabolic pathways, as well as on interaction with other chemicals, is important and should be continued. Regrettably, these data were not always available for the air pollutants evaluated during the update and revision of the guidelines. The process of evaluating guidelines and the impact of exposure to these chemicals on human health should continue and be revised as new information becomes available.

Several chemicals considered in this publication have been studied using animal cancer bioassays. The process is continuing and new information on the potential carcinogenicity of chemicals is rapidly appearing. Consequently, the status of chemicals is constantly being reassessed.

There is no clear consensus on appropriate methodology for the risk assessment of chemicals for which the critical effect may not have a threshold, such as genotoxic carcinogens and germ cell mutagens. A number of approaches based largely on characterization of dose–response have been adopted for assessment of such effects:

- quantitative extrapolation by mathematical modelling of the dose–response curve to estimate the risk at likely human intakes or exposures (low-dose risk extrapolation);
- relative ranking of potencies in the experimental range; and
- division of effect levels by an uncertainty factor.

Low-dose risk extrapolation has been accomplished by the use of mathematical models such as the Armitage-Doll multi-stage model. In more recently developed biological models, the different stages in the process of carcinogenesis have been incorporated and time to tumour has been taken into account *(20)*. In some cases, such as that of butadiene, uncertainty regarding the metabolism in humans and experimental animals precluded the choice of the appropriate animal model for low-dose risk extrapolation. In other cases where data permitted, attempts were made to incorporate the dose delivered to the target tissue into the dose–response analysis (physiologically based pharmacokinetic modelling).

During revision of the guidelines, other approaches to establishing guideline levels for carcinogens were considered. Such approaches involve the identification of a level of exposure at which the risk is known to be small and the application of uncertainty factors to derive a level of exposure at which the risk is accepted as being exceedingly small or negligible. This approach has been adopted in the United Kingdom, for example. It was agreed that such an approach might be applicable on a national or smaller scale, but that it was unlikely to be generally applicable.

Interpretation of risk estimates

The risk estimates presented in this book should *not* be regarded as being equivalent to the true cancer risk. It should be noted that crude expression of risk in terms of excess incidence or numbers of cancers per unit of the population at doses or concentrations much less than those on which the estimates are based may be inappropriate, owing to the uncertainties of quantitative extrapolation over several orders of magnitude. Estimated risks are believed to represent only the plausible upper bounds, and may vary widely depending on the assumptions on which they are based.

The presented quantitative risk estimates can provide policy-makers with rough estimates of risk that may serve well as a basis for setting priorities, balancing risks and benefits, and establishing the degree of urgency of public health problems among subpopulations inadvertently exposed to carcinogens. A risk management approach for compounds for which the critical effect is considered not to have a threshold involves eliminating or reducing exposure as far as practically or technologically possible. Characterization of the dose–response, as indicated in the procedures described above, can be used in conjunction with this approach to assess the need to reduce exposure.

Combined exposures

Exposure to combinations of air pollutants is inevitable. Data dealing with the effects of co-exposure to air pollutants are, however, very limited and it is not possible to recommend guidelines for such combinations. Of course, measures taken to control air pollution frequently lead to the reduction in concentrations of more than one pollutant. This is often achieved by controlling sources of pollutants rather than by focusing on individual pollutants.

ECOLOGICAL EFFECTS

The importance of taking an integrated view of both health and ecological effects in air quality management was recognized from the beginning of the project. Ecological effects may have a significant indirect influence on human health and wellbeing. For example, most of the major urban air pollutants are known to have adverse effects at low levels on plants, including food crops. A consultation group was therefore convened to consider the ecological effects on terrestrial vegetation of sulfur dioxide, nitrogen dioxide, and ozone and other photochemical oxidants. These substances are important both because of the high anthropogenic amounts produced and because of their wide distribution. They deserve special attention because of significant adverse effects on ecological systems in concentrations far below those known to be harmful to humans.

The pollutants selected for consideration here form only part of the vast range of air pollutants that have ecological effects. The project timetable permitted only an evaluation of adverse effects on terrestrial plant life, although effects on animal and aquatic ecosystems are also of great concern in parts of Europe. Nevertheless, even this limited evaluation clearly indicates the importance attached to the ecological effects of such pollutants in the European Region.

REFERENCES

1. *Assessing human health risks of chemicals: derivation of guidance values for health-based exposure limits.* Geneva, World Health Organization, 1994 (Environmental Health Criteria, No. 170).
2. CRUMP, K.S. A new method for determining allowable daily intakes. *Fundamental and applied toxicology*, **4**: 854–871 (1984).
3. US ENVIRONMENTAL PROTECTION AGENCY. Guidelines and methodology used in the preparation of health effect assessment chapters of the consent decree water quality criteria. *Federal register,* **45**: 79347–79357 (1980).
4. ALLEN, B.C. ET AL. Dose–response modeling for developmental toxicity. *Toxicologist,* **12**: 300 (1992).
5. ALLEN, B.C. ET AL. Comparison of quantitative dose response modeling approaches for evaluating fetal weight changes in segment II developmental toxicity studies. *Teratology,* **47**(5): 41 (1993).
6. FARLAND, W. & DOURSON, M. Noncancer health endpoints: approaches to quantitative risk assessment. *In*: Cothern, R., ed. *Comparative environmental risk assessment.* Boca Raton, FL, Lewis Publishers, 1992, pp. 87–106.
7. VETTORAZZI, G. *Handbook of international food regulatory toxicology. Vol. 1. Evaluations.* New York, SP Medical and Scientific Books, 1980.
8. *Guidelines for drinking-water quality. Vol. 1. Recommendations.* Geneva, World Health Organization, 1984.
9. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
10. *Air quality criteria and guides for urban air pollutants: report of a WHO Expert Committee*. Geneva, World Health Organization, 1972 (WHO Technical Report Series, No. 506).
11. PEAKALL, D.B. ET AL. Methods for quantitative estimation of risk from exposure to chemicals. *In*: Vouk, V.B. et al., ed. *Methods for estimating risk of chemical injury: human and non-human biota and ecosystems.* New York, John Wiley & Sons, 1985.
12. *Polychlorinated dibenzo*-para-*dioxins and polychlorinated dibenzofurans.* Lyons, International Agency for Research on Cancer, 1997 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 69).

13. ANDERSON, E.L. Quantitative approaches in use in the United States to assess cancer risk. *In*: Vouk, V.B. et al., ed. *Methods for estimating risk of chemical injury: human and non-human biota and ecosystems.* New York, John Wiley & Sons, 1985.
14. *Health assessment document for nickel.* Research Triangle Park, NC, US Environmental Protection Agency, 1985 (Final Report No. EPA-600/8-83-12F).
15. ANDERSON, E.L. ET AL. Quantitative approaches in use to assess cancer risk. *Risk analysis,* **3**: 277–295 (1983).
16. NATIONAL RESEARCH COUNCIL. *Drinking water and health.* Washington, DC, National Academy Press, 1977.
17. *Risk assessment and risk management of toxic substances. A report to the Secretary, Department of Health and Human Services.* Washington, DC, US Department of Health and Human Services, 1985.
18. *Health assessment document for chromium.* Washington, DC, US Environmental Protection Agency, 1984 (Final report EPA-600-8-83-014F).
19. *Health assessment document for inorganic arsenic.* Washington, DC, US Environmental Protection Agency, 1984 (Final report EPA-600-8-83-021F).
20. MOOLGAVKAR, S.H. ET AL. A stochastic two-stage model for cancer risk assessment. I. The hazard function and the probability of tumor. *Risk analysis,* **8**: 383–392 (1988).

Summary of the guidelines

The term "guidelines" in the context of this book implies not only numerical values (guideline values), but also any kind of guidance given. Accordingly, for some substances the guidelines encompass recommendations of a more general nature that will help to reduce human exposure to harmful levels of air pollutants. For some pollutants no guideline values are recommended, but risk estimates are indicated instead.

The numerical guideline values and the risk estimates for carcinogens (Tables 2–4) should be regarded as the shortest possible summary of a complex scientific evaluation process. Nevertheless, the information given in the tables should not be used without reference to the rationale given in the chapters on the respective pollutants. Scientific results are an abstraction of real situations, and this is even more true for numerical values and risk estimates based on such results. Numerical guideline values, therefore, are not to be regarded as separating the acceptable from the unacceptable, but rather as indications. They are proposed in order to help avoid major discrepancies in reaching the goal of effective protection against recognized hazards for human health and the environment. Moreover, numerical

Table 2. Guideline values for individual substances based on effects other than cancer or odour/annoyance

Substance	Time-weighted average	Averaging time
Cadmium	5 ng/m^3 [a]	annual
Carbon disulfide[b]	100 $\mu g/m^3$	24 hours
Carbon monoxide	100 mg/m^3 [c]	15 minutes
	60 mg/m^3 [c]	30 minutes
	30 mg/m^3 [c]	1 hour
	10 mg/m^3	8 hours
1,2-Dichloroethane[b]	0.7 mg/m^3	24 hours
Dichloromethane	3 mg/m^3	24 hours
	0.45 mg/m^3	1 week

Table 2 (contd)

Substance	Time-weighted average	Averaging time
Fluoride[d]	–	–
Formaldehyde	0.1 mg/m^3	30 minutes
Hydrogen sulfide[b]	150 μg/m^3	24 hours
Lead	0.5 μg/m^3	annual
Manganese	0.15 μg/m^3	annual
Mercury	1 μg/m^3	annual
Nitrogen dioxide	200 μg/m^3	1 hour
	40 μg/m^3	annual
Ozone	120 μg/m^3	8 hours
Particulate matter[e]	Dose–response	–
Platinum[f]	–	–
PCBs[g]	–	–
PCDDs/PCDFs[h]	–	–
Styrene	0.26 mg/m^3	1 week
Sulfur dioxide	500 μg/m^3	10 minutes
	125 μg/m^3	24 hours
	50 μg/m^3	annual
Tetrachloroethylene	0.25 mg/m^3	annual
Toluene	0.26 mg/m^3	1 week
Vanadium[b]	1 μg/m^3	24 hours

[a] The guideline value is based on the prevention of a further increase of cadmium in agricultural soils, which is likely to increase the dietary intake.

[b] Not re-evaluated for the second edition of the guidelines.

[c] Exposure at these concentrations should be for no longer than the indicated times and should not be repeated within 8 hours.

[d] Because there is no evidence that atmospheric deposition of fluorides results in significant exposure through other routes than air, it was recognized that levels below 1 μg/m^3, which is needed to protect plants and livestock, will also sufficiently protect human health.

[e] The available information for short- and long-term exposure to PM_{10} and $PM_{2.5}$ does not allow a judgement to be made regarding concentrations below which no effects would be expected. For this reason no guideline values have been recommended, but instead risk estimates have been provided (see Chapter 7, Part 3).

[f] It is unlikely that the general population, exposed to platinum concentrations in ambient air at least three orders of magnitude below occupational levels where effects were seen, may develop similar effects. No specific guideline value has therefore been recommended.

[g] No guideline value has been recommended for PCBs because inhalation constitutes only a small proportion (about 1–2%) of the daily intake from food.

[h] No guideline value has been recommended for PCDDs/PCDFs because inhalation constitutes only a small proportion (generally less than 5%) of the daily intake from food.

guidelines for different substances are not directly comparable. Variations in the quality and extent of the scientific information and in the nature of critical effects, although usually reflected in the applied uncertainty factor, result in guideline values that are only to a limited extent comparable between pollutants.

Owing to the different bases for evaluation, the numerical values for the various air pollutants should be considered in the context of the accompanying scientific documentation giving the derivation and scientific considerations. Any *isolated* interpretation of numerical data should therefore be avoided, and guideline values should be used and interpreted in conjunction with the information contained in the appropriate sections.

It is important to note that the approach taken in the preparation of the guidelines was to evaluate data on the health effects of individual compounds. Consequently, each chemical was considered in isolation. Pollutant mixtures can yield different toxic effects, but data are at present insufficient for guidelines relating to mixtures to be laid down. There is little emphasis on interaction between pollutants that might lead to additive or synergistic effects and on the environmental fate of pollutants, though there is growing evidence about the role of solvents in atmospheric photochemical processes leading to the formation or degradation of ozone, the formation of acid rain, and the propensity of metals and trace elements to accumulate in environmental niches. These factors militate strongly against allowing a rise in ambient pollutant levels. Many uncertainties still remain, particularly regarding the ecological effects of pollutants, and therefore efforts should be continued to maintain air quality at the best possible level.

GUIDELINE VALUES BASED ON NON-CANCER EFFECTS OTHER THAN CANCER

The guideline values for individual substances based on effects other than cancer and annoyance from odour are given in Table 2. The emphasis in the guidelines is placed on exposure, since this is the element that can be controlled to lessen dose and hence lessen the consequent health effect. When general ambient air levels are orders of magnitude lower than the guideline values, present exposures are unlikely to cause concern. Guideline values in those cases are directed only to specific release episodes or specific indoor pollution problems.

As stated earlier, the starting point for the derivation of guideline values was to define the lowest concentration at which adverse effects are observed. On the basis of the body of scientific evidence and judgements of uncertainty factors, numerical guideline values were established to the extent possible. Compliance with the guideline values does not, however, guarantee the absolute exclusion of undesired effects at levels below the guideline values. It means only that guideline values have been established in the light of current knowledge and that uncertainty factors based on the best scientific judgements have been incorporated, though some uncertainty cannot be avoided.

For some of the substances, a direct relationship between concentrations in air and possible toxic effects is very difficult to establish. This is especially true of those pollutants for which a greater body burden results from ingestion than from inhalation. For instance, available data show that for the general population the food chain is the critical route of non-occupational exposure to lead and cadmium, and to persistent organic pollutants such as dioxins and PCBs. On the other hand, emissions of these pollutants into air may contribute significantly to the contamination of food by these compounds. Complications of this kind were taken into consideration, and an attempt was made to develop guidelines that would also prevent those toxic effects of air pollutants that resulted from uptake by both ingestion and inhalation.

For certain compounds, such as organic solvents, the proposed health-related guidelines are orders of magnitude higher than current ambient levels. The fact that existing environmental levels for some substances are much lower than the guideline levels by no means implies that pollutant burdens may be increased up to the guideline values. Any level of air pollution is a matter of concern, and the existence of guideline values never means a licence to pollute.

Unfortunately, the situation with regard to actual environmental levels and proposed guideline values for some substances is just the opposite – guideline values are below existing levels in some parts of Europe. For instance, the guideline values recommended for major urban air pollutants such as nitrogen dioxide, ozone and sulfur dioxide point to the need for a significant reduction of emissions in some areas.

For substances with malodorous properties at concentrations below those where toxic effects occur, guideline values likely to protect the public from odour nuisance were established; these were based on data provided by expert panels and field studies (Table 3). In contrast to other air pollutants, odorous substances in ambient air often cannot be determined easily and

Table 3. Rationale and guideline values based on sensory effects or annoyance reactions, using an averaging time of 30 minutes

Substance	Detection threshold	Recognition threshold	Guideline value
Carbon disulfide[a] (index substance for viscose emissions)	200 μg/m³	–	20 μg/m³
Hydrogen sulfide[a]	0.2–2.0 μg/m³	0.6–6.0 μg/m³	7 μg/m³
Formaldehyde	0.03–0.6 mg/m³	–	0.1 mg/m³
Styrene	70 μg/m³	210–280 μg/m³	70 μg/m³
Tetrachloroethylene	8 mg/m³	24–32 mg/m³	8 mg/m³
Toluene	1 mg/m³	10 mg/m³	1 mg/m³

[a] Not re-evaluated for the second edition of the guidelines.

systematically by analytical methods because the concentrations are usually very low. Furthermore, odours in the ambient air frequently result from a complex mixture of substances and it is difficult to identify individual ones; future work may have to concentrate on odours as perceived by individuals rather than on separate odorous substances.

GUIDELINES BASED ON CARCINOGENIC EFFECTS

In establishing criteria upon which guidelines could be based, it became apparent that carcinogens and noncarcinogens would require different approaches. These approaches are determined by theories of carcinogenesis, which postulate that there is no threshold for effects (that is, that there is no safe level). Risk managers are therefore faced with two choices: either to prohibit a chemical or to regulate it at levels that result in an acceptable degree of risk. Indicative figures for risk and exposure assist the risk manager to reach the latter decision. Air quality guidelines are therefore indicated in terms of incremental unit risks (Table 4) in respect of those carcinogens that are considered to be genotoxic (see Chapter 2). To allow risk managers to judge the acceptability of risks, this edition of the guidelines has provided concentrations of carcinogenic air pollutants associated with an excess lifetime cancer risk of 1 per 10 000, 1 per 100 000 and 1 per 1 000 000.

For butadiene, there is substantial information on its mutagenic and carcinogenic activity. It has been shown that butadiene is mutagenic in both

Table 4. Carcinogenic risk estimates based on human studies[a]

Substance	IARC Group	Unit risk[b]	Site of tumour
Acrylonitrile[c]	2A	2×10^{-5}	lung
Arsenic	1	1.5×10^{-3}	lung
Benzene	1	6×10^{-6}	blood (leukaemia)
Butadiene	2A	–	multisite
Chromium (VI)	1	4×10^{-2}	lung
Nickel compounds	1	4×10^{-4}	lung
Polycyclic aromatic hydrocarbons (BaP)[d]	–	9×10^{-2}	lung
Refractory ceramic fibres	2B	1×10^{-6} (fibre/l)$^{-1}$	lung
Trichloroethylene	2A	4.3×10^{-7}	lung, testis
Vinyl chloride[c]	1	1×10^{-6}	liver and other sites

[a] Calculated with average relative risk model.

[b] Cancer risk estimates for lifetime exposure to a concentration of 1 μg/m³.

[c] Not re-evaluated for the second edition of the guidelines.

[d] Expressed as benzo[*a*]pyrene (based on a benzo[*a*]pyrene concentration of 1 μg/m³ in air as a component of benzene-soluble coke-oven emissions).

bacterial and mammalian systems, but metabolic activation into DNA-reactive metabolites is required for this activity. In general, metabolism of butadiene to epoxides in humans is significantly less than in mice and rats, with mice having the highest metabolic activity. Human cancer risk estimates for butadiene based on bioassays vary considerably depending on the animal species used, with risk estimates based on data in mice being 2–3 orders of magnitude higher than those based on rat data. At present, no definite conclusion can be made as to which animal species is most appropriate for human cancer risk estimates, and thus no guideline value is recommended for butadiene.

Separate consideration is given to risk estimates for asbestos (Table 5) and radon daughters (Table 6) because they refer to different physical units, and the risk estimates are indicated in the form of ranges.

Risk estimation for residential radon exposure has often been based on extrapolation of findings in underground miners. Several circumstances, however, make such estimates uncertain for the general population: exposure to other factors in the mines; differences in age and sex; size distribution of aerosols; the attached fraction of radon progeny; breathing rate; and

Table 5. Risk estimates for asbestos

Concentration	Range of lifetime risk estimates	
500 F*/m^3 (0.0005 F/ml)[a]	10^{-6}–10^{-5}	(lung cancer in a population where 30% are smokers)
	10^{-5}–10^{-4}	(mesothelioma)

[a] F* = fibres measured by optical methods.

Table 6. Risk estimates and recommended action level for radon progeny

Exposure	Lung cancer excess lifetime risk estimate	Recommended level for remedial action in buildings
1 Bq/m^3	3–6 × 10^{-5}	$\geq$ 100 Bq/m^3 (annual average)

route. Furthermore, uncertainties in the exposure–response exist, and possible differences in the relative risk estimates for smokers and non-smokers are not fully understood (see Chapter 8, Part 3).

For radon, a unit risk of approximately 3–6 × 10^{-5} per Bq/m^3 can be calculated assuming a life time risk of lung cancer of 3% (Table 6). This means that a person living in an average European house with 50 Bq/m^3 has a lifetime excess lung cancer risk of 1.5–3 × 10^{-3} Thus current levels of radon in dwellings and other buildings are of public health concern. In addition it should be noted that a lifetime lung cancer risk below about 10^{-4} could normally not be expected to be achievable because natural concentration of radon in ambient air outdoors is about 10 Bq/m^3. Therefore no numerical guideline value for radon is recommended.

It is important to note that quantitative risk estimates may give an impression of accuracy that they do not in fact have. An excess of cancer in a population is a biological effect and not a mathematical function, and uncertainties of risk estimation are caused not only by inadequate exposure data but also, for instance, by the fact that specific metabolic properties of agents are not reflected in the models. The guidelines do not indicate therefore that a specified lifetime risk is virtually safe or acceptable.

The decision on the acceptability of a certain risk should be taken by the national authorities in the context of a broader risk management process. Risk estimate figures should not be applied in isolation when regulatory decisions are being made; combined with data on exposure levels and individuals exposed, they may be a useful contribution to risk assessment. Risk assessment can then be used together with technological, economic and other considerations in the risk management process.

GUIDELINES BASED ON EFFECTS ON VEGETATION

Although the main objective of the air quality guidelines is the direct protection of human health, it was decided that ecological effects of air pollutants on vegetation should also be considered. The effects of air pollutants on the natural environment are of special concern when they occur at concentrations lower than those that damage human health. In such cases, air quality guidelines based only on effects on human health would not allow for environmental damage that might indirectly affect human wellbeing.

Ecologically based guidelines for preventing adverse effects on terrestrial vegetation were included in the first edition of this book, and guidelines were recommended for some gaseous air pollutants. Since that time, however, significant advances in the scientific understanding of the impacts of air pollutants on the environment have been made. For the updating and revision of the guidelines, the ecological effects of major air pollutants were considered in more detail within the framework of the Convention on Long-range Transboundary Air Pollution. This capitalizes on the scientific work undertaken since 1988 to formulate criteria for the assessment of the effects of air pollutants on the natural environment, such as critical levels and critical loads.

It should be understood that the pollutants selected (SO_2, NO_x and ozone/photochemical oxidants) (Table 7) are only a few of a larger category of air

Table 7. Guideline values for individual substances based on effects on terrestrial vegetation

Substance	Guideline value	Averaging time
SO_2: critical level	10–30 µg/m³ [a]	annual
critical load	250–1500 eq/ha/year [b]	annual
NO_x: critical level	30 µg/m³	annual
critical load	5–35 kg N/ha/year [b]	annual
Ozone: critical level	0.2–10 ppm·h [a, c]	5 days–6 months

[a] Depending on the type of vegetation (see Part III).

[b] Depending on the type of soil and ecosystem (see Part III).

[c] AOT: Accumulated exposure Over a Threshold of 40 ppb.

pollutants that may adversely affect the ecosystem, and that the effects considered are only part of the spectrum of ecological effects. Effects on aquatic ecosystems were not evaluated, nor were effects on animals taken into account. Nevertheless, the available information indicates the importance of these pollutants and of their effects on terrestrial vegetation in the European Region.

Use of the guidelines in protecting public health

When strategies to protect public health are under consideration, the air quality guidelines need to be placed in the perspective of total chemical exposure. The interaction of humans and the biosphere is complex. Individuals can be exposed briefly or throughout their lifetime to chemicals in air, water and food; exposures may be environmental and occupational. In addition, individuals vary widely in their response to exposure to chemicals; each person has a pre-existing status (for example, age, sex, pregnancy, pulmonary disease, cardiovascular disease, genetic make-up) and a lifestyle, in which such factors as exercise and nutrition play key roles. All these different elements may influence a person's susceptibility to chemicals. Various sensitivities also exist within the plant kingdom and need to be considered in protecting the environment.

The primary aim of these guidelines is to provide a uniform basis for the protection of public health and of ecosystems from adverse effects of air pollution, and to eliminate or reduce to a minimum exposure to those pollutants that are known or are likely to be hazardous. The guidelines are based on the scientific knowledge available at the time of their development. They have the character of recommendations, and it is not intended or recommended that they simply be adopted as standards. Nevertheless, countries may wish to transform the recommended guidelines into legally enforceable standards, and this chapter discusses ways in which this may be done. It is based on the report of a WHO working group (*1*). The discussion is limited to ambient air and does not include the setting of emission standards.

In the process of moving from a "guideline" or a "guideline value" to a "standard", a number of factors beyond the exposure–response relationship need to be taken into account. These factors include current concentrations of pollutants and exposure levels of a population, the specific mixture of air pollutants, and the specific social, economic and cultural conditions encountered. In addition, the standard-setting procedure may be influenced by the likelihood of implementing the standard. These considerations may lead to a standard above or below the respective guideline value.

DEFINITIONS

Several terms are in use to describe the tools available to manage ambient air pollution. To avoid confusion, definitions are needed for the terms used here – guideline, guideline value and standard – within this specific context.

Guideline

A guideline is defined as any kind of recommendation or guidance on the protection of human beings or receptors in the environment from adverse effects of air pollutants. As such, a guideline is not restricted to a numerical value but might also be expressed in a different way, for example as exposure–response information or as a unit risk estimate.

Guideline value

A guideline value is a particular form of guideline. It has a numerical value expressed either as a concentration in ambient air or as a deposition level, which is linked to an averaging time. In the case of human health, the guideline value provides a concentration below which no adverse effects or (in the case of odorous compounds), no nuisance or indirect health significance are expected, although it does not guarantee the absolute exclusion of effects at concentrations below the given value.

Standard

A standard is considered to be the level of an air pollutant, such as a concentration or a deposition level, that is adopted by a regulatory authority as enforceable. Unlike the case of a guideline value, a number of elements in addition to the effect-based level and the averaging time have to be specified in the formulation of a standard. These elements include:

- the measurement strategy
- the data handling procedures
- the statistics used to derive the value to be compared with the standard.

The numerical value of a standard may also include the permitted number of exceedings.

MOVING FROM GUIDELINES TO STANDARDS

The regulatory approach to controlling air pollution differs from country to country. Different countries have different political, regulatory and administrative approaches, and legislative and executive activities can be carried out at various levels such as national, regional and local. Fully effective air quality management requires a framework that guarantees a consistent derivation of air quality standards and provides a transparent basis for decisions with regard to risk-reducing measures and abatement strategies. In establishing such a framework, several issues should be considered, such as legal aspects, the protection of specific populations at risk, the role of stakeholders in the process, cost–benefit analysis, and control and enforcement measures.

Legal aspects

In setting air quality standards at the national or supranational level, a legislative framework usually provides the basis for the evaluation and decision-making process. The setting of standards strongly depends on the type of risk management strategy adopted. Such a strategy is influenced by country-specific sociopolitical considerations and/or supranational agreements.

Legislation and the format of air quality standards vary from country to country, but in general the following issues should be considered:

- identification and selection of pollutants to which the legislative instrument will apply;
- the process for making decisions about the appropriate standards;
- the numerical value of the standards for the various pollutants, applicable detection methods and monitoring methodology;
- actions to be taken to implement the standard, such as the definition of the time frame needed/allowed for achieving compliance with the standard, considering emission control measures and necessary abatement strategies; and
- identification of responsible enforcement authorities.

Depending on their position within a legislative framework, standards may or may not be legally binding. In some countries the national constitution contains provisions for the protection of public health and the environment. In general, the development of a legal framework on the basis of constitutional provisions comprises two regulatory actions. The first is the enactment of a formal legal instrument, such as an act, a law, an ordinance or a decree, and the second is the development of regulations, by-laws, rules and orders.

Air quality standards may be based solely on scientific and technical data on public health and environmental effects, but other aspects such as cost–benefit or cost–effectiveness may be also taken into consideration. In practice, there are generally several opportunities within a legal framework to address these economic aspects as well as other issues, such as technical feasibility, structural measures and sociopolitical considerations. These can be taken into account during the standard-setting procedure or at the level of designing appropriate measures to control emissions. This rather complicated process might result in several standards being set, such as an effect-oriented standard as a long-term goal and less stringent interim standards to be achieved within shorter periods of time.

Standards also depend on political choices as to which receptors in the environment should be protected and to what extent. Some countries have separate standards for the protection of public health and the environment. Moreover, the stringency of a standard can be influenced by provisions designed to take account of higher sensitivities of specific receptor groups, such as young children, sick or elderly people, or pregnant women. It might also be important to specify whether effects are considered for individual pollutants or for a combined exposure to several pollutants.

Air quality standards can set the reference point for emission control and abatement strategies on a national level. It should be recognized, however, that exposure to some pollutants is the result of long-range transboundary transport. In these cases adequate protection measures can only be achieved by appropriate international agreements.

Air quality standards should be regularly reviewed, and need to be revised as new scientific evidence on effects on public health and the environment emerges.

Standards often strongly influence the implementation of an air pollution control policy. In many countries, the exceeding of standards is linked to an obligation to develop action plans at the local, regional or national level to reduce air pollution levels. Such plans often address several pollution sources. Standards also play a role in environmental impact assessment procedures and in the provision of public information on the state of the environment. Provisions for such activities can be found in many national legal instruments.

Within national or supranational legislative procedures, the role of stakeholders in the process of standard-setting also needs to be considered. This is dealt with in more detail below.

Items to be considered in setting standards

Within established legal frameworks and using air quality guidelines as a starting point, development of standards involves consideration of a number of issues. These are in part determined by characteristics of populations or physical properties of the environment. A number of these issues are discussed below.

Adverse health effects

In setting a standard for the control of an environmental pollutant, the effects that the population is to be protected against need to be defined. A hierarchy of effects on health can be identified, ranging from acute illness and death through chronic and lingering diseases and minor and temporary ailments, to temporary physiological or psychological changes. The distinction between adverse and non-adverse effects poses considerable difficulties. Of course, more serious effects are generally accepted as adverse. As one considers effects that are either temporary and reversible, or involve biochemical or functional changes whose clinical significance is uncertain, judgements must be made as to which of these less serious effects should be considered adverse. With any definition of adversity, a significant degree of subjectivity and uncertainty remains. Judgements as to adversity may differ between countries because of factors such as different cultural backgrounds and different background levels of health status.

In some cases, the use of biomarkers or other indicators of exposure may provide a basis for standard-setting. Changes in such indicators, while not necessarily being adverse in themselves, may be predictors of significant effects on health. For example, the blood lead concentration can provide information on the likelihood of impairment of neurobehavioural development.

Special populations at risk

Sensitive populations or groups are defined here as those impaired by concurrent disease or other physiological limitations, and those with specific characteristics that make the health consequences of exposure more significant (such as the developmental phase in children or reduction in reserve capacity in the elderly). In addition, other groups may be judged to be at special risk because of their exposure patterns or due to an increased effective dose for a given exposure. Sensitive populations may vary from country to country owing to differences in the number of people lacking access to adequate medical care, in the existence of endemic disease, in the prevailing genetic factors, or in the prevalence of debilitating diseases, nutritional deficiencies or lifestyle factors. It is up to the politician to decide which specific groups at risk should be protected by the standards (and thus which should not be protected).

Exposure–response relationships

A key factor to be considered in developing standards is information about the exposure–response relationship for the pollutant concerned. For a number of pollutants an attempt has been made to provide exposure–response relationships in the revised version of the guidelines. For particulate matter and ozone, detailed tables specifying the exposure–response relationship are provided. The information included in these tables is derived from epidemiological studies of the effects of these pollutants on health. Such information is available for only a few of the pollutants considered in the guidelines. For known "no-threshold compounds" such as the carcinogen benzene, quantitative risk assessment methods provide estimates of risk at different exposure concentrations.

When developing standards, regulators should consider the degree of uncertainty about exposure–response relationships provided in the guidelines. Differences in the population structure, climate and geography that can have an impact on the prevalence, frequency and severity of effects may modify the exposure–response relationships provided in the guidelines.

Exposure characterization

An important factor to be considered in developing standards is that of how many people are exposed to concentrations of concern and the distribution of exposure among various population groups. Current distributions of exposure should be considered, together with those that are likely to occur should the standard be met. Besides using monitoring data, results of exposure modelling can be used at this stage. The origins of pollutants, including long-range transport and its contribution to ambient levels, should also be evaluated.

The extent to which ambient air quality estimates from monitoring networks or models correspond to personal exposure in the population is also a factor to be considered in the standard-setting. This will depend on the pollutant in question (for example, personal exposure to carbon monoxide is poorly characterized by fixed-site monitors) as well as on a number of local characteristics, including lifestyle, climatic conditions, spatial distribution of pollution sources and local determinants of pollution dispersion.

Other important exposure-related concerns include:

- how much of total human exposure is due to ambient, outdoor sources as opposed to indoor sources; and

- where multiple routes of exposure are important, how to apportion the regulatory burden among the different routes of exposure (such as lead from air sources versus lead from paint, water pipes, etc.).

These factors may vary substantially across countries. For example, indoor air pollution levels might be quite substantial in countries in which fossil and/or biomass fuels are used in homes.

Risk assessment

In general, the central question in developing air quality standards to protect public health or ecosystems is the degree of protection associated with different pollution levels at which standards might be established. In the framework of quantitative risk assessment, various proposals for standards can be considered in health or ecological risk models. These models provide a tool that is increasingly used to inform decision-makers about some of the possible consequences associated with various options for standards, or the reduction in adverse effects associated with moving from the current situation to a particular standard.

The first two steps in risk assessment, namely hazard identification and, in some cases, development of exposure–response relationships, have been provided in these guidelines and are discussed in greater detail in later chapters. The third step, exposure analysis, predicts changes in exposure associated with reductions in emissions from a specific source or groups of sources under different control scenarios. Instead of exposure estimates, ambient concentrations (based on monitoring or modelling) are often used as the inputs to a risk assessment. This is in part because of the availability of information on concentration–response relationships from epidemiological studies in which fixed-site monitors were used.

The final step in a regulatory risk assessment is the risk characterization stage, whereby exposure estimates are combined with exposure–response relationships to generate quantitative estimates of risk (such as how many individuals may be affected). Regulatory risk assessments are likely to result in different risk estimates across countries, owing to differences in exposure patterns and in the size and characteristics of sensitive populations and those at special risk.

It is important to recognize that there are many uncertainties at each stage of a regulatory risk assessment. The results of sensitivity and uncertainty analyses should be presented so as to characterize the impact of major uncertainties on the risk estimates. In addition, the methods used to conduct the risk assessments should be clearly described and the limitations and caveats associated with the analysis should be discussed.

Acceptability of risk

The role of a regulatory risk assessment in developing standards may differ from country to country, owing to differences in the legal framework and availability of information. Also, the degree of acceptability of risk may vary between countries because of differences in social norms, degree of adversity and risk perception among the general population and various stakeholders. How the risks associated with air pollution compare with those from other pollution sources or human activities may also influence risk acceptability.

In the absence of clearly identified thresholds for health effects for some pollutants, the selection of a standard that provides adequate protection of public health requires an exercise of informed judgement by the regulator. The acceptability of the risks and, therefore, the standard selected will depend on the effect, on the expected incidence and severity of the potential effects, on the size of the population at risk, and on the degree of scientific certainty that the effects will occur at any given level of pollution. For example, if a suspected health effect is severe and the size of the population at risk is large, a more cautious approach would be appropriate than if the effect were less troubling or if the exposed population were small.

Cost–benefit analysis

Two comprehensive techniques provide a framework for comparing monetarized costs and benefits of implementing legislation or policy: cost–effectiveness analysis and cost–benefit analysis. These two techniques differ in their treatment of benefits. In cost–benefit analysis, costs and benefits (for example, avoided harm, injury or damage) of implemented control measures are compared using monetary values. In cost–effectiveness analysis, the costs of control measures are reported in quantitative terms, such as cost per ton of pollutant or cost per exposure unit. That is, the benefits are described in their own physical, chemical or biological terms, such as reduced concentrations or emissions, or avoided cases of illness, crop losses or damage to ecosystems.

Analysis of control measures to reduce ambient pollutant levels

Control measures to reduce emissions of many air pollutants are known. Direct control measures at the source are readily expressed in monetary terms. Indirect control measures, such as alternative traffic plans or changes in public behaviour, may not all be measurable in monetary terms but their impact should be understood. Effective control measures should be designed to deal with secondary as well as primary pollutants.

Cost identification should include costs of investment, operation and maintenance, both for the present and for the future. Unforeseen effects, technical innovations and developments, and indirect costs arising during implementation of the regulation are additional complicating factors. Cost estimates derived in one geographical area may not be generally transferable to other areas.

Air quality assessment has to provide information about expected air quality, both with and without implementation of control measures. Typically, the assessment will be based on a combination of air quality monitoring data and dispersion modelling. These two assessment methods are complementary, and must be seen as equally important inputs to the assessment process.

For the assessment, several types of data have to be acquired:

- measured concentrations for relevant averaging times (hourly, daily, seasonal) with information on site classification;
- emission data from all significant sources, including emission conditions (such as stack height) and with sufficient information on spatial and temporal variation; and
- meteorological and topographical data relevant to dispersion of the emissions.

Defining the scope and quantifying the benefits

The air quality guidelines are based on health and ecosystem endpoints determined by consensus. This does not imply that other effects on health and the ecosystem that were not considered in the guidelines may not occur or are unimportant. After assessing the local situation, other health- and ecosystem-related benefit categories should be considered in the analysis.

It is a difficult and comprehensive task to quantify the benefit categories included in a cost–benefit analysis. Some indicators of morbidity, such as the use of medication, the number of hospital admissions or work days lost, can be quantified. Other effects, such as premature death or excess mortality, present more difficult problems. Wellbeing, the quality of life or the value of ecosystems may be very difficult to express in monetary terms. In different countries, values assigned to benefit categories might differ substantially owing to different cultural attitudes. Despite these uncertainties, it is better to include as many of the relevant benefit categories as possible, even if the economic assessment is uncertain or ambiguous. A clear understanding of the way in which the economic assessment has been undertaken is important and should be reported.

Comparison of benefits with and without control actions

This step involves combining the information on exposure–response relationships with that on air quality assessment, and applying the combined information to the population at risk. Additional data needed in this step include specification of the population at risk, and determination of the prevalence of the different health effects in the population at risk.

Comparison of costs and benefits

Monetary valuation of control actions and of health and environmental effects may be different in concept and vary substantially from country to country. In addition to variations in assessing costs, the relative value of benefit categories, such as benefits to health or building materials, will vary. Thus, the result of comparing costs and benefits in two areas with otherwise similar conditions may differ significantly.

The measures taken to reduce one pollutant may increase or decrease the concentration of other pollutants. These additional effects should be considered, even if they result from exposure to pollutants not under consideration in the primary analysis. Pollutant interactions pose additional complications. Interaction effects may possibly lead to double counting of costs, or to disregarding some costly but necessary action. The same argumentation can be used when estimating benefits.

Sensitivity and uncertainty analysis

Sensitivity analysis includes comparisons of the results of a particular cost–benefit analysis with that of other studies, recalculation of the whole chain of the analysis using other assumptions, or the use of ranges of values. Specifically, a range of values may be used, such as for value of statistical life. Knowledge of the costs of control measures tends to be better developed than that of the benefits to health and ecosystems, and thus costs tend to be more accurately estimated than benefits. In addition, costs tend to be overestimated and benefits underestimated. One important reason for underestimating the benefits is not considering some important benefit categories because of lack of information. Another reason is the variability of the databases available for monetary assessment of benefits.

Many uncertainties are connected with the steps of cost–benefit and cost–effectiveness analysis, such as exposure, exposure–response, control cost estimates and benefits valuation. The results of sensitivity and uncertainty analyses should be presented so as to characterize the impact of major uncertainties on the result of the analysis. In addition, the methods used to conduct the analysis should be clearly described, and the limitations and caveats associated with the analysis should be discussed. Transparency of the analysis is most important.

Involvement of stakeholders and public awareness

The development of standards should encompass a process involving stakeholders that ensures, as much as possible, social equity or fairness to all involved parties. It should also provide sufficient information to guarantee understanding by stakeholders of the scientific and economic consequences. A review by stakeholders of the standard-setting process, initiated at an early stage, is helpful. Transparency in the process of moving from air quality guidelines to standards helps the public to accept necessary measures.

The participation of all those affected by the procedure of standard-setting – industry, local authorities, nongovernmental organizations and the general public – at an early stage of standard derivation is strongly recommended. If these parties are involved in the process at an early stage their cooperation is more likely to be elicited.

Raising public awareness of the health and environmental effects of air pollution is also an important means to obtain public support for necessary control actions, such as with respect to vehicle emissions. Information about the quality of air (such as warnings of air pollution episodes) and the entailed risks (risk communication) should be published in the media to keep the public informed.

IMPLEMENTATION

The main objectives of the implementation of air quality standards are: (*a*) to define the measures needed to achieve the standards; and (*b*) to establish a suitable regulatory strategy and legislative instrument to achieve this goal. Long- as well as medium-term goals are likely to be needed.

The implementation process should ensure a mechanism for regular assessment of air quality, set up the abatement strategies, and establish the enforcement regulations. Also, the impact of control actions should be assessed, both for public health and environmental effects, for example by the use of epidemiological studies and integrated ecosystems monitoring. Epidemiological studies of the effects of air pollutants on health should be repeated as control measures are implemented. Changes to the mixture of air pollutants and in the composition of complex pollutants such as particulate matter may occur, and changes in exposure–response relationships should be expected.

Assessment of air quality

Air quality assessment has an important role to play within the implementation of an air quality management strategy. The goals of air quality assessment are to provide the air quality management process with relevant data through a proper characterization of the air pollution situation, using monitoring and/or modelling programs and projection of future air quality associated with alternative strategies. Dispersion models can be used very effectively in the design of the definitive monitoring network.

Monitoring methods

The monitoring method (automatic, semi-automatic or manual) adopted for each pollutant should be a standard or reference method, or be validated against such methods. The full description of the method should include information on the sampling and analytical method, on the quality assurance and quality control (internal and external) procedures and on the methods of data management, including data treatment, statistical handling of the data and data validation procedures.

Quality assurance/quality control procedures are an essential part of the measurement system, the aim being to reduce and minimize errors in both the instruments and management of the networks. These procedures should ensure that air quality measurements are consistent (and can be used to give a reliable assessment of ambient air quality) and harmonized over a scale as large as possible, especially in the area of the implementation of the standard.

Design of the monitoring network

An air quality monitoring network can consist of fixed and/or mobile monitoring stations. Although such a network is a fundamental tool for any air quality assessment, its limitations should be borne in mind.

In designing a monitoring network, a primary requirement is to have information about emissions from the dominant and/or most important sources of pollutants. Second, a pilot (or screening) study is needed to gain a good understanding of the geographical distribution of pollutants and to identify the areas with the highest concentrations. Such a screening study can be performed using dispersion models, with the emission inventory as input, combined with a monitoring study using inexpensive passive samplers in a rather dense network.

The selection strategy for site locations generally varies for different pollutants. The number and distribution of sampling sites required in any network depend on the area to be covered, the spatial variability of the emissions being measured, and the purpose for which the data should be used. Meteorological and topographical conditions as well as the density, type and strength of sources (mobile and stationary) must be considered.

Different types of monitoring station are likely to be needed to provide data at a regional or local level. In monitoring rural and urban areas, specific attention should be paid to sites affected by defined sources such as traffic and other "hot-spots". The representativeness of each site should be defined and assessed. Micro-scale conditions, including the buildings around the stations (street canyons), traffic intensity, the height of the sampling point, distances to obstacles, and the effects of the local sources must be kept in mind.

Air quality modelling

Air quality models are used to establish a relationship between emissions and air quality in a given area, such as a city or region. On the basis of emission data, of atmospheric chemistry, and of meteorological, topographical and geographical parameters, modelling gives an opportunity (*a*) to calculate the projected concentration or deposition of the pollutants in regions, and (*b*) to predict the air pollution level in those areas where air sampling is not performed. Measured concentrations should be used for evaluating and validating models, or even as input data. These measurements improve the accuracy of the concentrations calculated by models by allowing refinement and development of the modelling strategies adopted.

Abatement strategies

Abatement strategies are the set of measures to be taken to reduce pollutant emissions and therefore to improve air quality. Authorities should consider the measures necessary in order to meet the standards. An important factor in selecting abatement strategies is deciding the geographical scale of the area(s) that are considered not to meet the standard(s) and the geographical scale of the area for which control should be applied. In defining the geographical scale for abatement strategies, the extent of the transport of pollution from neighbouring areas should be considered. This may involve action at supranational, national, regional or local levels.

Supranational, national, regional and local actions form a hierarchy of approaches. Action at the supranational or national level is likely to be most effective in reducing background levels of air pollution. Local air quality management measures may be needed to address specific local problems, and such measures may need to be implemented urgently to deal with special pollution problems. National and supranational plans should specify the extent of the reduction in levels of air pollution that is required and the time-scale for achieving that reduction.

In addition to the comprehensive programme of emission control designed to reduce average pollution levels and the risk of high pollution episodes, short-term actions may be required for the period when the pollution episodes may occur. Such actions, however, should be considered to be applicable in a transitional period only or as a contingency plan. The objective of measures applied on a larger scale is to minimize the occurrence of local air pollution episodes. A link between control of emissions and ambient air quality is required and may need to be demonstrated. Emission-based air quality standards represent one possible step in this process.

Enforcement

The government of each country establishes the responsibilities for implementing air quality standards. Responsibilities for overseeing different aspects of compliance can be distributed among national, regional and local governments depending on the level at which it is necessary to take action.

Success in the enforcement of standards is influenced by the technology applied and the availability of financial resources to industry and government. Compliance with standards may be ensured by various approaches such as administrative penalties or economic incentives. Sufficient staff and other resources are needed to implement the policy actions effectively.

Periodic reports on compliance and trends in pollutant emissions and concentrations should be developed and disseminated to the public. These reports should also predict trends. It is important that the public be aware of the importance of meteorological factors in controlling pollution levels, as these may produce episodes of pollution that are not within the control of the regulatory authorities.

REFERENCE

1. *Guidance for setting air quality standards. Report on a WHO Working Group*. Copenhagen, WHO Regional Office for Europe, 1998 (document EUR/ICP/EHPM 02 01 02).

PART II

EVALUATION OF RISKS TO HUMAN HEALTH

Organic pollutants

5.1 Acrylonitrile

Exposure evaluation

On the basis of large-scale calculations using dispersion models, the average annual ambient air concentration of acrylonitrile in the Netherlands was estimated to be about 0.01 μg/m^3 *(1),* which is below the present detection limit of 0.3 μg/m^3 *(1, 2).* Production figures *(1)* indicate that, in 8 out of 10 European countries for which data are available, ambient concentrations of acrylonitrile are lower or markedly lower than this. Near industrial sites, air concentrations can exceed 100 μg/m^3 over a 24 hour period, but are usually less than 10 μg/m^3 at a distance of about 1 km. Acrylonitrile concentrations in the air at the workplace have exceeded 100 mg/m^3, but shift averages are usually in the range of 1–10 mg/m^3. Exposure from smoking is possible if acrylonitrile is used for tobacco fumigation, and could amount to 20–40 μg daily for an average smoker.

A more sensitive method of determination, with a detection limit below 0.1 μg/m^3, is required in order to examine concentrations in the ambient air and to allow populations at possible risk to be identified.

Health risk evaluation

Acute and noncancer chronic toxicity may occur at concentrations still reported in some industries. Subjective complaints were reported in acute exposure to 35 mg/m^3, and in chronic exposure to 11 mg/m^3, 4.2–7.2 mg/m^3 or 0.6–6 mg/m^3. Teratogenic effects in animals were observed at 174 mg/m^3 and carcinogenicity was shown in rats exposed for 2 years to 44 mg/m^3.

Twelve epidemiological studies investigating the relationship between acrylonitrile exposure and cancer are available; only five indicate a carcinogenic risk from exposure to acrylonitrile *(1).* Negative studies suffered from small cohort size, insufficient characterization of exposure, short follow-up times and relatively young cohorts. Although four of the remaining five epidemiological studies indicate a higher risk of lung cancer, and one study showed a higher mortality rate for liver, gall bladder and cystic duct cancer, all have problems with regard to methodology, definition and/or size of the population, existence of exposure to other carcinogens, and duration of the follow-up period.

In laboratory animals an increased incidence of tumours of the central nervous system, Zymbal gland, stomach, tongue, small intestine and mammary glands was observed at all doses tested *(3).* There is nevertheless a clear difference between animal and human studies concerning the tumorigenic response to acrylonitrile: no lung tumours have been produced in animals and no brain tumours have been observed in humans.

Acrylonitrile was placed in IARC Group 2A *(3)* on the basis of sufficient evidence of its carcinogenicity in experimental animals and limited evidence of its carcinogenicity in humans.

The epidemiological study by O'Berg *(4)* presents the clearest available evidence of acrylonitrile as a human lung carcinogen. Furthermore, in this study there were no confounding exposures to other carcinogenic chemicals during exposure to acrylonitrile. It was therefore used to make an estimate of the incremental unit risk. As this study has now been updated to the end of 1983 for cancer incidence and to the end of 1981 for overall mortality, the most recent data are used here *(5).* Of 1345 workers exposed to acrylonitrile, a total of 43 cases of cancer occurred versus 37.1 expected. Ten cases of lung cancer were observed versus 7.2 expected, based on the company rates. Lung cancer, which had been the focus of the previous report *(4),* remained in excess but not as high as before; 2 new cases occurred after 1976, with 2.8 expected. This means that the relative risk (RR) would be 10/7.2 = 1.4, significantly lower than in the previous report. On the assumption made by the US Environmental Protection Agency *(6)* for the first O'Berg study *(4)* that the 8-hour time-weighted average exposure was 33 mg/m^3 (15 ppm), and with an estimated work duration of 9 years, the average lifetime daily exposure (X) is estimated to be 930 μg/m^3 ($X = 33\ mg/m^3 \times 8/24 \times 240/365 \times 9/70$).

Using the average relative risk model, the lifetime unit risk (UR) for exposure to 1 μg/m^3 can be calculated to be 1.7×10^{-5} [$UR = P_o(RR - 1)/X = 0.04(1.4 - 1)/930$].

Using animal data, an upper-bound risk of cancer associated with a lifetime inhalation exposure to acrylonitrile was calculated from a rat inhalation study *(7)* to be 1.5×10^{-5} *(6).*

The calculated unit risk based on the human study is consistent with that of the animal study, although the human estimate is uncertain, particularly because of the lack of documentation on exposure.

Guidelines

Because acrylonitrile is carcinogenic in animals and there is limited evidence of its carcinogenicity in humans, it is treated as if it were a human carcinogen. No safe level can therefore be recommended. At an air concentration of 1 μg/m^3, the lifetime risk is estimated to be 2×10^{-5}.

References

1. *Criteriadocument over acrylonitril* [Acrylonitrile criteria document]. The Hague, Ministry of Housing, Spatial Planning and Environment, 1984 (Publikatiereeks Lucht, No. 29).
2. GOING, J.E. ET AL. *Environmental monitoring near industrial sites: acrylonitrile.* Washington, DC, US Environmental Protection Agency, 1979 (Report No. EPA-560/6-79-003).
3. *Chemicals, industrial processes and industries associated with cancer in humans. IARC Monographs, Volumes 1 to 29.* Lyons, International Agency for Research on Cancer, 1982 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Supplement 4), pp. 25–27.
4. O'BERG, M.T. Epidemiologic study of workers exposed to acrylonitrile. *Journal of occupational medicine,* **22**: 245–252 (1980).
5. O'BERG, M.T. ET AL. Epidemiologic study of workers exposed to acrylonitrile: an update. *Journal of occupational medicine,* **27**: 835–840 (1985).
6. *Health assessment document for acrylonitrile.* Washington, DC, US Environmental Protection Agency, 1983 (Final report EPA-600/8-82-007F).
7. QUAST, J.F. ET AL. *A two-year toxicity and oncogenicity study with acrylonitrile following inhalation exposure of rats. Final report.* Midland, MI, Dow Toxicology Research Laboratory, 1980.

5.2 Benzene

Exposure evaluation

Sources of benzene in ambient air include cigarette smoke, combustion and evaporation of benzene-containing petrol (up to 5% benzene), petrochemical industries, and combustion processes.

Mean ambient air concentrations of benzene in rural and urban areas are about 1 μg/m^3 and 5–20 μg/m^3, respectively. Indoor and outdoor air levels are higher near such sources of benzene emission as filling stations.

Inhalation is the dominant pathway for benzene exposure in humans. Smoking is a large source of personal exposure, while high short-term exposures can occur during refuelling of motor vehicles. Extended travel in motor vehicles with elevated air benzene levels (from combustion and evaporative emissions) produces exposures reported from various countries that are second only to smoking as contributors to the intensity of overall exposure. The contribution of this source to cumulative ambient benzene exposure and associated cancer risk comprises about 30% when the travel time is one hour, a duration not untypical for urban and suburban commuting by the general population.

Health risk evaluation

The most significant adverse effects from prolonged exposure to benzene are haematotoxicity, genotoxicity and carcinogenicity.

Chronic benzene exposure can result in bone marrow depression expressed as leukopenia, anaemia and/or thrombocytopenia, leading to pancytopenia and aplastic anaemia. Decreases in haematological cell counts and in bone marrow cellularity have been demonstrated in mice after inhalation of concentrations as low as 32 mg/m^3 for 25 weeks. Rats are less sensitive than mice. In humans, haematological effects of varying severity have occurred in workers occupationally exposed to high levels of benzene. Decreased red and white blood cell counts have been reported above median levels of approximately 120 mg/m^3, but not at 0.03–4.5 mg/m^3. Below 32 mg/m^3, there is only weak evidence of effects.

The genotoxicity of benzene has been extensively studied. Benzene does not induce gene mutations in *in vitro* systems, but several studies have demonstrated induction of both numerical and structural chromosomal aberrations, sister chromatid exchanges and micronuclei in experimental animals and humans after *in vivo* benzene exposure. Some studies in humans have demonstrated chromosomal effects at mean workplace exposures as low as 4–7 mg/m^3. The *in vivo* data indicate that benzene is mutagenic.

The carcinogenicity of benzene has been established both in humans and in laboratory animals. An increased mortality from leukaemia has been demonstrated in workers occupationally exposed. Several types of tumour, primarily of epithelial origin, have been induced in mice and rats after oral exposure and inhalation exposure at 320–960 mg/m^3; these include tumours in the Zymbal gland, liver, mammary gland and nasal cavity. Lymphomas/leukaemias have also been observed, but with lower frequency. The results indicate that benzene is a multisite carcinogen.

Because benzene is characterized as a genotoxic carcinogen and recent data gathered in humans and mice suggest mutagenic potential *in vivo*, establishment of exposure duration and concentration in the human exposure studies is of major importance for the calculation of cancer risk estimates. The Pliofilm cohort is the most thoroughly studied. It was noted that significant exposures to other substances at the studied facilities were probably not a complicating factor, but that exposure estimates for this cohort vary considerably. Three different exposure matrices have been used to describe the Pliofilm cohort, i.e. those reported by Crump & Allen *(1)*, by Rinsky et al. *(2)*, and a newer and more extensive one by Paustenbach et al. *(3)*. The main difference between the first two is that the exposure estimates by Crump & Allen are greater for the early years, during the 1940s. Paustenbach et al. have, among other things, considered short-term, high-level exposure, background concentrations and absorption through the skin, which leads to exposure levels 3–5 times higher than those calculated by Rinsky et al. Compared to the Crump & Allen estimates, Paustenbach et al. arrived at higher exposure estimates for some job classifications, and lower ones for some others.

Within the most recently updated Pliofilm cohort, Paxton et al. *(4, 5)* conducted an extended regression analysis with exposure description for the 15 leukaemia cases and 650 controls. They used all three exposure matrices, which gave estimates of 0.26–1.3 excess cancer cases among 1000 workers at a benzene exposure of 3.2 mg/m^3 (1 ppm) for 40 years (Table 8).

Crump *(7)* calculated unit risk estimates for benzene using the most recently updated data for the Pliofilm cohort and a variety of models

Table 8. Published leukaemia risk estimates for the Plioform cohort at two benzene exposure levels

Cases per 1000 workers exposed to:			
3.2 mg/m^3 (1 ppm)	**0.32 mg/m^3 (0.1 ppm)**	**Exposure matrix**	**Reference**
5.3	–	Rinsky et al. *(2)*	Brett et al. *(6)*
0.5–1.6	–	Rinsky et al. *(2)*	
		Crump & Allen *(1)*	Brett et al. *(6)*
1.3	0.12	Rinsky et al. *(2)*	Paxton et al. *(4, 5)*
0.26	0.026	Crump & Allen *(1)*	Paxton et al. *(4, 5)*
0.49	0.048	Paustenbach et al. *(3)*	Paxton et al. *(4, 5)*

(Table 9). Multiplicative risk models were found to describe the cohort data better than additive risk models and cumulative exposure better than weighted exposures. Dose–responses were essentially linear when the Crump & Allen exposure matrix was used but, according to the author, there was evidence of concentration-dependent nonlinearity in dose–responses derived using the Paustenbach et al. exposure matrix. In that case, the best-fitting model was quadratic.

As can be seen in Table 9, the concentration-dependent model gives a much lower risk estimate than the other models when the Paustenbach et al. exposure matrix is used. In such a model, the concentration of benzene is raised to the second power and thus given greater weight than the duration of exposure. Although there are biological arguments to support the use of a concentration-dependent model, many of the essential data are preliminary and need to be further developed and peer reviewed.

Models giving equal weight to concentration and duration of exposure have been preferred here for the derivation of a risk estimate. Using multiplicative risk estimates and a cumulative exposure model, Crump *(7)* calculated a unit risk for lifetime exposure of $1.4–1.5 \times 10^{-5}$ per ppb with the Paustenbach et al. exposure matrix, and of 2.4×10^{-5} per ppb with the Crump & Allen exposure matrix. If expressed in μg/m^3, the unit risk would thus range from 4.4×10^{-6} to 7.5×10^{-6}. With an additive model instead of a multiplicative model, the risk estimate would have been somewhat smaller. If similar linear extrapolations were done on the occupational cancer risk

Table 9. Model-dependent worker risk and lifetime unit risk estimates for exposure to benzene for the Plioform cohort by Crump *(7)* [a]

Risk estimate	Linear	Nonlinear	Intensity dependent	Exposure reference
Cases per 1000 workers exposed to 3.2 mg/m^3 (1 ppm)	5.1	5.0	5.1	Crump & Allen *(1)*
	3.8	2.9	0.036	Paustenbach et al. *(3)*
Unit risk per ppb	2.4×10^{-5}	2.4×10^{-5}	2.4×10^{-5}	Crump & Allen *(1)*
	1.5×10^{-5}	1.4×10^{-5}	1.7×10^{-10}	Paustenbach et al. *(3)*
Unit risk per µg/m^3 [b]	7.5×10^{-6}	7.5×10^{-6}	7.5×10^{-6}	Crump & Allen *(1)*
	4.7×10^{-6}	4.4×10^{-6}	5.3×10^{-11}	Paustenbach et al. *(3)*

[a] Multiplicative risk model, cumulative exposure.

[b] Calculated by converting ppb to µg/m^3.

estimates by Paxton et al. (Table 8), unit risks lower by up to about one order of magnitude would result.

Guidelines

Benzene is carcinogenic to humans and no safe level of exposure can be recommended. For purposes of guideline derivation, it was decided to use the 1994 risk calculation of Crump rather than to derive new estimates. It was recognized that this use of existing analyses of the most recently updated cohort ruled out the inclusion of certain of the analyses noted earlier.

The geometric mean of the range of estimates of the excess lifetime risk of leukaemia at an air concentration of 1 µg/m^3 is 6×10^{-6}. The concentrations of airborne benzene associated with an excess lifetime risk of 1/10 000, 1/100 000 and 1/1 000 000 are, respectively, 17, 1.7 and 0.17 µg/m^3.

References

1. CRUMP, K. & ALLEN, B. *Quantitative estimates of risk of leukemia from occupational exposure to benzene*. Washington, DC, US Department of Labor, 1984 (OSHA Docket H-059b, Exhibit 152, Annex B).
2. RINSKY, R.A. ET AL. Benzene and leukaemia. An epidemiologic risk assessment. *New England journal of medicine,* **316**: 1044–1050 (1987).
3. PAUSTENBACH, D.J. ET AL. Reevaluation of benzene exposure for the pliofilm (rubberworker) cohort (1936–1976). *Journal of toxicology and environmental health,* **36**: 177–231 (1992).

4. Paxton, M.B. et al. Leukaemia risk associated with benzene exposure in the pliofilm cohort: I. Mortality update and exposure distribution. *Risk analysis,* **14**: 147–154 (1994).
5. Paxton, M.B. et al. Leukaemia risk associated with benzene exposure in the pliofilm cohort. II. Risk estimates. *Risk analysis,* **14**: 155–161 (1994).
6. Brett, S.M. et al. Review and update of leukaemia risk potentially associated with occupational exposure to benzene. *Environmental health perspectives*, **82**: 267–281 (1989).
7. Crump, K.S. Risk of benzene-induced leukaemia: a sensitivity analysis of the pliofilm cohort with additional follow-up and new exposure estimates. *Journal of toxicology and environmental health*, **42**: 219–242 (1994).

5.3 Butadiene

Exposure evaluation

In a survey of butadiene monomer, polymer and end-user industries in the United States, the geometric mean concentration for full-shift exposure for all job categories was 0.098 ppm and the arithmetic mean was 2.12 ppm *(1)*.

Although data for ambient air levels in Europe are limited, reported concentrations in urban air generally ranged from less than 2 μg/m³ to 20 μg/m³ *(2, 3)*. Mean levels in indoor air in a small number of Canadian homes and offices were 0.3 μg/m³ *(4)*. Sidestream cigarette smoke contains 1,3-butadiene at approximately 0.4 mg/cigarette, and levels of butadiene in smoky indoor environments are typically 10–20 μg/m³ *(5)*.

Health risk evaluation

Irritation or effects on the central nervous system may be associated with acute exposure to high concentrations of butadiene. Nevertheless, carcinogenicity is considered to be the critical effect for the derivation of air quality guidelines.

1,3-Butadiene has induced a wide variety of tumours in rats and mice, with mice being considerably more sensitive than rats. There are widely divergent points of view as to which animal species – the rat or the mouse – is most appropriate for use in human risk assessments for butadiene *(6, 7)*.

Epidemiological studies, while relatively few in number, suggest that there is equivocal evidence for an association between exposure to butadiene and lymphohaematopoietic cancer. In 1992, IARC classified butadiene in Group 2A (probably carcinogenic to humans). Preliminary (unpublished) reports suggest, however, that there may be an association between butadiene exposure and leukaemia in workers in the synthetic rubber industry.

The genotoxicity of butadiene has been studied in a variety of *in vitro* and *in vivo* mutagenicity assays, and the data overwhelmingly suggest that the induction of cancer requires the metabolism of butadiene to its DNA-reactive metabolites. Butadiene is mutagenic in both bacterial and mammalian systems. The butadiene metabolites epoxybutene and diepoxybutane are also carcinogenic and genotoxic *in vivo*. Butadiene is metabolized to epoxides to a significantly lesser extent in human tissues than in mice and rats. The differences between mice and rats observed *in vitro* are supported by *in vivo* studies, indicating that mice form very high levels of epoxides compared to rats when exposed to butadiene. In general, the data support the conclusion that the metabolism of butadiene in humans is more similar to that in rats, a relatively insensitive species to butadiene carcinogenicity, than to that in mice, a highly sensitive species. It should be recognized, however, that inter-individual differences in butadiene metabolism may exist that will influence the extent to which butadiene epoxides are formed.

In the only published human study, of 40 individuals occupationally exposed to butadiene at levels typical of an industrial setting (1–3 ppm), there were no significant increases in chromosome aberrations, micronucleus formation or sister chromatid exchanges in peripheral blood lymphocytes *(8)* compared to controls (30 individuals). This observation is of particular interest since butadiene concentrations as low as 6.25 ppm increased the occurrence of the same indicators of genetic damage in the bone marrow and peripheral blood lymphocytes of mice.

Several different risk assessments have been conducted for butadiene, and a number of these for occupational exposures to butadiene have been summarized by the US Occupational Safety and Health Administration *(9)*. The estimates in these risk assessments were based on different assumptions. Some were adjusted for absorbed dose, since changes in butadiene absorption will occur in animals with changes in the inhaled concentration *(10)*. For the most part, they were based on the multistage model. There was considerable variation in human cancer risk estimates depending on the animal species used for the calculations, with those based on tumour data in mice being 100–1000 times higher than those based on tumour data in rats.

Unit risk estimates for cancer associated with continuous lifetime exposure to butadiene in ambient air have been reported *(11–13)*. Values estimated by the Californian Air Resources Board in 1992 *(11)*, based on adjustment of dose for absorption *(10)* and tumour incidence in mice *(14)* and rats *(15)*, were 0.0098 and 0.8 per ppm, respectively. The value estimated by the US Environmental Protection Agency's Integrated Risk Information System (IRIS), based on linearized multistage modelling of data from an earlier, limited US National Toxicology Program (NTP) bioassay in mice, was 2.8×10^{-4} per $\mu g/m^3$ *(12)*. Values estimated by the National Institute of Public Health and Environmental Protection (RIVM) in the Netherlands, based on linearized multistage modelling of the incidence of lymphocytic lymphoma and haemangiosarcomas of the heart in mice in the most recent NTP bioassay *(14)*, were in the range $0.7–1.7 \times 10^{-5}$ per $\mu g/m^3$ *(13)*.

Estimates of human cancer risk could be improved by the inclusion of mechanistic information such as *in vivo* toxicokinetic data, genotoxicity data, and data from the recent epidemiology reassessment. For example, new data on levels of butadiene epoxides in blood and tissues in laboratory animals *(16–18)* could be used to replace the earlier absorption data *(10)*. Additionally, physiologically based pharmacokinetic models developed since earlier attempts to apply this approach to risk assessment have been greatly improved, most notably by the incorporation of model parameters that have been experimentally measured rather than empirically estimated. None the less, none of the models published to date incorporates the necessary information on the formation, removal and distribution of diepoxybutane.

Guidelines

Quantitative cancer risk estimates vary widely, in particular depending on the test species used. No definitive conclusions can yet be made as to which species should be used for risk estimates. New, as yet unpublished epidemiological data might have an impact on the risk estimates and hence on the derivation of a guideline value. In the light of these considerations, no guideline value can be recommended at this time.

References

1. FAJEN, J.M. ET AL. Industrial exposure to 1,3-butadiene in monomer, polymer, and end user industries. *In*: Sorsa, M. et al., ed. *Butadiene and styrene: assessment of health hazards.* Lyons, International Agency for Research on Cancer, 1993 (IARC Scientific Publications, No. 127), pp. 3–13.
2. NELIGAN, R.E. Hydrocarbons in the Los Angeles atmosphere. *Archives of environmental health,* **5**: 581–591 (1962).
3. COTE, I.L. & BAYARD, S.P. Cancer risk assessment of 1,3-butadiene. *Environmental health perspectives,* **86**: 149–153 (1990).
4. BELL, R.W. ET AL. *The 1990 Toronto Personal Exposure Pilot (PEP) Study*. Toronto, Ministry of the Environment, 1991.
5. LÖFROTH, G. ET AL. Characterization of environmental tobacco smoke. *Environmental science and technology,* **23**: 610–614 (1989).
6. BOND, J.A. ET AL. Epidemiological and mechanistic data suggest that 1,3-butadiene will not be carcinogenic to humans at exposures likely to be encountered in the environment or workplace. *Carcinogenesis,* **16**: 165–171 (1995).
7. MELNICK, R.L. & KOHN, M.C. Mechanistic data indicate that 1,3-butadiene is a human carcinogen. *Carcinogenesis,* **16**: 157–163 (1995).
8. SORSA, M. ET AL. Human cytogenetic biomonitoring of occupational exposure to 1,3-butadiene. *Mutation research,* **309**: 321–326 (1994).

9. US Occupational Safety and Health Administration. Occupational exposure to 1,3 butadiene: proposed rule and notice of hearing. *Federal register*, **55**: 32747 (1990).
10. Bond, J.A. et al. Species differences in the disposition of inhaled butadiene. *Toxicology and applied pharmacology,* **84**: 617–627 (1986).
11. *Proposed identification of 1,3-butadiene as a toxic air contaminant. Part B: health assessment.* Sacramento, California Air Resources Board, 1992.
12. *Integrated Risk Information System (IRIS)* (http://www.epa.gov/ngispgm3/iris/). Cincinnati, OH, US Environmental Protection Agency (accessed 18 September 2000).
13. Slooff, W. et al. *Exploratory report: 1,3-butadiene.* Bilthoven, National Institute of Public Health and Environmental Protection (RIVM), 1994 (Report No. 7104010333).
14. Melnick, R.L. et al. Carcinogenicity of 1,3-butadiene in C57Bl/6 × C3HF1 mice at low exposure concentrations. *Cancer research,* **50**: 6592–6599 (1990).
15. Owen, P.E. et al. Inhalation toxicity studies with 1,3-butadiene. 3. Two year toxicity/carcinogencity study in rats. *American Industrial Hygiene Association journal,* **48**: 407–413 (1987).
16. Himmelstein, M.W. et al. Comparison of blood concentrations of 1,3-butadiene and butadiene epoxides in mice and rats exposed to 1,3-butadiene by inhalation. *Carcinogenesis,* **15**: 1479–1486 (1994).
17. Himmelstein, M.W. et al. High concentrations of butadiene epoxides in livers and lungs of mice compared to rats exposed to 1,3-butadiene. *Toxicology and applied toxicology,* **132**: 281–288 (1995).
18. Thornton-Manning, J.R. et al. Disposition of butadiene monoepoxide and butadiene diepoxide in various tissues of rats and mice following a low-level inhalation exposure to 1,3-butadiene. *Carcinogenesis,* **16**: 1723–1731 (1995).

5.4 Carbon disulfide

Exposure evaluation

Inhalation represents the main route of entry of carbon disulfide into the human organism. Values in the vicinity of viscose rayon plants range from 0.01 mg/m^3 to about 1.5 mg/m^3, depending mostly on the distance from the source.

Health risk evaluation

A summary of the most relevant concentration–response findings is given in Table 10.

In the light of numerous epidemiological studies, it is very difficult to establish the exact exposure–time relationship. During the approximate period 1955–1965, carbon disulfide concentrations in viscose rayon plants averaged about 250 mg/m^3; they were subsequently reduced to 50–150 mg/m^3 and more recently to 20–30 mg/m^3. It is thus practically impossible to evaluate the long-term (five or more years) exposure level in a retrospective study. Moreover, most exposure data in occupational studies are not reliable, owing to poor measurement methodology. It is necessary to keep this in mind also when studying Table 10.

At exposure levels of 30 mg/m^3 and above, observable adverse health effects have been well established. The coronary heart disease rate increases at levels of 30–120 mg/m^3 of carbon disulfide after an exposure of more than 10 years. Effects on the central and peripheral nervous systems and the vascular system have been established in the same range of concentrations after long-term exposure. Functional changes of the central nervous system have even been observed at lower concentrations (20–25 mg/m^3).

Some authors claim to have observed adverse health effects in workers exposed to 10 mg/m^3 of carbon disulfide for 10–15 years. Because of the lack of reliable retrospective data on exposure levels, however, the dose–response relationship governing these findings is difficult to establish.

Guidelines

The lowest concentration of carbon disulfide at which an adverse effect was observed in occupational exposure was about 10 mg/m^3, which may be equivalent to a concentration in the general environment of 1 mg/m^3. In

Table 10. Some concentration–response relationships in occupational exposure to carbon disulfide

Carbon disulfide concentration (mg/m^3)	Duration of exposure (years)	Symptoms and signs	Reference
500–2500	0.5	Polyneuritis, myopathy, acute psychosis	*1*
450–1000	< 0.5	Polyneuritis, encephalopathy	*2*
200–500	1–9	Increased ophthalmic pressure	*3*
60–175	5	Eye burning, abnormal pupillary light reactions	*4*
31–137	10	Psychomotor and psychological disturbances	*5*
29–118	15	Polyneuropathy, abnormal EEG, conduction velocity slowed, psychological changes	*6, 7*
29–118	10	Increase in coronary mortality, angina pectoris, slightly higher systolic and diastolic blood pressure	*8–11*
40–80	2	Asthenospermia, hypospermia, teratospermia	*12*
22–44	> 10	Arteriosclerotic changes and hypertension	*13*
30–50	> 10	Decreased immunological reactions	*14*
30	3	Increase in spontaneous abortions and premature births	*15*
20–25	< 5	Functional disturbances of the central nervous system	*16, 17*
10	10–15	Sensory polyneuritis, increased pain threshold	*18*
10	10–15	Depressed blood progesterone, increased estriol, irregular menstruation	*19*

selecting the size of the protection (safety) factor, the expected variability in the susceptibility of the general population was taken into account, and a protection factor of 10 was considered appropriate. This leads to the recommendation of a guideline value of 100 μg/m^3, with an averaging time of 24 hours. It is believed that below this value adverse health effects of environmental exposure to carbon disulfide (outdoor or indoor) are not likely to occur.

If carbon disulfide is used as the index substance for viscose emissions, odour perception is not to be expected when carbon disulfide peak concentration is kept below one tenth of its odour threshold value, i.e. below 20 μg/m^3. Based on the sensory effects of carbon disulfide, a guideline value of 20 μg/m^3 (average time 30 minutes) is recommended.

References

1. Vigliani, E.C. Chronic carbon disulfide poisoning: a report on 100 cases. *Medicina del lavoro,* **37**: 165–193 (1946).
2. Vigliani, E.C. Carbon disulphide poisoning in viscose rayon factories. *British journal of industrial medicine,* **11**: 235–244 (1954).
3. Maugeri, U. et al. La oftalmodinamografia nella intossicazione solofocarbonica professionalle [Ophthalmodynamography in occupational carbon disulfide poisoning]. *Medicina del lavoro,* **57**: 730–740 (1966).
4. Savic, S. Influence of carbon disulfide on the eye. *Archives of environmental health,* **14**: 325–326 (1967).
5. Hänninen, H. Psychological picture of manifest and latent carbon disulphide poisoning. *British journal of industrial medicine,* **28**: 374–381 (1971).
6. Seppäläinen, A.M. & Tolonen, M. Neurotoxicity of long-term exposure to carbon disulfide in the viscose rayon industry: a neurophysiological study. *Work, environment, health,* **11**: 145–153 (1974).
7. Tolonen, M. Chronic subclinical carbon disulfide poisoning. *Work, environment, health,* **11**: 154–161 (1974).
8. Tolonen, M. et al. A follow-up study of coronary heart disease in viscose rayon workers exposed to carbon disulphide. *British journal of industrial medicine,* **32**: 1–10 (1975).
9. Hernberg, S. et al. Excess mortality from coronary heart disease in viscose rayon workers exposed to carbon disulfide. *Work, environment, health,* **10**: 93–99 (1973).
10. Nurminen, M. Survival experience of a cohort of carbon disulphide exposed workers from an eight-year prospective follow-up period. *International journal of epidemiology,* **5**: 179–185 (1976).

11. HERNBERG, S. ET AL. Coronary heart disease among workers exposed to carbon disulphide. *British journal of industrial medicine,* **27**: 313–325 (1970).
12. LANCRANJAN, I. ET AL. Changes of the gonadic function in chronic carbon disulfide poisoning. *Medicina del lavoro,* **60**: 556–571 (1969).
13. GAVRILESCU, N. & LILIS, R. Cardiovascular effects of long-extended carbon disulphide exposure. *In:* Brieger H. & Teisinger J., ed. *Toxicology of carbon disulphide.* Amsterdam, Excerpta Medica, 1967, pp. 165–167.
14. KAŠIN, L.M. [Overall immunological reactivity and morbidity of workers exposed to carbon disulfide]. *Gigiena i sanitarija,* **30**: 331–335 (1965) [Russian].
15. PETROV, M.V. [Course and termination of pregnancy in women working in the viscose industry]. *Pediatrija, akušerstvo i ginekologija,* **3**: 50–52 (1969) [Russian].
16. GILIOLI, R. ET AL. Study of neurological and neurophysiological impairment in carbon disulphide workers. *Medicina del lavoro,* **69**: 130–143 (1978).
17. CASSITO, M.G. ET AL. Subjective and objective behavioural alterations in carbon disulphide workers. *Medicina del lavoro,* **69**: 144–150 (1978).
18. MARTYNOVA, A.P. ET AL. [Clinical, hygienic and experimental investigations of the action on the body of small concentrations of carbon disulfide]. *Gigiena i sanitarija,* **5**: 25–28 (1976) [Russian].
19. VASILJEVA, I.A. [Effect of low concentrations of carbon disulfide and hydrogen sulfide on the menstrual function in women and on the estrous cycle under experimental conditions]. *Gigiena i sanitarija,* **38**: 24–27 (1973) [Russian].

5.5 Carbon monoxide

Exposure evaluation

Global background concentrations of carbon monoxide range between 0.06 mg/m^3 and 0.14 mg/m^3 (0.05–0.12 ppm). In urban traffic environments of large European cities, the 8-hour average carbon monoxide concentrations are generally lower than 20 mg/m^3 (17 ppm) with short-lasting peaks below 60 mg/m^3 (53 ppm). Carbon monoxide concentrations inside vehicles are generally higher than those measured in ambient outdoor air. The air quality data from fixed-site monitoring stations seem to reflect rather poorly short-term exposures of various urban population groups, but appear to reflect better longer averaging times, such as 8 hours.

In underground and multistorey car parks, road tunnels, enclosed ice arenas and various other indoor microenvironments, in which combustion engines are used under conditions of insufficient ventilation, the mean levels of carbon monoxide can rise above 115 mg/m^3 (100 ppm) for several hours, with short-lasting peak values that can be much higher. In homes with gas appliances, peak carbon monoxide concentrations of up to 60–115 mg/m^3 (53–100 ppm) have been measured. Environmental tobacco smoke in dwellings, offices, vehicles and restaurants can raise the 8-hour average carbon monoxide concentration to 23–46 mg/m^3 (20–40 ppm).

Carbon monoxide diffuses rapidly across alveolar, capillary and placental membranes. Approximately 80–90% of the absorbed carbon monoxide binds with haemoglobin to form carboxyhaemoglobin (COHb), which is a specific biomarker of exposure in blood. The affinity of haemoglobin for carbon monoxide is 200–250 times that for oxygen. During an exposure to a fixed concentration of carbon monoxide, the COHb concentration increases rapidly at the onset of exposure, starts to level off after 3 hours, and reaches a steady state after 6–8 hours of exposure. The elimination half-life in the fetus is much longer than in the pregnant mother.

In real-life situations, the prediction of individual COHb levels is difficult because of large spatial and temporal variations in both indoor and outdoor carbon monoxide concentrations.

Health risk evaluation

The binding of carbon monoxide with haemoglobin to form COHb reduces the oxygen-carrying capacity of the blood and impairs the release of oxygen from haemoglobin to extravascular tissues. These are the main causes of tissue hypoxia produced by carbon monoxide at low exposure levels. At higher concentrations the rest of the absorbed carbon monoxide binds with other haem proteins such as myoglobin, and with cytochrome oxidase and cytochrome P-450 *(1, 2)*. The toxic effects of carbon monoxide become evident in organs and tissues with high oxygen consumption such as the brain, the heart, exercising skeletal muscle and the developing fetus.

Severe hypoxia due to acute carbon monoxide poisoning may cause both reversible, short-lasting neurological deficits and severe, often delayed neurological damage. The neurobehavioural effects include impaired coordination, tracking, driving ability, vigilance and cognitive performance at COHb levels as low as 5.1–8.2% *(3–5)*.

In apparently healthy subjects, maximal exercise performance has decreased at COHb levels as low as 5%. The regression between the percentage decrease in maximal oxygen consumption and the percentage increase in COHb concentration appears to be linear, with a fall in oxygen consumption of approximately one percentage point for each percentage point rise in COHb level above 4% *(1, 6)*.

In controlled human studies involving patients with documented coronary artery disease, mean postexposure COHb levels of 2.9–5.9% (corresponding to postexercise COHb levels of 2.0–5.2%) have been associated with a significant shortening in the time to onset of angina, with increased electrocardiographic changes and with impaired left ventricular function during exercise *(7–11)*. In addition, ventricular arrhythmias may be increased significantly at the higher range of mean postexercise COHb levels *(12, 13)*. Epidemiological and clinical data indicate that carbon monoxide from recent smoking and environmental or occupational exposures may contribute to cardiovascular mortality and the early course of myocardial infarction *(1)*. According to one study there has been a 35% excess risk of death from arteriosclerotic heart disease among smoking and nonsmoking tunnel officers, in whom the long-term mean COHb levels were generally less than 5% *(13)*. Current data from epidemiological studies and experimental animal studies indicate that common environmental exposures to carbon monoxide do not have atherogenic effects on humans *(1, 14)*.

During pregnancy, endogenous production of carbon monoxide is increased so that maternal COHb levels are usually about 20% higher than the non-pregnant values. At steady state, fetal COHb levels are up to 10–15% higher than maternal COHb levels *(1, 15)*. There is a well established and probably causal relationship between maternal smoking and low birth weight at fetal COHb levels of 2–10%. In addition, maternal smoking seems to be associated with a dose-dependent increase in perinatal deaths and with behavioural effects in infants and young children *(15)*.

In contrast with most other man-made air pollutants at very high concentrations (well above ambient levels), carbon monoxide causes a large number of acute accidental and suicidal deaths in the general population.

Guidelines

In healthy subjects, endogenous production of carbon monoxide results in COHb levels of 0.4–0.7%. During pregnancy, elevated maternal COHb levels of 0.7–2.5%, mainly due to increased endogenous production, have been reported. The COHb levels in non-smoking general populations are usually 0.5–1.5%, owing to endogenous production and environmental exposures. Nonsmokers in certain occupations (car drivers, policemen, traffic wardens, garage and tunnel workers, firemen, etc.) can have long-term COHb levels of up to 5%, and heavy cigarette smokers have COHb levels of up to 10% *(1, 2, 15)*. Well trained subjects engaging in heavy exercise in polluted indoor environments can increase their COHb levels quickly up to 10–20%. In indoor ice arenas, epidemic carbon monoxide poisonings have recently been reported.

To protect nonsmoking, middle-aged and elderly population groups with documented or latent coronary artery disease from acute ischaemic heart attacks, and to protect the fetuses of nonsmoking pregnant women from untoward hypoxic effects, a COHb level of 2.5% should not be exceeded.

The following guidelines are based on the Coburn-Foster-Kane exponential equation, which takes into account all the known physiological variables affecting carbon monoxide uptake *(16)*. The following guideline values (ppm values rounded) and periods of time-weighted average exposures have been determined in such a way that the COHb level of 2.5% is not exceeded, even when a normal subject engages in light or moderate exercise:

- 100 mg/m^3 (90 ppm) for 15 minutes
- 60 mg/m^3 (50 ppm) for 30 minutes
- 30 mg/m^3 (25 ppm) for 1 hour
- 10 mg/m^3 (10 ppm) for 8 hours

References

1. *Air quality criteria for carbon monoxide*. Washington, DC, US Environmental Protection Agency, 1991 (Publication EPA-600/B-90/045F).
2. ACGIH Chemical Substances TLV Committee. Notice of intended change – carbon monoxide. *Applied occupational and environmental hygiene*, **6**: 621–624 (1991).
3. ACGIH Chemical Substances TLV Committee. Notice of intended change – carbon monoxide. *Applied occupational and environmental hygiene*, **6**: 896–902 (1991).
4. Putz, V.R. The effects of carbon monoxide on dual-task performance. *Human factors*, **21**: 13–24 (1979).
5. Benignus, V.A. et al. Effect of low level carbon monoxide on compensatory tracking and event monitoring. *Neurotoxicology and teratology*, **9**: 227–234 (1987).
6. Bascom, R. et al. Health effects of outdoor air pollution (Part 2). *American journal of respiratory and critical care medicine*, **153**: 477–498 (1996).
7. Anderson, E.W. et al. Effect of low-level carbon monoxide exposure on onset and duration of angina pectoris: a study in ten patients with ischemic heart disease. *Annals of internal medicine*, **79**: 46–50 (1973).
8. Kleinman, M.T. et al. Effects of short–term exposure to carbon monoxide in subjects with coronary artery disease. *Archives of environmental health*, **44**: 361–369 (1989).
9. Allred, E.N. et al. Short-term effects of carbon monoxide exposure on the exercise performance of subjects with coronary artery disease. *New England journal of medicine*, **321**: 1426–1432 (1989).
10. Sheps, D.S. et al. Lack of effect of low levels of carboxyhemoglobin on cardiovascular function in patients with ischemic heart disease. *Archives of environmental health*, **42**: 108–116 (1987).
11. Adams, K.F. et al. Acute elevation of blood carboxyhemoglobin to 6% impairs exercise performance and aggravates symptoms in patients with ischemic heart disease. *Journal of the American College of Cardiology*, **12**: 900–909 (1988).
12. Sheps, D.S. et al. Production of arrhythmias by elevated carboxyhemoglobin in patients with coronary artery disease. *Annals of internal medicine*, **113**: 343–351 (1990).
13. Stern, F.B. et al. Heart disease mortality among bridge and tunnel officers exposed to carbon monoxide. *American journal of epidemiology*, **128**: 1276–1288 (1988).
14. Smith, C.J. & Steichen, T.J. The atherogenic potential of carbon monoxide. *Atherosclerosis*, **99**: 137–149 (1993).

15. LONGO, L.D. The biological effects of carbon monoxide on the pregnant woman, fetus, and newborn infant. *American journal of obstetrics and gynecology*, **129**: 69–103 (1977).
16. COBURN, R.F. ET AL. Considerations of the physiological variables that determine the blood carboxyhemoglobin concentration in man. *Journal of clinical investigation*, **44**: 1899–1910 (1965).

5.6 1,2-Dichloroethane

Exposure evaluation

Rural or background atmospheric concentrations in western Europe and North America are approximately 0.2 μg/m^3, and the limited data available on indoor concentrations show that they are about the same. Average levels in cities vary from 0.4 μg/m^3 to 1.0 μg/m^3, increasing to 6.1 μg/m^3 near petrol stations, parking garages and production facilities.

Health risk evaluation

Human studies point to effects on the central nervous system and the liver, but the limited data do not allow a definitive conclusion regarding a LOAEL or NOAEL. In animals, long-term inhalation exposure (> 6 months) to 1,2-dichloroethane levels of approximately 700 mg/m^3 and above has been shown to result in histological changes in the liver *(1–3).* The same animal studies reported no adverse histological changes in the liver and kidneys of guinea pigs and rats at levels of about 400 mg/m^3. Findings concerning effects on reproduction are contradictory.

Animal data suggest a NOAEL in laboratory animals of 400 mg/m^3 and a LOAEL of 700 mg/m^3.

With regard to mutagenicity as an endpoint and to the causal connections between DNA damage and the initiation of carcinogenicity, 1,2-dichloroethane has been shown to be weakly mutagenic in *Salmonella typhimurium,* both in the absence and in the presence of microsomal activation systems. It has also been demonstrated to be mutagenic in other test species and in *in vitro* tests using mammalian cells.

In a lifetime study in rats and mice in which 1,2-dichloroethane was administered by gavage, it caused tumours at multiple sites in both species. In the only inhalation study performed *(4),* exposure to 1,2-dichloroethane did not result in an increased tumour incidence. The negative results obtained in this study, however, do not detract from the positive findings of the oral study *(5, 6)* when differences in total dose, exposure time and pharmacokinetics are considered.

1,2-Dichloroethane was evaluated in 1979 by IARC as a chemical for which there is sufficient evidence of carcinogenicity in experimental animals and inadequate evidence in humans *(7)*. To date there are two publications giving quantitative carcinogenic risk estimates based on animal data. One, developed by the National Institute of Public Health in the Netherlands on the basis of oral exposure of rats by gavage *(6),* indicates a lifetime risk of one in a million from exposure to 0.48 μg/m^3 *(8),* which corresponds to a unit risk of about 2×10^{-6}. The US Environmental Protection Agency *(9)* has estimated an incremental unit risk of 2.6×10^{-5} on the basis of data from gavage studies and of 1×10^{-6} on the basis of a negative inhalation study.

Guidelines

Evidence of carcinogenicity in animals is sufficient on the basis of oral ingestion data. However, animal inhalation data do not at present provide positive evidence. Because of deficiencies in extrapolation from oral data to inhalation, the two risk estimates available are not used in the guidelines.

For noncarcinogenic endpoints, data from animal studies imply a NOAEL of about 400 mg/m^3 and suggest a LOAEL of about 700 mg/m^3. A protection (safety) factor of 1000 is considered appropriate in extrapolation of animal data to the general population. In selecting such a large protection factor, variations in exposure time, the limitations of the database and the fact that a no-effect level in humans cannot be established are of decisive importance. The resulting value of 0.7 mg/m^3 for continuous exposure (averaging time 24 hours) is recommended as a guideline value. Since this value is above current environmental levels and present exposures are not of concern to health, this guideline relates only to accidental release episodes or specific indoor pollution problems.

References

1. Spencer, H.C. et al. Vapour toxicity of ethylene dichloride determined by experiments on laboratory animals. *Archives of industrial hygiene and occupational medicine,* **4**: 482–493 (1951).
2. Heppel, L.A. et al. The toxicology of 1,2-dichloroethane (ethylene dichloride). V. The effect of daily inhalations. *Journal of industrial hygiene and toxicology,* **28**: 113–120 (1946).
3. Hofmann, H.T. et al. Zur Inhalationstoxizität von 1,1- und 1,2-Dichloräthan [On the inhalation toxicity of 1,1- and 1,2-dichloroethane]. *Archiv für Toxikologie,* **27**: 248–265 (1971).
4. Spreafico, F. et al. Pharmacokinetics of ethylene dichloride in rats treated by different routes and its long-term inhalatory toxicity. *Banbury reports,* **5**: 107–129 (1980).
5. US National Cancer Institute. *Bioassay of 1,2-dichloroethane for possible carcinogenicity.* Bethesda, MD, US Department of Health, Education and Welfare, 1978 (DHEW Publication No. (NIH) 78-1305).

6. WARD, J.M. The carcinogenicity of ethylene dichloride in Osborne-Mendel rats and B6C3F1 mice. *Banbury reports,* **5**: 35–53 (1980).
7. 1,2-Dichloroethane. *In: Some halogenated hydrocarbons.* Lyons, International Agency for Research on Cancer, 1979 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 20), pp. 429–448.
8. BESEMER, A.C. ET AL. *Criteriadocument over 1,2-dichloorethaan* [1,2-Dichloroethane criteria document]. The Hague, Ministry of Housing, Spatial Planning and Environment, 1984 (Publikatiereeks Lucht, No. 30).
9. *Health assessment document for 1,2-dichloroethane (ethylene dichloride).* Washington, DC, US Environmental Protection Agency, 1985 (Report EPA-600/8-84-0067).

5.7 Dichloromethane

Exposure evaluation

Mean outdoor concentrations of dichloromethane are generally below 5 μg/m^3 *(1–4)*. Significantly higher concentrations (by at least one order of magnitude) may occur close to industrial emission sources. Indoor air concentrations are variable but tend to be about three times greater than outdoor values *(5, 6)*. Under certain circumstances, much higher values (up to 4000 μg/m^3) may be recorded indoors, particularly with use of paint stripping solutions *(7)*. Exposures of the general population occur principally through the use of dichloromethane-containing consumer products. Exposure in outdoor air, water *(8–12)* and food *(13–15)* is low.

Health risk evaluation

The critical effects of dichloromethane include effects on the central nervous system, the production of carboxyhaemoglobin (COHb) and carcinogenicity. The impairment of behavioural or sensory responses may occur in humans following acute inhalation exposure at levels exceeding 1050 mg/m^3 (300 ppm) for short durations, and the effects are transient. The cytochrome P-450-related oxidative pathway resulting in carbon monoxide production is saturable, producing maximum blood COHb levels of ≤ 9%. Nevertheless, these COHb levels are sufficiently high to induce acute effects on the central nervous system, and it thus appears that such effects are probably due to COHb production. Dichloromethane does not appear to cause serious effects in humans at those relatively high levels reported in occupational settings.

Although there is no convincing evidence of cancer incidence associated with occupational exposure, the available data have limitations and are considered inadequate to assess human carcinogenicity. In male and female mice and male and female rats, the National Toxicology Program's bioassays led to the conclusion of clear evidence of carcinogenicity in mice, clear evidence in female rats and equivocal evidence in male rats *(16)*. IARC has classified dichloromethane as showing sufficient evidence of carcinogenicity in experimental animals (Group 2B) *(17)*.

The health risks of exposure to dichloromethane have been considered in detail by an International Programme on Chemical Safety (IPCS) expert group. Given the data on interspecies differences in metabolism and comparative cancer risks, that group concluded that carcinogenicity was not the critical endpoint for risk assessment purposes. It is therefore concluded that the formation of COHb is a more direct indication of a toxic effect, that it can be monitored, and that it is therefore more suitable as a basis for the derivation of a guideline. Furthermore, it is unlikely that ambient air exposures represent a health concern with reference to any cancer endpoint, since concentrations of dichloromethane in ambient air are orders of magnitude lower than levels associated with direct adverse effects on the central nervous system or on COHb production in humans.

The application of physiologically based pharmacokinetic models to the available animal data lead to small risk estimates *(18, 19)*. These risk estimates are much lower than the recommended guideline value using COHb formation, and were therefore not employed in guideline derivation.

Guidelines

The selected biological endpoint of interest is the formation of COHb, which is measured in the blood of normal subjects at levels of 0.50–1.5% of total haemoglobin. In heavy smokers, the level of COHb may range up to 10%. Carbon monoxide from various sources may contribute to the formation of COHb. Since overall levels in many cases approach the recommended maximum of 3%, it is prudent to minimize any additional amounts of COHb contributed from dichloromethane. It was thus concluded that no more than 0.1% additional COHb should be formed from dichloromethane exposure. This corresponds to the analytical reproducibility of the method applied to measure COHb at the level of concern. This maximum allowable increase in COHb corresponds to a 24-hour exposure to dichloromethane at a concentration of 3 mg/m^3. Consequently, a guideline value of 3 mg/m^3 is recommended. In addition, the weekly average concentration should not exceed one seventh (0.45 mg/m^3) of this 24-hour guideline, given the half-life of COHb.

References

1. DANN, T. *Measurement of volatile organic compounds in Canada 1991–1992*. Ottawa, Environment Canada, 1993.
2. MCALLISTER, R. ET AL. *Non-methane organic compound program. Final Report, Vol. II.* Research Triangle Park, NC, US Environmental Protection Agency, 1989 (Report No. EPA-450/4-89-005).
3. SHIKIYA, J. ET AL. Ambient monitoring of selected halogenated hydrocarbons and benzene in the California South Coast Air Basin. *Proceedings of the Air Pollution Control Association 77th Annual Meeting*, Vol. 1, Paper 84e–1.1. Pittsburg, PA, Air Pollution Control Association, 1984.

4. HARKOV, R. ET AL. Comparison of selected volatile organic compounds during the summer and winter at urban sites in New Jersey. *Science of the total environment,* **38**: 259–274 (1984).
5. WALLACE, L. ET AL. The Los Angeles TEAM Study: personal exposures, indoor–outdoor air concentrations, and breath concentrations of 25 volatile organic compounds. *Journal of exposure analysis and environmental epidemiology*, **1**: 157–192 (1991).
6. CHAN, C. C. ET AL. Determination of organic contaminants in residential indoor air using an adsorption-thermal desorption technique. *Journal of the Air and Waste Management Association*, **40**: 62–67 (1990).
7. OTSON, R. ET AL. Dichloromethane levels in air after application of paint removers. *American Industrial Hygiene Association journal,* **42**: 56–60 (1981).
8. MCGEORGE, L. ET AL. Implementation and results of a mandatory statewide program for organic contaminant analysis of delivered water. *Proceedings of Water Quality Technology Conference,* **15**: 71–102 (1987).
9. *Priority Substances List Assessment Report. Dichloromethane*. Ottawa, Environment Canada and Health Canada, 1993.
10. OTSON, R. ET AL. Volatile organic compounds in water at thirty Canadian potable water treatment facilities. *Journal of the Association of Official Analytical Chemists*, **65**: 1370–1374 (1982).
11. OTSON, R. Purgeable organics in Great Lakes raw and treated water. *International journal of environmental analytical chemistry*, **31**: 41–53 (1987).
12. STAPLES, C.A. ET AL. Assessment of priority pollutant concentrations in the United States using STORET database. *Environmental toxicology and chemistry*, **4**: 131–142 (1985).
13. FERRARIO, J.B. ET AL. Volatile organic pollutants in biota and sediments of Lake Pontchartrain. *Bulletin of environmental contamination and toxicology*, **43**: 246–255 (1985).
14. HEIKES, D.L. Environmental contaminants in table-ready foods from the total diet program of the Food and Drug Administration. *Advances in environmental science and technology,* **23**: 31–57 (1990).
15. PAGE, B.D. & CHARBONNEAU, C.F. Headspace gas chromatographic determination of residual methylene chloride in decaffeinated tea and coffee with electronic conductivity detection. *Journal of the Association of Official Analytical Chemists*, **67**: 757–761 (1984).
16. NATIONAL TOXICOLOGY PROGRAM. *Toxicology and carcinogenesis studies of dichloromethane (methylene chloride) (CAS No. 75-09-2) in F344/N rats and B6C3F1 mice (inhalation studies).* Research Triangle Park, NC, US Department of Health and Human Services, 1986 (Document No. NTP-TRS-306).

17. *Some halogenated hydrocarbons and pesticide exposure*. Lyons, International Agency for Research on Cancer, 1986 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 41), pp. 43–85.
18. *Methylene chloride (dichloromethane): human risk assessment using experimental animal data*. Brussels, European Centre for Ecotoxicology and Toxicology of Chemicals, 1988 (Technical Report No.32).
19. DANKOVIC, D.A. & BAILER, A.J. The impact of exercise and intersubject variability on dose estimates for dichloromethane derived from a physiologically based pharmacokinetic model. *Fundamental and applied toxicology*, **22**: 20–25 (1994).

5.8 Formaldehyde

Exposure evaluation

The major route of exposure to formaldehyde is inhalation. Table 11 shows the contribution of the various atmospheric environments to non-occupational air levels. Indoor air concentrations are several orders of magnitude higher than levels in ambient air. Owing to the extremely high concentrations of formaldehyde in tobacco smoke, smoking constitutes a major source of formaldehyde *(1)*.

Health risk evaluation

Predominant symptoms of formaldehyde exposure in humans are irritation of the eyes, nose and throat, together with concentration-dependent discomfort, lachrymation, sneezing, coughing, nausea, dyspnoea and finally death (Table 12) .

Damage to the nasal mucosa, such as squamous cell metaplasia and mild dysplasia of the respiratory epithelium, have been reported in humans, but

Table 11. Average exposure concentrations to formaldehyde and contribution of various atmospheric environments to average exposure to formaldehyde

Source	Concentration (mg/m^3)	Exposure (mg/day)
Ambient air (10% of time; 2 m^3/day)	0.001–0.02	0.002–0.04
Indoor air		
Home (65% of time; 10 m^3/day)		
– conventional	0.03–0.06	0.3–0.6
– mobile home	0.1	1.0
– environmental tobacco smoke	0.05–0.35	0.5–3.5
Workplace (25% of time; 8 m^3/day)		
– without occupational exposure [a]	0.03–0.06	0.2–0.5
– with occupational exposure	1.0	8.0
– environmental tobacco smoke	0.05–0.35	0.4–2.8
Smoking (20 cigarettes/day)	60–130	0.9–2.0 [b]

[a] Assuming the normal formaldehyde concentration in conventional buildings.

[b] Total amount of formaldehyde in smoke from 20 cigarettes.

Source: World Health Organization *(2)*.

Table 12. Effects of formaldehyde in humans after short-term exposure

Concentration range or average (mg/m³)	Time range or average	Health effects in general population
0.03	Repeated exposure	Odour detection threshold (10th percentile) [a]
0.18	Repeated exposure	Odour detection threshold (50th percentile) [a]
0.6	Repeated exposure	Odour detection threshold (90th percentile) [a]
0.1–3.1	Single and repeated exposure	Throat and nose irritation threshold
0.6–1.2	Single and repeated exposure	Eye irritation threshold
0.5–2.0	3–5 hours	Decreased nasal mucus flow rate
2.4	40 minutes on 2 successive days with 10 minutes of moderate exercise on second day	Postexposure (up to 24 hours) headache
2.5–3.7	–[b]	Biting sensation in eyes and nose
3.7	Single and repeated exposure	Decreased pulmonary function only at heavy exercise
5–6.2	30 minutes	Tolerable for 30 minutes with lachrymation
12–25	–[b]	Strong lachrymation, lasting for 1 hour
37–60	–[b]	Pulmonary oedema, pneumonia, danger to life
60–125	–[b]	Death

[a] Frequency of effect in population.

[b] Time range or average unspecified.

these findings may have been confounded by concomitant exposures to other substances *(3)*.

There is convincing evidence of high concentrations of formaldehyde being capable of inducing nasal cancer in rats and possibly in mice *(3)*. Formaldehyde has been shown to be genotoxic in a variety of *in vitro* and *in vivo* systems *(3)*. There is also epidemiological evidence of associations between relatively high occupational exposure to formaldehyde and both nasopharyngeal and sinonasal cancers *(3–7)*.

There is substantial variation in individual responses to formaldehyde in humans *(1–3)*. Significant increases in signs of irritation occur at levels above 0.1 mg/m^3 in healthy subjects. At concentrations above 1.2 mg/m^3, a progression of symptoms and effects occurs. Lung function of healthy nonsmokers and asthmatics exposed to formaldehyde at levels up to 3.7 mg/m^3 was generally unaltered *(8–10)*. It is assumed that in these studies the observed effects were more related to peak concentrations than to mean values.

There is some evidence of formaldehyde inducing pathological and cytogenetic changes in the nasal mucosa of humans. Reported mean exposures ranged from 0.02 mg/m^3 to 2.4 mg/m^3, with peaks between 5 mg/m^3 and 18 mg/m^3. Epidemiological studies suggest a causal relationship between exposure to formaldehyde and nasopharyngeal cancer, although the conclusion is tempered by the small numbers of observed and expected cases *(3–6)*. There are also epidemiological observations of an association between relatively high occupational exposures to formaldehyde and sinonasal cancer *(7)*. IARC *(3)* has interpreted the available cancer data as limited evidence for the carcinogenicity of formaldehyde in humans, and classified formaldehyde in Group 2A.

Formaldehyde is a nasal carcinogen in rats. A highly significant incidence of nasal cancer was found in rats exposed to a level of 16.7 mg/m^3, but the dose–response curve was nonlinear, the risk being disproportionately low at low concentrations. It also appears that the dose–response curves were nearly identical for neoplastic changes, cell turnover, DNA–protein cross-links and hyperproliferation, when the relationship between non-neoplastic and neoplastic lesions in the nasal respiratory epithelium was analysed. This close concordance indicates an association among the observed cytotoxic, genotoxic and carcinogenic effects. It is thus likely that hyperproliferation induced by cytotoxicity plays a significant role in the formation of nasal tumours by formaldehyde.

Despite differences in the anatomy and physiology of the respiratory tract between rats and humans, the respiratory tract defence mechanisms are similar. It is therefore reasonable to assume that the response of the human respiratory tract mucosa to formaldehyde will be similar to that of the rat. Thus, if the respiratory tract tissue is not repeatedly damaged, exposure of humans to low, noncytotoxic concentrations of formaldehyde can be assumed to be associated with a negligible cancer risk. This is consistent with epidemiological findings of excess risks of nasopharyngeal and sinonasal cancers associated with concentrations above about 1 mg/m^3.

Simultaneous exposure of humans to formaldehyde and other upper respiratory tract toxicants, such as acrolein, acetaldehyde, crotonaldehyde, furfural, glutaraldehyde and ozone, may lead to additive or synergistic effects, in particular with respect to sensory irritation and possibly also regarding cytotoxic effects on the nasal mucosa *(3, 11–16)*.

Guidelines

The lowest concentration that has been associated with nose and throat irritation in humans after short-term exposure is 0.1 mg/m^3, although some individuals can sense the presence of formaldehyde at lower concentrations.

To prevent significant sensory irritation in the general population, an air quality guideline value of 0.1 mg/m^3 as a 30-minute average is recommended. Since this is over one order of magnitude lower than a presumed threshold for cytotoxic damage to the nasal mucosa, this guideline value represents an exposure level at which there is a negligible risk of upper respiratory tract cancer in humans.

References

1. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
2. *Formaldehyde.* Geneva, World Health Organization, 1989 (Environmental Health Criteria, No. 89).
3. Formaldehyde. *In*: *Wood dust and formaldehyde.* Lyons, International Agency for Research on Cancer, 1995 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 62), pp. 217–362.
4. Blair, A. et al. Epidemiologic evidence on the relationship between formaldehyde exposure and cancer. *Scandinavian journal of work, environment and health*, **16**: 381–393 (1990).
5. Partanen, T. Formaldehyde exposure and respiratory cancer – a meta-analysis of the epidemiologic evidence. *Scandinavian journal of work, environment and health*, **19**: 8–15 (1993).
6. McLaughlin, J.K. Formaldehyde and cancer: a critical review. *International archives of occupational and environmental health*, **66**: 295–301 (1994).

7. HANSEN, J. & OLSEN, J.H. Formaldehyde and cancer morbidity among male employees in Denmark. *Cancer causes and control*, **6**: 354–360 (1995).
8. SAUNDER, L.R. ET AL. Acute pulmonary response to formaldehyde exposure in healthy nonsmokers. *Journal of occupational medicine*, **28**: 420–424 (1986).
9. SAUNDER, L.R. ET AL. Acute pulmonary response of asthmatics to 3.0 ppm formaldehyde. *Toxicology and industrial health*, **3**: 569–578 (1987).
10. GREEN, D.J. ET AL. Acute response to 3.0 ppm formaldehyde in exercising healthy nonsmokers and asthmatics. *American review of respiratory diseases*, **135**: 1261–1266 (1987).
11. CASSEE, F.R. & FERON, V.J. Biochemical and histopathological changes in nasal epithelium of rats after 3-day intermittent exposure to formaldehyde and ozone alone or in combination. *Toxicology letters*, **72**: 257–268 (1994).
12. LAM, C.-W. ET AL. Depletion of nasal mucosal glutathione by acrolein and enhancement of formaldehyde-induced DNA–protein cross-linking by simultaneous exposure to acrolein. *Archives of toxicology*, **58**: 67–71 (1985).
13. CHANG, J.C.F. & BARROW C.S. Sensory irritation tolerance and cross-tolerance in F-344 rats exposed to chlorine or formaldehyde gas. *Toxicology and applied pharmacology*, **76**: 319–327 (1984).
14. BABIUK, C. ET AL. Sensory irritation response to inhaled aldehydes after formaldehyde pretreatment. *Toxicology and applied pharmacology*, **79**: 143–149 (1985).
15. GRAFSTRÖM, R.C. ET AL. Genotoxicity of formaldehyde in cultured human bronchial fibroblasts. *Science*, **228**: 89–91 (1985).
16. GRAFSTRÖM, R.C. ET AL. Mutagenicity of formaldehyde in Chinese hamster lung fibroblasts: synergy with ionizing radiation and N-nitroso-N-methylurea. *Chemical–biological interactions*, **86**: 41–49 (1993).

5.9 Polycyclic aromatic hydrocarbons

Exposure evaluation

Polycyclic aromatic hydrocarbons (PAHs) are formed during incomplete combustion or pyrolysis of organic material and in connection with the worldwide use of oil, gas, coal and wood in energy production. Additional contributions to ambient air levels arise from tobacco smoking, while the use of unvented heating sources can increase PAH concentrations in indoor air. Because of such widespread sources, PAHs are present almost everywhere. PAHs are complex mixtures of hundreds of chemicals, including derivatives of PAHs, such as nitro-PAHs and oxygenated products, and also heterocyclic PAHs. The biological properties of the majority of these compounds are as yet unknown. Benzo[*a*]pyrene (BaP) is the PAH most widely studied, and the abundance of information on toxicity and occurrence of PAHs is related to this compound. Current annual mean concentrations of BaP in major European urban areas are in the range 1–10 ng/m^3. In rural areas, the concentrations are < 1 ng/m^3 *(1–5)*.

Food is considered to be the major source of human PAH exposure, owing to PAH formation during cooking or from atmospheric deposition of PAHs on grains, fruits and vegetables. The relative contribution of airborne PAH pollutants to food levels (via fallout) has not been well characterized *(6)*.

Health risk evaluation

Data from animal studies indicate that several PAHs may induce a number of adverse effects, such as immunotoxicity, genotoxicity, carcinogenicity and reproductive toxicity (affecting both male and female offspring), and may possibly also influence the development of atherosclerosis. The critical endpoint for health risk evaluation is the well documented carcinogenicity of several PAHs *(7)*.

BaP is by far the most intensively studied PAH in experimental animals. It produces tumours of many different tissues, depending on the species tested and the route of application. BaP is the only PAH that has been tested for carcinogenicity following inhalation, and it produced lung tumours in hamsters, the only species tested. Induction of lung tumours in rats and hamsters has also been documented for BaP and several other PAHs following direct application, such as intratracheal instillation into the pulmonary tissue. The lung carcinogenicity of BaP can be enhanced by coexposure to other substances such as cigarette smoke, asbestos and probably also airborne particles. Several studies have shown that the benzene-soluble fraction, containing 4- to 7-ring PAHs of condensates from car exhausts, domestic coal-stove emissions and tobacco smoke, contains nearly all the carcinogenic potential of PAHs from these sources *(8)*.

Because several PAHs have been shown to be carcinogenic, and many more have been shown to be genotoxic in *in vitro* assays, a suitable indicator for the carcinogenic fraction of the large number of PAHs in ambient air is desirable. The most appropriate indicator for the carcinogenic PAHs in air seems to be BaP concentrations, given present knowledge and the existing database. Assessment of risks to health of a given mixture of PAHs using this indicator approach would entail, first, measurement of the concentration of BaP in a given mixture present in a medium such as air. Then, assuming that the given mixture resembles that from coke ovens, the unit risk estimate is applied in tandem with the measured BaP air concentration to obtain the lifetime cancer risk at this exposure level.

The proportions of different PAHs detected in different emissions and workplaces sometimes differ widely from each other and from PAH profiles in ambient air. Nevertheless, the profiles of PAHs in ambient air do not seem to differ very much from one area to another, although large variations may be seen under special conditions. Furthermore, the carcinogenicity of PAH mixtures may be influenced by synergistic and antagonistic effects of other compounds emitted together with PAHs during incomplete combustion. It should also be recognized that in ambient air the carcinogenic 4- to 7-ring PAHs (representing the majority of PAHs) are preferentially attached to particles and only a minor fraction, depending on the temperature, exists as volatiles. A few studies indicate that the toxicokinetic properties of inhaled BaP attached to particles are different from those of pure BaP alone. Virtually nothing is known about other PAHs in this respect.

Risk assessments and potency assessments of various individual PAHs and complex mixtures of PAHs have been attempted. BaP is the only PAH for which a database is available, allowing a quantitative risk assessment. Risk assessment of BaP is, however, hampered by the poor quality of the data sets available *(9)*.

Attempts to derive relative potencies of individual PAHs (relative to BaP) have also been published, and the idea of summarizing the contributions from each of the selected PAHs into a total BaP equivalent dose (assuming their carcinogenic effects to be additive) has emerged *(10, 11)*. There are doubts, however, about the scientific justification for these procedures.

Risk estimates considered in the United States for coke-oven emissions were used in the first edition of these guidelines. Using a linearized multistage model, the most plausible upper-bound individual lifetime unit risk estimate associated with a continuous exposure to 1 $\mu g/m^3$ of benzene-soluble compounds of coke-oven emissions in ambient air was approximately 6.2×10^{-4}. Using BaP as an indicator of general PAH mixtures from emissions of coke ovens and similar combustion processes in urban air, and a reported value of 0.71% BaP in the benzene-soluble fraction of coke oven emissions, a lifetime risk of respiratory cancer of 8.7×10^{-5} per ng/m^3 was calculated *(1)*.

From the lung tumour rates obtained in a recent rat inhalation study with coal tar/pitch condensation aerosols, containing two different levels of BaP, a lifetime tumour risk of 2×10^{-5} per ng/m^3 for BaP as a constituent of a complex mixture was calculated using a linearized multistage model *(12)*.

Guidelines

No specific guideline value can be recommended for PAHs as such in air. These compounds are typically constituents of complex mixtures. Some PAHs are also potent carcinogens, which may interact with a number of other compounds. In addition, PAHs in air are attached to particles, which may also play a role in their carcinogenicity. Although food is thought to be the major source of human exposure to PAHs, part of this contamination may arise from air pollution with PAHs. The levels of PAHs in air should therefore be kept as low as possible.

In view of the difficulties in dealing with guidelines for PAH mixtures, the advantages and disadvantages of using a single indicator carcinogen to represent the carcinogenic potential of a fraction of PAH in air were considered. Evaluation of, for example, BaP alone will probably underestimate the carcinogenic potential of airborne PAH mixtures, since co-occurring substances are also carcinogenic. Nevertheless, the well studied common constituent of PAH mixtures, BaP, was chosen as an indicator, although the limitations and uncertainties in such an approach were recognized.

To set priorities with respect to control, an excess lifetime cancer risk, expressed in terms of the BaP concentration and based on observations in coke-oven workers exposed to mixtures of PAHs, is presented here. It must be emphasized that the composition of PAHs to which coke-oven workers are exposed may not be similar to that in ambient air, although it was noted that similar risks have been derived from studies of individuals exposed to other mixtures containing PAHs. Having also taken into consideration some recent animal data from which a unit risk of the same order of magnitude can be derived, it was concluded that the occupational epidemiology data should serve as the basis for the risk estimate.

Based on epidemiological data from studies in coke-oven workers, a unit risk for BaP as indicator air constituent for PAHs is estimated to be 8.7×10^{-5} per ng/m^3, which is the same as that established by WHO in 1987. The corresponding concentrations of BaP producing excess lifetime cancer risks of 1/10 000, 1/100 000 and 1/1 000 000 are 1.2, 0.12 and 0.012 ng/m^3, respectively.

References

1. Polynuclear aromatic hydrocarbons (PAH). *In*: *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987, pp. 105–117.
2. *Toxicological profile for polycyclic aromatic hydrocarbons (PAHs): update.* Atlanta, GA, Agency for Toxic Substances and Disease Registry, 1994.
3. BAEK, S.O. ET AL. A review of atmospheric polycyclic aromatic hydrocarbons: sources, fate and behavior. *Water, air, and soil pollution,* **60**: 279–300 (1991).
4. PFEFFER, H.U. Ambient air concentrations of pollutants at traffic-related sites in urban areas of North Rhine-Westphalia, Germany. *Science and the total environment,* **146/147**: 263–273 (1994).
5. NIELSEN, T. ET AL. *Traffic PAH and other mutagens in air in Denmark.* Copenhagen, Danish Environmental Protection Agency, 1995 (Miljøprojekt No. 285).
6. DE VOS, R.H. ET AL. Polycyclic aromatic hydrocarbons in Dutch total diet samples (1984–1986). *Food chemistry and toxicology,* **28**: 263–268 (1990).
7. *Polynuclear aromatic compounds. Part 1. Chemical, environmental and experimental data.* Lyons, International Agency for Research on Cancer, 1983 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 32).
8. POTT, F. & HEINRICH, U. Relative significance of different hydrocarbons for the carcinogenic potency of emissions from various incomplete combustion processes. *In*: Vainio, H. et al., ed. *Complex mixtures and cancer risk.* Lyons, International Agency for Research on Cancer, 1990, pp. 288–297 (IARC Scientific Publications, No. 104).
9. COLLINS, J.F. ET AL. Risk assessment for benzo[*a*]pyrene. *Regulatory toxicology and pharmacology,* **13**: 170–184 (1991).
10. NISBET, I.C.T. & LAGOY, K. Toxic equivalency factors (TEFs) for polycyclic aromatic hydrocarbons (PAHs). *Regulatory toxicology and pharmacology,* **16**: 290–300 (1992).

11. RUGEN, P.J. ET AL. Comparative carcinogenicity of the PAHs as a basis for acceptable exposure levels (AELs) in drinking water. *Regulatory toxicology and pharmacology,* **9**: 273–283 (1989).
12. HEINRICH, U. ET AL. Estimation of a lifetime unit lung cancer risk for benzo[*a*]pyrene based on tumour rates in rats exposed to coal tar/pitch condensation aerosol. *Toxicology letters,* **72**: 155–161 (1994).

5.10 Polychlorinated biphenyls

Exposure evaluation

Analysis of polychlorinated biphenyls (PCBs) should be performed by congener-specific methods. The method of quantifying total PCBs, by comparing the sample peak pattern with that of a commercial mixture, is accurate only when the sample under investigation has been directly contaminated by a commercial mixture. Because of substantial differences in PCB patterns between biological samples and technical products, however, this method leads to errors in the quantification of biological samples and also to differences between laboratories owing to the use of different standard mixtures. As a consequence, data have to be interpreted with great care. Comparisons can only be made between data either from the same laboratory, using the same validated technique and the same standards over a longer period, or from different laboratories when very strict interlaboratory controls have been applied. Indications of trends can only be obtained when these considerations are taken into account.

Food

Food is the main source of human intake of PCBs; intake through drinking-water is negligible.

The daily intake of total PCBs in Sweden was recently estimated at 0.05 µg/kg body weight (BW), with a 50% contribution from fish *(1)*. This is markedly lower than an earlier Finnish estimate of 0.24 µg/kg BW *(2)*, and might reflect the decreasing trends in PCB levels in Nordic food. Recent data from the Nordic countries indicate that the current average daily intake in toxic equivalents of dioxin-like PCBs may be slightly above 1 pg/kg BW *(3, 4)*.

If the contributions of PCDDs and PCDFs are also taken into account, the daily intake in toxic equivalents would be in the range 2–6 pg/kg BW for many European countries and the United States *(5)*. For certain risk groups, such as fishermen from the Baltic Sea and Inuits in the Arctic who consume large amounts of contaminated fatty fish, the intake may be up to four times higher.

Air

PCB levels have been shown to be higher in indoor air than in ambient air. Inhalation exposure to PCBs, assuming an indoor air level of 3 ng/m^3 in an uncontaminated building and an inhaled volume of 20 m^3 of air per day for adults, is approximately 0.001 μg/kg BW per day. In contaminated buildings concentrations above 300 ng/m^3 have been found, corresponding to a daily dose of at least 0.1 μg/kg BW. In buildings using PCB-containing sealants, levels up to 7500 ng/m^3 have been found (corresponding to a daily dose of 2.5 μg/kg BW). In ambient air there is a wide variation in the measurements from nonindustrialized (e.g. 0.003 ng/m^3) and industrial/urban areas (e.g. 3 ng/m^3). The levels of dioxin-like PCBs cannot be estimated owing to the lack of congener-specific analytical data.

Health risk evaluation

In 1990, the Joint FAO/WHO Expert Committee on Food Additives concluded that, owing to the limitations of the available data, it was impossible to establish a precise numerical value for a tolerable intake of total PCBs for humans *(6)*. IARC concluded that available studies suggested an association between human cancer and exposure to PCBs *(7)*. Overall, PCBs were classified as probably carcinogenic to humans (Group 2A), although several national governments are employing tolerable daily intakes (TDIs) for PCBs for the purpose of risk management.

In Germany a TDI for PCB of 1–3 μg/kg BW has been suggested. It was also recommended that, for precautionary reasons, the proportional daily intake via indoor air should not exceed 10% of the TDI for long periods. On this basis an action level for source removal of 3000 ng/m^3 has been derived. For concentrations between 3000 ng/m^3 and 10 000 ng/m^3 (that is, between 3 μg/m^3 and 10 μg/m^3) a concrete health risk is not assumed. However, mitigation measures should be undertaken as soon as possible to reduce the level to 300 ng/m^3, below which concentrations are thought to be of no concern. Source removal should also be undertaken if levels are found to be between 300 and 3000 ng/m^3 *(8)*.

Neurobehavioural and hormonal effects have been observed in infants exposed to background concentrations of PCBs, prenatally and/or through breastfeeding. The clinical significance of these observations is, however, unclear.

On average, the contribution from inhalation exposure is approximately 1% of the dietary intake but may approach that intake in certain extreme situations (areas close to sources or contaminated indoor air).

Exposures to dioxin-like PCBs can be converted to toxic equivalents using the WHO/IPCS interim toxic equivalent factors *(9)* and subsequently be assessed using the TDI for TCDD. In 1992, WHO established a TDI for TCDD of 10 pg/kg BW. This was derived on the basis of TCDD-induced liver cancer in rats *(10)* for which a NOAEL of 1 ng/kg BW per day, corresponding to a liver concentration of 540 ng/kg on a wet-weight basis, was calculated. Owing to toxicokinetic differences between humans and rats, this would correspond to a daily intake in humans of 100 pg/kg BW, to which value an uncertainty factor of 10 (to cover inter-individual variation) was applied. Although not explicitly stated, the TDI can be looked on as applicable to the total intake of toxic equivalents derived from PCDDs, PCDFs and other dioxin-like compounds that act by the same mechanisms and cause similar types of toxicity.

For the average consumer, the daily intake of dioxin-like PCBs determined as toxic equivalents would be 10–30% of the TDI. When the contribution from the PCDDs and PCDFs is taken into account, the intake would increase to 20–60%. There are, however, groups with specific dietary habits (such as a high intake of contaminated food) or occupational exposure that may exceed the TDI for PCDDs and PCDFs.

The WHO human milk exposure study *(11)* indicated that the daily intake in toxic equivalents of PCDDs and PCDFs in breastfeeding infants in industrialized countries ranged from about 20 pg/kg BW in less industrialized areas to about 130 pg/kg BW in highly industrialized areas. This indicates intakes 2–13 times higher than the TDI. When the contribution from dioxin-like PCBs is taken into account, the intakes may be up to 2 times higher. It has been noted, however *(12)*, that the TDI should not be applied to such infants because the TDI concept relates to a dose ingested throughout a lifetime. The quantity of PCDDs and PCDFs ingested over a 6-month breastfeeding period would be less than 5% of the quantity ingested over a lifetime.

Guidelines

An air quality guideline for PCBs is not proposed because direct inhalation exposures constitute only a small proportion of the total exposure, in the order of 1–2% of the daily intake from food. WHO has not developed a TDI for total PCB exposure. Owing to the multiplicity of mechanisms underlying PCB-induced health effects, there may not be a scientifically sound rationale to set such a TDI. Average ambient air concentrations of PCBs are estimated to be 3 ng/m^3 in urban areas. Although this air concentration is only a minor contributor to direct human exposure, it is a major contributor to contamination of the food chain. It would also be possible to perform such calculations using toxic equivalents for dioxin-like PCBs in ambient air, but no such analytical data have been published.

Although indoor air levels of PCBs are generally very low, in certain instances levels of up to several μg/m^3 have been detected. For people living or working in such buildings, exposure to PCBs via air could contribute significantly to the overall PCB exposure.

Because of the potential importance of the indirect contribution of PCBs in air to total human exposure, it is important to control known sources as well as to identify new sources.

References

1. Darnerud, P.O. et al. Bakgrund till de reviderade kostråden. PCB och dioxiner i fisk [Background to the revised dietary guidelines. PCB and dioxins in fish]. *Vår föda,* **47**: 10–21 (1995).
2. Moilanen, R. et al. Average total dietary intakes of organochlorine compounds from the Finnish diet. *Zeitung für Lebensmittel Untersuchung und Forschung,* **182**: 484–488 (1986).
3. Færden, K. *Dioksiner i næringsmidler*. [Dioxins in food]. Oslo, National Food Control Authority, 1991 (SNT-Rapport No. 4).
4. Svenson, B.G. et al. Exposure to dioxins and dibenzofurans through consumption of fish. *New England journal of medicine*, 324; 8–1 (1991).
5. *International toxicity equivalency factors (I–TEF) method of risk assessment for complex mixtures of dioxins and related compounds.* Brussels, North Atlantic Treaty Organization, 1988 (Report No. 176).
6. *Evaluation of certain food additives and contaminants. Thirty-fifth report of the Joint FAO/WHO Expert Committee on Food Additives*. Geneva, World Health Organization, 1990 (WHO Technical Report Series, No. 789).
7. *Overall evaluations of carcinogenicity: an updating of IARC monographs volumes 1 to 42*. Lyons, International Agency for Research on Cancer, 1987 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Supplement 7).
8. Rosskamp, E. Polychloriente Biphenyle in der Innerraumluft – Sachstand. *Bundesgesundheitsblatt,* **35**: 434 (1992).
9. Ahlborg, U.G. et al. Toxic equivalency factors for dioxin-like PCBs. Report on a WHO-ECEH and IPCS consultation, December 1993. *Chemosphere,* **28**: 1049–1067 (1994).
10. Kociba, R.J. et al. Results of a two year chronic toxicity and oncogenicity study of 2,3,7,8-tetrachlordibenzo-*p*-dioxin (TCDD) in rats. *Toxicology and applied pharmacology,* **46**: 279–303 (1978).

11. *Levels of PCBs, PCDDs and PCDFs in human milk. Second round of WHO coordinated exposure study.* Geneva, World Health Organization, 1992 (Environmental Health Series, No.3).
12. AHLBORG, U.G. ET AL., ED. Special issue: tolerable daily intake of PCDDs and PCDFs. *Toxic substances journal,* **12**: 101–131 (1992).

5.11 Polychlorinated dibenzodioxins and dibenzofurans

Exposure evaluation

Food is the main source of human intake of polychlorinated dibenzodioxins (PCDDs) and dibenzofurans (PCDFs); intake through drinking-water is negligible. Calculated as toxic equivalents, average intakes in European countries have been estimated to be in the range 1.5–2 pg/kg body weight (BW) per day *(1–3)*. Very recent data from the Nordic countries indicate that this figure today may be slightly less than 1 pg/kg BW per day *(4, 5)*. For the United States, intake estimates are in the range 1–3 pg/kg BW per day *(6)*.

If the contributions of dioxin-like polychlorinated biphenyls (PCBs) are taken into account, and using the WHO toxic equivalency factors (TEFs) for PCBs *(7, 8)*, the toxic equivalent intake would be in the range 2–6 pg/kg BW per day. For certain risk groups, such as fishermen from the Baltic Sea and Inuits in the Arctic, intakes may be considerably higher.

Inhalation exposure to PCDDs and PCDFs is generally low. Assuming an ambient air toxic equivalent level of 0.1 pg/m^3 and an inhaled volume of air of 20 m^3/day for adults, inhalation intake would amount to about 0.03 pg/kg BW per day *(9, 10)*. Certain industrial and urban areas, however, as well as areas close to major sources, may have up to 20 times higher air concentrations. The contribution to the total toxic equivalents of dioxin-like PCBs from ambient air cannot be calculated owing to lack of congener-specific data. Under special circumstances, for example indoor air highly contaminated from coated particle boards containing PCBs, inhalation exposure may reach 1 pg/kg BW per day *(11)*.

Although present concentrations of PCDDs and PCDFs in ambient air do not present a health hazard through direct human exposure, these concentrations will lead to deposition of PCDDs and PCDFs followed by uptake through the food chain.

Health risk evaluation

In 1990, WHO established a tolerable daily intake (TDI) for TCDD of 10 pg/kg BW *(12)*. This was based on TCDD-induced liver cancer in rats *(13)* for which the NOAEL was 1 ng/kg BW. Owing to toxicokinetic differences between humans and rats, this corresponded to a daily intake in humans of 100 pg/kg BW, to which value an uncertainty factor of 10 (to cover inter-individual variation) was applied.

Since then, new data on hormonal, reproductive and developmental effects at low doses in animal studies (rats and monkeys) have been published, and the health risk of dioxins was therefore reassessed in 1998 *(14, 15)*. It was concluded that the human data do not lend themselves to be used as the basis for setting a TDI, but they were nevertheless considered to constitute an important reference for comparison with a health risk assessment based on animal data. Consequently, the TDI was based on animal data. It was further decided that body burdens should be used to scale doses across species. Human daily intakes corresponding to body burdens similar to those associated with LOAELS in rats and monkeys could be estimated to be in the range of 14–37 pg/kg BW per day. By applying an uncertainty factor of 10 to this range of LOAELs, a TDI expressed as a range of 1–4 pg toxic equivalent per kg BW was established for dioxins and dioxin-like compounds.

The TDI represents a tolerable daily intake for lifetime exposure, and occasional short-term excursions above the TDI would have no health consequences provided that the averaged intake over long periods was not exceeded. Although not explicitly stated, the TDI can be looked on as applicable to the total intake of toxic equivalents, via both the oral and inhalation routes, derived from PCDDs and PCDFs and other dioxin-like compounds that act by the same mechanisms and cause similar types of toxicity.

The average daily intake by all routes of exposure to PCDDs and PCDFs, calculated as toxic equivalents, is in the same range as the current TDI. When the contribution from dioxin-like PCBs is taken into account, the intake increases by a factor of 2–3. There are, however, groups with specific dietary habits (such as a high intake of contaminated food) or occupational exposure, that may have exposures in excess of the TDI for PCDDs and PCDFs.

The daily intake of PCDDs and PCDFs in breastfed infants in industrialized countries has been calculated in toxic equivalents to range from about 20 pg/kg BW in less industrialized areas up to about 130 pg/kg BW in more industrialized areas. When the contribution from dioxin-like PCBs is taken into account, the intakes may be up to twice these figures. This indicates intakes being far above the TDI. WHO noted, however, that the TDI should not be applied to breastfed infants because the concept of TDI relates to a dose ingested throughout a lifetime *(14)*. In general, the quantity of PCDDs and PCDFs ingested over a 6-month breastfeeding period would be less than 5% of the quantity ingested over a lifetime.

The contribution from inhalation exposure is on average approximately 1% of the dietary intake, but may in certain extreme situations (areas close to point emission sources or contaminated indoor air) approach the dietary intake.

Guidelines

An air quality guideline for PCDDs and PCDFs is not proposed because direct inhalation exposures constitute only a small proportion of the total exposure, generally less than 5% of the daily intake from food.

Urban ambient toxic equivalent air concentrations of PCDDs and PCDFs are estimated to be about 0.1 pg/m^3. However, large variations have been measured. Although such an air concentration is only a minor contributor to direct human exposure, it is a major contributor to contamination of the food chain. It is difficult, however, to calculate indirect exposure from contamination of food via deposition from ambient air. Mathematical models are being used in the absence of experimental data, but these models require validation. Air concentrations of 0.3 pg/m^3 or higher are indications of local emission sources that need to be identified and controlled.

Although indoor air levels of PCDDs and PCDFs are generally very low, in certain instances, toxic equivalent levels of up to 3 pg/m^3 have been detected. Such levels will constitute an exposure ranging from 25% up to 100% of the current TDI of 1–4 pg toxic equivalent per kg BW (corresponding to 60–240 pg toxic equivalent per day for a 60-kg person).

Owing to the potential importance of the indirect contribution of PCDDs and PCDFs in air to the total human exposure to these compounds through deposition and uptake in the food chain, measures should be undertaken to further reduce emissions to air from known sources. For risk reduction, it is important to control known sources as well as to identify new sources.

References

1. BECK, H. ET AL. PCDD and PCDF body burden from food intake in the Federal Republic of Germany. *Chemosphere,* **18**: 417–424 (1989).
2. MINISTRY OF AGRICULTURE FISHERIES AND FOOD. *Dioxins in food*. London, H. M. Stationery Office, 1992 (Food Surveillance Paper, No. 31).

3. THEELEN, R.M.C. Modeling of human exposure to TCDD and I–TEQ in the Netherlands: background and occupational. *In:* Gallo, M.A. et al., ed. *Biological basis for risk assessment of dioxins and related compounds.* New York, Cold Spring Harbor Laboratory Press, 1991, pp. 277–290 (Banbury Report, No. 35).
4. FÆRDEN, K. *Dioksiner i næringsmidler*. [Dioxins in food]. Oslo, National Food Control Authority, 1991 (SNT-Rapport No. 4).
5. SVENSON, B.G. ET AL. Exposure to dioxins and dibenzofurans through consumption of fish. *New England journal of medicine,* **324**: 8–12 (1991).
6. *Health assessment document for 2,3,7,8–tetrachlorodibenzo-*p*-dioxin (TCDD) and related compounds.* Washington, DC, US Environmental Protection Agency, 1994 (Final report EPA-600/BP-92-001c).
7. AHLBORG, U.G. ET AL. Impact of polychlorinated dibenzo-*p*-dioxins, dibenzofurans and biphenyls on human and environmental health, with special emphasis on application of the toxic equivalency factor concept. *European journal of pharmacology,* **228**: 179–199 (1992).
8. *International toxicity equivalency factors (I–TEF) method of risk assessment for complex mixtures of dioxins and related compounds.* Brussels, North Atlantic Treaty Organization, 1988 (Report No. 176).
9. WEVERS, M. ET AL. Concentrations of PCDDs and PCDFs in ambient air at selected locations in Flanders. *Organohalogen compounds,* **12**: 123–126 (1993).
10. DUARTE-DAVIDSON, R. ET AL. Polychlorinated dibenzo-*p*-dioxins (PCDDs) and furans (PCDFs) in urban air and deposition. *Environmental science and pollution research,* **1**: 262–270 (1994).
11. BALFANZ, E. ET AL. Sampling and analysis – polychlorinated biphenyls (PCB) in indoor air due to permanently elastic sealants. *Chemosphere*, **26**: 871–880 (1993).
12. AHLBORG, U.G. ET AL., ED. Special issue: tolerable daily intake of PCDDs and PCDFs. *Toxic substances journal,* **12**: 101–131 (1992).
13. KOCIBA, R.J. ET AL. Results of a two year chronic toxicity and oncogenicity study of 2,3,7,8-tetrachlordibenzo-*p*-dioxin (TCDD) in rats. *Toxicology and applied pharmacology,* **46**: 279–303 (1978).
14. VAN LEEUWEN, F.X.R. & YOUNES, M.M. Assessment of the health risk of dioxins: re-evaluation of the tolerable daily intake (TDI). *Food additives and contaminants*, **17**: 223–240 (2000).
15. VAN LEEUWEN, F.X.R. ET AL. Dioxins: WHO's Tolerable Daily Intake (TDI) revisited. *Chemosphere*, **40**: 1095–1101 (2000).

5.12 Styrene

Exposure evaluation

Concentrations of styrene in rural ambient air are generally less than 1 μg/m³, while indoor air in such locations may contain several μg/m³. Levels in polluted urban areas are generally less than 20 μg/m³ but can be much higher in newly built houses containing styrene-based materials.

Health risk evaluation

Potentially critical effects for the derivation of a guideline for styrene are considered to be carcinogenicity/genotoxicity and neurological effects, including effects on development.

Styrene in its pure form has an odour detection threshold of 70 μg/m³. Its pungent odour is recognized at concentrations three to four times greater than this threshold value.

The value of the available evidence for an association between exposure to styrene and small increases in lymphatic and haematopoietic cancers observed in workers in some studies is limited by concurrent exposure to other substances, lack of specificity and absence of dose–response. In limited studies in animals, there is little evidence that styrene is carcinogenic. IARC has classified styrene in group 2B *(1)*.

Styrene was genotoxic *in vivo* and *in vitro* following metabolic activation. In cytogenetic studies on peripheral lymphocytes of workers in the reinforced plastics industry, there were increased rates of chromosomal aberrations at mean levels of styrene of more than 120 mg/m³ (> 20 ppm). Elevated levels of single-strand breaks and styrene-7,8-oxide adducts in DNA and haemoglobin have also been observed. Although these genotoxic effects have been observed at relatively low concentrations, they were not considered as critical endpoints for development of a guideline, in view of the equivocal evidence of carcinogenicity for styrene.

The available data, although limited, indicate that neurotoxicity in the form of neurological developmental impairments is among the most sensitive of endpoints. In the offspring of rats exposed to styrene at a concentration of 260 mg/m³ (60 ppm) there were effects on behaviour and biochemical parameters in the brain *(2)*.

Guidelines

Although genotoxic effects in humans have been observed at relatively low concentrations, they were not considered as critical endpoints for development of a guideline, in view of the equivocal evidence for the carcinogenicity of styrene.

In occupationally exposed populations, subtle effects such as reductions in visuomotor accuracy and verbal learning skills *(3–5)* and subclinical effects on colour vision have been observed at concentrations as low as 107–213 mg/m^3 (25–50 ppm) *(6–10)*. Taking the lower number of this range for precautionary reasons, adjusting this to allow for conversion from an occupational to a continuous pattern of exposure (a factor of 4.2), and incorporating a factor of 10 for inter-individual variation and 10 for use of a LOAEL rather than a NOAEL results in a guideline of 0.26 mg/m^3 (weekly average). This value should also be protective for the developmental neurological effects observed in animal species.

The air quality guideline could also be based on the odour threshold. In that case, the peak concentration of styrene in air should be kept below the odour detection threshold level of 70 µg/m^3 as a 30-minute average.

References

1. *Some industrial chemicals*. Lyons, International Agency for Research on Cancer, 1994, pp. 233–320 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 60).
2. KISHI, R. ET AL. Neurochemical effects in rats following gestational exposure to styrene. *Toxicology letters*, **63**: 141–146 (1992).
3. HÄRKÖNEN, H. Exposure–response relationship between styrene exposure and central nervous function. *Scandinavian journal of work, environment and health,* **4**: 53–59 (1978).
4. LINDSTRÖM, K. ET AL. Relationships between changes in psychological performances and styrene exposure in work with reinforced plastics. *Työ ja ihminen,* **6**: 181–194 (1992) [Swedish].
5. MUTTI, A. ET AL. Exposure–effect and exposure–response relationships between occupational exposure to styrene and neuropsychological functions. *American journal of industrial medicine,* **5**: 275–286 (1984).
6. GOBBA, F. ET AL. Acquired dyschromatopsia among styrene-exposed workers. *Journal of occupational medicine,* **33**: 761–765 (1991).
7. FALLAS, C. ET AL. Subclinical impairment of colour vision among workers exposed to styrene. *British journal of industrial medicine,* **49**: 679–682 (1992).

8. GOBBA, F. & CAVALLERI, A. Kinetics of urinary excretion and effects on colour vision after exposure to styrene. *In:* Sorsa, M. et al., ed. *Butadiene and styrene: assessment of health hazards.* Lyons, International Agency for Research on Cancer, 1993 (IARC Scientific Publication, No. 127), pp. 79–88.
9. CHIA, S.E. ET AL. Impairment of colour vision among workers exposed to low concentrations of styrene. *American journal of industrial medicine,* **26**: 481–488 (1994).
10. EGUCHI, T. ET AL. Impaired colour discrimination among workers exposed to styrene: relevance to a urinary metabolite. *Occupational and environmental medicine,* **52**: 534–538 (1995).

5.13 Tetrachloroethylene

Exposure evaluation

Ambient air concentrations of tetrachloroethylene are generally less than 5 μg/m^3 in urban areas and typically less than 1 μg/m^3 in rural areas. Indoor concentrations are generally less than 5 μg/m^3. Indoor tetrachloroethylene air levels may rise to more than 1 mg/m^3 in close proximity to dry-cleaning operations where tetrachloroethylene is used as a cleaning solvent or in homes where dry-cleaned clothing is often worn. Inhalation of tetrachloroethylene is the major route of exposure in the general population.

Health risk evaluation

The main health effects of concern are cancer and effects on the central nervous system, liver and kidneys. Tetrachloroethylene is classified by IARC as a Group 2A carcinogen (probably carcinogenic to humans) *(1)*.

In carcinogenicity studies, an increased incidence of adenomas and carcinomas was observed in the livers of exposed mice. There is suggestive evidence from mechanistic studies that humans are less sensitive to the development of these tumours following tetrachloroethylene exposure. A low incidence of kidney tumours has been reported among male rats. It can be concluded from this small and statistically non-significant increase, together with the data related to a possible mechanism of induction, that the result in male rats is equivocal evidence only for a risk of renal cancer in humans. The significance for humans of the increased incidences of mononuclear-cell leukaemias, as observed in a study in F344 rats, is unclear owing to the lack of understanding of the mechanism underlying the formation of this cancer type, which has a high background incidence.

Epidemiological studies in humans show positive associations between exposure to tetrachloroethylene and risks for oesophageal and cervical cancer and non-Hodgkin lymphoma. Confounding factors cannot be ruled out and the statistical power of the studies is limited. These studies therefore provide only limited evidence for the carcinogenicity of tetrachloroethylene in humans *(1)*.

From the weight of the evidence from mutagenicity studies, it can be concluded that tetrachloroethylene is not genotoxic. Several *in vitro* studies indicate that conjugation of tetrachloroethylene with reduced glutathione, a minor biotransformation route demonstrated to occur in rodents, produces renal metabolites that are mutagenic in *Salmonella typhimurium* TA 100 *(1)*. In the absence of further data on this point, the significance of the latter results for humans is uncertain.

Short-term exposure studies in volunteers (duration 1 or 5 days) have shown effects on the central nervous system at a concentration of > 678 mg/m^3 *(2–5)*. A recent study of dry-cleaning workers with long-term exposure showed that renal effects may develop at lower exposure concentrations, with the reported onset of renal damage occurring following exposure to a median concentration of 102 mg/m^3 (range, trace to 576 mg/m^3) *(6)*.

Although the results of carcinogenicity studies in experimental animals are available, those of adequate long-term toxicity studies are not. A chronic LOAEL of 678 mg/m^3 (100 ppm) for the systemic toxicity (in kidney and liver) of tetrachloroethylene in mice can be derived from the National Toxicology Program carcinogenicity study in this species *(7)*.

Use of existing physiologically based pharmacokinetic models for derivation of a guideline value based on kidney effects is not considered feasible because these models do not contain the kidney or kidney-specific metabolism as a component. As yet it is therefore unknown what an appropriate internal dose measure would be.

Guidelines

Given the limitations of the weight of the epidemiological evidence, and the uncertainty of the relevance to humans of the induction of tumours in animals exposed to tetrachloroethylene, the derivation of a guideline value is at present based on non-neoplastic effects rather than on carcinogenicity as the critical endpoint.

On the basis of a long-term LOAEL for kidney effects of 102 mg/m^3 in dry-cleaning workers, a guideline value of 0.25 mg/m^3 is calculated. In deriving this guideline value, the LOAEL is converted to continuous exposure (dividing by a factor of 4.2, 168/40) and divided by an uncertainty factor of 100 (10 for use of an LOAEL and 10 for intraspecies variation). Recognizing that some uncertainty in the LOAEL exists because the effects observed at this level are not clear-cut, and because of fluctuations in exposure levels, an alternative calculation was made based on the LOAEL in mice of 680 mg/m^3, and using an appropriate uncertainty factor of 1000. This calculation yields a guideline value of 0.68 mg/m^3.

On the basis of the overall health risk evaluation, a guideline of 0.25 mg/m^3 is currently established. However, the concern about a possible carcinogenic effect of tetrachloroethylene exposure in humans should be addressed through in-depth risk evaluation in the near future.

References

1. *Dry cleaning, some chlorinated solvents and other industrial chemicals*. Lyons, International Agency for Research on Cancer, 1995 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 63).
2. *Toxicological profile for tetrachloroethylene*. Atlanta, GA, Agency for Toxic Substances and Disease Registry, 1993 (Public Health Service Report, No. TP-92/18).
3. *Tetrachloroethylene*. Geneva, World Health Organization, 1984 (Environmental Health Criteria, No. 31).
4. HAKE, C.L. & STEWART, R.D. Human exposure to tetrachloroethylene: inhalation and skin contact. *Environmental health perspectives,* **21**: 231–238 (1977).
5. ALTMANN, L. ET AL. Neurophysiological and psychological measurements reveal effects of acute low-level organic solvent exposure in humans. *International archives of occupational and environmental health,* **62**: 493–499 (1990).
6. MUTTI, A. ET AL. Nephropathies and exposure to perchloroethylene in dry-cleaners. *Lancet,* **340**: 189–193 (1992).
7. *Technical report on the toxicology carcinogenesis studies of tetrachloroethylene (perchloroethylene) in F344/N rats and B6C3F1 mice (inhalation studies)*. Research Triangle Park, NC, National Toxicology Program, 1986 (NTP Technical Report, No. 311).

5.14 Toluene

Exposure evaluation

Mean ambient air concentrations of toluene in rural areas are generally less than 5 μg/m^3, while urban air concentrations are in the range 5–150 μg/m^3. Concentrations may be higher close to industrial emission sources.

Health risk evaluation

Toluene in its pure form has an odour detection threshold of 1 mg/m^3 (*1, 2*). Its odour is recognized at concentrations about ten times greater than this threshold value (*1–3*).

The acute and chronic effects of toluene on the central nervous system are the effects of most concern. Toluene may also cause developmental decrements and congenital anomalies in humans, and these effects are supported by findings of studies on animals, for example fetal development retardation, skeletal anomalies, low birth weight and developmental neurotoxicity. The potential effects of toluene on reproduction and hormone balance in women, coupled with findings of hormone imbalances in exposed males, are also of concern. Limited information suggests an association between occupational toluene exposure and spontaneous abortions. Both the human and animal data indicate that toluene is ototoxic at elevated exposures. Sensory effects have also been found. Toluene has minimal effects on the liver and kidney, except in cases of toluene abuse. There has been no indication that toluene is carcinogenic in bioassays conducted to date, and the weight of available evidence indicates that it is not genotoxic.

The lowest level of chronic occupational toluene exposure unequivocally associated with neurobehavioural functional decrements is 332 mg/m^3 (88 ppm) *(4, 5)*. Effects on the central nervous system in humans are supported by findings in exposed animals. For example, rat pups exposed to either 100 or 500 ppm 1–28 days after birth demonstrated histo-pathological changes in the hippocampus *(6)*. Women occupationally exposed to toluene at an average concentration of 332 mg/m^3 (88 ppm) incurred higher spontaneous abortion rates and menstrual function disturbances *(7–9)*. The interpretation of these observations was hampered, however, by confounding factors *(10)*. Men occupationally exposed to toluene at 5–25 ppm have also been shown to exhibit hormonal changes.

With regard to short-term exposure, subjective effects have been reported at 100 ppm (6-hour exposure) while symptoms at lower levels cannot be ruled out. Numerous confounding factors, however, need to be considered.

Exposure data related to central nervous system endpoints were best characterized in certain occupational studies and these data have been employed in the derivation of the guideline. A NOAEL for chronic effects of toluene has not been identified.

Guidelines

The LOAEL for effects on the central nervous system from occupational studies is approximately 332 mg/m^3 (88 ppm). A guideline value of 0.26 mg/m^3 is established from these data, adjusting for continuous exposure (dividing by a factor of 4.2) and dividing by an uncertainty factor of 300 (10 for inter-individual variation, 10 for use of a LOAEL rather than a NOAEL, and an additional factor of 3 given the potential effects on the developing central nervous system). This guideline value should be applied as a weekly average. This guideline value should also be protective for reproductive effects (spontaneous abortions).

The air quality guideline could also be based on the odour threshold. In this case, the peak concentrations of toluene in air should be kept below the odour detection threshold level of 1 mg/m^3 as a 30-minute average.

References

1. Hellman, T.M. & Small, F.H. Characterization of petrochemical odors. *Chemical engineering progress*, **69**: 75–77 (1973).
2. Nauš, A. Cichové prahy nekterých prumyslových látek [Olfactory thresholds of industrial substances]. *Pracovni lekarstvi*, **34**: 217–218 (1982).
3. Hellman, T.M. & Small, F.H. Characterization of the odor properties of 101 petrochemicals using sensory methods. *Journal of the Air Pollution Control Association*, **24**: 979–982 (1974).
4. Foo, S.C. et al. Chronic neurobehavioural effects of toluene. *British journal of industrial medicine,* **47**: 480–484 (1990).
5. Foo, S.C. et al. Neurobehavioural effects in occupational chemical exposure. *Environmental research,* **60**: 267–273 (1993).
6. Slomianka, L. et al. The effect of low-level toluene exposure on the developing hippocampal region of the rat: histological evidence and volumetric findings. *Toxicology,* **62**: 189–202 (1990).
7. Ng, T.P. et al. Risk of spontaneous abortion in workers exposed to toluene. *British journal of industrial medicine,* **49**: 804–808 (1992).

8. LINDBOHM, M.-L. ET AL. Spontaneous abortions among women exposed to organic solvents. *American journal of industrial medicine,* **17**: 449–463 (1990).
9. NG, T.P. ET AL. Menstrual function in workers exposed to toluene. *British journal of industrial medicine,* **49**: 799–803 (1992).
10. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).

5.15 Trichloroethylene

Exposure evaluation

The average ambient air concentrations of trichloroethylene are less than 1 μg/m³ in rural areas and up to 10 μg/m³ in urban areas. Concentrations in indoor air are typically similar, although higher concentrations can be expected in certain areas, such as in proximity to industrial operations. Inhalation of airborne trichloroethylene is the major route of exposure for the general population.

Health risk evaluation

The main health effects of concern with trichloroethylene are cancer, and effects on the liver and the central nervous system.

Studies in animals and humans show that the critical organs or systems for noncarcinogenic effects are the liver and the central nervous system. The dose–response relationship for these effects is insufficiently known, making it difficult to assess the health risk for the occurrence of these effects in case of long-term exposure to low levels of trichloroethylene.

IARC has classified trichloroethylene as a Group 2A carcinogen (probably carcinogenic to humans). This classification was based on sufficient evidence in animals and limited evidence in humans *(1)*.

The available data suggest that trichloroethylene may have a weak genotoxic action *in vivo*. Several of the animal carcinogenicity studies show limitations in design. In mice, increased incidences of adenomas and carcinomas in lungs and liver were observed *(2–5)*. In two rat studies, the incidence of testicular tumours was increased *(6, 7)*. Evidence from mechanistic studies suggests that humans are likely to be less susceptible to developing tumours as a result of exposure to trichloroethylene. Nevertheless, the relevance of the observed increase in lung tumours in mice and testicular tumours in rats for human cancer risks cannot be excluded. The results of the mechanistic studies do not provide full elucidation or guidance on this point.

Positive associations between exposure to trichloroethylene and risks for cancer of the liver and biliary tract and non-Hodgkin lymphomas were observed in epidemiological studies on cancer in humans. Confounding cannot be ruled out. A quantitative risk estimate cannot be made from these human data. The increased tumours in lungs and testes observed in animal bioassays are considered to be the best available basis for the risk evaluation. However, it cannot be conclusively established whether a threshold with regard to carcinogenicity in the action of trichloroethylene may be assumed. Therefore, linear extrapolation from the animal tumour data is used, providing a conservative approach to the estimation of human cancer risk.

Using the data on the incidence of pulmonary adenomas in B3C6F1 mice and on pulmonary adenomas/carcinomas in Swiss mice *(2)*, unit risks of 9.3×10^{-8} and 1.6×10^{-7}, respectively, can be calculated by applying the linearized multistage model. Applying the same model on the incidence of Leydig cell tumours in the testes of rats, a unit risk of 4.3×10^{-7} can be derived *(6)*.

Physiologically based pharmacokinetic models have been developed for trichloroethylene. Use of these models for cancer risk estimates is not considered feasible because it is not known what an appropriate internal dose measure would be.

Guidelines

Because the available evidence indicates that trichloroethylene is genotoxic and carcinogenic, no safe level can be recommended. On the basis of the most sensitive endpoint, Leydig cell tumours in rats, a unit risk estimate of 4.3×10^{-7} per µg/m^3 can be derived. The ranges of ambient air concentrations of trichloroethylene corresponding to an excess lifetime risk of 1:10 000, 1:100 000 and 1:1 000 000 are 230, 23 and 2.3 µg/m^3, respectively.

References

1. *Dry cleaning, some chlorinated solvents and other industrial chemicals*. Lyons, International Agency for Research on Cancer, 1995 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 63).
2. Maltoni, C. et al. Long-term carcinogenicity bioassays on trichloroethylene administered by inhalation to Sprague-Dawley rats and Swiss mice and B3C6F$_1$ mice. *Annals of the New York Academy of Science,* **534**: 316–342 (1988).
3. Fukuda, K. et al. Inhalation carcinogenicity of trichloroethylene in mice and rats. *Industrial health,* **21**: 243–254 (1983).
4. *Technical report on the carcinogenesis bioassay of trichloroethylene, CAS No. 79–01–6*. Bethesda, MD, US Department of Health, Education and Welfare, 1976 (DHEW Publication, No. 76–802; National Cancer Institute Technical Report, No. 2).

5. *Technical report on the carcinogenesis studies of trichloroethylene (without epichlorohydrin) in F344/N rats and B6C3F1 mice (gavage studies)*. Research Triangle Park, NC, National Toxicology Program, 1990 (NTP Technical Report, No. 243).
6. MALTONI, C. ET AL. Experimental research on trichloroethylene carcinogenesis. *In:* Maltoni, C. & Mehlman, M.A., ed. *Archives of research on industrial carcinogenesis 5*. Princeton, NJ, Princeton Science Publishers, 1986.
7. *Technical report on the carcinogenesis studies of trichloroethylene in four strains of rats (ACI, August, Marshall, Osborne-Mendel) (gavage studies)*. Research Triangle Park, NC, National Toxicology Program, 1988 (NTP Technical Report, No. 273).

5.16 Vinyl chloride

Exposure evaluation

Calculations based on dispersion models indicate that 24-hour average concentrations of 0.1–0.5 μg/m^3 exist as background levels in much of western Europe, but such concentrations are below the current detection limit (approximately 1.0 μg/m^3). In the vicinity of vinyl chloride (VC) and polyvinyl chloride (PVC) production facilities 24-hour concentrations can exceed 100 μg/m^3, but are generally less than 10 μg/m^3 at distances greater than 1 km from plants. The half-time of VC in the air is calculated to be 20 hours; this figure is based on measured rates of reaction with hydroxyl radicals and their concentrations in the air *(1)*.

Health risk evaluation

There is sufficient evidence of carcinogenicity of VC in humans and experimental animals *(2)*. Extrapolation (or rather interpolation) to lower exposure levels can be made, based on knowledge or assumptions about the dose and time-dependence of risk. As seen in the low exposure data of Maltoni et al. *(3)*, a linear dose–response relationship accords well with the animal data for haemangiosarcoma. The finding of at least three cases of haemangiosarcoma in PVC processors as compared with about 100 in VC or PVC production workers is compatible with a linear relationship. The average exposures in the production industry were about 100 times lower than those in the polymerization industry, but the workforce was 10 times larger.

Data from a cohort study *(4)* and an analysis of the incidence of haemangiosarcoma in the United States and western Europe *(5)* suggest that the risk of haemangiosarcoma increases as the second or third power of time from onset of exposure. Using a model in which the risk increases as t^3 during exposure and as t^2 subsequently, estimates of the relative risk in various exposure circumstances can be calculated and used to convert limited-duration exposure risks into lifetime exposure risks.

Estimates of cancer risk can be made from the data relating to the cohort studied by Nicholson et al. *(4)*. A group of 491 workers at two long-established PVC production plants was studied. One plant began operations in 1936 and the other in 1946. Each cohort member had a minimum of 5 years' employment; the average work duration was 18 years. It is estimated that the average VC exposure was 2050 mg/m^3. The overall standardized mortality rate (SMR) for cancer was 142 (28 observed; 19.7 expected); that for liver and biliary cancer was 2380 (10 observed; 0.42 expected). Using the liver cancer data, the estimated lifetime risk of death from VC exposure is 3.6×10^{-4} per mg/m^3, or [(23.8–1) × 0.003/ (2050 mg/m^3) × 2.8 × 70/18], where 0.003 is the lifetime risk of death from liver biliary cancer in white American males, 2.8 is the working week–total week conversion and 70/18 the work period–lifetime conversion. Since there are an equal number of cancers at other sites (averaging over 12 cohorts), the excess cancer risk is 7.2×10^{-4} per mg/m^3. If the total cancer SMR is used directly, the risk is 4.5×10^{-4} per mg/m^3, or [(1.42–1) × 0.2/ (2050 mg/m^3) × 2.8 × 70/18], which is in good agreement with the above. The average of the two estimates indicates that a 10^{-6} cancer risk occurs at exposures of 1.7 µg/m^3.

The risk of cancer from VC can be calculated from data on the United States population exposed in the Equitable Environmental Health study *(6).* This study identified 10 173 workers who were employed for one or more years in 37 (of 43) VC and PVC production plants. The average duration of employment before 1973 was 8.7 years. Using the data of Barnes *(7),* a weighted exposure of 650 ppm (1665 mg/m^3) was estimated. Considering the total population at risk to be 12 000, the unit exposure lifetime risk from an average exposure of 9 years is 0.75×10^{-5} per mg/m^3, or [(150/12 000) × (1/1665)].

Using a linear dose–response relationship converting to a lifetime exposure (assuming that one half of the workers began exposure at the age of 20 and one half at the age of 30), the continuous lifetime haemangiosarcoma risk is 4.7×10^{-4} per mg/m^3, or [$0.75 \times 10^{-5} \times 2.8 \times 22.4$], where 2.8 is the ratio of the air volume inhaled in a full week (20 m^3 × 7) to that in a working week (10 m^3 × 5) and 22.4 is the average conversion to a lifetime for a ten-year exposure beginning at an average age of 25 years, taking into account the time course of haemangiosarcoma. (Without explicit consideration of the time course, the multiplier would be 70/9 = 7.8.) A 10^{-6} risk occurs at a concentration of 2.1 µg/m^3.

Assuming that the number of cancers in other sites may equal that of haemangiosarcomas, the best estimate for excess cancer risk is that a 10^{-6} risk occurs as a result of continuous lifetime exposure to 1.0 µg/m^3.

The risks estimated from epidemiological studies are the most relevant for human exposures. The above estimate from human angiosarcoma incidences is a conservative one from the point of view of health, because of the use of a model that assumes that the haemangiosarcoma risk continues to increase throughout the lifetime of an exposed individual.

These risk estimates are in agreement with those made by others. The US Environmental Protection Agency has estimated that 11 cancer deaths per year would result from 4.6×10^{-6} people being exposed to 0.017 ppm (43 μg/m^3) *(8)*: this translates to a 10^{-6} lifetime risk at 0.25 μg/m^3. A Dutch criteria document, on the basis of animal data, estimates that a 10^{-6} risk occurs at 0.035 μg/m^3 *(1).*

One cautionary note should be sounded: the particular sensitivity of newborn rats to VC, referred to above, suggests that risks may be much greater in childhood than those estimated from adult exposures. By the age of 10 years, however, the latter risks should prevail.

Guidelines

Vinyl chloride is a human carcinogen and the critical concern with regard to environmental exposures to VC is the risk of malignancy. No safe level can be indicated. Estimates based on human studies indicate a lifetime risk from exposure to 1 μg/m^3 to be 1×10^{-6}.

References

1. *Criteriadocument over vinylchloride* [Vinyl chloride criteria document]. The Hague, Ministry of Housing, Spatial Planning and Environment, 1984 (Publikatiereeks Lucht, No. 34).
2. *Some monomers, plastics and synthetic elastomers, and acrolein.* Lyons, International Agency for Research on Cancer, 1979 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 19).
3. Maltoni, C. et al. Carcinogenicity bioassays of vinyl chloride monomer: a model of risk assessment on an experimental basis. *Environmental health perspectives,* **41**: 3–29 (1981).
4. Nicholson, W.J. et al. Occupational hazards in the VC-PVC industry. *In:* Jarvisalo, P. et al., ed. *Industrial hazards of plastics and synthetic elastomers.* New York, Alan R. Liss, 1984 (Progress in Clinical and Biological Research, Vol. 141), pp. 155–176.
5. Nicholson, W.J. et al. Trends in cancer mortality among workers in the synthetic polymers industry. *In:* Jarvisalo, P. et al., ed. *Industrial hazards of plastics and synthetic elastomers.* New York, Alan R. Liss, 1984, pp. 65–78 (Progress in Clinical and Biological Research, Vol. 141).
6. *Epidemiological study of vinyl chloride workers.* Rockville, MD, Equitable Environmental Health, Inc., 1978.

7. BARNES, A.W. Vinyl chloride and the production of PVC. *Proceedings of the Royal Society of Medicine,* **69**: 277–281 (1976).
8. KUZMACH, A.M. & MCGAUGHY, R.E. *Quantitative risk assessment for community exposure to vinyl chloride.* Washington, DC, US Environmental Protection Agency, 1975.

CHAPTER 6

Inorganic pollutants

6.1 Arsenic

Exposure evaluation

There are many arsenic compounds, both organic and inorganic, in the environment. Airborne concentrations of arsenic range from 1 ng/m^3 to 10 ng/m^3 in rural areas and from a few nanograms per cubic metre to about 30 ng/m^3 in noncontaminated urban areas. Near emission sources, such as nonferrous metal smelters and power plants burning arsenic-rich coal, concentrations of airborne arsenic can exceed 1 µg/m^3.

Health risk evaluation

Inorganic arsenic can have acute, subacute and chronic effects, which may be either local or systemic. Lung cancer is considered to be the critical effect following inhalation. An increased incidence of lung cancer has been seen in several occupational groups exposed to inorganic arsenic compounds. Some studies also show that populations near emission sources of inorganic arsenic, such as smelters, have a moderately elevated risk of lung cancer. Information on the carcinogenicity of arsenic compounds in experimental animals was considered inadequate to make an evaluation *(1, 2)*.

A significant number of studies concerning occupational exposure to arsenic and the occurrence of cancer have been described. Unit risks derived by the US Environmental Protection Agency (EPA) Carcinogen Assessment Group in 1984 *(3)* were not changed until 1994 *(4)*. They form five sets of data involving two independently exposed populations of workers in Montana and Tacoma smelters in the United States, ranging from 1.25×10^{-3} to 7.6×10^{-3}, a weighted average of these five estimates giving a composite estimate of 4.29×10^{-3}.

A WHO Working Group on Arsenic *(5)* conducted a quantitative risk assessment, assuming a linear relationship between the cumulative arsenic dose and the relative risk of developing lung cancer. Risk estimates for lung cancer from inorganic arsenic exposure were based on the study by Pinto et al. *(6)* of workers at the Tacoma smelter. The lifetime risk of lung cancer was calculated to be 7.5×10^{-3} per microgram of airborne arsenic per cubic metre.

The second study relating to the quantitative risk assessment included a large number of the 8047 males employed as smelting workers at the Montana copper smelter *(7)*. Exposures to airborne arsenic levels were estimated to average 11.17, 0.58 and 0.27 mg/m^3 in the high-, medium- and low-exposure areas. Unit risks for these three groups were calculated to be 3.9×10^{-3}, 5.1×10^{-3} and 3.1×10^{-3}, respectively.

Assuming that the risk estimation based on the Tacoma study was higher because of the urine measurements made, it may have underestimated the actual inhalation exposure; the unit risk was considered to be 4×10^{-3}.

In 1994, Viren & Silvers *(8)*, using updated results from the cohort mortality study in the Tacoma smelter workers together with findings from a cohort study of 3619 Swedish smelter workers, developed other unit risk estimates. A unit risk of 1.28×10^{-3} was estimated for the Tacoma smelter cohort and 0.89×10^{-3} for the Swedish cohort. Pooling these new estimates with the EPA's earlier estimates from the Montana smelter yielded a composite unit risk of 1.43×10^{-3} (Table 13). This value is three times lower than the EPA estimate *(4)* and two times lower than the value assumed in the first edition of *Air quality guidelines for Europe (9)*.

Table 13. Updated unit risk estimates

Risk update	Smelter population	Estimated unit risk: Study	Estimated unit risk: Cohort	Estimated unit risk: Pooled
Pooled estimate using updated Swedish and Tacoma cohorts	Tacoma, 1987	1.28×10^{-3}	1.28×10^{-3}	1.07×10^{-3}
	Ronnskar, 1989:			
	– workers hired pre-1940	0.46×10^{-3}	0.89×10^{-3}	
	– workers hired 1940 and later	1.71×10^{-3}	–	
Updated Tacoma cohort with original EPA estimates for Montana cohort	Tacoma, 1987 (updated results supersede earlier estimates)		1.28×10^{-3}	1.81×10^{-3}
	Montana, 1984 (new estimates not available, 1984 EPA estimates apply)		2.56×10^{-3}	
Pooled across all smelter cohorts	Ronnskar, 1989		0.89×10^{-3}	1.43×10^{-3}
	Tacoma, 1987		1.28×10^{-3}	
	Montana, 1984		2.56×10^{-3}	

Source: Viren & Silvers *(8)*.

Guidelines

Arsenic is a human carcinogen. Present risk estimates have been derived from studies in exposed human populations in Sweden and the United States. When assuming a linear dose–response relationship, a safe level for inhalation exposure cannot be recommended. At an air concentration of 1 $\mu g/m^3$, an estimate of lifetime risk is 1.5×10^{-3}. This means that the excess lifetime risk level is 1:10 000, 1:100 000 or 1:1 000 000 at an air concentration of about 66 ng/m^3, 6.6 ng/m^3 or 0.66 ng/m^3, respectively.

References

1. WOOLSON, E.A. Man's perturbation of the arsenic cycle. *In:* Lederer, W.H. & Fensterheim, R.J., ed. *Arsenic: industrial, biomedical and environmental perspectives. Proceedings of the Arsenic Symposium, Gaithersburg, MD.* New York, Van Nostrand Reinhold, 1983.
2. *Overall evaluations of carcinogenicity: an updating of IARC monographs volumes 1 to 42*. Lyons, International Agency for Research on Cancer, 1987 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Supplement 7).
3. *Health assessment document for inorganic arsenic.* Washington, DC, US Environmental Protection Agency, 1984 (Final report EPA-600-8-83-021F).
4. INTEGRATED RISK INFORMATION SYSTEM (IRIS). *Carcinogenicity assessment for lifetime exposure to arsenic* (http://www.epa.gov/ngispgm3/iris/subst/0278.htm#I.B). Cincinnati, OH, US Environmental Protection Agency (accessed January 1994).
5. *Arsenic*. Geneva, World Health Organization, 1981 (Environmental Health Criteria, No. 18).
6. PINTO, S.S. ET AL. Mortality experience in relation to a measured arsenic trioxide exposure. *Environmental health perspectives*, **19**: 127–130 (1977).
7. LEE-FELDSTEIN, A. Arsenic and respiratory cancer in man: follow-up of an occupational study. *In:* Lederer, W.H. & Fensterheim, R.J., ed. *Arsenic: industrial, biomedical and environmental perspectives. Proceedings of the Arsenic Symposium, Gaithersburg, MD.* New York, Van Nostrand Reinhold, 1983, pp. 245–254.
8. VIREN, J.R. & SILVERS, A. Unit risk estimates for airborne arsenic exposure: an updated view based on recent data from two copper smelter cohorts. *Regulatory toxicology and pharmacology*, **20**: 125–138 (1994).
9. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).

6.2 Asbestos

Exposure evaluation

Actual indoor and outdoor concentrations in air range from below one hundred to several thousand fibres per m^3.

Health risk evaluation

On the basis of the evidence from both experimental and epidemiological studies, it is clear that asbestos inhalation can cause asbestosis, lung cancer and mesothelioma. The evidence that ingested asbestos causes gastrointestinal or other cancers is insufficient. Furthermore, the carcinogenic properties of asbestos are most probably due to its fibre geometry and remarkable integrity; other fibres with the same characteristics may also be carcinogenic.

Current environmental concentrations of asbestos are not considered a hazard with respect to asbestosis. However, a risk of mesothelioma and lung cancer from the current concentrations cannot be excluded.

In 1986 a WHO Task Group expressed reservations about the reliability of risk assessment models applied to asbestos risk. Its members suggested that such models can only be used to obtain a broad approximation of the lung cancer risk of environmental exposures to asbestos and "that any number generated will carry a variation over many orders of magnitude". The same was found to be true for estimates of the risk of mesothelioma. The same document stated: "In the general population the risks of mesothelioma and lung cancer attributable to asbestos cannot be quantified reliably and probably are undetectably low." *(1).*

The following estimates of risk are based on the relatively large amount of evidence from epidemiological studies concerning occupational exposure. Data from these studies have been conservatively extrapolated to the much lower concentrations found in the general environment. Although there is evidence that chrysotile is less potent than amphiboles, as a precaution chrysotile has been attributed the same risk in these estimates.

Mesothelioma

A formula by which the excess incidence of mesothelioma can be approximated has been derived by Peto *(2)*. Fibre concentration, duration of exposure and time since first exposure are parameters incorporated in this model, which assumes a linear dose–response relationship. Peto verified this model from data on an urban population exposed for its whole life and on workers exposed for many decades. In both cases, duration of exposure is assumed to be equal or close to time since first exposure. The data show that the incidence of mesothelioma is proportional to the fibre concentration to which the workers were exposed and to time since first exposure for both workers and the general population. Starting from this relationship, one may calculate the risk of lifetime exposure to environmental concentrations from the incidence of mesothelioma in occupational populations exposed to much higher concentrations, but for a shorter time.

Apart from incomplete knowledge about the true workplace exposure, a further complication arises from the fact that workplace concentrations were measured by means of an optical microscope, counting only fibres longer than 5 µm and thicker than, say, 0.5 µm. In this chapter all fibre concentrations based on optical microscopy are marked F^*/m^3 and risk estimates will be based on F^*/m^3. If concentrations measured by optical microscopy are to be compared with environmental fibre concentrations measured by scanning electron microscopy, a conversion factor has to be used: $2\ F/m^3 = 1\ F^*/m^3$.

Several studies have been performed to calculate the risk of mesothelioma resulting from nonoccupational exposure to asbestos. Lifetime exposure to $100\ F^*/m^3$ has been estimated by various authors to carry differing degrees of mesothelioma risk (see Table 14). The risk estimates in Table 14 differ by a factor of 4. A "best" estimate may be 2×10^{-5} for $100\ F^*/m^3$.

An independent check of this risk estimate can be made by calculating the incidence of mesothelioma in the general population, based on a hypothetical

Table 14. Estimates of mesothelioma risk resulting from lifetime exposure to asbestos

Risk of mesothelioma from $100\ F^*/m^3$	Values in original publication (risk for fibre concentration indicated)	Reference
1.0×10^{-5}	1.0×10^{-4} for $1000\ F^*/m^3$	*(3)*
$\sim 2.0 \times 10^{-5}$	1.0×10^{-4} for $(130–800)\ F^*/m^3$	*(4)*
$\sim 3.9 \times 10^{-5}$	1.56×10^{-4} for $400\ F^*/m^3$	*(5, 6)*
$\sim 2.4 \times 10^{-5}$	2.75×10^{-3} (females) 1.92×10^{-3} (males) for 0.01 F/ml	*(7)*

average asbestos exposure 30–40 years ago *(8)*. If the latter had been 200–500 F*/m^3 (corresponding to about 400–1000 F/m^3 as measured today), the resulting lifetime risk of mesothelioma would be (4–10) × 10^{-5}. With the average United States death rate of 9000 × 10^{-6} per year, this would give 0.4–0.9 mesothelioma cases each year per million persons from past environmental asbestos exposure. The reported mesothelioma incidence in the United States ranges from 1.4 × 10^{-6} per year to 2.5 × 10^{-6} per year according to various authors *(5, 8)*. Thus, the calculated risk figures would account for only part of the observed incidence. Nevertheless, other factors that may account for this discrepancy must be considered.

- Uncertainties in the risk extrapolations result from the lack of reliable exposure data in the cohort studies, errors in the medical reports, and necessary simplifications in the extrapolation model itself *(7)*. Furthermore, the amount of past ambient exposure can only be an educated guess.

- The incidence of nonoccupational mesotheliomas is calculated from the difference between the total of observed cases and the number of those probably related to occupational exposure. Neither of these two figures is exactly known. Moreover, the influence of other environmental factors in the generation of mesothelioma is unknown.

In the light of these uncertainties, the result obtained by using the risk estimate can be considered to be in relatively good agreement with the annual mesothelioma death rate based on national statistical data.

Lung cancer

Unlike mesothelioma, lung cancer is one of the most common forms of cancer. As several exogenous noxious agents can be etiologically responsible for bronchial carcinoma, the extrapolation of risk and comparison between different studies is considerably complicated. In many epidemiological studies, the crucial effect of smoking has not been properly taken into account.

Differentiation of the observed risks according to smoking habits has been carried out, however, in the cohort of North American insulation workers studied by Hammond et al. *(9)*. This study suggests that the relative risk at a given time is approximately proportional to the cumulative amount of fine asbestos dust received up to this point, for both smokers and nonsmokers. The risks for non-asbestos-exposed nonsmokers and smokers must therefore be multiplied by a factor that increases in proportion to the cumulative exposure.

The dose–response relationship in the case of asbestos-induced lung cancer can be described by the following equation *(7)*.

$$I_L \text{ (age, smoking, fibre dose)} = I_L^o \text{ (age, smoking)} [l + K_L \times C_f \times d]$$

This equation could also be written as:

$$K_L = [(I_L/I_L^o) - 1]/C_f \times d = \text{(relative risk} - 1)/\text{(cumulative exposure)}$$

where:

K_L = a proportionality constant, which is a measure of the carcinogenic potency of asbestos

C_f = fibre concentration

d = duration of exposure in years

I_L = lung cancer incidence, observed or projected, in a population exposed to asbestos concentration C_f during time d

I_L^o = lung cancer incidence expected in a group without asbestos exposure but with the same age and smoking habits (this factor includes age dependence).

There are several studies that allow the calculation of K_L. Liddell *(10, 11)* has done this in an interesting and consistent manner. The results are given in Table 15.

Taking the data in Table 15 as a basis, a reasonable estimate for K_L is 1.0 per 100 F*years/ml. For a given asbestos exposure, the risk for smokers is about 10 times that for nonsmokers *(9)*. In extrapolating from workers to the general public, a factor of 4 for correction of exposure time has to be applied to K_L.

The incidence of lung cancer in the general population exposed to 100 F^*/m^3 is calculated as follows:

$$I_L = I_L^o(l + 4 \times 0.01 \times 10^{-4} \text{ F}^*/\text{ml} \times 50 \text{ years})$$

or

$$I_L = I_L^o(l + 2 \times 10^{-4} \text{ F}^*/\text{ml})$$

Table 15. Increase in the relative risk of lung cancer, as shown by different studies

K_L per 100 F*year/ml	Type of activity	Reference
0.04	mining and milling	*(12)*
0.045	mining and milling	*(13)*
0.06	friction material	*(14)*
0.1	factory processes	*(15, 16)*
(M) 0.4–1.1	factory processes	
(F) 2.7 [a]	factory processes	*(17)*[b]
0.2	asbestos-cement	*(18)*
0.07	textiles (before 1951)	*(19)*
0.8 [a]	textiles (after 1950)	
6(M) 1.6 [a]	textiles	*(20)*
1.6	textiles	*(21)*[c]
1.1	insulation products	*(22)*[b]
1.5	insulation	*(23)*[b]

[a] Fewer than 10 cases of lung cancer expected (i.e. small cohort).

[b] Inadequate knowledge of actual fibre concentrations.

[c] Same factory as in *(20)*, but larger cohort.

Source: Liddell *(10)*.

The extra risk is $I_L - I_L^o$. Values for I_L^o are about 0.1 for male workers and 0.01 for male nonsmokers *(5)*.

Lifetime exposure to 100 F*/m³ (lifetime assumed to be 50 years since, in a lifetime of 70 years, the first 20 years without smoking probably do not make a large contribution) is therefore estimated as follows.

Status	Risk of lung cancer per 100 000	Range (using the highest and lowest values of K_L from Table 15)
Smokers	2.0	0.08–3.2
Nonsmokers	0.2	0.008–0.32

This risk estimate can be compared, when adjusted to 100 F*/m^3, with estimates for male smokers made by other authors or groups:

Breslow (National Research Council) *(6)*: 7.3×10^{-5}

Schneiderman et al. *(4)*: $(14–1.4) \times 10^{-5}$

US Environmental Protection Agency *(7)*: 2.3×10^{-5}.

A fibre concentration of 100 F*/m^3 (about 200 F/m^3 as seen by scanning electron microscope) thus gives a total risk of $(2 + 2) \times 10^{-5}$ for smokers or 2.2×10^{-5} for nonsmokers.

Guidelines

Asbestos is a proven human carcinogen (IARC Group 1). No safe level can be proposed for asbestos because a threshold is not known to exist. Exposure should therefore be kept as low as possible.

Several authors and working groups have produced estimates indicating that, with a lifetime exposure to 1000 F/m^3 (0.0005 F*/ml or 500 F*/m^3, optically measured) in a population of whom 30% are smokers, the excess risk due to lung cancer would be in the order of 10^{-6}–10^{-5}. For the same lifetime exposure, the mesothelioma risk for the general population would be in the range 10^{-5}–10^{-4}. These ranges are proposed with a view to providing adequate health protection, but their validity is difficult to judge. An attempt to calculate a "best" estimate for the lung cancer and mesothelioma risk is described above.

References

1. *Asbestos and other natural mineral fibres.* Geneva, World Health Organization, 1986 (Environmental Health Criteria, No. 53).
2. PETO, J. Dose and time relationships for lung cancer and mesothelioma in relation to smoking and asbestos exposure. *In:* Fischer, M. & Meyer, E., ed. *Zur Beurteilung der Krebsgefahr durch Asbest* [Assessment of the cancer risk of asbestos]. Munich, Medizin Verlag, 1984.
3. AURAND, K. & KIERSKI, W.-S., ED. *Gesundheitliche Risiken von Asbest. Eine Stellungnahme des Bundesgesundheitsamtes Berlin* [Health risks of asbestos. A position paper of the Federal Health Office, Berlin]. Berlin, Dietrich Reimer Verlag, 1981 (BgA-Berichte, No. 4/81).
4. SCHNEIDERMAN, M.S. ET AL. *Assessment of risks posed by exposure to low levels of asbestos in the general environment.* Berlin, Dietrich Reimer Verlag, 1981 (BgA-Bericht, No. 4/81).

5. National Research Council. *Asbestiform fibers: nonoccupational health risks.* Washington, DC, National Academy Press, 1984.
6. Breslow, L. et al. Letter. *Science,* **234**: 923 (1986).
7. *Airborne asbestos health assessment update.* Research Triangle Park, NC, US Environmental Protection Agency, 1985 (Publication EPA-600/8-84-003F).
8. Enterline, P.E. Cancer produced by nonoccupational asbestos exposure in the United States. *Journal of the Air Pollution Control Association,* **33**: 318–322 (1983).
9. Hammond, E.C. et al. Asbestos exposure, cigarette smoking and death rates. *Annals of the New York Academy of Sciences,* **330**: 473–490 (1979).
10. Liddell, F.D.K. Some new and revised risk extrapolations from epidemiological studies on asbestos workers. *In:* Fischer, M. & Meyer, E., ed. *Zur Beurteilung der Krebsgefahr durch Asbest* [Assessment of the cancer risk of asbestos]. Munich, Medizin Verlag, 1984.
11. Liddell, F.D.K. & Hanley, J.A. Relations between asbestos exposure and lung cancer SMRs in occupational cohort studies. *British journal of industrial medicine,* **42**: 389–396 (1985).
12. McDonald, J.C. et al. Dust exposure and mortality in chrysotile mining, 1910–1975. *British journal of industrial medicine,* **37**: 11–24 (1980).
13. Nicholson, W.J. et al. Long-term mortality experience of chrysotile miners and millers in Thetford Mines, Quebec. *Annals of the New York Academy of Sciences,* **330**: 11–21 (1979).
14. Berry, G. & Newhouse, M.L. Mortality of workers manufacturing friction materials using asbestos. *British journal of industrial medicine,* **40**: 1–7 (1983).
15. Henderson, V.L. & Enterline, P.E. Asbestos exposure: factors associated with excess cancer and respiratory disease mortality. *Annals of the New York Academy of Sciences,* **330**: 117–126 (1979).
16. Enterline, P. et al. Mortality in relation to occupational exposure in the asbestos industry. *Journal of occupational medicine,* **14**: 897–903 (1972).
17. Newhouse, M.L. & Berry, G. Patterns of mortality in asbestos factory workers in London. *Annals of the New York Academy of Sciences,* **330**: 53–60 (1979).
18. Weill, H. et al. Influence of dose and fiber type on respiratory malignancy risk in asbestos cement manufacturing. *American review of respiratory diseases,* **120**: 345–354 (1979).
19. Peto, J. Lung cancer mortality in relation to measured dust levels in an asbestos textile factory. *In:* Wagner, J.C., ed. *Biological effects of mineral fibres.* Lyons, International Agency for Research on Cancer, 1980 (IARC Scientific Publications, No. 30).

20. Dement, J.M. et al. Estimates of dose–response for respiratory cancer among chrysotile asbestos textile workers. *Annals of occupational hygiene,* **26**: 869–887 (1982).
21. Fry, J.S. et al. Respiratory cancer in chrysotile production and textile manufacture. *Scandinavian journal of work, environment and health,* **9**: 68–70 (1983).
22. Seidman, H. et al. Short-term asbestos work exposure and long-term observation. *Annals of the New York Academy of Sciences,* **330**: 61–67 (1979).
23. Selikoff, J.J. et al. Mortality experience of insulation workers in the United States and Canada, 1943–1976. *Annals of the New York Academy of Sciences,* **330**: 91–116 (1979).

6.3 Cadmium

Exposure evaluation

It is not possible to carry out a dose–response analysis for cadmium in air solely on the basis of epidemiological data collected in the general population, since the latter is exposed to cadmium mainly via food or tobacco smoking. In addition, the recently reported renal effects in areas of Belgium and the Netherlands polluted by cadmium refer to historical contamination of the environment. Assuming, however, that the only route of exposure is by inhalation, an indirect estimate of the risk of renal dysfunction or lung cancer can be made on the basis of data collected in industrial workers.

Health risk evaluation

Pooled data from seven studies, in which the relationships between the occurrence of tubular proteinuria and cumulative cadmium exposure were examined, show that the prevalence of tubular dysfunction (background level 2.4%) increases sharply at a cumulative exposure of more than 500 μg/m^3-years (8% at 400 μg/m^3-years, 50% at 1000 μg/m^3-years and > 80% at more than 4500 μg/m^3-years) *(1)*. Some studies suggest that a proportion of workers with cumulative exposures of 100–400 μg/m^3-years might develop tubular dysfunction (prevalences increasing from 2.4% to 8.8%, at cumulative exposures above 200 μg/m^3-years). These estimates agree well with that derived from the kinetic model of Kjellström *(2)*, which predicted that the critical concentration of 200 mg/kg in the renal cortex will be reached in 10% of exposed workers after 10 years of exposure to 50 μg/m^3 and in 1% after 10 years of exposure to 16 μg/m^3 (cumulative exposures of 500 and 160 μg/m^3-years, respectively).

With respect to the risk of lung cancer, two risk estimates have been made, one based on the long-term rat bioassay data of Takenaka et al. *(3)* and the other on the epidemiological data of Thun et al. *(4)*. Modelling of these data yielded risk estimates that did not agree. On the basis of the Takenaka data, the unit risk is 9.2×10^{-2} per μg/m^3; the human data yielded a unit risk of 1.8×10^{-3} per μg/m^3. In general, the use of human data is more reliable because of species variation in response. Nevertheless, there is evidence from recent studies that this latter unit risk might be substantially overestimated owing to confounding by concomitant exposure to arsenic.

Some uncertainty exists with regard to the thresholds of exposure associated with effects on the kidney. This is primarily due to the limited number of subjects, methodological differences and inaccuracies in exposure data. An overall assessment of the data from industrial workers suggests that, to prevent tubular dysfunction, the 8-hour exposure level for cadmium should not exceed 5 $\mu g/m^3$. This corresponds to a cumulative exposure of 225 $\mu g/m^3$-years. Adopting the lowest estimate of the critical cumulative exposure to airborne cadmium (100 $\mu g/m^3$-years), extrapolation to continuous lifetime exposure results in a permissible concentration of about 300 ng/m^3.

Cadmium in ambient air is transferred to soil by wet or dry deposition and can enter the food chain. However, the rate of transfer from soil to plant depends on numerous factors (type of soil and plant, soil pH, use of fertilizers, meteorology, etc.) and is impossible to predict.

Present average concentrations of cadmium in the renal cortex in the general population in Europe at the age of 40–60 years are in the range 15–40 mg/kg. These values are only 4–12 times lower than the critical levels estimated in cadmium workers for the induction of tubular dysfunction (180 mg/kg) and very close to the critical level of 50 mg/kg estimated by the Cadmibel study in Belgium *(5)*. Any further increase in the dietary intake of cadmium owing to an accumulation of the metal in agricultural soils will further narrow the gap to these critical levels. It is thus imperative to maintain a zero balance for cadmium in agricultural soils by controlling and restricting inputs from fertilizers (including sewage sludge) and atmospheric emissions. Since emissions from industry are currently decreasing, attention must be focused on the emissions from waste incineration, which are likely to increase in the future.

Guidelines

IARC has classified cadmium and cadmium compounds as Group 1 human carcinogens, having concluded that there was sufficient evidence that cadmium can produce lung cancers in humans and animals exposed by inhalation *(6)*. Because of the identified and controversial influence of concomitant exposure to arsenic in the epidemiological study, however, no reliable unit risk can be derived to estimate the excess lifetime risk for lung cancer.

Cadmium, whether absorbed by inhalation or via contaminated food, may give rise to various renal alterations. The lowest estimate of the cumulative exposure to airborne cadmium in industrial workers leading to an increased risk of renal dysfunction (low-molecular-weight proteinuria) or lung cancer is 100 $\mu g/m^3$-years for an 8-hour exposure. Extrapolation to a continuous lifetime exposure gives a value of around 0.3 μg/m^3. Existing levels of cadmium in the air of most urban or industrial areas are around one-fiftieth of this value.

The finding of renal effects in areas contaminated by past emissions of cadmium indicates that the cadmium body burden of the general population in some parts of Europe cannot be further increased without endangering renal function. To prevent any further increase of cadmium in agricultural soils likely to increase the dietary intake of future generations, a guideline of 5 ng/m^3 is established.

References

1. Thun, M. et al. Scientific basis for an occupational standard for cadmium. *American journal of industrial medicine,* **20**: 629–642 (1991).
2. Kjellström, T. Critical organs, critical concentrations, and whole body dose–response relationships. *In:* Friberg, L. et al., ed. *Cadmium and health: a toxicological and epidemiological appraisal. Vol. 2. Effects and response.* Boca Raton, FL, CRC Press, 1986.
3. Takenaka, S. et al. Carcinogenicity of cadmium chloride aerosols in Wistar rats. *Journal of the National Cancer Institute,* **70**: 367–371 (1983).
4. Thun, M. et al. Mortality among a cohort of U.S. cadmium production workers – an update. *Journal of the National Cancer Institute,* **74**: 325–333 (1985).
5. Buchet, J.P. et al. Renal effects of cadmium body burden of the general population. *Lancet*, **336**: 699–702 (1990).
6. *Beryllium, cadmium, mercury, and exposure in the glass manufacturing industry.* Lyons, International Agency for Research on Cancer, 1993 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 58).

6.4 Chromium

Exposure evaluation

Chromium is ubiquitous in nature. Available data, generally expressed as total chromium, show a concentration range of 5–200 ng/m³. There are few valid data on the valency and bioavailability of chromium in the ambient air.

Health risk evaluation

Chromium(III) is recognized as a trace element that is essential to both humans and animals. Chromium(VI) compounds are toxic and carcinogenic, but the various compounds have a wide range of potencies. As the bronchial tree is the major target organ for the carcinogenic effects of chromium(VI) compounds, and cancer primarily occurs following inhalation exposure, uptake in the respiratory organs is of great significance with respect to the cancer hazard and the subsequent risk of cancer in humans. IARC has stated that for chromium and certain chromium compounds there is sufficient evidence of carcinogenicity in humans (Group 1) *(1)*.

A large number of epidemiological studies have been carried out on the association between human exposure to chromates and the occurrence of cancer, particularly lung cancer, but only a few of these include measurements of exposure *(2–8)*. Measurements were made mainly at the time that the epidemiological studies were performed, whereas the carcinogenic effect is caused by exposure dating back 15–30 years. Hence, there is a great need for studies that include historical data on exposure.

Four sets of data for chromate production workers can be used for the quantitative risk assessment of chromium(VI) lifetime exposure *(3, 5–9)*. The average relative risk model is used in the following to estimate the incremental unit risk.

Using the study performed by Hayes et al. on chromium production workers *(3)*, several cohorts were investigated by Braver et al. *(8)* for cumulative exposure to chromium(VI) in terms of μg/m³-years (cumulative exposure = usual exposure level in μg/m³ × average duration of exposure). Average lifetime exposures for two cohorts can be calculated from the cumulative exposures of 670 and 3647 μg/m³-years, as 2 μg/m³ and 11.4 μg/m³, respectively ($X = \mu g/m^3 \times 8/24 \times 240/365 \times (\text{No. of years})/70$).

The relative risk (RR) for these two cohorts, calculated from observed and expected cases of lung cancer, was 1.75 and 3.04. On the basis of the vital statistics data, the background lifetime probability of death due to lung cancer (P_0) is assumed to be 0.04. The risks (unit risk, UR) associated with a lifetime exposure to 1 μg/m^3 can therefore be calculated to be 1.5×10^{-2} and 7.2×10^{-3}, respectively (UR = P_0(RR–1)/X). The arithmetic mean of these two risk estimates is 1.1×10^{-2}.

A risk assessment can also be made on the basis of the study carried out by Langård et al. on ferrochromium plant workers in Norway *(5, 10)*. The chromium concentration to which the workers were exposed is not known, but measurements taken in 1975 showed a geometric mean value of about 530 μg/m^3. Assuming that the content of chromium(VI) in the sample was 19% and previous concentrations were at least as high as in 1975, the ambient concentration would have been about 100 μg/m^3. On the assumption that occupational exposure lasted for about 22 years, the average lifetime exposure can be determined as 6.9 μg/m^3 (X = 100 μg/m^3 × 8/24 × 240/365 × 22/70).

When workers in the same plant who were not exposed to chromium were used as a control population, the relative risk of lung cancer in chromium-exposed workers was calculated to be 8.5. The lifetime unit risk is therefore 4.3×10^{-2}.

Since earlier exposures must have been much higher than the values measured in 1975, the calculated unit risk of 4.3×10^{-2} can only be considered as an upper-bound estimate. The highest relative incidence ever demonstrated in chromate workers in Norway is about 38, at an exposure level for chromium(VI) of about 0.5 mg/m^3 *(6, 7)*. This relative rate is based on the incidence of bronchial cancer of 0.079 in the total Norwegian male population, irrespective of smoking status. If the average exposure duration is about 7 years, the average lifetime daily exposure is calculated to be 11 μg/m^3 (X = 500 μg/m^3 × 8/24 × 240/365 × 7/70). The incremental unit risk was calculated to be 1.3×10^{-1}. This very high lifetime risk may be due to the relatively small working population.

Differences in the epidemiological studies cited may suggest that the different hexavalent chromium compounds have varying degrees of carcinogenic potency.

The estimated lifetime risks based on various epidemiological data sets, in the range of 1.3×10^{-1} to 1.1×10^{-2}, are relatively consistent. As a best estimate, the geometric mean of the risk estimates of 4×10^{-2} may be taken as the incremental unit risk resulting from a lifetime exposure to chromium(VI) at a concentration of 1 μg/m^3.

Using some other studies and different risk assessment models, the US Environmental Protection Agency (EPA) estimated the lifetime cancer risk due to exposure to chromium(VI) to be 1.2×10^{-2}. This estimate placed chromium(VI) in the first quartile of the 53 compounds evaluated by the EPA Carcinogen Assessment Group for relative carcinogenic potency *(11)*.

Guidelines

Information on the speciation of chromium in ambient air is essential since, when inhaled, only hexavalent chromium is carcinogenic in humans. The available data are derived from studies among chromium(VI)-exposed workers. When assuming a linear dose–response relationship between exposure to chromium(VI) compounds and lung cancer, no safe level of chromium(VI) can be recommended. At an air concentration of chromium(VI) of 1 μg/m^3, the lifetime risk is estimated to be 4×10^{-2}.

It should be noted that chromium concentration in air is often expressed as total chromium and not chromium(VI). The concentrations of chromium(VI) associated with an excess lifetime risk of 1:10 000, 1:100 000 and 1:1 000 000 are 2.5 ng/m^3, 0.25 ng/m^3 and 0.025 ng/m^3, respectively.

References

1. *Chromium, nickel and welding*. Lyons, International Agency for Research on Cancer, 1990 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 49), pp. 463–474.
2. Machle, W. & Gregorius, F. Cancer of the respiratory system in the United States chromate–producing industry. *Public health reports*, **63**: 1114–1127 (1948).
3. Hayes, R.B. et al. Mortality in chromium chemical production workers: a prospective study. *International journal of epidemiology*, **8**: 365–374 (1979).
4. Mancuso, T.F. Consideration of chromium as an industrial carcinogen. *In*: Hutchinson, T.C., ed. *Proceedings of the International Conference on Heavy Metals in the Environment, Toronto, 1975*. Toronto, Institute for Environmental Studies, 1975, pp. 343–356.
5. Langård, S. et al. Incidence of cancer among ferrochromium and ferrosilicon workers. *British journal of industrial medicine*, **37**: 114–120 (1980).

6. Langård, S. & Vigander, T. Occurrence of lung cancer in workers producing chromium pigments. *British journal of industrial medicine*, **40**: 71–74 (1983).
7. Langård, S. & Norseth, T. A cohort study of bronchial carcinomas in workers producing chromate pigments. *British journal of industrial medicine*, **32**: 62–65 (1975).
8. Braver, E.R. et al. An analysis of lung cancer risk from exposure to hexavalent chromium. *Teratogenesis, carcinogenesis and mutagenesis*, **5**: 365–378 (1985).
9. Hayes, R. et al. Cancer mortality among a cohort of chromium pigment workers. *American journal of industrial medicine*, **16**: 127–133 (1989).
10. Langård, S. et al. Incidence of cancer among ferrochromium and ferrosilicon workers: an extended follow up. *British journal of industrial medicine*, **47**: 14–19 (1990).
11. *Health assessment document for chromium.* Washington, DC, US Environmental Protection Agency, 1984 (Final report EPA-600-8-83-014F).

6.5 Fluoride

Exposure evaluation

Exposure of the general European population to fluoride in its various chemical forms is highly variable. In heavily industrialized urban areas, typical daily inhalation intakes are in the range 10–40 μg/day (0.5–2 $\mu g/m^3$), and in some cases are as high as 60 μg/day (3 $\mu g/m^3$). Fluorides are emitted to the atmosphere in both gaseous and particulate forms, but studies typically only report total fluoride content.

The main sources of fluoride intake by humans are food and water. Except for occupational exposure, exposure to fluoride by inhalation is negligible.

Regarding occupational exposure, the daily amount of fluoride inhaled, assuming a total respiratory rate of 10 m^3 during a working day, could be 10–25 mg when the air concentration is at the most frequent exposure limits of 1–2.5 mg/m^3.

Health risk evaluation

The most important long-term adverse effect of fluorides on human populations is endemic skeletal fluorosis. The beneficial effect is prevention of caries, as a result both of fluoride incorporation into developing teeth and post-eruptive exposure of enamel to adequate levels of fluoride. It is therefore of crucial importance to gather information on fluoride sources in the diet, especially water, the etiology of early skeletal fluorosis as related to bone mineralization, and dose–response relationships *(1).*

The earliest reports of skeletal fluorosis appeared from industries where exposure of workers to 100–500 $\mu g/m^3$ per 8-hour day for more than 4 years led to severe skeletal changes. Skeletal fluorosis has also been diagnosed in persons living in areas with excessive fluoride in soil, water, dust or plants *(1)*.

In one study, bronchial hyperreactivity was the main health effect at a mean fluoride concentration of 0.56 mg/m^3 and a mean particulate fluoride concentration of 0.15 mg/m^3 *(2)*. In a longitudinal study performed on 523 aluminium potroom workers, total fluoride was the most important risk factor among the exposure variables. In this study, the risk of developing asthmatic symptoms such as dyspnoea and wheezing was 3.4 and 5.2 times higher in the medium- and high-exposure groups, respectively, than in the low-exposure group. Exposure to other pollutants was limited and did not appear to confound the results *(3)*.

Children living in the vicinity of a phosphate processing facility who were exposed to concentrations of about 100–500 μg/m^3 exhibited an impairment of respiratory function. It is not known, however, whether the concentrations were gaseous or total fluoride. In another study, no effects on respiratory function were observed at gaseous fluoride levels of up to 16 μg/m^3.

There is no evidence that atmospheric deposition of fluorides results in significant exposure through other routes, such as through contamination of soil and consequently groundwater.

Guidelines

For exposure of the general population to fluoride, reference exposure levels have been derived by applying a "benchmark dose" approach to a variety of animal and human exposure studies. The 1-hour reference exposure level to protect against any respiratory irritation is about 0.6 mg/m^3, and the level to protect against severe irritation from a once-in-a-lifetime release is about 1.6 mg/m^3 *(4)*.

Data from various sources indicate that prolonged exposure of humans (workers and children) to fluoride concentrations of 0.1–0.5 mg/m^3 leads to impairment of pulmonary function and skeletal fluorosis. No effects have been found at levels of up to 16 μg/m^3 gaseous fluoride. However, the available information does not permit the derivation of an air quality guideline value for fluoride(s).

Skeletal fluorosis is associated with a systemic uptake exceeding 5 mg/day in a relatively sensitive section of the general population. Systemic uptake from food and fluoridated water is about 3 mg/day. It is highly unlikely that ambient air concentrations of fluorides could pose any material risk of fluorosis.

It has been recognized that fluoride levels in ambient air should be less than 1 μg/m^3 to prevent effects on livestock and plants. These concentrations will also sufficiently protect human health.

References

1. *Fluorine and fluorides*. Geneva, World Health Organization, 1984 (Environmental Health Criteria, No. 36).

2. Saric, M. et al. The role of atopy in potroom workers' asthma. *American journal of industrial medicine,* **9**: 239–242 (1986).
3. Kongerud, J. & Samuelsen, S.O. A longitudinal study of respiratory symptoms in aluminum potroom workers. *American review of respiratory diseases,* **144**: 10–16 (1991).
4. Alexeef, G.V. et al. Estimation of potential health effects from acute exposure to hydrogen fluoride using a "benchmark dose" approach. *Risk analysis*, **13**: 63–69 (1993).

6.6 Hydrogen sulfide

Exposure evaluation

Typical symptoms and signs of hydrogen sulfide intoxication are most often caused by relatively high concentrations in occupational exposures. There are many occupations where there is a potential risk of hydrogen sulfide intoxication and, according to the US National Institute for Occupational Safety and Health *(1),* in the United States alone approximately 125 000 employees are potentially exposed to hydrogen sulfide. Low-level concentrations can occur more or less continuously in certain industries, such as in viscose rayon and pulp production, at oil refineries and in geothermal energy installations.

In geothermal areas there is a risk of exposure to hydrogen sulfide for the general population *(2).* The biodegradation of industrial wastes has been reported to cause ill effects in the general population *(2).* An accidental release of hydrogen sulfide into the air surrounding industrial facilities can cause very severe effects, as at Poza Rica, Mexico, where 320 people were hospitalized and 22 died *(2).* The occurrence of low-level concentrations of hydrogen sulfide around certain industrial installations is a well known fact.

Health risk evaluation

The first noticeable effect of hydrogen sulfide at low concentrations is its unpleasant odour. Conjunctival irritation is the next subjective symptom and can cause so-called "gas eye" at hydrogen sulfide concentrations of 70–140 mg/m^3. Table 16 shows the established dose–effect relationships for hydrogen sulfide.

The hazards caused by high concentrations of hydrogen sulfide are relatively well known, but information on human exposure to very low concentrations is scanty. Workers exposed to hydrogen sulfide concentrations of less than 30 mg/m^3 are reported to have rather diffuse neurological and mental symptoms *(4)* and to show no statistically significant differences when compared with a control group. On the other hand, changes in haem synthesis have been reported at hydrogen sulfide concentrations of less than 7.8 mg/m^3 (1.5–3 mg/m^3 average) *(5).* It is not known whether the inhibition is caused by the low concentrations or by the cumulative effects of occasional peak concentrations. Most probably, at concentrations below 1.5 mg/m^3 (1 ppm), even with exposure for longer periods, there are very few detectable health hazards in the toxicological sense. The malodorous

Table 16. Hydrogen sulfide: established dose–effect relationships

Hydrogen sulfide concentration		Effect	Reference
mg/m³	ppm		
1400–2800	1000–2000	Immediate collapse with paralysis of respiration	*(2)*
750–1400	530–1000	Strong central nervous system stimulation, hyperpnoea followed by respiratory arrest	*(2)*
450–750	320–530	Pulmonary oedema with risk of death	*(2)*
210–350	150–250	Loss of olfactory sense	*(3)*
70–140	50–100	Serious eye damage	*(3)*
15–30	10–20	Threshold for eye irritation	*(3)*

property of hydrogen sulfide is a source of annoyance for a large proportion of the general population at concentrations below 1.5 mg/m^3, but from the existing data it cannot be concluded whether any health effects result. The need for epidemiological studies on possible effects of long-term, low-level hydrogen sulfide exposure is obvious. A satisfactory biological exposure indicator is also needed.

Guidelines

The LOAEL of hydrogen sulfide is 15 mg/m^3, when eye irritation is caused. In view of the steep rise in the dose–effect curve implied by reports of serious eye damage at 70 mg/m^3, an uncertainty factor of 100 is recommended, leading to a guideline value of 0.15 mg/m^3 with an averaging time of 24 hours. A single report of changes in haem synthesis at a hydrogen sulfide concentration of 1.5 mg/m^3 should be borne in mind.

In order to avoid substantial complaints about odour annoyance among the exposed population, hydrogen sulfide concentrations should not be allowed to exceed 7 µg/m^3, with a 30-minute averaging period.

When setting concentration limits in ambient air, it should be remembered that in many places hydrogen sulfide is emitted from natural sources.

References

1. *Occupational exposure to hydrogen sulfide.* Cincinnati, OH, US Department of Health, Education, and Welfare, 1977 (DHEW Publication (NIOSH) No. 77-158).
2. *Hydrogen sulfide.* Geneva, World Health Organization, 1981 (Environmental Health Criteria, No. 19).
3. Savolainen, H. Nordiska expertgruppen för gränsvärdesdokumentation. 40. Dihydrogensulfid [Nordic expert group for TLV evaluation. 40. Hydrogen sulfide]. *Arbeta och hälsa,* **31**: 1–27 (1982).
4. Kangas, J. et al. Exposure to hydrogen sulfide, mercaptans and sulfur dioxide in pulp industry. *American Industrial Hygiene Association journal,* **45**: 787–790 (1984).
5. Tenhunen, R. et al. Changes in haem synthesis associated with occupational exposure to organic and inorganic sulphides. *Clinical science,* **64**: 187–191 (1983).

6.7 Lead

Exposure evaluation

Average air lead levels are usually below 0.15 µg/m³ at nonurban sites. Urban air lead levels are typically between 0.15 and 0.5 µg/m³ in most European cities *(1–3)*. Additional routes of exposure must not be neglected, such as lead in dust, a cause of special concern for children.

The relationship between air lead exposure and blood lead has been shown to exhibit downward curvilinearity if the range of exposures is sufficiently large. At lower levels of exposure, the deviation from linearity is negligible and linear models of the relationship between intake and blood lead are satisfactory approximations.

The level of lead in blood is the best available indicator of current and recent past environmental exposure, and may also be a reasonably good indicator of lead body burden with stable exposures. Biological effects of lead will, therefore, be related to blood lead as an indicator of internal exposure.

Health risk evaluation

Table 17 summarizes LOAELs for haematological and neurological effects in adults. Cognitive effects in lead workers have not been observed at blood lead levels below 400 µg/l *(4, 5)*. Reductions in nerve conduction velocity were found in lead workers at blood levels as low as 300 µg/l *(6–8)*. Elevation of free erythrocyte protoporphyrin has been observed at blood levels of 200–300 µg/l. Delta-aminolaevulinic acid dehydrase (ALAD) inhibition is likely to occur at blood levels of about 100 µg/l *(9)*. Because of its uncertain biological significance relative to the functional reserve capacity of the haem biosynthetic system, ALAD inhibition is not treated as an adverse effect here.

Table 18 summarizes LOAELs for haematological, endocrinological and neurobehavioural endpoints in children. Reduced haemoglobin levels have been found at concentrations in blood of around 400 µg/l. Haematocrit values below 35% have not been reported at blood levels below 200 µg/l *(10)*; this is also true for several enzyme systems, which may be of clinical significance.

Central nervous system effects, as assessed by neurobehavioural endpoints, appear to occur at levels below 200 µg/l. Consistent effects have been

Table 17. Summary of LOAELs for lead-induced health effects in adults		
LOAEL at given blood lead level (µg/l)	**Haem synthesis, haematological and other effects**	**Effects on the nervous system**
1000–1200		Encephalopathic signs and symptoms
800	Frank anaemia	
500	Reduced haemoglobin production	Overt subencephalopathic neurological symptoms, cognition impairment
400	Increased urinary ALA and elevated coproporphyrin	
300		Peripheral nerve dysfunction (slowed nerve conduction velocities)
200–300	Erythrocyte protoporphyrin elevation in males	
150–200	Erythrocyte protoporphyrin elevation in females	

Table 18. Summary of LOAELs for lead-induced health effects in children		
LOAEL at given blood lead level (µg/l)	**Haem synthesis, haematological and other effects**	**Effects on the nervous system**
800–1000		Encephalopathic signs and symptoms
700	Frank anaemia	
400	Increased urinary delta-aminolaevulinic acid and elevated coproporphyrin	
250–300	Reduced haemoglobin synthesis	
150–200	Erythrocyte protoporphyrin elevation	
100–150	Vitamin D3 reduction	Cognitive impairment
100	ALAD inhibition	Hearing impairment

reported for global measures of cognitive functioning, such as the psychometric IQ, to be associated with blood lead levels of 100–150 µg/l *(11, 12)*. Some epidemiological studies have indicated effects at blood lead levels below 100 µg/l. Existing animal studies do provide qualitative support for the claim of lead as the causative agent *(12)*.

Guidelines

Guidelines for lead in air will be based on the concentration of lead in blood. Critical effects to be considered in the adult organism include elevation of free erythrocyte protoporphyrin, whereas for children cognitive deficit, hearing impairment and disturbed vitamin D metabolism *(13, 14)* are taken as the decisive effects. All of these effects are considered adverse. A critical level of lead in blood of 100 µg/l is proposed. It should be stressed that all of these values are based on population studies yielding group averages, which apply to the individual child only in a probabilistic manner. Although some lead salts have been found to be carcinogenic in animals, the evidence for a carcinogenic potential in humans is inadequate and will, therefore, not be considered here.

For the derivation of a guideline value, the following arguments have been considered.

- Currently measured "baseline" blood lead levels of minimal anthropogenic origin are probably in the range10–30 µg/l.

- Various international expert groups have determined that the earliest adverse effects of lead in populations of young children begin at 100–150 µg/l. Although it cannot be excluded that population effects may occur below this range, it is assumed to be prudent to derive a guideline value based on the lowest value in this range (100 µg/l).

- It can be assumed that inhalation of airborne lead is a significant route of exposure for adults (including pregnant women) but is of less significance for young children, for whom other pathways of exposure such as ingested lead are generally more important.

- It appears that 1 µg lead per m^3 air directly contributes approximately 19 µg lead per litre blood in children and about 16 µg per litre blood in adults, although it is accepted that the relative contribution from air is less significant in children than in adults. These values are approximations, recognizing that the relationships are curvilinear in nature and will apply principally at lower blood lead levels.

- It must be taken into account that, in typical situations, an increase of lead in air also contributes to increased lead uptake by indirect environmental pathways. To correct for uptake by other routes as well, it is assumed that 1 μg lead per m^3 air would contribute to 50 μg lead per litre blood.

- It is recommended that efforts be made to ensure that at least 98% of an exposed population, including preschool children, have blood lead levels that do not exceed 100 μg/l. In this case, the median blood lead level would not exceed 54 μg/l. On this basis, the annual average lead level in air should not exceed 0.5 μg/m^3. This proposal is based on the assumption that the upper limit of nonanthropogenic blood is 30 μg/l. These estimates are assumed to protect adults also.

- To prevent further increases of lead in soils and consequent increases in the exposure of future generations, air lead levels should be kept as low as possible.

Since both direct and indirect exposure of young children to lead in air occurs, the air guidelines for lead should be accompanied by other preventive measures. These should specifically take the form of monitoring the lead content of dust and soils arising from lead fallout. The normal hand-to-mouth behaviour of children with regard to dust and soil defines these media as potentially serious sources of exposure. A specific monitoring value is not recommended. Some data indicate that lead fallout in excess of 250 μg/m^2 per day will increase blood lead levels.

References

1. Delumyea, R. & Kalivretenos, A. Elemental carbon and lead content of fine particles from American and French cities of comparable size and industry, 1985. *Atmospheric environment*, **21**: 1643–1647 (1987).
2. Ministerium für Umwelt, Raumordnung und Landwirtschaft des Landes NW. *Luftreinhaltung in Nordrhein-Westfalen. Eine Erfolgsbilanz der Luftreinhalteplanung 1975–1988*. Bonn, Bonner Universitätsdruckerei, 1989.
3. Ducoffre, G. et al. Lowering time trend of blood lead levels in Belgium since 1978. *Environmental research*, **51**: 25–34 (1990).
4. Stollery, B.T. et al. Cognitive functioning in lead workers. *British journal of industrial medicine*, **46**: 698–707 (1989).
5. Stollery, B.T. et al. Short term prospective study of cognitive functioning in lead workers. *British journal of industrial medicine*, **48**: 739–749 (1991).

6. SEPPÄLÄINEN, A.M. ET AL. Subclinical neuropathy at "safe" levels of lead exposure. *Archives of environmental health*, **30**: 180–183 (1975).
7. SEPPÄLÄINEN, A.M. & HERNBERG, S. A follow-up study of nerve conduction velocities in lead-exposed workers. *Neurobehavioral toxicology and teratology*, **4**: 721–723 (1982).
8. DAVIS, J.M. & SVENDSGAARD, D.J. Nerve conduction velocity and lead: a critical review and meta-analysis. *In*: Johnson, B.L., ed. *Advances in neurobehavioral toxicology*. Chelsea, Lewis Publishers, 1990, pp. 353–376.
9. HERNBERG, S. & NIKKANEN, J. Enzyme inhibition by lead under normal urban conditions. *Lancet*, **1**: 63–64 (1970).
10. SCHWARTZ, J. ET AL. Lead-induced anemia: dose–response relationships and evidence for a threshold. *American journal of public health*, **80**: 165–168 (1990).
11. SCHWARTZ, J. Low-level lead exposure and children's IQ: a meta-analysis and search for a threshold. *Environmental research*, **65**: 42–55 (1994).
12. *Inorganic lead*. Geneva, World Health Organization, 1995 (Environmental Health Criteria, No. 165).
13. MAHAFFEY, K.R. ET AL. Association between age, blood lead concentration, and serum 1,25-dihydroxycholealciferol levels in children. *American journal of clinical nutrition*, **35**: 1327–1331 (1982).
14. ROSEN, J.F. ET AL. Reduction in 1,25-dihydroxyvitamin D in children with increased lead absorption. *New England journal of medicine*, **302**: 1128–1131 (1980).

6.8 Manganese

Exposure evaluation

In urban and rural areas without significant manganese pollution, annual averages are mainly in the range of 0.01–0.07 μg/m^3; near foundries the level can rise to an annual average of 0.2–0.3 μg/m^3 and, where ferro- and silico-manganese industries are present, to more than 0.5 μg/m^3, with individual 24-hour concentrations sometimes exceeding 10 μg/m^3 *(1, 2)*.

Health risk evaluation

The toxicity of manganese varies according to the route of exposure. By ingestion, manganese has relatively low toxicity at typical exposure levels and is considered a nutritionally essential trace element. By inhalation, however, manganese has been known since the early nineteenth century to be toxic to workers. Manganism is characterized by various psychiatric and movement disorders, with some general resemblance to Parkinson's disease in terms of difficulties in the fine control of some movements, lack of facial expression, and involvement of underlying neuroanatomical (extrapyramidal) and neurochemical (dopaminergic) systems *(3–5)*. Respiratory effects such as pneumonitis and pneumonia and reproductive dysfunction such as reduced libido are also frequently reported features of occupational manganese intoxication. The available evidence is inadequate to determine whether or not manganese is carcinogenic; some reports suggest that it may even be protective against cancer. Based on this mixed but insufficient evidence, the US Environmental Protection Agency has concluded that manganese is not classifiable as to human carcinogenicity *(6)*. IARC has not evaluated manganese *(7)*.

Several epidemiological studies of workers have provided consistent evidence of neurotoxicity associated with low-level manganese exposure. Sufficient information was available to develop a benchmark dose using the study by Roels et al. *(3)*, thereby obviating the need to account for a LOAEL to NOAEL extrapolation. With regard to exposure, both lifetime integrated respirable dust concentrations as well as current respirable dust concentrations were considered. Correlation between effects and exposure was strongest for eye–hand coordination with current concentration of respirable dust. From the data of Roels et al. *(3)*, lower 95% confidence limits of the best concentration estimate giving respectively a 10% effect ($BMDL_{10}$) of 74 μg/m^3 and a 5% effect ($BMDL_5$) of 30 μg/m^3 were calculated *(8)*. Taking a conservative approach, the lower 95% confidence limit of the $BMDL_5$ values was chosen as representative of the NOAEL.

$BMDL_5$ values for the other exposure measures (time-integrated and average concentration of respirable dust) are not substantially different *(5)*.

In evaluating the potential health risks associated with inhalation exposure to manganese, various uncertainties must be taken into consideration. Virtually all of the human health evidence is based on healthy, adult male workers; other, possibly more sensitive populations have not been adequately investigated. Also, the potential reproductive and developmental toxicity of inhaled manganese has not been fully investigated.

Guidelines

Based on neurotoxic effects observed in occupationally exposed workers and using the benchmark approach, an estimated NOAEL (the lower 95% confidence limit of the $BMDL_5$) of 30 $\mu g/m^3$ was obtained. A guideline value for manganese of 0.15 $\mu g/m^3$ was derived by dividing by a factor of 4.2 to adjust for continuous exposure and an uncertainty factor of 50 (10 for interindividual variation and 5 for developmental effects in younger children). This latter factor was chosen by analogy with lead where neurobehavioural effects were found in younger children at blood lead levels five times lower than in adults and supported by evidence from studies of experimental animals. The adjustment for continuous exposure was considered sufficient to account for long-term exposure based on knowledge of the half-time of manganese in the brain. The guideline value should be applied as an annual average.

References

1. *Reevaluation of inhalation health risks associated with methylcyclopentadienyl manganese tricarbonyl (MMT) in gasoline*. Washington, DC, US Environmental Protection Agency, 1994.
2. PACE, T. G. & FRANK, N. H. *Procedures for estimating probability of non-attainment of a PM_{10} NAAQS using total suspended particulate or inhalable particulate data*. Research Triangle Park, NC, US Environmental Protection Agency, 1983.
3. ROELS, H. A. ET AL. Assessment of the permissible exposure level to manganese in workers exposed to manganese dioxide dust. *British journal of industrial medicine*, **49**: 25–34 (1992).
4. IREGREN, A. Psychological test performance in foundry workers exposed to low levels of manganese. *Neurotoxicology and teratology*, **12**: 673–675 (1990).
5. MERGLER, D. ET AL. Nervous system dysfunction among workers with long-term exposure to manganese. *Environmental research*, **64**: 151–180 (1994).

6. Integrated Risk Information System (IRIS). *Carcinogenicity assessment for lifetime exposure to manganese* (http://www.epa.gov/ngispgm3/iris/subst/0373.htm#II). Cincinnati, OH, US Environmental Protection Agency (accessed 25 May 1988).
7. Boffetta, P. Carcinogenicity of trace elements with reference to evaluations made by the International Agency for Research on Cancer. *Scandinavian journal of work, environment and health*, **19** (Suppl. 1): 67–70 (1993).
8. Slob, W. et al. *Review of the proposed WHO air quality guideline for manganese*. Bilthoven, National Institute of Public Health and Environmental protection (RIVM), 1996 (Report No. 6135100001).

6.9 Mercury

Exposure evaluation

In areas remote from industry, atmospheric levels of mercury are about 2–4 ng/m^3, and in urban areas about 10 ng/m^3. This means that the daily amount absorbed into the bloodstream from the atmosphere as a result of respiratory exposure is about 32–64 ng in remote areas, and about 160 ng in urban areas. However, this exposure to mercury from outdoor air is marginal compared to exposure from dental amalgams, given that the estimated average daily absorption of mercury vapour from dental fillings varies between 3000 and 17 000 ng.

Health risk evaluation

Sensitive population groups

With regard to exposure to mercury vapour, sensitive population groups have not been conclusively identified from epidemiological, clinical or experimental studies. Nevertheless, the genetic expression of the enzyme catalase, which catalyses the oxidation of mercury vapour to divalent mercuric ion, varies throughout populations. Swiss and Swedish studies have revealed a gene frequency of the order of 0.006 for this trait *(1, 2)*. Thus 30–40 per million of the population are almost completely lacking catalase activity (homozygotes) and 1.2% are heterozygotes with a 60% reduction in catalase activity. Information is lacking on the degree to which other enzymes in the blood are able to take over the oxidation.

Effects on the kidney of inorganic mercury and phenylmercury are believed to occur first in a subgroup of individuals whose susceptibility may be genetically determined, although the proportion of this subgroup in the general population is unknown. Virtually nothing is known about the relative sensitivity at different stages of the life cycle to mercury vapour or inorganic cationic compounds, except that the developing rat kidney is less sensitive than the mature tissue to inorganic mercury *(3)*.

The prenatal stage appears to be the period of life when sensitivity to methylmercury is at its greatest; neuromotor effects in exposed Iraqi populations indicated that sensitivity at this time is at least three times greater than that in adults *(4)*.

Mercury vapour

Time-weighted air concentrations are the usual means of assessing human exposure. Reported air values depend on the type of sampling. Static sampling generally gives lower values than personal sampling. In order to convert the air concentrations quoted in Table 19 to equivalent concentrations in ambient air, two factors have to be taken into account. First, the air concentrations listed in Table 19 were measured in the working environment using static samplers. The conversion factor may vary, depending on exposure conditions. The values shown should be increased by a factor of 3 to correspond to the true air concentrations inhaled by the workers as determined by personal samplers. Second, the total amount of air inhaled at the workplace per week is assumed to be 50 m^3 (10 m^3/day × 5 days) whereas the amount of ambient air inhaled per week would be 140 m^3 (20 m^3/day × 7 days). Thus the volume of ambient air inhaled per week is approximately three times the volume inhaled at the workplace. Thus, to convert the workplace air concentrations quoted in Table 19 to equivalent ambient air concentrations, they should first be multiplied by 3 to convert to actual concentrations in the workplace, and divided by 3 to correct for the greater amount of ambient air inhaled per week by the average adult. It follows that the mercury vapour concentrations quoted in Table 19 are approximately equivalent to ambient air concentrations.

Table 19. Concentrations of total mercury in air and urine at which effects are observed at a low frequency in workers subjected to long-term exposure to mercury vapour

Observed effect [a]	Mercury level		Reference
	Air [b] (µg/m^3)	Urine (µg/litre)	
Objective tremor	30	100	*(5)*
Renal tubular effects; changes in plasma enzymes	15 [c]	50	*(6)*
Nonspecific symptoms	10–30	25–100	*(5)*

[a] These effects occur with low frequency in occupationally exposed groups. Other effects have been reported, but air and urine levels are not available.

[b] The air concentrations measured by static air samplers are taken as a time-weighted average, assuming 40 hours per week for long-term exposure (at least five biological half-times, equivalent to 250 days).

[c] Calculated from the urine concentration, assuming that a mercury concentration in air of 100 µg/m^3 measured by static samplers is equivalent to a mercury concentration of 300 µg/litre in the urine.

Since these figures are based on observations in humans, an uncertainty factor of 10 would seem appropriate. However, the LOAELs in Table 19 are rough estimates of air concentrations at which effects occur at a "low frequency". Because it seems unlikely that such effects would occur in occupationally exposed workers at air concentrations as low as one half of those given in Table 19, it seems appropriate to use an uncertainty factor of 20. Thus, the estimated guideline for mercury concentration in air would be 1 μg/m^3.

Inorganic compounds

Cationic forms of inorganic mercury are retained in the lungs about half as efficiently as inhaled mercury vapour (40% versus 80% retained); thus the estimated guideline providing adequate protection against renal tubular effects would be twice as high as that for mercury vapour.

Methylmercury compounds

It does not seem appropriate to set air quality guidelines for methylmercury compounds. Inhalation of this form of mercury, if it is present in the atmosphere, would make a negligible contribution to total human intake. Nevertheless, mercury in the atmosphere may ultimately be converted to methylmercury following deposition on soils or sediments in natural bodies of water, leading to an accumulation of that form of mercury in aquatic food chains. In this situation, guidelines for food intake would be appropriate, such as those recommended by the Joint FAO/WHO Expert Committee on Food Additives.

Guidelines

It is necessary to take into account the different forms of mercury in the atmosphere and the intake of these forms of mercury from other media. The atmosphere and dental amalgam are the sole sources of exposure to mercury vapour, whereas the diet is the dominant source of methylmercury compounds.

Current levels of mercury in outdoor air, except for regional "hot spots", are typically in the order of 0.005–0.010 μg/m^3 and thus are marginal compared to exposure from dental amalgam. The exposure to mercury from outdoor air at these air levels is not expected to have direct effects on human health.

The predominant species of mercury present in air, Hg^0, is neither mutagenic nor carcinogenic. Exposure to airborne methylmercury is 2–3 orders of magnitude below the food-related daily intake and will, in this context, be regarded as insignificant. It is thus only possible to derive a numerical guideline for inhalation of inorganic mercury, by including mercury vapour and divalent mercury.

The LOAELs for mercury vapour are around 15–30 μg/m³. Applying an uncertainty factor of 20 (10 for uncertainty due to variable sensitivities in higher risk populations and, on the basis of dose–response information, a factor of 2 to extrapolate from a LOAEL to a likely NOAEL), a guideline for inorganic mercury vapour of 1 μg/m³ as an annual average has been established. Since cationic inorganic mercury is retained only half as much as the vapour, the guideline also protects against mild renal effects caused by cationic inorganic mercury. Present knowledge suggests, however, that effects on the immune system at lower exposures cannot be excluded.

An increase in ambient air levels of mercury will result in an increase in deposition in natural bodies of water, possibly leading to elevated concentrations of methylmercury in freshwater fish. Such a contingency might have an important bearing on acceptable levels of mercury in the atmosphere. Unfortunately, the limited knowledge of the global cycle and of the methylation and bioaccumulation pathways in the aquatic food chain does not allow any quantitative estimates of risks from these post-depositional processes. Therefore, an ambient air quality guideline value that would fully prevent the potential for adverse health impacts of post-depositional methylmercury formation cannot be proposed. To prevent possible health effects in the near future, however, ambient air levels of mercury should be kept as low as possible.

References

1. AEBI, H. Investigation of inherited enzyme deficiencies with special reference to acatalasia. *In:* Crow, J.F. & Neel, J.V., ed. *Proceedings from 3rd International Congress of Human Genetics, Chicago 1966.* Baltimore, MD, Johns Hopkins, 1967, p.189.
2. PAUL, K.G. & ENGSTEDT, L.M. Normal and abnormal catalase activity in adults. *Scandinavian journal of clinical laboratory investigation,* **10**: 26 (1958).
3. DASTON, G.P. ET AL. Toxicity of mercuric chloride to the developing rat kidney. I. Post-natal ontogeny of renal sensitivity. *Toxicology and applied pharmacology,* **71**: 24–41(1983).
4. AL-SHAHRISTANI, H. & SHIHAB, K.M. Variation of biological half-life of methylmercury in man. *Archives of environmental health*, **18**: 342–344 (1974).

5. *Inorganic mercury*. Geneva, World Health Organization, 1991 (Environmental Health Criteria, No. 118).
6. CÁRDENAS, A. ET AL. Markers of early renal changes induced by industrial pollutants. I. Application to workers exposed to mercury vapour. *British journal of industrial medicine,* **50**: 17–27 (1993).

6.10 Nickel

Exposure evaluation

Nickel is present throughout nature and is released into air and water both from natural sources and as a result of human activity.

In nonsmokers, about 99% of the estimated daily nickel absorption stems from food and water; for smokers the figure is about 75%. Nickel levels in the ambient air are in the range 1–10 ng/m^3 in urban areas, although much higher levels (110–180 ng/m^3) have been recorded in heavily industrialized areas and larger cities. There is, however, limited information on the species of nickel in ambient air.

Consumer products made from nickel alloys and nickel-plated items lead to cutaneous contact exposure.

Exposure to nickel levels of 10–100 mg/m^3 have been recorded for occupational groups, with documented increased cancer risk. Exposure levels in the refining industry are currently usually less than 1–2 mg/m^3, often less than 0.5 mg/m^3. Experimental and epidemiological data indicate that the nickel species in question is important for risk estimation.

Health risk evaluation

Allergic skin reactions are the most common health effect of nickel, affecting about 2% of the male and 11% of the female population. Nickel content in consumer products and possibly in food and water are critical for the dermatological effect. The respiratory tract is also a target organ for allergic manifestations of occupational nickel exposure.

Work-related exposure in the nickel-refining industry has been documented to cause an increased risk of lung and nasal cancers. Inhalation of a mixture of oxidic, sulfidic and soluble nickel compounds at concentrations higher than 0.5 mg/m^3, often considerably higher, for many years has been reported *(1)*.

Nickel has a strong and prevalent allergenic potency. There is no evidence that airborne nickel causes allergic reactions in the general population, although this reaction is well documented in the working environment. The key criterion for assessing the risk of nickel exposure is its carcinogenic potential.

In general, nickel compounds give negative results in short-term bacterial mutagenicity tests because of limited uptake. Nevertheless, they show a wide range of transformation potencies in mammalian cell assays, depending mainly on their bioavailability.

Both green nickel oxide and the subsulfide have caused tumours in animal inhalation studies. In addition, nickel monoxide (not further specified) and an alloy with 66.5% nickel and 12.5% chromium caused tumours following tracheal instillation. A corresponding instillation with an alloy of 26.8% nickel and 16.2% chromium had no such effect, indicating that it was nickel and not chromium that caused the tumours. Injection-site tumours in a number of organs are found with many particulate nickel compounds. The tumorigenic potency varies with chemical composition, solubility and particle surface properties *(2, 3)*.

Epidemiological evidence from the nickel-refining industry indicates that sulfidic, oxidic and soluble nickel compounds are all carcinogenic. Exposure to metallic nickel has not been demonstrated to cause cancer in workers.

Several theories have been suggested for the mechanisms of nickel tumorigenesis. All of these assume that the nickel ion is the ultimate active agent. On the basis of the underlying concept that all nickel compounds can generate nickel ions that are transported to critical sites in target cells, IARC has classified nickel compounds as carcinogenic to humans (Group 1) and metallic nickel as possibly carcinogenic to humans (Group 2B) *(4)*.

On the basis of one inhalation study *(5)*, the US Environmental Protection Agency (EPA) classified nickel subsulfide as a class A carcinogen and estimated the maximum likelihood incremental unit risk to be $1.8–4.1 \times 10^{-3}$ *(6)*. This study, however, involves only exposure to nickel subsulfide. It is not known whether this compound is present in ambient air, but since it is probably one of the most potent nickel compounds, this risk estimate may represent an upper limit, if accepted. WHO estimated an incremental unit risk of 4×10^{-4} per $\mu g/m^3$ calculated from epidemiological results *(7)*.

On the basis of epidemiological studies, EPA classified nickel dust as a class A carcinogen and estimated the lifetime cancer risk from exposure to nickel dust to be 2.4×10^{-4}. This estimate placed nickel in the third quartile of the 55 substances evaluated by the EPA Carcinogen Assessment Group with regard to their relative carcinogenic potency *(8)*. Assuming a content of 50% of nickel subsulfide in total dust, a unit risk of 4.8×10^{-4} was estimated for this compound.

An estimate of unit risk can be given on the basis of the report of lung cancer in workers first employed between 1968 and 1972 and followed through to 1987 in Norway *(9, 10)*. Using the estimated risk of 1.9 for this group and an exposure of 2.5 mg/m^3, a lifetime exposure of 155 µg/m^3 and a unit risk of 3.8×10^{-4} per µg/m^3 can be calculated.

Guidelines

Even if the dermatological effects of nickel are the most common, such effects are not considered to be critically linked to ambient air levels.

Nickel compounds are human carcinogens by inhalation exposure. The present data are derived from studies in occupationally exposed human populations. Assuming a linear dose–response, no safe level for nickel compounds can be recommended.

On the basis of the most recent information of exposure and risk estimated in industrial populations, an incremental risk of 3.8×10^{-4} can be given for a concentration of nickel in air of 1 µg/m^3. The concentrations corresponding to an excess lifetime risk of 1:10 000, 1:100 000 and 1: 1 000 000 are about 250, 25 and 2.5 ng/m^3, respectively.

References

1. Report of the International Committee on Nickel Carcinogenesis in Man. *Scandinavian journal of work, environment and health*, **16**: 1–82 (1990).
2. Sunderman, F.W., Jr. Search for molecular mechanisms in the genotoxicity of nickel. *Scandinavian journal of work, environment and health*, **19**: 75–80 (1993).
3. Costa, M. et al. Molecular mechanisms of nickel carcinogenesis. *Science of the total environment*, **148**: 191–200 (1994).
4. Nickel and nickel compounds. *In: Chromium, nickel and welding*. Lyons, International Agency for Research on Cancer, 1990, pp. 257–445 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 49).
5. Ottolenghi, A.D. et al. Inhalation studies of nickel sulfide in pulmonary carcinogenesis of rats. *Journal of the National Cancer Institute*, **54**: 1165–1172 (1974).
6. Integrated Risk Information System (IRIS). *Reference concentration (RfC) for inhalation exposure for nickel subsulfide* (http://www.epa.gov/ngispgm3/iris/subst/0273.htm).Cincinnati, OH, US Environmental Protection Agency (accessed 1 April 1987).
7. Nickel. *In: Air Quality Guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23), pp. 285–296.

8. INTEGRATED RISK INFORMATION SYSTEM (IRIS). *Reference concentration (RfC) for inhalation exposure for nickel refinery dust* (http://www.epa.gov/ngispgm3/iris/subst/0272.htm). Cincinnati, OH, US Environmental Protection Agency (accessed 1 April 1987).
9. ANDERSEN, A. Recent follow–up of nickel refinery workers in Norway and respiratory cancer. *In:* Nieboer, E. & Nriagu, J.O., ed. *Nickel and human health*. New York, Wiley, 1992, pp. 621–628.
10. ANDERSEN, A. ET AL. Exposure to nickel compounds and smoking in relation to incidence of lung and nasal cancer among nickel refinery workers. *Occupational and environmental medicine,* **53**: 708–713 (1996).

6.11 Platinum

Exposure evaluation

There is currently very little information on levels of exposure to soluble platinum compounds in the general environment, and there are no authenticated observations on adverse health effects in the population resulting from such exposure. The available data derived from air sampling and from dust deposition of total platinum are limited. Ambient air concentrations of platinum compounds that would occur in different scenarios have been estimated using dispersion models developed by the US Environmental Protection Agency *(1)*. Ambient air concentrations of total platinum in various urban exposure situations, assuming an average emission rate of approximately 20 ng/km from the monolithic three-way catalyst, were estimated. These concentrations are lower by a factor of 100 than those estimated for the old, pellet-type catalyst. In the exposure conditions studied, estimated ambient air concentrations of platinum ranged from 0.05 pg/m^3 to 0.09 ng/m^3. The WHO Task Group on Environmental Health Criteria for Platinum considered that environmental contamination with platinum from the monolithic three-way catalyst is likely to be very low or negligible *(2)*. The Group concluded that platinum-containing exhaust emissions from such catalysts most probably do not pose a risk for adverse health effects in the general population but it was recommended that, to be on the safe side, the possibility should be kept under review.

A recently completed pilot study sought to acquire information on direct and indirect sources and emissions of platinum group metals in the United Kingdom environment *(3)*. With regard to emissions from motor vehicle catalytic converters, samples of road dusts and soils were collected from areas with high and low traffic flows, for platinum and lead estimation. Higher levels of platinum were found in dusts and soils at major road intersections and on roads with high traffic densities, indicating traffic as the source of platinum at these sites *(4)*.

As platinum in road dust is at least partially soluble, it may enter the food chain so that diet may also be a major source of platinum intake in the non-industrially exposed population. This is suggested by the total diet study carried out in Australia in Sydney, an area of high traffic density, and Lord Howe Island, an area with very low traffic density. Blood platinum levels were similar in the two locations *(5)*.

In early studies on platinum-exposed workers, exposure levels were high. Values ranged from 0.9 to 1700 μg/m^3 in four British platinum refineries, giving rise to symptoms in 57% of the exposed workers *(6)*. Following the adoption of an occupational exposure limit with a threshold limit value (TLV) for soluble platinum salts of 2 μg/m^3 as an 8-hour time-weighted average, the incidence of platinum salt hypersensitivity has fallen, but sensitization in workers has still been observed. Thus, in a cross-sectional survey, skin sensitization was reported in 19% of 65 workers in a platinum refinery, where analysis of airborne dust showed levels of soluble platinum of 0.08–0.1 μg/m^3 in one department and less than 0.05 μg/m^3 in other areas *(7)*. In another plant with air levels generally below 0.08 μg/m^3, 20% of exposed workers were sensitized *(8)*. It is possible, however, that short, sharp exposures to concentrations above the TLV could have been responsible for some of these effects. In a 4-month study in a United States platinum refinery with a high prevalence of rhinitis and asthma, workplace concentrations exceeded the occupational limit of 2 μg/m^3 for 50–75% of the time *(9)*. The risk of developing platinum salt sensitivity appears to be correlated with exposure intensity, the highest incidence occurring in groups with the highest exposure, although no unequivocal concentration–effect relationship can be deduced from the reported studies.

Health risk evaluation

There is no convincing evidence for sensitization or for other adverse health effects following exposure to metallic platinum. Exposure to the halogenated platinum complexes already described has given rise to sensitization following occupational exposure to platinum concentrations in air greater than the TLV of 2 μg/m^3, and may have caused sensitization reactions at concentrations down to and even below the limit of detection in workplace monitoring of 0.05 μg/m^3. Furthermore, as subsequent exposure to minute concentrations of these platinum salts may lead to a recurrence of the health effects shown in Table 20 in previously sensitized subjects, it is not possible to define a no-effect level for these platinum compounds.

Because the correlation between platinum exposure concentration and the development of sensitization is unknown, the WHO Task Group *(2)* considered that a recommendation for a reduction in the occupational exposure limit cannot at present be justified. It did, however, recommend that the occupational exposure limit of 2 μg/m^3 be changed from an 8-hour time-weighted average to a ceiling value, and that personal sampling devices be used in conjunction with area sampling to determine more correctly the true platinum exposure. Should it be ascertained unequivocally that sensitization has occurred in workers consistently exposed to platinum

Table 20. Concentration–effect data for platinum

Concentration range	Average duration of exposure	Frequency of health effects in the general population	Health effects in susceptible groups
Airborne dust level for soluble platinum salts above the TLV time-weighted average of 2 μg/m³	Varies from weeks to years	No data available	In some occupationally exposed individuals: conjunctivitis, rhinitis, cough, wheeze, dyspnoea, asthma, contact dermatitis, urticaria, mucous membrane inflammation
Airborne dust level for soluble platinum salts < 0.05 μg/m³			Possibility that the above effects cannot be excluded Recurrence of the above effects in subjects previously sensitized Conversion to positive skin-prick test

levels below the current exposure limit of 2 μg/m³, and that intermittent, short exposures above this level had not taken place, there would be strong grounds for reducing the exposure limit.

The degree of solubilization and perhaps conversion to halide complexes of platinum particulate matter emitted into the general environment is not known, but is likely to be small. The prevalence of asthma in industrialized communities is increasing markedly. While there are no observations to suggest that platinum (emitted from vehicle catalytic converters or from industrial sources, deposited and in part converted in the general environment into halide salts) may act as an etiological agent, it would be inappropriate in the present state of knowledge to propose a no-effect level. From observations following occupational exposure, a value of 0.05 μg/m³ for soluble platinum salts may be considered as a tentative LOAEL. Platinum levels in air in the general environment are at least three orders of magnitude below this figure.

While *cis*-platin, an IARC Group 2A carcinogen, is released into the environment following medical use, there are no grounds for considering this platinum compound or its analogues as significant atmospheric pollutants.

Guidelines

In occupational settings, sensitization reactions have been observed for soluble platinum down to the limit of detection of 0.05 μg/m^3. However, these effects have occurred only in individuals previously sensitized by higher exposure levels. It is unlikely that the general population exposed to ambient concentrations of soluble platinum, which are at least three orders of magnitude lower, will develop similar effects. At present no specific guideline value is recommended but further studies are required, in particular on the speciation of platinum in the environment.

References

1. ROSNER, G. & MERGET, R. Allergenic potential of platinum compounds. *In:* Dayan, A.D. et al., ed. *Immunotoxicity of metals and immunotoxicology.* New York, Plenum Press, 1990, pp. 93–101.
2. *Platinum.* Geneva, World Health Organization, 1991 (Environmental Health Criteria, No. 125).
3. FARAGO, M..E. *Platinum group metals in the environment: their use in vehicle exhaust catalysts and implications for human health in the UK. A report prepared for the Department of the Environment.* London, IC Consultants, 1995.
4. FARAGO, M.E. ET AL. Platinum metal concentrations in urban road dust and soil in the United Kingdom. *Fresenius journal of analytical chemistry*, **354**: 660–663 (1996).
5. VAUGHAN, G.T. & FLORENCE, T.M. Platinum in the human diet, blood, hair and excreta. S*cience of the total environment*, **111**: 47–58 (1992).
6. HUNTER, D. ET AL. Asthma caused by the complex salts of platinum. *British journal of industrial medicine*, **2**: 92–98 (1945).
7. BOLM-AUDORFF, U. ET AL. On the frequency of respiratory allergies in a platinum processing factory. *In:* Baumgartner, E. et al., ed. *Industrial change – occupational medicine facing new questions. Report of the 28th Annual Meeting of the German Society for Occupational Medicine, Innsbruck, Austria, 4–7 May, 1988.* Stuttgart, Gentner Verlag, 1988, pp. 411–416.
8. MERGET, R. ET AL. Asthma due to the complex salts of platinum – a cross sectional survey of workers in a platinum refinery. *Clinical allergy,* **18**: 569–580 (1988).
9. CALVERLEY, A.E. ET AL. Platinum salt sensitivity in refinery workers. Incidence and effects of smoking and exposure. *Occupational and environmental medicine,* **52**: 661–666 (1995).

6.12 Vanadium

Exposure evaluation

The natural background level of vanadium in air in Canada has been reported to be in the range 0.02–1.9 ng/m^3 *(1)*. Vanadium concentrations recorded in rural areas varied from a few nanograms to tenths of a nanogram per m^3, and in urban areas from 50 ng/m^3 to 200 ng/m^3. In cities during the winter, when fuel oil with a high vanadium content was used for heating, concentrations as high as 2000 ng/m^3 were reported. Air pollution by industrial plants may be less than that caused by power stations and heating equipment.

The concentrations of vanadium in workplace air (0.01–60 mg/m^3) are much higher than those in the general environment.

Health risk evaluation

The acute and chronic effects of vanadium exposure on the respiratory system of occupationally exposed workers should be regarded as the most significant factors when establishing air quality guidelines. Most of the clinical symptoms reported reflect irritative effects of vanadium on the upper respiratory tract, except at higher concentrations (above 1 mg vanadium per m^3), when more serious effects on the lower respiratory tract are observed. Clinical symptoms of acute exposure are reported *(2)* in workers exposed to concentrations ranging from 80 µg to several mg vanadium per m^3, and in healthy volunteers *(3)* exposed to concentrations of 56–560 $\mu g/m^3$ (Table 21).

A study of occupationally exposed groups provides data reasonably consistent with those obtained from controlled acute human exposure experiments, suggesting that the LOAEL for acute exposure can be considered to be 60 $\mu g/m^3$.

Chronic exposure to vanadium compounds revealed a continuum in the respiratory effects, ranging from slight changes in the upper respiratory tract, with irritation, coughing and injection of pharynx, detectable at 20 $\mu g/m^3$, to more serious effects such as chronic bronchitis and pneumonitis, which occurred at levels above 1 mg/m^3. Occupational studies illustrate the concentration–effect relationship at low levels of exposure *(4–6)*, showing increased prevalence of irritative symptoms of the upper respiratory tract; this suggests that 20 $\mu g/m^3$ can be regarded as the LOAEL for

Table 21. Respiratory effects after acute and chronic exposures to low levels of vanadium

Type of exposure	Vanadium compound	Concentration in µg/m^3 Compound	Vanadium	Symptoms	Reference
Acute					
Boiler cleaning	V_2O_5 V_2O_3	523	80	Changes in parameters of lung functions	*(2)*
Clinical study (experimental 8-hour exposure)	V_2O_5	1000	560	Respiratory irritation: persistent and frequent cough, expiratory wheezes	*(3)*
	V_2O_5	200	112	Persistent cough (7- 1 0 days)	
	V_2O_5	100	56	Slight cough for 4 days	
Chronic					
Vanadium refinery	V_2O_5	536	300	Respiratory irritation: cough, sputum, nose and throat irritation, injected pharynx	*(4)*
Vanadium refinery	V_2O_5	18-71	10-40	Irritative changes of mucous membranes of upper respiratory tract	*(5)*
Vanadium processing	V_2O_5 V_2O_3	—	1.2-12.0	Respiratory irritation: injected pharynx	*(6)*

chronic exposure (Table 21). There are no conclusive data on the health effects of exposure to airborne vanadium at present concentrations in the general population, and a susceptible subpopulation is not known. Vanadium is a potent respiratory irritant, however, which would suggest that asthmatics should be considered a special group at risk.

There are no well documented animal data to support findings in human studies, although one study reported systemic and local respiratory effects in rats at levels of 3.4–15 μg/m³ *(7)*.

Guidelines

Available data from occupational studies suggest that the LOAEL of vanadium can be assumed to be 20 μg/m³, based on chronic upper respiratory tract symptoms. Since the adverse nature of the observed effects on the upper respiratory tract were minimal at this concentration, and a susceptible subpopulation has not been identified, a protection factor of 20 was selected. It is believed that below 1 μg/m³ (averaging time 24 hours) environmental exposure to vanadium is not likely to have adverse effects on health.

The available evidence indicates that the current vanadium levels generally found in industrialized countries are not in the range associated with potentially harmful effects.

References

1. Committee on Biologic Effects of Atmospheric Pollutants. *Vanadium.* Washington, DC, National Academy of Sciences, 1974.
2. Lees, R.E.M. Changes in lung function after exposure to vanadium compounds in fuel oil ash. *British journal of industrial medicine*, **37**: 253–256 (1980).
3. Zenz, C. & Berg, B.A. Human responses to controlled vanadium pentoxide exposure. *Archives of environmental health,* **14**: 709–712 (1967).
4. Lewis, C.E. The biological effects of vanadium. II. The signs and symptoms of occupational vanadium exposure. *AMA archives of industrial health,* **19**: 497–503 (1959).
5. Kiviluoto, M. et al. Effects of vanadium on the upper respiratory tract of workers in a vanadium factory. *Scandinavian journal of work, environment and health,* **5**: 50–58 (1979).
6. Nishiyama, K. et al. [A survey of people working with vanadium pentoxide]. *Shikoku igaku zasshi,* **31**: 389–393 (1977) [Japanese].
7. Paznych, V.M. [Maximum permissible concentration of vanadium pentoxide in the atmosphere]. *Gigiena i sanitarija,* **31**: 6–12 (1966) [Russian].

CHAPTER 7

Classical pollutants

7.1 Nitrogen dioxide

Exposure evaluation

Levels of nitrogen dioxide vary widely because a continuous baseline level is frequently present, with peaks of higher levels superimposed. Natural background annual mean concentrations are in the range 0.4–9.4 μg/m^3. Outdoor urban levels have an annual mean range of 20–90 μg/m^3 and hourly maxima in the range 75–1015 μg/m^3. Levels indoors where there are unvented gas combustion appliances may average more than 200 μg/m^3 over a period of several days. A maximum 1-hour peak may reach 2000 μg/m^3. For briefer periods, even higher concentrations have been measured.

Critical concentration–response data

Monotonic concentration–response data are available only from a few animal studies. Thus, this section will focus on lowest-observed-effect levels and their interpretation.

Short-term exposure effects

Available data from animal toxicology experiments rarely indicate the effects of acute exposure to nitrogen dioxide concentrations of less than 1880 μg/m^3 (1 ppm). Normal healthy people exposed at rest or with light exercise for less than 2 hours to concentrations of more than 4700 μg/m^3 (2.5 ppm) experience pronounced decrements in pulmonary function; generally, such people are not affected at less than 1880 μg/m^3 (1 ppm). One study showed that the lung function of people with chronic obstructive pulmonary disease is slightly affected by a 3.75-hour exposure to 560 μg/m^3 (0.3 ppm) *(1)*. A wide range of findings in asthmatics has been reported; one study observed no effects from a 75-minute exposure to 7520 μg/m^3 (4 ppm) *(2)*, whereas others showed decreases in FEV_1 after 10 minutes of exercise during exposure to 560 μg/m^3 (0.3 ppm) *(3)*.

Asthmatics are likely to be the most sensitive subjects, although uncertainties exist in the health database. The lowest concentration causing effects on pulmonary function was reported from two laboratories that exposed mild asthmatics for 30–110 minutes to 560 μg/m^3 (0.3 ppm) during intermittent exercise. However, neither of these laboratories was able to replicate these responses with a larger group of asthmatic subjects. One of these studies indicated that nitrogen dioxide can increase airway reactivity to cold air in asthmatics. At lower concentrations, the pulmonary function of asthmatics was not changed significantly.

Nitrogen dioxide increases bronchial reactivity as measured by pharmacological bronchoconstrictor agents in normal and asthmatic subjects, even at levels that do not affect pulmonary function directly in the absence of a bronchoconstrictor. Asthmatics appear to be more susceptible. For example, some but not all studies show increased responsiveness to bronchoconstrictors at nitrogen dioxide levels as low as 376–560 μg/m^3 (0.2–0.3 ppm); in other studies, higher levels had no such effect. Because the actual mechanisms are not fully defined and nitrogen dioxide studies with allergen challenges showed no effects at the lowest concentration tested (190 μg/m^3; 0.1 ppm), full evaluation of the health consequences of the increased responsiveness to bronchoconstrictors is not yet possible.

Long-term exposure effects

Studies with animals have clearly shown that several weeks to months of exposure to nitrogen dioxide concentrations of less than 1880 μg/m^3 (1 ppm) cause a plethora of effects, primarily in the lung but also in other organs, such as the spleen, liver and blood. Both reversible and irreversible lung effects have been observed. Structural changes range from a change in cell types in the tracheobronchial and pulmonary regions (lowest reported level 640 μg/m^3) to emphysema-like effects (at concentrations much higher than ambient). Biochemical changes often reflect cellular alterations (lowest reported levels for several studies 380–750 μg/m^3 (0.2–0.4 ppm) but isolated cases at lower effective concentrations). Nitrogen dioxide levels as low as 940 μg/m^3 (0.5 ppm) also increase susceptibility to bacterial and viral infection of the lung *(4)*.

There are no epidemiological studies that can be confidently used quantitatively to estimate long-term nitrogen dioxide exposure durations or concentrations likely to be associated with the induction of unacceptable health risks in children or adults. Because homes with gas cooking appliances have peak nitrogen dioxide levels that are in the same range as levels causing effects in some animal and human clinical studies, epidemiological studies evaluating the effects of nitrogen dioxide exposures in such homes have been of much interest. In general, epidemiological studies on adults and on infants under 2 years showed no significant effect of the use of gas cooking appliances on respiratory illness; nor do the few available studies of infants and adults show any associations between pulmonary function changes and gas stove use. However, children aged 5–12 years are estimated to have a 20% increased risk for respiratory symptoms and disease for each increase in nitrogen dioxide concentration of 28.3 μg/m^3 (2-week average) where the weekly average concentrations are in the range 15–128 μg/m^3 or possibly higher. Nevertheless, the observed effects cannot clearly be attributed to either the repeated short-term high-level peak exposures or long-term exposures in the range of the stated weekly averages (or possibly both).

As hinted at by the indoor studies, the results of outdoor studies tend to point consistently toward increased respiratory symptoms, their duration, and/or lung function decrements being qualitatively associated in children with long-term ambient nitrogen dioxide exposures. Outdoor epidemiology studies, as with indoor studies, however, provide little evidence for the association of long-term ambient exposures with health effects in adults. None of the available studies yields confident estimates of long-term exposure–effect levels, but available results are most clearly suggestive of respiratory effects in children at annual average nitrogen dioxide concentrations of 50–75 μg/m^3 or higher.

Health risk evaluation

Small, statistically significant, reversible effects on lung function and airway responsiveness have been observed in mild asthmatics during a 30-minute exposure to nitrogen dioxide concentrations of 380–560 μg/m^3 (0.2–0.3 ppm). The sequelae of repetitive exposures of such individuals or the impact of single exposures on more severe asthmatics are not known. In most animal experiments, however, 1–6 months of exposure to 560–940 μg/m^3 are required to produce changes in lung structure, lung metabolism and lung defences against bacterial infection. Thus, it is prudent to avoid exposures in humans, because repetitive exposures in animals lead to adverse effects. Animal toxicology studies of lung host defence and morphology suggest that peak concentrations contribute more to the toxicity of nitrogen dioxide than does duration, although duration is still important. Nitrogen dioxide puts children at increased risk of respiratory illness. This is of concern because repeated lung infections in children can cause lung damage later in life.

Nitrogen dioxide presents a dilemma with respect to guidelines. It is clear that the public should be protected from excessive exposure, but the recommendation of a guideline is complicated owing to the difficulties posed by the uncertainties in exposure–response relationships for both acute (< 3-hour) and long-term exposure, and the uncertainties in establishing an appropriate margin of protection. Studies of asthmatics exposed to 380–560 μg/m^3 indicate a change of about 5% in pulmonary function and an increase in airway responsiveness to bronchoconstrictors. Asthmatics are more susceptible to the acute effects of nitrogen dioxide: they have a higher baseline airway responsiveness. Thus, a nitrogen-dioxide-induced increase in airway responsiveness is expected to have clinical implications for exaggerated responses to a variety of provocative agents, such as cold air, allergies or exercise. Concern about asthmatics is also enhanced, considering the increase in the number of asthmatics in many countries (many countries have 4–6% asthmatics). A number of epidemiological studies of relatively large populations exposed indoors to peak levels of nitrogen dioxide from gas-combustion appliances have not provided consistent evidence of adverse pulmonary function effects. In one study, elderly women who used gas stoves had a high prevalence of asthma. Nevertheless, the human clinical studies of function and airway reactivity do not show monotonic concentration responses, and the studies are not internally consistent. Animal studies do not provide substantial evidence of biochemical, morphological or physiological effects in the lung following a single acute exposure to concentrations in the range of the lowest-observed-effect level in humans. On the other hand, the mild asthmatics chosen for the controlled exposure studies do not represent all asthmatics, and there are likely to be some individuals with greater sensitivity to nitrogen dioxide. Furthermore, subchronic and chronic animal studies do show significant morphological, biochemical and immunological changes.

The epidemiological studies discussed show increased risk of respiratory illness in children at an increase in nitrogen dioxide level of about 30 $\mu g/m^3$; most studies measured 2-week averages on personal samplers. It is not known, however, whether the effect was related to this 2-week average, the actual pattern (baseline and peaks) over the 2 weeks, the peaks over the 2 weeks, or some other index for a longer time-frame prior to the study measurement. It is also not possible to clearly discern the relative contributions of indoor and outdoor levels of nitrogen dioxide.

Guidelines

Despite the large number of acute controlled exposure studies on humans, several of which used multiple concentrations, there is no evidence for a clearly defined concentration–response relationship for nitrogen dioxide exposure. For acute exposures, only very high concentrations (1990 $\mu g/m^3$; > 1000 ppb) affect healthy people. Asthmatics and patients with chronic obstructive pulmonary disease are clearly more susceptible to acute changes in lung function, airway responsiveness and respiratory symptoms. Given the small changes in lung function (< 5% drop in FEV_1 between air and nitrogen dioxide exposure) and changes in airway responsiveness reported in several studies, 375–565 $\mu g/m^3$ (0.20–0.30 ppm) is a clear lowest-observed-effect level. A 50% margin of safety is proposed because of the reported statistically significant increase in response to a bronchoconstrictor (increased airway responsiveness) with exposure to 190 $\mu g/m^3$ and a meta-analysis suggesting changes in airway responsiveness below 365 μg/m³. (The significance of the response at 190 μg/m³ (100 ppb) has been questioned on the basis of an inappropriate statistical analysis.)

On the basis of these human clinical data, a 1-hour guideline of 200 μg/m³ is proposed. At double this recommended guideline (400 μg/m³) there is evidence to suggest possible small effects in the pulmonary function of asthmatics. Should the asthmatic be exposed either simultaneously or sequentially to nitrogen dioxide and an aeroallergen, the risk of an exaggerated response to the allergen is increased. At 50% of the suggested guideline (100 μg/m³, 50 ppb) there have been no studies of acute response in 1 hour.

Although there is no particular study or set of studies that clearly support selection of a specific numerical value for an annual average guideline, the database nevertheless indicates a need to protect the public from chronic nitrogen dioxide exposure. For example, indoor air studies with a strong nitrogen dioxide source, such as gas stoves, suggest that an increment of about 30 μg/m³ (2-week average) is associated with a 20% increase in lower respiratory illness in children aged 5–12 years. However, the affected children had a pattern of indoor exposure that included peak exposures higher than those typically encountered outdoors. Thus the results cannot be readily extrapolated quantitatively to the outdoor situation. Outdoor epidemiological studies have found qualitative evidence of ambient exposures being associated with increased respiratory symptoms and lung function decreases in children (most clearly suggestive at annual average concentrations of 50–75 μg/m³ or higher and consistent with findings from indoor studies), although they do not provide clear exposure–response information for nitrogen dioxide. In these epidemiological studies, nitrogen dioxide has appeared to be a good indicator of the pollutant mixture. Furthermore, animal toxicological studies show that prolonged exposures can cause decreases in lung host defences and changes in lung structure. On these grounds, it is proposed that a long-term guideline for nitrogen dioxide be established. Selecting a well supported value based on the studies reviewed has not been possible, but it has been noted that a prior review conducted for the Environmental Health Criteria document on nitrogen oxides recommended an annual value of 40 μg/m³ *(5)*. In the absence of support for an alternative value, this figure is recognized as an air quality guideline.

References

1. Morrow, P.E. & Utell, M.J. *Responses of susceptible subpopulations to nitrogen dioxide*. Cambridge, MA, Health Effects Institute, 1989 (Research Report, No. 23).

2. LINN, W.S. & HACKNEY, J.D. *Short-term human respiratory effects of nitrogen dioxide: determination of quantitative dose–response profiles, phase II. Exposure of asthmatic volunteers to 4 ppm NO_2*. Atlanta, GA, Coordinating Research Council, Inc., 1984 (Report No. CRC-CAPM-48-83-02).
3. ROGER, L.J. ET AL. Pulmonary function, airway responsiveness, and respiratory symptoms in asthmatics following exercise in NO_2. *Toxicology and industrial health*, **6**: 155–171 (1990).
4. EHRLICH, R. & HENRY M.C. Chronic toxicity of nitrogen dioxide. I. Effect on resistance to bacterial pneumonia. *Archives of environmental health*, **17**: 860–865 (1968).
5. *Nitrogen oxides*. Geneva, World Health Organization, 1997 (Environmental Health Criteria, No. 188).

7.2 Ozone and other photochemical oxidants

Exposure evaluation

Ozone and other photochemical oxidants are formed by the action of short-wavelength radiation from the sun on nitrogen dioxide. In the presence of volatile organic compounds, the equilibrium favours the formation of higher levels of ozone. Background levels of ozone, mainly of anthropogenic origin, are in the range 40–70 μg/m³ (0.02–0.035 ppm) but can be as high as 120–140 μg/m³ (0.06–0.07 ppm) for 1 hour. In Europe, maximum hourly ozone concentrations may exceed 300 μg/m³ (0.15 ppm) in rural areas and 350 μg/m³ (0.18 ppm) in urbanized regions. Submaximal levels (80–90% of maximum) can occur for 8–12 hours a day for many consecutive days.

Health risk evaluation

Ozone toxicity occurs in a continuum in which higher concentrations, longer exposure duration and greater activity levels during exposure cause greater effects. Short-term acute effects include respiratory symptoms, pulmonary function changes, increased airway responsiveness and airway inflammation. These health effects were statistically significant at a concentration of 160 μg/m³ (0.08 ppm) for 6.6-hour exposures in a group of healthy exercising adults, with the most sensitive subjects experiencing functional decrements of > 10% within 4–5 hours *(1)*. Controlled exposures of heavily exercising adults or children to an ozone concentration of 240 μg/m³ (0.l2 ppm) for 2 hours have also been observed to produce decrements in pulmonary function *(2, 3)*. There is no question that substantial acute adverse effects occur with 1 hour of exercising exposure at concentrations of 500 μg/m³ or higher, particularly in susceptible individuals or subgroups.

Field studies in children, adolescents and young adults have indicated that pulmonary function decrements can occur as a result of short-term exposure to ozone concentrations of 120–240 μg/m³ and higher. Mobile laboratory studies using ambient air containing ozone have observed associations between changes in pulmonary function in children or asthmatics and ozone concentrations of 280–340 μg/m³ (0.14–0.17 ppm) with exposures lasting several hours. Respiratory symptoms, especially cough, have been associated with ozone concentrations as low as 300 μg/m³ (0.15 ppm).

Ozone exposure has also been reported to be associated with increased hospital admissions for respiratory causes and exacerbation of asthma. That these effects are observed both with exposures to ambient ozone (and co-pollutants) and with controlled exposures to ozone alone demonstrates that the functional and symptomatic responses can be attributed primarily to ozone.

A number of studies evaluating rats and monkeys exposed to ozone for a few hours or days have shown alterations in the respiratory tract in which the lowest-observed-effect levels were in the range 160–400 μg/m^3 (0.08–0.2 ppm). These included the potentiation of bacterial lung infections, inflammation, morphological alterations in the lung, increases in the function of certain lung enzymes active in oxidant defences, and increases in collagen content. Long-term exposure to ozone in the range 240–500 μg/m^3 (0.12–0.25 ppm) causes morphological changes in the epithelium and interstitium of the centriacinar region of the lung, including fibrotic changes.

Guidelines

The selection of guidelines for ambient ozone concentrations is complicated by the fact that detectable responses occur at or close to the upper limits of background concentrations. At ozone levels of 200 μg/m^3 and lower (for exposure periods of 1–8 hours) there are statistically significant decrements in lung function, airway inflammatory changes, exacerbations of respiratory symptoms and symptomatic and functional exacerbations of asthma in exercising susceptible people. Functional changes and symptoms as well as increased hospital admissions for respiratory causes are also observed in population studies. Thus it is not possible to base the guidelines on a NOAEL or a LOAEL with an uncertainty factor of more than a small percentage. Thus, selection of a guideline has to be based on the premise that some detectable functional responses are of little or no health concern, and that the number of responders to effects of concern are too few to represent a group warranting protection from exposures to ambient ozone.

In the case of respiratory function responses, a judgement could be made that ozone-related reductions in FEV_1, for example, of < 10% were of no clinical concern. In the case of visits to clinics or emergency departments or hospital admissions for respiratory diseases, it would be necessary to determine how many cases per million population would be needed to constitute a group warranting societal protection. In the case of asthmatic children needing extra medication in response to elevated ozone concentrations, it would be necessary to conclude that medication will be available to sufficiently ameliorate their distress and thereby prevent more serious consequences.

On such a basis, a guideline value for ambient air of 120 μg/m^3 for a maximum period of 8 hours per day is established as a level at which acute effects on public health are likely to be small.

For those public health authorities that cannot accept such levels of health risk, an alternative is to select explicitly some other level of acceptable exposure and associated risk. Tables 22 and 23 summarize the ambient ozone concentrations that are associated with specific levels of response among specified population subgroups. Although chronic exposure to ozone can cause effects, quantitative information from humans is inadequate to estimate the degree of protection from chronic effects offered by this guideline. In any case, the ozone concentration at which any adverse health outcome is expected will vary with the duration of the exposure and the volume of air that is inhaled during the exposure.

Thus, the amount of time spent outdoors and the typical level of activity are factors that should be considered in risk evaluation. Table 22 summarizes the ozone levels at which two representative adverse health outcomes,

Table 22. Health outcomes associated with controlled ozone exposures

Health outcome	Ozone concentration (μg/m^3) at which the health effect is expected	
	Averaging time 1 hour	Averaging time 8 hours
Change in FEV_1 (active, healthy, outdoors, most sensitive 10% of young adults and children):		
5%	250	120
10%	350	160
20%	500	240
Increase in inflammatory changes (neutrophil influx) (healthy young adults at > 40 litres/minute outdoors)		
2-fold	400	180
4-fold	600	250
8-fold	800	320

Table 23. Health outcomes associated with changes in ambient ozone concentration in epidemiological studies

Health outcome	Change in ozone concentration (μg/m³)	
	Averaging time 1 hour	Averaging time 8 hours
Increase in symptom exacerbations among adults or asthmatics (normal activity):		
25%	200	100
50%	400	200
100%	800	300
Increase in hospital admissions for respiratory conditions:[a]		
5%	30	25
10%	60	50
20%	120	100

[a] Given the high degree of correlation between the 1-hour and 8-hour ozone concentration in field studies, the reduction in health risk associated with decreasing 1-hour or 8-hour ozone levels should be almost identical.

based on controlled exposure experiments, may be expected. The concentrations presented in this table have been established by experts on the basis of collective evidence from numerous studies and linear extrapolation in a few cases where data were limited.

Epidemiological data show relationships between changes in various health outcomes and changes in the peak daily ambient ozone concentration. Two examples of such relationships are shown in Table 23. Short-term increases in levels of ambient ozone are associated both with increased hospital admissions with a respiratory diagnosis and respiratory symptom exacerbations, both in healthy people and in asthmatics. These observations may be used to quantify expected improvements in health outcomes that may be associated with lowering the ambient ozone concentration. The values presented in the table assume a linear relationship between ozone concentration and health outcome. Uncertainties exist, however, concerning the forms of these relationships and it is unclear whether similar response slopes can be expected at widely different ambient ozone levels. In the event that such relationships are curvilinear (concave), the benefits of lowering the ozone concentration are likely to be greater when the average ambient level is higher. Consequently, if the ambient ozone concentration is already low, the benefits of lowering the concentration may be less than would be suggested by Table 23. Another important area of uncertainty is the degree to which other pollutants influence these relationships.

The first edition of *Air quality guidelines for Europe (4)* recommended a 1-hour guideline value of 150–200 μg/m^3. Although recent research does not indicate that this guideline would necessarily be erroneous, the 8-hour guideline would protect against acute 1-hour exposures in this range and thus it is concluded that a 1-hour guideline is not necessary. Furthermore, the health problems of greatest concern (increased hospital admissions, exacerbations of asthma, inflammatory changes in the lung, and structural alterations in the lung) are more appropriately addressed by a guideline value that limits average daily exposure, and consequently inhaled dose and dose rate, rather than one designed to cover the rare short-duration deteriorations in air quality that may be associated with unusual meteorological conditions.

A guideline for peroxyacetyl nitrate is not warranted at present since it does not seem to pose a significant health problem at levels observed in the environment.

References

1. Horstman, D.H. et al. Ozone concentration and pulmonary response relationships for 6.6-hour exposures with five hours of moderate exercise to 0.08, 0.10, and 0.12 ppm. *American review of respiratory disease*, **142**: 1158–1163 (1990).
2. McDonnell, W.F. et al. Pulmonary effects of ozone exposure during exercise: dose–response characteristics. *Journal of applied physiology: respiratory and environmental exercise physiology*, **54**: 1345–1352 (1983).
3. Gong, H. Jr et al. Impaired exercise performance and pulmonary function in elite cyclists during low-level ozone exposure in a hot environment. *American review of respiratory disease*, **134**: 726–733 (1986).
4. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).

7.3 Particulate matter

Exposure evaluation

Data on exposure levels to airborne inhalable particles are still limited for Europe. Data have mostly been obtained from studies not directly aimed at providing long-term distributions of exposure data for large segments of the population. Nevertheless, it seems that in northern Europe, PM_{10} levels (particulate matter in which 50% of particles have an aerodynamic diameter of less than 10 µm) are low, with winter averages even in urban areas not exceeding 20–30 µg/m^3. In western Europe, levels seem to be higher at 40–50 µg/m^3, with only small differences between urban and non-urban areas. Levels in some central and eastern European locations from which data are available appear nowadays to be only a little higher than those measured in cities such as Amsterdam and Berlin. As a result of the normal day-to-day variation in PM_{10} concentrations, 24-hour averages of 100 µg/m^3 are regularly exceeded in many areas in Europe, especially during winter inversions.

Health risk evaluation

A variety of methods exist to measure particulate matter in air. For the present evaluation, studies have been highlighted in which particulate matter exposure was expressed as the thoracic fraction (~ PM_{10}) or size fractions or constituents thereof. Practically speaking, at least some data are also available on fine particles ($PM_{2.5}$), sulfates and strong aerosol acidity. Health effect studies conducted with (various forms of) total suspended particulates or black smoke as exposure indicators have provided valuable additional information in recent years. They are, however, less suitable for the derivation of exposure–response relationships for particulate matter, because total suspended particulates include particles that are too large to be inhaled or because the health significance of particle opacity as measured by the black smoke method is uncertain.

Recent studies suggest that short-term variations in particulate matter exposure are associated with health effects even at low levels of exposure (below 100 µg/m^3). The current database does not allow the derivation of a threshold below which no effects occur. This does not imply that no threshold exists; epidemiological studies are unable to define such a threshold, if it exists, precisely.

At low levels of (short-term) exposure (defined as 0–100 µg/m^3 for PM_{10}), the exposure–response curve fits a straight line reasonably well. There are indications from studies conducted in the former German Democratic Republic and in China, however, that at higher levels of exposure (several hundreds of $\mu g/m^3$ PM_{10}) the curve is shallower, at least for effects on mortality. In the London mortality studies, there was also evidence of a curvilinear relationship between black smoke and daily mortality, the slope becoming shallower at higher levels of exposure. Estimates of the magnitude of effect occurring at low levels of exposure should therefore not be used to extrapolate to higher levels outside the range of exposures that existed in most of the recent acute health effect studies.

Although there are now many studies showing acute effect estimates of PM_{10} that are quantitatively reasonably consistent, this does not imply that particle composition or size distribution within the PM_{10} fraction is unimportant. Limited evidence from studies on dust storms indicates that such PM_{10} particles are much less toxic than those associated with combustion sources. Recent studies in which PM_{10} size fractions and/or constituents have been measured suggest that the observed effects of PM_{10} are in fact largely associated with fine particles, strong aerosol acidity or sulfates (which may serve as a proxy for the other two) and not with the coarse (PM_{10} minus $PM_{2.5}$) fraction.

Traditionally, particulate matter air pollution has been thought of as a primarily urban phenomenon. It is now clear that in many areas of Europe, urban–rural differences in PM_{10} are small or even absent, indicating that particulate matter exposure is widespread. Indeed, several of the health effect studies reviewed in this chapter were conducted in rural or semirural rather than urban areas. This is not to imply that exposure to primary, combustion-related particulate matter may not be higher in urban areas. At present, however, data are lacking on the specific health risks of such exposures.

Evidence is emerging also that long-term exposure to low concentrations of particulate matter in air is associated with mortality and other chronic effects, such as increased rates of bronchitis and reduced lung function. Two cohort studies conducted in the United States suggest that life expectancy may be shortened by more than a year in communities exposed to high concentrations compared to those exposed to low concentrations. This is consistent with earlier results from cross-sectional studies comparing age-adjusted mortality rates across a range of long-term average concentrations. Again, such effects have been suggested to be associated with long-term average exposures that are low, starting at a concentration of fine particulate matter of about 10 $\mu g/m^3$. Whereas such observations require further corroboration, preferably also from other areas in the world, these new studies suggest that the public health implications of particulate matter exposure may be large.

Evaluation of the effects of short-term exposure on mortality and morbidity

Table 24 shows the summary estimates of relative increase in daily mortality, respiratory hospital admissions, reporting of bronchodilator use, cough and lower respiratory symptoms, and changes in peak expiratory flow associated with a 10 μg/m^3 increase in PM_{10} or $PM_{2.5}$, as reported in studies in which PM_{10} and/or $PM_{2.5}$ concentrations were actually measured (as opposed to being inferred from other measures such as coefficient of haze, black smoke or total suspended particulates). The database for parameters other than PM_{10} is still limited, but for the reasons noted above, it is very important to state that even though the evaluation of (especially the short-term) health effects is largely expressed in terms of PM_{10}, future regulations and monitoring activities should place emphasis on (appropriate representations of) the respiratory fraction in addition to, or even preferred to, PM_{10} *(1)*.

It is important to realize that at present it is not known what reduction in life expectancy is associated with daily mortality increases related to particulate matter exposure. If effects are restricted to people in poor health, effects on age at death may be small.

Table 24. Summary of relative risk estimates for various endpoints associated with a 10 μg/m^3 increase in the concentration of PM_{10} or $PM_{2.5}$

Endpoint	Relative risk for $PM_{2.5}$ (95% confidence interval)	Relative risk for PM_{10} (95% confidence interval)
Bronchodilator use		1.0305 (1.0201–1.0410)
Cough		1.0356 (1.0197–1.0518)
Lower respiratory symptoms		1.0324 (1.0185–1.0464)
Change in peak expiratory flow (relative to mean)		–0.13% (–0.17% to –0.09%)
Respiratory hospital admissions		1.0080 (1.0048–1.0112)
Mortality	1.015 (1.011–1.019)	1.0074 (1.0062–1.0086)

The effect estimates in Table 24 can be used with considerable reservation to estimate, for a population of a given size and mortality and morbidity experience, how many people would be affected over a short period of time with increased particulate matter levels. The reservation stems from the finding that for some of the estimated effects, there was no evidence of heterogeneity between studies in the magnitude of the effect estimate. An investigation of the reasons for heterogeneity is beyond the scope of this chapter. As a consequence, the pooled effect estimates may not be applicable in all possible circumstances.

For illustrative purposes, Table 25 contains an estimate of the effect of a 3-day episode with daily PM_{10} concentrations averaging 50 μg/m³ and 100 μg/m³ on a population of 1 million people. Table 25 makes it clear that, in a population of that size, the number of people dying or having to be admitted to hospital as a result of particulate matter exposure is small relative to the additional number of "person-days" of increased medication use and/or increased respiratory symptoms due to exposure to particulate matter.

Whereas these calculations should be modified according to the size, mortality and morbidity experience of populations of interest and, where possible, for factors contributing to the heterogeneity in the effect estimates, they do provide some insight into the public health consequences of certain exposures to particulate matter.

Table 25. Estimated number of people (in a population of 1 million) experiencing health effects over a period of 3 days characterized by a mean PM_{10} concentration of 50 or 100 μg/m³

Health effect indicator	No. of people affected by a three-day episode of PM_{10} at:	
	50 μg/m³	100 μg/m³
No. of deaths	4	8
No. of hospital admissions due to respiratory problems	3	6
Person-days of bronchodilator use	4 863	10 514
Person-days of symptom exacerbation	5 185	11 267

Evaluation of the effects of long-term exposure on mortality and morbidity

The most convincing information on long-term effects of particulate matter exposure on mortality is provided by two recent cohort studies. Relative risk estimates for total mortality from the first study *(2)*, expressed per 10 μg/m³, were 1.10 for inhalable particles (measured as either PM_{15} or PM_{10}), 1.14 for fine particles ($PM_{2.5}$) and 1.33 for sulfates. Relative risk estimates for total mortality from the second study *(3)*, expressed per 10 μg/m³, were 1.07 for fine particles ($PM_{2.5}$) and 1.08 for sulfates. Sulfate levels used in the second study (range 3.6–23.6 μg/m³) may have been inflated owing to sulfate formation on filter material used in earlier studies. The first study included one of the high-sulfate communities (Steubenville), yet the range of sulfate levels in this study was much lower (4.8–12.8 μg/m³), possibly owing to the more adequate measurement methods employed in this study.

Long-term effects of particulate matter exposure on morbidity have been demonstrated in the Harvard 24 cities study among children *(4, 5)*. Expressed per 10 μg/m³, the relative risks for bronchitis were 1.34 for $PM_{2.1}$, 1.29 for PM_{10}, and 1.96 for sulfate particles. The corresponding changes in FEV_1 were –1.9% ($PM_{2.1}$), –1.2% (PM_{10}) and –3.1% (sulfate particles). Whereas such mean changes are clinically unimportant, the proportion of children having a clinically relevant reduced lung function (forced vital capacity (FVC) or FEV_1 < 85% of predicted) was increased by a factor of 2–3 across the range of exposures *(5)*. A recent study from Switzerland *(6)* has shown significant reductions in FEV_1 of –1.0% per 10 μg/m³ PM_{10}.

Table 26 provides a summary of the current knowledge of effects of long-term exposure to particulate matter on morbidity and mortality endpoints.

Using the risk estimates presented in Table 26, Table 27 provides estimates of the number of people experiencing health effects associated with long-term exposure to particulate matter, using similar assumptions about population size and morbidity as in Table 25. Specifically, a population size of one million has been assumed, 20% of whom are children, with a baseline prevalence of 5% for bronchitis symptoms among children (that is, 10 000 children are assumed to have bronchitis symptoms) and with a baseline prevalence of 3% of children (6000 children) having a lung function (FVC or FEV_1) lower than 85% of predicted.

In addition, the impact of long-term exposures to particulate matter on total mortality can be estimated. The number of persons surviving to a

Table 26. Summary of relative risk estimates for effects of long-term exposure to particulate matter on the morbidity and mortality associated with a 10 μg/m³ increase in the concentration of $PM_{2.5}$ or PM_{10}

Endpoint	Relative risk for $PM_{2.5}$ (95% confidence interval)	Relative risk for PM_{10} (95% confidence interval)
Death *(2)*	1.14 (1.04–1.24)	1.10 (1.03–1.18)
Death *(3)*	1.07 (1.04–1.11)	
Bronchitis *(4)*	1.34 (0.94–1.99)	1.29 (0.96–1.83)
Percentage change in FEV_1, children *(5)* [a]	–1.9% (–3.1% to –0.6%)	–1.2% (–2.3% to –0.1%)
Percentage change in FEV_1, adults *(6)*		–1.0% (not available)

[a] For $PM_{2.1}$ rather than $PM_{2.5}$.

Table 27. Estimated number of children (out of 200 000 in a population of 1 million) experiencing health effects per year due to long-term exposure to a $PM_{2.5}$ concentration of 10 or 20 μg/m³ above a background level of 10 μg/m³

Health effect indicator	No. of children affected per year at $PM_{2.5}$ concentrations above background of:	
	10 μg/m³	20 μg/m³
No. of additional children with bronchitis symptoms	3350	6700
No. of additional children with lung function (FVC or FEV_1) below 85% of predicted	4000	8000

certain age will be smaller in a population exposed to higher concentrations, and the difference will depend on the age group. If the mortality structure of Dutch males is taken as a basis for calculation, and if the assumptions used in the construction of Table 25 are applied, in each birth cohort of 100 000 men the number of survivors exposed to pollution increased by 10 μg/m³ (PM_{10}) will be reduced by 383 men before the age of 50, by 1250 men before the age of 60 and by 3148 men before the age of 70. An increase in the long-term exposure of 20 μg/m^3 (PM_{10}) corresponds to an estimated reduction of the number of men surviving to a certain age in the cohorts by, respectively, 764, 2494 or 6250 men.

Guidelines

The weight of evidence from numerous epidemiological studies on short-term responses points clearly and consistently to associations between concentrations of particulate matter and adverse effects on human health at low levels of exposure commonly encountered in developed countries. The database does not, however, enable the derivation of specific guideline values at present. Most of the information that is currently available comes from studies in which particles in air have been measured as PM_{10}. There is now also a sizeable body of information on fine particulate matter ($PM_{2.5}$) and the latest studies are showing that this is generally a better predictor of health effects than PM_{10}. Evidence is also emerging that constituents of $PM_{2.5}$ such as sulfates are sometimes even better predictors of health effects than $PM_{2.5}$ *per se*.

The large body of information on studies relating day-to-day variations in particulate matter to day-to-day variations in health provides quantitative estimates of the effects of particulate matter that are generally consistent. The available information does not allow a judgement to be made of concentrations below which no effects would be expected. Effects on mortality, respiratory and cardiovascular hospital admissions and other health variables have been observed at levels well below 100 μg/m^3, expressed as a daily average PM_{10} concentration. For this reason, no guideline value for short-term average concentrations is recommended either. Risk managers are referred to the risk estimates provided in the tables for guidance in decision-making regarding standards to be set for particulate matter.

The body of information on long-term effects is still smaller. Some studies have suggested that long-term exposure to particulate matter is associated with reduced survival, and a reduction of life expectancy in the order of 1–2 years. Other recent studies have shown that the prevalence of bronchitis symptoms in children, and of reduced lung function in children and adults, are associated with particulate matter exposure. These effects have been observed at annual average concentration levels below 20 μg/m^3 (as $PM_{2.5}$) or 30 μg/m^3 (as PM_{10}). For this reason, no guideline value for long-term average concentrations is recommended. Risk managers are referred to the risk estimates provided in the tables for guidance in decision-making regarding standards to be set for particulate matter.

References

1. LIPPMANN, M. & THURSTON, G.D. Sulphate concentrations as an indicator of ambient particulate matter air pollution for health risk evaluations. *Journal of exposure analysis and environmental epidemiology*, **6**: 123–146 (1996).
2. DOCKERY, D.W. ET AL. An association between air pollution and mortality in six U.S. cities. *New England journal of medicine*, **329**: 1753–1759 (1993).
3. POPE, C.A. III. ET AL. Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults. *American journal of respiratory and critical care medicine*, **151**: 669–674 (1995).
4. DOCKERY, D.W. ET AL. Health effects of acid aerosols on North American children: respiratory symptoms. *Environmental health perspectives*, **104**: 500–505 (1996).
5. RAIZENNE, M. ET AL. Health effects of acid aerosols on North American children: pulmonary function. *Environmental health perspectives*, **104**: 506–514 (1996).
6. ACKERMANN-LIEBRICH, U. ET AL. Lung function and long-term exposure to air pollutants in Switzerland. *American journal of respiratory and critical care medicine*, **155**: 122–129 (1997).

7.4 Sulfur dioxide

Exposure evaluation

In much of western Europe and North America, concentrations of sulfur dioxide in urban areas have continued to decline in recent years as a result of controls on emissions and changes in fuel use. Annual mean concentrations in such areas are now mainly in the range 20–60 μg/m^3 (0.007–0.021 ppm), with daily means seldom more than 125 μg/m^3 (0.044 ppm). In large cities where coal is still widely used for domestic heating or cooking, however, or where there are poorly controlled industrial sources, concentrations may be 5–10 times these values. Peak concentrations over shorter averaging periods, of the order of 10 minutes, can reach 1000–2000 μg/m^3 (0.35–0.70 ppm) in some circumstances, such as the grounding of plumes from major point sources or during peak dispersion conditions in urban areas with multiple sources.

Health risk evaluation

Short-term exposures (less than 24 hours)

The most direct information on the acute effects of sulfur dioxide comes from controlled chamber experiments on volunteers. Most of these studies have been for exposure periods ranging from a few minutes up to 1 hour, but the exact duration is not critical because responses occur very rapidly, within the first few minutes after commencement of inhalation; continuing the exposure further does not increase effects *(1–3)*. The effects observed include reductions in FEV_1 or other indices of ventilatory capacity, increases in specific airway resistance, and symptoms such as wheezing or shortness of breath. Such effects are enhanced by exercise, which increases the volume of air inspired thereby allowing sulfur dioxide to penetrate further into the respiratory tract *(4, 5)*.

A wide range of sensitivity has been demonstrated, both among normal individuals and among those with asthma, who form the most sensitive group *(1, 4, 6, 7)*. Continuous exposure–response relationships, without any clearly defined threshold, are evident. To develop a guideline value, the minimum concentrations associated with adverse effects in the most extreme circumstances, that is with asthmatic patients exercising in chambers, have been considered. An example of an exposure–response relationship for such subjects, expressed in terms of reductions in FEV_1 after a 15-minute exposure, comes from a study by Linn et al. (*8*). Only small changes, not regarded as of clinical significance, were seen at 572 μg/m^3 (0.2 ppm); reductions representing about 10% of baseline FEV_1 occurred at about 1144 μg/m^3 (0.4 ppm); and reductions of about 15% occurred at about 1716 μg/m^3 (0.6 ppm). The response was not greatly influenced by the severity of asthma. These findings are consistent with those reported from other exposure studies. In one early series, however, a small change in airway resistance was reported in two of the asthmatic patients at 286 μg/m^3 (0.1 ppm).

Exposure over a 24-hour period

Information on effects of exposure averaged over a 24-hour period is derived mainly from epidemiological studies in which the effects of sulfur dioxide, particulate matter and other associated pollutants are considered *(9)*. Exacerbation of symptoms among panels of selected sensitive patients occurred consistently when the sulfur dioxide concentration exceeded 250 μg/m^3 (0.087ppm) in the presence of particulate matter. Such findings have related mainly to situations in which emissions from the inefficient burning of coal in domestic appliances have been the main contributor to the pollution complex. Several more recent studies, involving the mixed industrial and vehicular sources that now dominate, have consistently demonstrated effects on mortality (total, cardiovascular and respiratory) *(10–18)* and hospital emergency admissions *(14, 19–22)* for total respiratory causes and chronic obstructive pulmonary disease at lower levels of exposure (mean annual levels below 50 μg/m^3; daily levels usually not exceeding 125 μg/m^3). These results have been shown, in some instances, to persist when levels of black smoke and total suspended particulate matter were controlled for, while in other studies no attempts were made to separate the effects of the pollutants. No obvious threshold levels could so far be identified in those studies.

Long-term exposure

A similar situation arises in respect of effects of long-term exposures, expressed as annual averages. Earlier assessments examined findings on the prevalence of respiratory symptoms, respiratory illness frequencies, or differences in lung function values in localities with contrasting concentrations of sulfur dioxide and particulate matter, largely in the coal-burning era. The LOAEL of sulfur dioxide was judged to be 100 μg/m^3 (0.035 ppm) annual average, together with particulate matter. More recent studies related to industrial sources, or to the changed urban mixture, have shown adverse effects below this level, but a major difficulty in interpretation is that long-term effects are liable to be affected not only by current conditions but also by the qualitatively and quantitatively different pollution of earlier years. Cohort studies of differences in mortality between areas with contrasting pollution levels indicate that there is a closer association with particulate matter than with sulfur dioxide *(23, 24).*

Guidelines

Short-term exposures

Controlled studies with exercising asthmatics indicate that some asthmatics experience changes in pulmonary function and respiratory symptoms after periods of exposure as short as 10 minutes. Based on this evidence, it is recommended that a value of 500 μg/m³ (0.175 ppm) should not be exceeded over averaging periods of 10 minutes. Because exposure to sharp peaks depends on the nature of local sources, no single factor can be applied to this value in order to estimate corresponding guideline values over somewhat longer periods, such as an hour.

Exposure over a 24-hour period and long-term exposure

Day-to-day changes in mortality, morbidity or lung function related to 24-hour average concentrations of sulfur dioxide are necessarily based on epidemiological studies in which people are in general exposed to a mixture of pollutants, which is why guideline values for sulfur dioxide have previously been linked with corresponding values for particulate matter. This approach led to a previous guideline value of 125 μg/m³ (0.04 ppm) as a 24-hour average, after applying an uncertainty factor of 2 to the LOAEL. In more recent studies, adverse effects with significant public health importance have been observed at much lower levels of exposure. Nevertheless, there is still uncertainty as to whether sulfur dioxide is the pollutant responsible for the observed adverse effects or, rather, a surrogate for ultrafine particles or some other correlated substance. There is no basis for revising the 1987 guidelines for sulfur dioxide *(9)* and thus the following guidelines are recommended:

24 hours:	125 μg/m³
annual:	50 μg/m³

It should be noted that, unlike in the 1987 guidelines, these values for sulfur dioxide are no longer linked with particles.

References

1. LAWTHER, P.J. ET AL. Pulmonary function and sulphur dioxide: some preliminary findings. *Environmental research*, **10**: 355–367 (1975).

2. SHEPPARD, D. ET AL. Exercise increases sulfur dioxide induced bronchoconstriction in asthmatic subjects. *American review of respiratory disease*, **123**: 486–491 (1981).
3. LINN, W.S. ET AL. Asthmatics responses to 6-hr sulfur dioxide exposures on two successive days. *Archives of environmental health*, **39**: 313–319 (1984).
4. DEPARTMENT OF HEALTH. *Advisory Group on the Medical Aspects of Air Pollution Episodes. Second report: sulphur dioxide, acid aerosols and particulates*. London, H.M. Stationery Office, 1992.
5. BETHEL R.A. ET AL. Effect of exercise rate and route of inhalation on sulfur dioxide induced bronchoconstriction in asthmatic subjects. *American review of respiratory disease*, **128**: 592–596 (1983).
6. NADEL, J.A. ET AL. Mechanism of bronchoconstriction during inhalation of sulfur dioxide. *Journal of applied physiology*, **20**: 164–167 (1965).
7. HORSTMAN, D.H. ET AL. The relationship between exposure duration and sulphur dioxide induced bronchoconstriction in asthmatic subjects. *American Industrial Hygiene Association journal*, **49**: 38–47 (1988).
8. LINN, W.S. ET AL. Replicated dose–response study of sulfur dioxide effects in normal, atopic and asthmatic volunteers. *American review of respiratory disease*, **136**: 1127–1134 (1987).
9. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
10. SPIX, C. ET AL. Air pollution and daily mortality in Erfurt, East Germany, 1980–1989. *Environmental health perspectives*, **101**: 518–526 (1993).
11. WIETLISBACH, V. ET AL. Air pollution and daily mortality in three Swiss urban areas. *Social and preventive medicine*, **41**: 107–115 (1996).
12. SCHWARTZ, J. & DOCKERY, D.W. Increased mortality in Philadelphia associated with daily air pollution concentrations. *American review of respiratory disease*, **145**: 600–604 (1992).
13. SUNYER, J. ET AL. Air pollution and mortality in Barcelona. *Journal of epidemiology and community health*, **50** (Suppl.): S76–S80 (1996).
14. DAB, W. ET AL. Short-term respiratory health effects of ambient air pollution: results of the APHEA project in Paris. *Journal of epidemiology and community health*, **50** (Suppl.): S42–S46 (1996).
15. ZMIROU, D. ET AL. Short-term effects of air pollution on mortality in the city of Lyons, France 1985–1990. *Journal of epidemiology and community health*, **50** (Suppl.): S30–S35 (1996).
16. TOULOUMI, G. ET AL. Daily mortality and air pollution from particulate matter, sulphur dioxide and carbon monoxide in Athens, Greece: 1987–1991. A time-series analysis within the APHEA project. *Journal of epidemiology and community health*, **50** (Suppl.): S47–S51 (1996).

17. ANDERSON, H.R. ET AL. Air pollution and daily mortality in London: 1987–92. *British medical journal*, **312**: 665–669 (1996).
18. KATSOUYANNI, K. ET AL. Short-term effects of air pollution on health: a European approach using epidemiologic time series data. *European respiratory journal*, **8**: 1030–1038 (1995).
19. SCHWARTZ, J. & MORRIS, R. Air pollution and hospital admissions for cardiovascular disease in Detroit, Michigan. *American journal of epidemiology*, **142**: 23–35 (1995).
20. SUNYER, J. ET AL. Air pollution and emergency room admissions for chronic obstructive pulmonary diseases. *American journal of epidemiology*, **134**: 277–286 (1991).
21. PONCE DE LEON, A. ET AL. The effects of air pollution on daily hospital admission for respiratory disease in London:1987–88 to 1991–92. *Journal of epidemiology and community health*, **50** (Suppl.): S63–S70 (1996).
22. SCHOUTEN, J.P. ET AL. Short-term effects of air pollution on emergency hospital admissions for respiratory disease: results of the APHEA project in two major cities in the Netherlands, 1977–89. *Journal of epidemiology and community health*, **50** (Suppl.): S22–S29 (1996).
23. DOCKERY, D.W. ET AL. An association between air pollution and mortality in six U.S. cities. *New England journal of medicine*, **329**: 1753–1759 (1993).
24. POPE, C.A. III. ET AL. Particulate air pollution as a predictor of mortality in a prospective study of U.S. adults. *American journal of respiratory and critical care medicine*, **151**: 669–674 (1995).

CHAPTER 8

Indoor air pollutants

8.1 Environmental tobacco smoke

Exposure evaluation

Environmental tobacco smoke (ETS) is a dynamic complex mixture of thousands of compounds in particulate and vapour phases, and cannot be measured directly as a whole. Instead, various marker compounds, such as nicotine and respirable suspended particulates (RSPs), are used to quantify environmental exposure. In the United States, nicotine concentrations in homes where smoking occurs typically range from less than 1 μg/m^3 to over 10 μg/m^3 (*1*). Concentrations in offices where people smoke typically range from near zero to over 30 μg/m^3. Levels in restaurants, and especially bars, tend to be even higher, and concentrations in confined spaces such as cars can be higher still. Measurements of ETS-associated RSPs in homes where people smoke range from a few μg/m^3 to over 500 μg/m^3, while levels in offices are generally less than 100 μg/m^3 and those in restaurants can exceed 1 mg/m^3. ETS levels are directly related to smoker density; in countries with a higher smoking prevalence, average ETS levels could be higher.

In Western societies, with adult smoking prevalences of 30–50%, it is estimated that over 50% of homes are occupied by at least one smoker, resulting in a high prevalence of ETS exposure in children and other nonsmokers. A large percentage of nonsmokers are similarly exposed at work.

Health risk evaluation

ETS has been shown to increase the risks for a variety of health effects in nonsmokers exposed at typical environmental levels. The pattern of health effects from ETS exposure produced in adult nonsmokers is consistent with the effects known to be associated with active cigarette smoking. Chronic exposures to ETS increase lung cancer mortality *(1–5)*. In addition, the combined evidence from epidemiology and studies of mechanisms leads to the conclusion that ETS increases the risk of morbidity and mortality from cardiovascular disease in nonsmokers, especially those with chronic exposure *(4, 6–11)*. ETS also irritates the eyes and respiratory tract. In infants and young children, ETS increases the risk of pneumonia, bronchitis, bronchiolitis and fluid in the middle ear *(1, 2, 13, 14)*. In asthmatic children, ETS increases the severity and frequency of asthma attacks *(12)*. Furthermore, as with active smoking, ETS reduces birth weight in the offspring of nonsmoking mothers *(15)*.

Other health effects have also been associated with ETS exposure, but the evidence is not as conclusive. In adults, there is strong suggestive evidence that ETS increases mortality from sinonasal cancer *(16, 17)*. In infants, recent evidence suggests that ETS is a risk factor for sudden infant death syndrome *(18–22)*.

Populations at special risk for the adverse health effects of ETS are young children and infants, asthmatics, and adults with other risk factors for cardiovascular disease. Levels of exposure where these effects have been observed are indicated by nicotine levels of 1–10 $\mu g/m^3$ (nicotine has been demonstrated to be a reliable marker of ETS levels).

Because of the extensive prevalence of ETS exposure and the high incidence of some of the health effects associated with ETS exposure, such as cardiovascular disease in adults and lower respiratory tract infections in children, even small increases in relative risks can translate into substantial levels of mortality and morbidity on a population basis.

Based on the combined evidence from several studies, WHO has estimated that some 9–13% of all cancer cases can be attributed to ETS in a nonsmoking population of which 50% are exposed to ETS. The proportion of lower respiratory illness in infants attributed to ETS exposure can be estimated at 15–26%, assuming that 35% of the mothers smoke at home. Those estimates, when applied to the European population, will result in approximately 3000–4500 cases of cancer in adults per year, and between 300 000 and 550 000 episodes of lower respiratory illness in infants per year, which are expected to be related to ETS exposure *(23)*.

Comparable results were calculated for nonsmokers in the United States *(1)*. The US Environmental Protection Agency (EPA) recently estimated that ETS causes 3000 lung cancer deaths in adult nonsmokers (roughly 100 million people who have never smoked and long-term former smokers) in the United States each year. The EPA also estimated that ETS is responsible for between 150 000 and 300 000 lower respiratory tract infections annually in the roughly 5.5 million children under 18 months of age, and that it exacerbates asthma in about 20% of asthmatic children. These estimates are based on a large quantity of human data from actual exposure levels, and involve no high-to-low-dose or animal-to-human extrapolations; thus confidence in these estimates is considered high.

Quantitative population estimates for cardiovascular disease mortality are less certain than those for lung cancer. The main reasons for greater quantitative uncertainty in estimates for cardiovascular disease are that (*a*) there are fewer epidemiological data available (in particular, there are few data for males, which is especially critical because males have a very different baseline risk of cardiovascular disease than females), and (*b*) there are more risk factors for cardiovascular disease that need to be adjusted for to obtain a reliable risk estimate. In general, the relative risk estimates for cardiovascular disease from ETS exposure are similar to those for lung cancer; however, the baseline risk of death from cardiovascular disease in nonsmokers is at least 10 times higher than the risk of lung cancer. Therefore, the population risks could be roughly 10 times higher as well. Thus, while there is more confidence in the presented estimates for lung cancer, the public health impact of ETS is expected to be substantially greater for cardiovascular disease.

Guidelines

ETS has been found to be carcinogenic in humans and to produce a substantial amount of morbidity and mortality from other serious health effects at levels of 1–10 $\mu g/m^3$ nicotine (taken as an indicator of ETS). Acute and chronic respiratory health effects on children have been demonstrated in homes with smokers (nicotine 1–10 $\mu g/m^3$) and even in homes with occasional smoking (0.1–1 $\mu g/m^3$). There is no evidence for a safe exposure level. The unit risk of cancer associated with lifetime ETS exposure in a home where one person smokes is approximately 1×10^{-3}.

References

1. *Respiratory health effects of passive smoking: lung cancer and other disorders*. Washington, DC, US Environmental Protection Agency, 1992 (EPA/600/6-90/006F).
2. *The health consequences of involuntary smoking. A report of the Surgeon General*. Washington, DC, US Department of Health and Human Services, 1986 (DHHS Publication No. (PHS) 87–8398).
3. *Tobacco smoking*. Lyons, International Agency for Research on Cancer, 1986 (IARC Monographs on the Evaluation of the Carcinogenic Risk of Chemicals to Humans, Vol. 38).
4. *Environmental tobacco smoke in the workplace: lung cancer and other health effects*. Cincinnati, OH, National Institute for Occupational Safety and Health, 1991 (Current Intelligence Bulletin, No. 54).
5. FONTHAM, E.T.H. ET AL. Environmental tobacco smoke and lung cancer in nonsmoking women: a multicenter study. *Journal of the American Medical Association*, **271**: 1752–1759 (1994).
6. GLANTZ, S.A. & PARMLEY, W.W. Passive smoking and heart disease: mechanisms and risk. *Journal of the American Medical Association*, **273**: 1047–1053 (1995).

7. WELLS, A.J. Passive smoking as a cause of heart disease. *Journal of the American College of Cardiology*, **24**: 546–554 (1994).
8. TAYLOR, A.E. ET AL. Environmental tobacco smoke and cardiovascular disease: a position paper from the Council on Cardiopulmonary and Critical Care, American Heart Association. *Circulation*, **86**: 1–4 (1992).
9. KRISTENSEN, T.S. Cardiovascular diseases and the work environment: a critical review of the epidemiologic literature on chemical factors. *Scandinavian journal of work, environment and health*, **15**: 245–264 (1989).
10. NATIONAL INSTITUTE FOR OCCUPATIONAL SAFETY AND HEALTH. *Posthearing Brief submitted to OSHA Docket No. H–122 (Submission #527)*. Washington, DC, Occupational Safety and Health Administration, 1995.
11. LAW, M.R. ET AL. Environmental tobacco smoke exposure and ischaemic heart disease: an evaluation of the evidence. *British medical journal*, **315**: 973–980 (1997).
12. CHILMONCZYK, B.A. ET AL. Association between exposure to environmental tobacco smoke and exacerbations of asthma in children. *New England journal of medicine*, **328**: 1665–1669 (1993).
13. STRACHAN, D.P. ET AL. Passive smoking, salivary cotinine concentrations, and middle ear effusion in 7-year-old children. *British medical journal*, **298**: 1549–1552 (1989).
14. RYLANDER, E. ET AL. Parental smoking, urinary cotinine, and wheezing bronchitis in children. *Epidemiology*, **6**: 289–293 (1995).
15. *The health benefits of smoking cessation. A report of the Surgeon General.* Washington, DC, US Department of Health and Human Services, 1990 (DHHS Publication No. (CDC) 90–8416).
16. TREDANIEL, J. ET AL. Environmental tobacco smoke and the risk of cancer in adults. *European journal of cancer*, **29A**: 2058–2068 (1993).
17. ZHENG, W. ET AL. Risk factors for cancers of the nasal cavity and paranasal sinuses among white men in the United States. *American journal of epidemiology*, **138**: 965–972 (1994).
18. HOFFMAN, H.J. & HILLMAN, L.S. Epidemiology of the sudden infant death syndrome: maternal, neonatal, and postnatal risk factors. *Clinics in perinatology*, **19**: 717–737 (1992).
19. SLOTKIN, T.A. ET AL. Loss of neonatal hypoxia tolerance after prenatal nicotine exposure: implications for sudden infant death syndrome. *Brain research bulletin*, **38**: 69–75 (1995).
20. MITCHELL, E.A. ET AL. Smoking and the sudden infant death syndrome. *Pediatrics*, **91**: 893–896 (1993).
21. KLONOFF-COHEN, H.S. ET AL. The effect of passive smoking and tobacco smoke exposure through breast milk on sudden infant death

syndrome. *Journal of the American Medical Association*, **273**: 795–798 (1995).

22. SCRAGG, R. ET AL. Bed sharing, smoking, and alcohol in the sudden infant death syndrome. *British medical journal*, **307**: 1312–1318 (1993).
23. WHO EUROPEAN CENTRE FOR ENVIRONMENT AND HEALTH. *Concern for Europe's tomorrow. Health and the environment in the WHO European Region.* Stuttgart, Wissenschaftliche Verlagsgesellschaft, 1995.

8.2 Man-made vitreous fibres

Exposure evaluation

Airborne concentrations during the installation of insulation comprising man-made vitreous fibres (MMVF) are in the range 10^5–2×10^6 fibres/m^3 *(1)*, which is generally higher than the concentrations of about 10^5 fibres/m^3 reported for production plants *(2)*. Little information is available on ambient concentrations of MMVF. A few limited studies of MMVF in outdoor air have reported concentrations ranging from 2 fibres/m^3 in a rural area to 1.7×10^3 fibres/m^3 near a city *(3–5)*. These levels are estimated to represent a very small percentage of the total fibre and total suspended particulate concentrations in the ambient air.

Health risk evaluation

MMVF of diameters greater than 3 µm can cause transient irritation and inflammation of the skin, eyes and upper airways *(6)*.

The deep lung penetration of various MMVF varies considerably, as a function of the nominal diameter of the material. For the six categories of MMVF considered here (continuous filament fibre glass, glass wool fibres, rock wool fibres, slag wool fibres, refractory ceramic fibres and special purpose fibres (glass microfibres)), the potential for deep lung penetration is greatest for refractory ceramic fibres and glass microfibres; both of these materials are primarily used in industrial applications.

In two large epidemiological studies, there have been excesses of lung cancer in rock/slag wool production workers, but not in glass wool, glass microfibre or continuous filament production workers. There have been no increases in the incidence of mesotheliomas in epidemiological studies of MMVF production workers *(7, 8)*. Although concomitant exposure to other substances may have contributed to the observed increase in lung cancer in the rock/slag wool production sector, available data are consistent with the hypothesis that the fibres themselves are the principal determinants of risk. Increases in tumour incidence have not been observed in inhalation studies in animals exposed to rock/slag wool, glass wool or glass microfibre, though they have occurred following intracavitary administration. Available data concerning the effects of continuous filament in animals are limited.

Several types of refractory ceramic fibre have been clearly demonstrated to be carcinogenic in inhalation studies in animal species, inducing dose-related increased incidence of pulmonary tumours and mesotheliomas in rats and hamsters *(9–11)*. Increased tumour incidence has also been observed following intratracheal *(12)* and intrapleural and intraperitoneal *(13)* administration in animals.

Though uses of refractory ceramic fibres are restricted primarily to the industrial environment, a unit cancer risk for lung tumours for refractory ceramic fibres has been calculated as 1×10^{-6} per fibre/l (for fibre length > 5 μm, and aspect ratio (ratio of fibre length to fibre diameter) of 3:1 as determined by optical microscopy) based on inhalation studies in animals *(14)*.

Guidelines

IARC classified rock wool, slag wool, glass wool and ceramic fibres in Group 2B (possibly carcinogenic to humans) while glass filaments were not considered classifiable as to their carcinogenicity to humans (Group 3) *(15)*. Recent data from inhalation studies in animals strengthen the evidence for the possible carcinogenicity of refractory ceramic fibres in humans.

Though uses of refractory ceramic fibres are restricted primarily to the industrial environment, the unit risk for lung tumours is 1×10^{-6} per fibre/l. The corresponding concentrations of refractory ceramic fibres producing excess lifetime risks of 1/10 000, 1/100 000 and 1/1 000 000 are 100, 10 and 1 fibre/l, respectively.

For most other MMVF, available data are considered inadequate to establish air quality guidelines.

References

1. ESMEN, N.A. ET AL. Exposure of employees to man-made mineral vitreous fibres: installation of insulation materials. *Environmental research,* **28**: 386–398 (1982).
2. DODGSON, J. ET AL. Estimates of past exposure to respirable man-made mineral fibres in the European insulation wool industry. *Annals of occupational hygiene*, **31**: 567–582 (1987).
3. BALZER, J.L. Environmental data: airborne concentrations found in various operations. *In: Occupational exposure to fibrous glass. Proceedings of a symposium, College Park, Maryland, June 26–27, 1974.* Washington, DC, US Department of Health, Education and Welfare, 1976.
4. HOHR, D. Transmissionselektronenmikroskopische Untersuchung: faserformige Staube in den Aussenluft [Investigation by means of transmission electron microscopy: fibrous particles in the ambient air]. *Staub Reinhaltung der Luft*, **45**: 171–174 (1985).

5. DOYLE, P. ET AL. *Mineral fibres (man-made vitreous fibres). Priority substances list assessment report.* Ottawa, Environment Canada and Health Canada, 1993.
6. *Man-made mineral fibres*. Geneva, World Health Organization, 1988 (Environmental Health Criteria, No. 77).
7. MARSH, G. ET AL. Mortality among a cohort of US man-made mineral fibre workers: 1985 follow-up. *Journal of occupational medicine*, **32**: 594–604 (1990).
8. SIMONATO, L. ET AL. The International Agency for Research on Cancer historical cohort study of MMMF production workers in seven European countries: extension of the follow up. *Annals of occupational hygiene*, **31**: 603–623 (1987).
9. DAVIS, J.M.G. ET AL. The pathogenicity of long versus short fibre sample of amosite asbestos administered to rats by inhalation and intraperitoneal injection. *British journal of experimental pathology*, **67**: 415–430 (1986).
10. MAST, R.W. ET AL. A multiple dose chronic inhalation toxicity study of kaolin refractory ceramic fiber (RFC) in male Fischer 344 rats. [Abstract no. 63]. *Toxicologist*, **13**: 43 (1993).
11. POTT, F. & ROLLER, M. Carcinogenicity of synthetic fibres in experimental animals: its significance for workers. *Journal of occupational health and safety – Australia and New Zealand*, **12**: 333–339 (1996).
12. POTT, F. ET AL. Significance of durability of mineral fibers for their toxicity and carcinogenic potency in the abdominal cavity of rats and the low sensitivity of inhalation studies. *Environmental health perspectives*, **102** (Suppl. 5): 145–150 (1994).
13. POTT, F. ET AL. Tumours by the intraperitoneal and interpleural routes and their significance for the classification of mineral fibres. *In*: Brown, R.C. et al., ed. *Mechanisms in fibre carcinogenesis*. New York & London, Plenum Press, 1991 (NATO ASI Series, Series A, Life Sciences, Vol. 223), pp. 547–565.
14. *Health-based tolerable intakes/concentrations and tumorigenic doses/concentrations for priority substances.* Ottawa, Health Canada, 1996.
15. *Man-made mineral fibres and radon.* Lyons, International Agency for Research on Cancer, 1988 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 43).

8.3 Radon

Exposure evaluation

Exposure to radon and radon progeny is the dominant source of exposure to ionizing radiation in most countries. The radon levels vary considerably between dwellings, and depend primarily on the inflow of soil gas and the type of building material. As shown in Table 28, arithmetic mean concentrations in European countries range from about 20 Bq/m^3 to 100 Bq/m^3, with even higher levels in some regions. The geometric mean concentrations are generally about 20–50% lower because of the skewed distribution of radon levels.

Health risk evaluation

A few recent case-control studies provide evidence on lung cancer risks related to residential radon exposure. In general, the exposure assessment was based on radon measurements in the homes of the people being studied, covering residential periods of about 10–30 years *(1)*. Some of the studies indicate increased relative risks for lung cancer by estimated time-weighted residential radon level or cumulative exposure, but the picture is not fully coherent. It should be realized, however, that most studies lacked an adequate statistical power. The largest of the studies, with analyses over the widest range of exposure, showed a clear increase in risk with estimated exposure to radon, which appeared consistent with a linear relative risk model *(2)*. The interaction between radon exposure and smoking with regard to lung cancer exceeded additivity and was close to a multiplicative effect.

To date, risk estimation for residential radon exposure has often been based on extrapolation of findings in underground miners. Several circumstances make such estimates uncertain for the general population, however, including the possible influence of other exposure factors in the mines and differences in age, sex, size distribution of aerosols, the attached fraction of radon progeny, breathing rate and route *(3, 4)*. Furthermore, the relevance is not fully understood of the apparent inverse effect of exposure rate observed in miners and the possible difference in relative risk estimates for nonsmokers and smokers *(5)*.

It is of interest to compare risk estimates based on the nationwide Swedish study on residential radon exposure and lung cancer *(2)* with those obtained from miners. Fig. 1 shows the estimated attributable proportion of lung

Table 28. Radon levels in dwellings of some European countries

				Radon concentration (Bq/m^3)					
Country	**Number of Houses sampled**	**Period and duration of exposure**	**Sample characteristics**	**Average**	**Geometric mean**	**Geometric mean SD**[a]	**Percentage over 200 Bq/m^3**	**Percentage over 400 Bq/m^3**	**Reference**
Belgium	300	1984–1990 3 months to 1 year	Population -based (selected acquaintances)	48	37	1.9	1.7	0.3	[b]
Czecho slovakia	1200	1982 random grab sampling	–	140	–	–	–	–	*(7)*
Denmark	496	1985–1986 6 months	random	47	29	2.2	2.2	< 0.4	*(8)*
Finland	3074	1990–1991 1 year	random	123	84	2.1	12.3	3.6	*(9)*
France	1548	1982–1991 3 months (using open alpha track detectors)	biased (not stratified)	85	52	2.3	7.1	2.3	*(10)*
Germany	7500	1978–1984 3 months 1991–1993 1 year	random	50	40	–	1.5–2.5	0.5–1	*(11,12)*

Greece	73	1988 6 months	–	52	–	–	–	–	*(7)*
Hungary	122	1985–1987 2.5 years	preliminary survey	55	42 (median)	–	–	–	[c]
Ireland	1259	1985–1989 6 months	random	60	34	2.5	3.8	1.6	*(13)*
Italy	4866	1989–1994 1 year	stratified random	75	62	2.0	4.8	1.0	*(14)*
Luxembourg	2500	1991	–	–	65	–	–	–	*(7)*
Netherlands	1000	1982–1984 1 year	random	29	24 (median)	1.6	–	–	*(7,15)*
Norway	7525	1987–1989 6 months	random	60	32	–	5.0	1.6	*(16)*
Portugal	4200	1989–1990 1–3 months	volunteers in a selected group (high school students)	81	37	–	8.6	2.6	*(17)*
Spain	1555–2000	winter of 1988– 1989 grab sampling	random	86	41–43	2.6–3.7	–	4	*(7,18)*
Sweden	1360	1982–1992 3 months in heating season	random	108	56	–	14	4.8	*(19)*

Table 28. (contd)

Switzerland	1540	1982–1990 3 months (mainly in winter)	biased (not stratified)	70	–	–	5.0	–	*(20)*
United Kingdom	2093	1986–1987 1 year	random	20.5	15	2.2	0.5	0.2	*(21)*

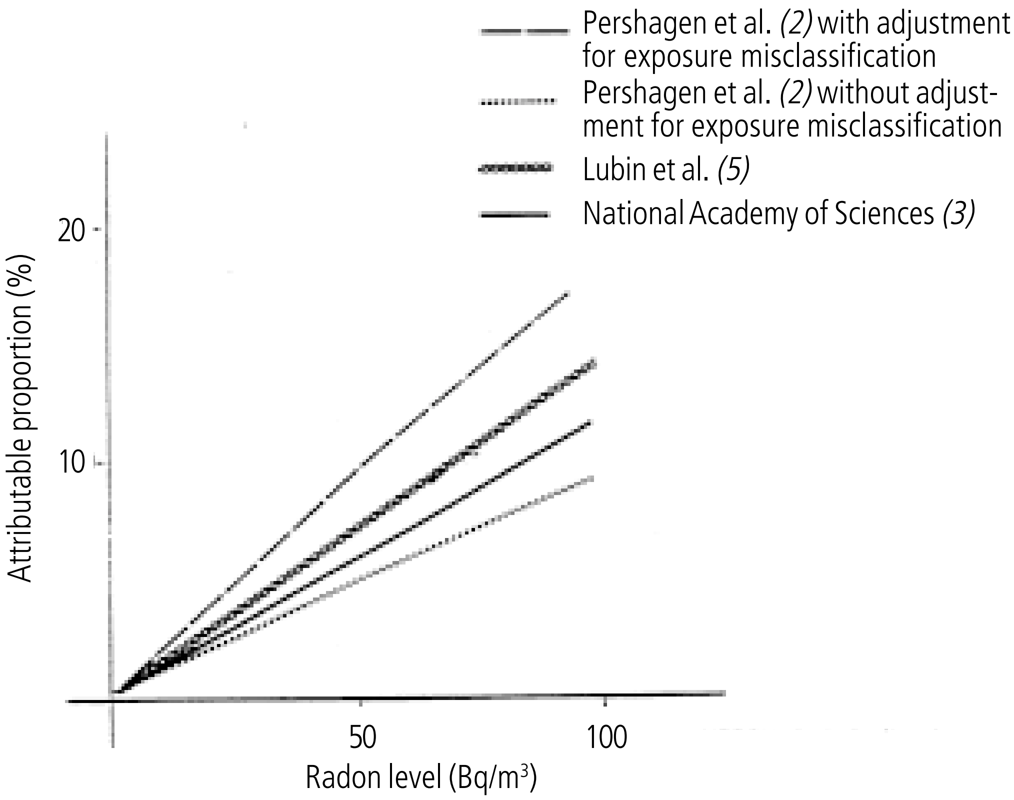
[a] SD = Standard deviation.

[b] A. Poffjin, personal communication.

[c] L. Sztanyik & I. Nikl, personal communication.

Source: Bochicchio et al. *(22)*.

Fig. 1. Estimated attributable proportion of lung cancer related to residential radon exposure based on the national Swedish study and extrapolations from miners



cancer related to residential radon, using risk estimates from the Swedish study and assuming a linear relative risk model. Imprecision in the exposure estimation leads to attenuation of the exposure–response relationship, and it has been indicated that this may have led to an underestimation of the risk by a factor of up to about 2 *(6)*. It is suggested that the true values lie between the unadjusted and adjusted estimates.

Fig. 1 also gives estimates of attributable proportion based on extrapolations from underground miners, after adjusting for dosimetric differences between mines and homes. As an example, the radon concentration distribution in western Germany, with an arithmetic mean of 50 Bq/m^3, leads to an attributable proportion of 7% (95% confidence interval: 1–29%) using the model in Lubin et al. *(5)*, and 6% (95% confidence interval: 2–17%) using that of the National Academy of Sciences *(3)*. Corresponding values based on the Swedish residential study are 5% and 9%, respectively, without and with adjustment for exposure misclassification.

Table 29 shows population risk estimates under three different assumptions with regard to population exposure, taken to represent long-term residential exposure in European countries with relatively high, medium and low residential radon concentrations. The estimated attributable proportion of lung cancer related to residential radon exposure ranges from 2–5% in low-exposure areas to 9–17% in high-exposure areas.

Table 29 also shows estimated excess lifetime deaths from lung cancer related to residential radon. Assuming that lung cancer deaths constitute 3% of total deaths, it is estimated that around 600–1500 excess lung cancer deaths occur per million people exposed on average to

Table 29. Attributable proportion of lung cancer related to long-term residential radon exposure in regions with high, medium and low indoor concentrations [a]

	Concentration		
	High	**Medium**	**Low**
Radon concentration			
Arithmetic mean (Bq/m^3)	100	50	25
> 200 Bq/m^3	15%	1.5%	0.75%
> 400 Bq/m^3	5%	0.5%	0.25%
Proportion of all lung cancers attributable to the exposure			
Total	9–17% [b]	5–9%	2–5%
> 200 Bq/m^3	4–6%	0.4–0.6%	0.2–0.3%
> 400 Bq/m^3	2–3%	0.2–0.3%	0.1–0.15%
Excess lifetime lung cancer deaths (per million) [c]			
Total	2700–5100	1500–2700	600–1500
> 200 Bq/m^3	1200–1800	120–180	60–90
> 400 Bq/m^3	600–900	60–90	30–45

[a] A linear relative risk model is assumed and a multiplicative interaction between radon and other risk factors for lung cancer, including smoking.

[b] The range in estimated attributable proportion is based on assessment of the uncertainty due to imprecision in exposure estimates of the observed exposure–response relationship *(6)*.

[c] It is assumed that lung cancer deaths constitute 3% of total deaths.

Source: Pershagen et al. *(2)*.

25 Bq/m^3 over their lifetime. For an average exposure of 100 Bq/m^3, the corresponding estimate ranges from 2700 to 5100 excess lung cancer deaths per million people exposed.

Guidelines

Radon is a known human carcinogen (classified by IARC as Group 1 *(23)*) with genotoxic action. No safe level of exposure can be determined. Quantitative risk estimates may be obtained from a recent large residential study, which are in general agreement with a linear extrapolation of risks observed in miners. The risk estimates obtained in the studies conducted among miners and the recent study from Sweden *(2)* would correspond to a unit risk of approximately 3–6 × 10^{-5} per Bq/m^3, assuming a lifetime risk of lung cancer of 3%. This means that a person living in an average European house with 50 Bq/m^3 has a lifetime excess lung cancer risk of 1.5–3 × 10^{-3}. Similarly, a person living in a house with a high radon concentration of 1000 Bq/m^3 has a lifetime excess lung cancer risk of 30–60 × 10^{-3} (3–6%), implying a doubling of background lung cancer risk.

Current levels of radon in dwellings and other buildings are of public health concern. A lifetime lung cancer risk below about 1 × 10^{-4} cannot be expected to be achievable because natural concentration of radon in ambient outdoor air is about 10 Bq/m^3. No guideline value for radon concentration is recommended. Nevertheless, the risk can be reduced effectively based on procedures that include optimization and evaluation of available control techniques. In general, simple remedial measures should be considered for buildings with radon progeny concentrations of more than 100 Bq/m^3 equilibrium equivalent radon as an annual average, with a view to reducing such concentrations wherever possible.

References

1. *Indoor air quality – a risk-based approach to health criteria for radon indoors*. Copenhagen, WHO Regional Office for Europe, 1996 (document EUR/ICP/CEH 108(A)).
2. Pershagen, G. et al. Residential radon exposure and lung cancer in Sweden. *New England journal of medicine*, **330**: 159–164 (1994).
3. National Academy of Sciences. *Health risks of radon and other internally deposited alpha-emitters*. Washington, DC, National Academy Press, 1988.
4. National Research Council. *Comparative dosimetry of radon in mines and houses*. Washington, DC, National Academy Press, 1991.

5. LUBIN, J.H. ET AL. *Radon and lung cancer – a joint analysis of 11 underground miners studies*. Washington DC, National Cancer Institute, 1994 (NIH publication No. 94-3644).
6. LAGARDE, F. ET AL. Residential radon and lung cancer in Sweden: risk analysis accounting for random error in the exposure assessment. *Health physics*, **72**: 269–276 (1997).
7. UNITED NATIONS SCIENTIFIC COMMITTEE ON THE EFFECTS OF ATOMIC RADIATION. *Report to the General Assembly, with scientific annexes.* New York, United Nations, 1993.
8. ULBAK, K. ET AL. Results from the Danish indoor radiation survey. *Radiation protection dosimetry*, **24**: 402–405 (1988).
9. ARVELA, H. ET AL. *Otantatutkimus asuntojen radonista Suomessa* [Radon in a sample of Finnish houses]. Helsinki, Finnish Centre for Radiation and Nuclear Safety, 1993.
10. RANNOU, A. ET AL. *Campagnes de mesure de l'irradiation naturelle gamma et radon en France. Bilan de 1977 a 1990.* Rapport SEGR No. 10, 1992.
11. URBAN, M. ET AL. *Bestimmung der Strahlenbelastung der Bevölkerung durch Radon und dessen kurzlebige Zerfallsprodukte in Wohnhäuser und im Freien*. Karlsruhe, Kernforschungszentrum, 1985.
12. CZARWINSKI, R. ET AL. Investigations of the radon concentrations in buildings of Eastern Germany. *Annals of the Association of Belgian Radioprotection*, **19**: 175–188 (1994).
13. MCLAUGHLIN, J.P. & WASIOLEK, P. Radon levels in Irish dwellings. *Radiation protection dosimetry*, **24**: 383–386 (1988).
14. BOCHICCHIO F. ET AL. Results of the representative Italian national survey on radon indoors. *Health physics,* **71**: 743–750 (1996).
15. PUT, L.W. ET AL. Survey of radon concentrations in Dutch dwellings. *Science of the total environment*, **45**: 441–448 (1985).
16. STRAND, T. ET AL. Radon in Norwegian dwellings. *Radiation protection dosimetry*, **45**: 503–508 (1992).
17. FAÍSCA, M.C. ET AL. Indoor radon concentrations in Portugal – a national survey. *Radiation protection dosimetry*, **45**: 465–467 (1992).
18. QUINDOS, L.S. et al. National survey of indoor radon in Spain. *Environment international*, **17**: 449–453 (1991).
19. SWEDJEMARK, G.A. ET AL. Radon levels in the 1988 Swedish housing stock. *In*: Proceedings of Indoor Air '93, Vol. 4, pp. 491–496. Helsinki, Indoor Air '93, 1993.
20. SURBECK, H. & VÖLKLE, H. Radon in Switzerland. *In*: *Proceedings of International Symposium on Radon and Radon Reduction Technology, Philadelphia, 1991, Vol 3, paper VI–3.* Research Triangle Park, NC, US Environmental Protection Agency, 1991.

21. WRIXON, A.D. ET AL. *Natural radiation exposure in UK dwellings. NRPB–R190.* Oxford, National Radiological Protection Board, 1988.
22. BOCHICCHIO F. ET AL. *Radon in indoor air. Report No. 15, European Collaborative Action: indoor air quality and its impact on man*. Luxembourg, Office for Official Publications of the European Communities, 1995.
23. *Man-made mineral fibres and radon.* Lyons, International Agency for Research on Cancer, 1988 (IARC Monographs on the Evaluation of Carcinogenic Risks to Humans, Vol. 43).

PART III

EVALUATION OF ECOTOXIC EFFECTS

General approach

In the context of the updating and revision of these guidelines, the ecological effects of major air pollutants were considered in more detail. This was undertaken in cooperation with the Working Group on Effects under the United Nations Economic Commission for Europe (ECE) Convention on Long-range Transboundary Air Pollution, capitalizing on the scientific work undertaken since 1988 to formulate criteria for the assessment of the effects of air pollutants on the natural environment.

The evaluation for the guidelines focused on the ecological effects of sulfur dioxide (including sulfur and total acid deposition), nitrogen dioxide (and other nitrogen compounds including ammonia) and ozone, which were thought to be currently of greatest concern across Europe. A number of other atmospheric contaminants are known to have ecological effects, but were not considered by the working groups. In the case of metals and persistent organic pollutants, levels of soil contamination or bioaccumulation leading to adverse effects have been proposed, but methods of linking these to atmospheric concentrations or depositions have not yet been developed. In the case of fluorides and particles, ecological effects are no longer of widespread concern in Europe, although air quality criteria have been proposed in the past by other bodies, and new criteria for fluorides are currently under consideration by certain national governments.

USE OF THE GUIDELINES IN PROTECTING THE ENVIRONMENT

Although the main objective of the guidelines is the direct protection of human health, the WHO strategy for health for all recognizes the importance of protecting the environment in terms of benefits to human health and wellbeing. Resolution WHA42.26 of the World Health Assembly and resolutions 42/187 and 42/186 of the United Nations General Assembly recognize the interdependence of health and the environment.

Ecologically based guidelines for preventing adverse effects on terrestrial vegetation were included for the first time in the first edition of *Air quality guidelines for Europe* in 1987, and guidelines were recommended for some gaseous air pollutants. Since that time, however, significant advances have been made in the scientific understanding of the impacts of air pollutants on the environment. The realization that soils play an important role in mediating both the direct and indirect effects of air pollutants on terrestrial and freshwater ecosystems has led to the development and acceptance of the joint concepts of critical levels and critical loads within the framework of the ECE Convention on Long-range Transboundary Air Pollution.

At the ECE Workshop on Critical Loads for Sulphur and Nitrogen, held at Skokloster, Sweden *(1)* and at a workshop on critical levels held at Bad Harzburg, Germany *(2)*, the following definitions were agreed on.

Critical level is the concentration of pollutants in the atmosphere above which direct adverse effects on receptors such as plants, ecosystems or materials may occur according to present knowledge.

Critical load is a quantitative estimate of an exposure, in the form of deposition, to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge.

The critical levels and loads approach is essentially a further development of the first edition of these guidelines published in 1987. There are several fundamental differences between conventional environmental objectives, critical levels and critical loads (Table 30).

Critical levels relate to direct effects on plant physiology, growth and vitality, and are expressed as atmospheric concentrations or cumulative exposures over a given averaging time. Typically, critical levels are based on effects observed over periods of from one day to several years. Critical loads relate to effects on ecosystem structure and functioning, and are expressed as annual depositions of mass or acidity. Typically, critical loads relate to the potential effects over periods of decades. In the case of sulfur and nitrogen compounds, critical levels can be directly related to critical loads when the deposition velocity for a given vegetation type is known. Nevertheless, while critical levels provide effects thresholds for relatively short-term exposures, and are not aimed at providing complete protection of all plants in all situations from adverse effects, critical loads provide the long-term deposition below which we are sure that adverse ecosystem effects will not occur.

Both critical levels and critical loads may be used to indicate the state of existing or required environmental protection, and they have been used by

Table 30. Differences between conventional environmental objectives, critical levels and critical loads

Conventional objectives	Critical levels	Critical loads
Effects are generally experienced at the organism level	Effects are experienced from organism to ecosystem levels	Effects are usually manifested at the ecosystem level
Objectives are established on the basis of laboratory tests	Objectives are established by laboratory or controlled environmental and field studies	Ecosystem studies are required to establish values
Lethality or physiological effects are the usual response used in setting objectives	Physiological, growth and ecosystem effects are caused by direct or indirect mechanisms	Ecosystem effects are caused by direct (abiotic change) or indirect (biotic interaction) mechanisms
Environmental objectives are set well below known effects to provide some margin of safety	Objectives are set as close to effect thresholds as possible	Objectives are set as close to effect thresholds as possible
No beneficial effects are likely to occur in the environment at any level	Changes may occur that are deemed beneficial (such as increased growth)	Changes may occur that are deemed beneficial (such as increased productivity)
Environmental damage from exceedances is usually observed within a short time	Environmental damage usually results from short- to medium-term exceedances	Environmental damage usually results from long-term (years, decades) exceedances and may be cumulative

ECE to define air pollutant emission control strategies for the whole of Europe. They are being or may be used in a series of protocols relating to the control of sulfur dioxide, nitrogen oxides, total nitrogen (including oxidized and reduced species) and ozone. Full use has been made in this publication of the data that underpin these protocols. The proposed guidelines cover the same range of air pollutants and are aimed at a wide range of vegetation types and ecosystems. Individual species, vegetation types and ecosystems may vary in their sensitivity to a given pollutant, and this sensitivity may also depend on other factors such as soil type or climate. When possible, therefore, different values of critical loads or levels are defined, depending on the relevant factors. When this approach is not possible, values are based on protecting the most sensitive type of vegetation or ecosystem for which good quality data are available.

There is thus a sound scientific basis for expecting that adverse ecological and economic effects may occur when the guidelines recommended below are exceeded. There is a possibility that adverse effects might also occur at exposures below these guidelines, but there is considerable uncertainty over this and it was decided to recommend values with a sound scientific basis rather than to incorporate arbitrary uncertainty factors. Critical levels and critical loads thus fulfil the primary aim of air quality guidelines in providing the best available sound scientific basis for the protection of vegetation from significant effects.

To carry out an assessment based on the guidelines, due consideration has to be given to the various problems caused by air pollution and their impact on the stock that may be at risk. The requirements for the former are often different from those needed to assess the risks to human health. Nevertheless, methodologies have been developed that can assess the risks of damage to vegetation and ecosystems.

Because of the different definition of critical loads and critical levels, the variable nature of the ecological impacts caused by different pollutants, and the different types of scientific evidence available, it is not possible to use a single methodology to derive the air quality guidelines presented in this section. For critical levels, the methods used rely on analysis either of experimental studies in the laboratory or in field chambers, or of field studies along pollution gradients. For critical loads, the methods used rely on analysis of field experiments, comparisons of sites with different deposition rates, or modelling. Where possible, data from a combination of sources are used to provide the strongest support for the proposed guidelines. Uncertainties in defining guidelines can arise (*a*) because of the limited availability of appropriate data; (*b*) because the data exist only for specific vegetation types and climates and therefore may not be representative of all areas of Europe; or (*c*) because exposure patterns in experimental chambers may not be representative of those under field conditions.

In the field, pollutants are never present in isolation, while the same pollutant may have several impacts simultaneously (for example, exposure to sulfur dioxide can cause direct effects on leaf physiology and contribute to long-term acidification, while deposition of nitrogen can cause both acidification and eutrophication). Currently, knowledge of the impacts of pollutant combinations is inadequate to define critical loads or levels for such combined impacts, and thus the guidelines are recommended for the ecological effects of individual pollutants. When applying these guidelines in ecological risk assessment, the possibility of such combined impacts should be considered. Furthermore, when considering an area of mixed vegetation types or ecosystems, several guidelines may apply. Thus ecological risk assessment applying the critical levels and loads approach must be aimed at identifying or protecting the most sensitive element of the environment.

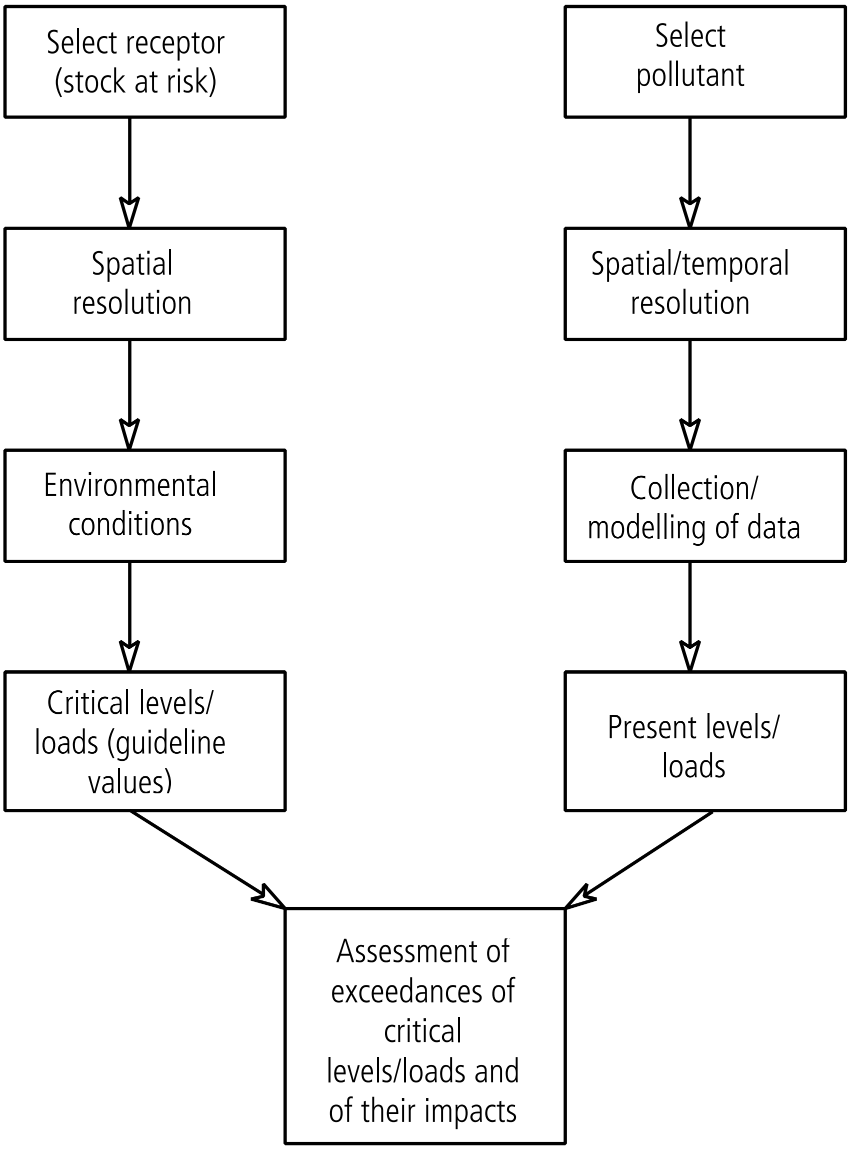
A simple overview of the elements of how critical levels and critical loads can be used is given in Fig. 2. The left- and right-hand pathways indicate the requirements, enabling finally the comparison of critical levels or critical loads with ambient air concentrations (present levels) or pollutant depositions (present loads) on broad spatial scales. The left-hand pathway depicts the steps needed to obtain a geographical distribution of critical levels and loads over European ecosystems.

Since critical levels and critical loads indicate the sensitivity of receptors (such as individual plant species or ecosystems) to air pollutants, an important step in the critical levels/loads application pathway consists of the geographical determination and mapping of the receptors and their sensitivities, at as fine a spatial resolution as possible.

Critical levels are in most cases formulated in such a way that a certain receptor type (such as forests or crops) has the same critical level value throughout Europe. In these cases, the resulting sensitivity maps look uniform over large areas. More recent developments in critical levels research attempt to incorporate environmental conditions into the assessment. The incorporation of such modifying factors – such as water availability, which influences the opening of the stomata and thus the uptake of gaseous pollutants by plants – can lead to a higher degree of differentiation in the mapping of sensitivities.

Critical loads are also allocated to certain receptor types, such as forests, bogs, heathlands, grasslands or lakes, but the spatial differentiation is generally more advanced than in the case of critical levels. It is often possible to take into account environmental conditions such as soil characteristics, water conditions, precipitation amounts, land use and management practices. The result is a critical load map with a high spatial variation in sensitivities.

Fig. 2. Critical levels/loads application pathway



The right-hand pathway in Fig. 2 depicts steps to ensure comparability of present levels/loads with critical levels/loads. The comparison with present ambient air concentrations or present depositions can only be made if the spatial resolution is compatible with the mapped critical levels/loads. The regional distribution of ambient air concentrations and depositions can be modelled to reflect data measured by national and/or international monitoring networks over Europe. Subject to the spatial resolution of these modelled data, comparisons of critical levels/loads with present levels/loads can be made at finer or coarser spatial resolutions. At the European level, present levels/loads are currently modelled for grid cells with a size of 150 km × 150 km or 50 km × 50 km by the ECE Co-operative

Programme for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (EMEP). In the case of depositions, compatibility with the mapped critical loads can be achieved by establishing cumulative frequency distributions of the critical loads occurring in the grid cell. A low percentile value (such as 5) of these distributions can be chosen for comparison with the present loads. If, in the framework of effect-orientated pollutant emission reduction strategies, the present levels or loads are reduced to critical levels or a 5-percentile value of the critical loads distribution, respectively, the protection of most sensitive receptors is reliably estimated to be high (for example achieving potential protection of 95% of the ecosystems in a grid cell).

The left- and right-hand pathways of Fig. 2 finally lead to the assessment of exceedances of critical levels/loads. Exceedances of critical levels/loads are interpreted in a qualitative rather than a quantitative manner, in that the probability of damage is considered to be non-zero whenever critical levels/loads are exceeded. Thus, the exceedance of critical levels/loads implies non-sustainable stress, which can lead to damage at any point in time and to an extent depending on the amount of excess pollution. Research is continuing to determine quantitative regional relationships between the actual excess pollution and the expected damage. Exposure–response relationships for sensitive receptors, established in experimental or field studies and modified for prevailing environmental conditions, may tentatively be used to quantify the consequences of excess pollution. However, research results are considered to lack the robustness needed to allow applications to European ecosystems as a whole.

REFERENCES

1. Nilsson, J. & Grennfelt, P., ed. *Critical loads for sulphur and nitrogen.* Copenhagen, Nordic Council of Ministers, 1993 (Miljørapport No. 15).
2. *Final report of the Critical Levels Workshop, Bad Harzburg, Germany, 14–18 March 1988*. Berlin, Federal Environment Agency, 1988.

Effects of sulfur dioxide on vegetation: critical levels

Since the publication of the first edition of the *Air quality guidelines for Europe* in 1987 *(1)*, the relative importance of sulfur dioxide as a phytotoxic pollutant in Europe has diminished to some extent, owing to falling emissions in many areas. In terms of understanding the basic mechanisms of direct injury by sulfur dioxide, and threshold concentrations for adverse effects, advances have been made in demonstrating the significance of very low concentrations on growth and yield and on changing plant sensitivity to other environmental stresses. New work has also provided information that can be utilized to introduce new guidelines for protecting lichens against sulfur dioxide and forests against acid mists.

A number of studies have provided valuable data for several major agricultural crops, based on fumigations, filtrations and transect studies *(2–4)*. These new data confirm the annual guideline value of 30 μg/m^3 as an annual mean concentration (Table 31). However, it is recommended that this value should also not be exceeded as a mean concentration for the winter months (October–March inclusive) in view of the abundant evidence for increased sensitivity of crops growing slowly under winter conditions. It is recommended that the 24-hour air quality guideline for all species be abandoned, in view of further evidence confirming that peak concentrations are not significant compared with the accumulated dose.

A lower air quality guideline of 20 μg/m^3 is now recommended for forests and natural vegetation, as both an annual and winter mean concentration (Table 31). This is based on new evidence of periods of high sensitivity of conifers during needle elongation and the longevity of many of the species concerned as well as their being unmanaged or minimally managed, which renders them more sensitive to pollution stress *(4–6)*.

New data have confirmed concerns over low-temperature stress contributing to greater sulfur dioxide sensitivity in forests. Further justification for modifying the air quality guideline to take account of interactions with low temperature is given by evidence of sulfate mists enhancing frost sensitivity.

Table 31. Guidelines for the effects of sulfur dioxide on vegetation: critical levels

Vegetation category	Guideline (µg/m³)	Time period [a]	Constraints
Agricultural crops	30	Annual and winter mean	
Forests and natural vegetation	20	Annual and winter mean	
Forests and natural vegetation	15	Annual and winter mean	Accumulated temperature sum above +5 °C is < 1000 °C·days per year
Lichens	10	Annual mean	
Forests	1.0 sulfate particulate[b]	Annual mean	Where ground level cloud is present ≥ 10% of time

[a] Where annual and winter mean concentrations are indicated, the higher value should be used to define exceedance. Winter is defined as October to March inclusive.
[b] Air quality guideline only applies in areas of oceanic Europe where calcium and magnesium concentrations in cloud or mist are less than the combined ionic concentrations of H^+ and NH_4^+.

A field study of Norway spruce at different altitudes in the Ore Mountains of Czechoslovakia has been used to develop a model, from which the accumulated temperature sum above +5 °C of < 1000 °C·days per year is used as a threshold for lowering mean annual and winter sulfur dioxide concentrations to 15 µg/m³ for protecting forests and natural vegetation. This lower concentration is now recommended as a WHO air quality guideline for regions below this threshold temperature sum (Table 31). It should be recognized, however, that this guideline is based on field studies in a region where the temperatures recorded were above those pertaining in some areas of northern Europe, and thus it is possible that in even more extreme environments a lower guideline is required.

The 1987 edition of the guidelines considered only the effects of sulfur dioxide on higher plants. Many sensitive lichen and bryophyte species have disappeared from large areas of Europe with only moderately elevated sulfur dioxide concentrations. Annual mean concentrations of 30 µg/m³ are associated with the eradication of the most sensitive lichen taxa. On the basis of new field studies, it is recommended that an air quality guideline of 10 μg/m³ annual mean (Table 31) be established for lichens *(7–9)*.

In the 1987 edition, no consideration was given to direct impacts of acid precipitation on above-ground plant organs. It is now recognized that mists can contain solute concentrations up to ten times those of rain, and can thus have a direct impact on vegetation. Since mists and clouds occur most frequently at high altitudes, and are intercepted with particular efficiency by forests, trees are likely to be the most sensitive receptors. Experiments on young trees, backed up by field observations, show significant effects of acid mists on leaf surface structure at pH 3.5, which is equivalent to 150 μmol/l sulfate. Because of the difficulties of measuring sulfate concentrations in cloud water, a guideline has been set based on the equivalent particulate sulfate concentration. A guideline of 1.0 μg/m³ particulate sulfate as an annual mean is recommended for trees where ground level cloud is present 10% or more of the time (Table 31). This guideline only applies, however, when calcium and magnesium concentrations in cloud do not exceed hydrogen and ammonium ion concentrations, because no data exist to establish a guideline under other conditions. This restriction excludes areas such as the Mediterranean region, eastern Europe and the Alps.

These guidelines do not take into account that sulfur dioxide increases sensitivity to other stresses, with the exception of low temperatures for forests and natural vegetation. Given further knowledge of its effects on stresses such as drought, pathogens and pests, it is possible that the guidelines may require further modification in the future. The 24-hour mean guideline has been abolished, but this is on the basis of knowledge on higher plants. The inclusion of lichens in these new guidelines may warrant future considerations of a short-term guideline for these organisms, if knowledge indicates the necessity for this. The new guideline for acid mists has similarly been set for forests only, and the effects on other receptors may also warrant future attention.

REFERENCES

1. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
2. JÄGER, H.J. ET AL., ED. *Effects of air pollution on agricultural crops in Europe*. Brussels, European Commission, 1993 (Air Pollution Research Report, No. 46).

3. KRUPA, S.G. & ARNDT, U. Special issue on the Hohenheim Long Term Experiment. *Environmental pollution*, **68**: 193–478 (1990).
4. MCLEOD, A. R. & SKEFFINGTON, R. A. The Liphook Forest Fumigation Project – an overview. *Plant, cell and environment*, **18**: 327–336 (1995).
5. SCHULZE, E.D. ET AL. Forest decline and air pollution; a study of spruce *(Picea abies)* on acid soils. Springer-Verlag, 1989 (Ecological Studies, No. 77).
6. MCLEOD, A. R. ET AL. Enhancement of nitrogen deposition to forest trees exposed to SO_2. *Nature*, **347**: 272–279 (1990).
7. FIELDS, R.F. Physiological responses of lichens to air pollutant fumigations. *In*: Nash, T.H. & Wirth, V., ed. *Lichens, bryophytes and air quality*. Berlin, Cramer, 1988, pp. 175–200.
8. RICHARDSON, D.H.A. Understanding the pollution sensitivity of lichens. *Botanical journal of the Linnean Society*, **96**: 31–43 (1988).
9. WINNER, W.E. Responses of bryophytes to air pollution. *In*: Nash, T.H. & Wirth, V., ed. *Lichens, bryophytes and air quality*. Berlin, Cramer, 1988, pp. 141–173.

Effects of nitrogen-containing air pollutants: critical levels

EFFECT EVALUATION

Various forms of nitrogen pollute the air, mainly nitric oxide (NO), nitrogen dioxide (NO_2) and ammonia (NH_3) as dry deposition, and nitrate (NO_3^-) and ammonium (NH_4^+) as wet deposition. Other contributions are from occult deposition (fog, clouds, aerosols), peroxyacetylnitrate (PAN), dinitrogen pentoxide (N_2O_5), nitrous oxide (N_2O) and amines. Since the publication of the *Air quality guidelines for Europe* in 1987 *(1)* there have been significant advances in knowledge of the impacts of nitrogen oxides (NO_x, i.e. NO_2 and NO) and NH_3 on vegetation.

In the present evaluation, attention is mainly paid to direct effects on plants caused by an exposure duration of between one hour and one year. The long-term impact (more than one year) on vegetation and the nitrogen cycle is discussed in Chapter 14, while the contribution of nitrogen-containing air pollutants to soil acidification is evaluated in Chapter 13. The properties of PAN are discussed in Chapter 12. The role of NO_x and N_2O in atmospheric chemistry (formation and depletion of ozone in the troposphere and stratosphere, respectively) and relations with climate change are not considered.

The reason for defining critical levels for NO, NO_2 and NH_3 is the recent evidence from monitoring and mapping that these are the dominant forms of nitrogen deposition in many parts of the world, and that several important effects of these compounds are not covered by the critical loads for nitrogen or acidity.

The critical levels are based on a survey of published evidence of physiological and ecologically important effects on plants *(2–7)*. Biochemical changes have only been used as additional indicators of potentially relevant ecological responses. The current survey has considered that, in an ecological context, growth stimulation and reduction are both potentially negative responses. For instance, both NO_x and NH_y (i.e. NH_3 and NH_4^+) generally cause an increase in the shoot:root ratio, which may or may not be beneficial.

Responses to nitrogenous pollutants can be further modified and exacerbated by interactions with other environmental factors, including frost, drought and pest organisms. These interactions generally include increased susceptibility to these factors, which may in turn lead to major ecological changes.

The method of estimating critical levels is different for NO_x and NH_3, but both are based on a 95% protection level (neglecting the 5% lowest effective exposures).

GAPS IN KNOWLEDGE

There have been important developments in the use of critical level and critical load approaches for setting air quality guidelines. With regard to the critical levels of nitrogen-containing air pollutants, however, there are several areas where improvements are urgently required.

- The guidelines for the critical levels of NO_x and NH_3 are intended to apply to all classes of vegetation and under all environmental conditions. However, more information is needed to quantify the range of sensitivity.

- The guideline for NH_3 is based on research performed in temperate climates on a limited range of soil types. To a lesser extent this applies to NO_x as well. Caution is required when critical levels are considered for plants in very different conditions, for example in tropical and subtropical zones.

- There is a need to understand further the long-term impacts on growth of changes in biochemical parameters.

- There is growing awareness of the physiological importance of NO, and this is reflected in the new incorporation of this compound in the guideline for NO_x. Comparisons of the phytotoxicity of NO and NO_2 are scarce and still not conclusive with regard to their relative degree of toxicity.

- The relevance of the emission of NH_3 from plants should be investigated in more detail in order to establish its potential importance in nitrogen budgets.

GUIDELINES

Evidence exists that NH_4^+ (and NO_3^-) in rain, clouds and fog can have significant direct effects on vegetation, but current knowledge is still insufficient to arrive at critical levels for those compounds. It is assumed that NO and NO_2 act in an additive manner.

A strong case can be made for the provision of critical levels for short-term exposures. There are insufficient data to provide these levels with confidence at present, but current evidence suggests values of about 75 μg/m³ for NO_x and 270 μg/m³ for NH_3 as 24-hour means.

Interactive effects between NO_2 and sulfur dioxide and/or ozone have been reported frequently *(8–13)*. From a review of recent literature, however, it was concluded that the lowest effective levels for NO_2 are approximately equal to those for combination effects (although in general, at concentrations near to its effect threshold, NO_2 causes growth stimulation if it is the only pollutant, while in combination with sulfur dioxide and/or ozone it results in growth inhibition).

Critical levels for a 1-year period are recommended to cover relatively long-term effects. The critical level for NO_x (NO and NO_2, added in ppb and expressed as NO_2 in μg/m³) is 30 μg/m³ as an annual mean. The critical level for NH_3 is 8 μg/m³ as an annual mean.

REFERENCES

1. *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23).
2. Ashenden, T.W. et al. *Critical loads of N & S deposition to semi-natural vegetation*. Bangor, Institute for Terrestrial Ecology, 1990 (Report Project T07064L5).
3. Bobbink, R. et al. Critical loads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna. *In*: Grennfelt, P. & Thörnelöf, E., ed. *Critical loads for nitrogen*. Copenhagen, Nordic Council of Ministers, 1992, pp. 111–159.
4. Fangmeijer, A. et al. Effects of atmospheric ammonia on vegetation – a review. *Environmental pollution*, **86**: 43–82 (1994).
5. Grennfelt, P. & Thörnelöf, E., ed. *Critical loads for nitrogen.* Copenhagen, Nordic Council of Ministers, 1993 (Miljørapport No. 41).
6. Guderian, R. *Critical levels for effects of NO_x*. Geneva, United Nations Economic Commission for Europe, 1988.

7. SCHULZE, E.D. ET AL. *Forest decline and air pollution; a study of spruce (*Picea abies*) on acid soils*. Heidelberg, Springer-Verlag, 1989 (Ecological Studies, No. 77).
8. ADAROS, G. ET AL. Concurrent exposure to SO_2 alters the growth and yield responses of wheat and barley to low concentrations of CO_2. *New phytologist*, **118**: 581–591 (1991).
9. ADAROS, G. ET AL. Single and interactive effects of low levels of O_3, SO_2 and NO_2 on the growth and yield of spring rape. *Environmental pollution*, **72**: 269–286 (1991).
10. CAPE, J.N. ET AL. Sulfate and ammonium in mist impair the frost hardening of red spruce seedlings. *New phytologist*, **118**: 119–126 (1991).
11. CAPORN, T.M. ET AL. Canopy photosynthesis of CO_2-enriched lettuce (*Lactuca sativa* L.). Response to short term changes in CO_2, temperature and oxides of nitrogen. *New phytologist*, **126**: 45–52 (1994).
12. ITO, O. ET AL. *Effects of NO_2 and O_3 alone or in combination on kidney bean plants. II. Amino acid pool size and composition*. Tokyo, National Institute of Environmental Studies, 1984 (Research Report No. 66).
13. VAN DE GEIJN, S.C. ET AL. Problems and approaches to integrating the concurrent impacts of elevated CO_2, temperature, UVb radiation and O_3 on crop production. *In*: Buxton, D.R. et al., ed. Madison, WI, Crop Science of America, 1993, pp. 333–338.

Effects of ozone on vegetation: critical levels

EFFECT EVALUATION

The revision of the air quality guidelines for ozone builds on the progress made to define critical levels to protect crops and tree species. Guidelines for other photochemical oxidants, such as peroxyacetylnitrate and hydrogen peroxide, are not recommended because of the low levels of these pollutants observed in Europe, and because data concerning their effects on plants in Europe are very limited. Research in recent years has mainly advanced our understanding of the exposure, uptake and effects of ozone *(1, 2)*.

Ozone concentrations vary widely both in space and in time, and in order to quantitatively relate ozone exposure to effects it is necessary to summarize the concentration pattern during the exposure period in a biologically meaningful way *(3, 4)*. From results of exposure–response studies with open-top chambers, it is concluded that mean concentrations are not appropriate to characterize ozone exposure. This is mainly because (*a*) the effect of ozone results from the cumulative exposure; and (*b*) not all concentrations are equally effective, higher concentrations having greater effects than lower concentrations. Ozone exposure is therefore expressed as the sum of all 1-hour mean concentrations above a cut-off concentration of 40 ppb. It is emphasized that 40 ppb should not be regarded as a lower concentration limit for biological effects, since some biological effects may occur below this value; rather, it is a cut-off concentration used to calculate an exposure index that is strongly related to biological responses, and hence to the degree of risk to sensitive vegetation.

The use of 40 ppb as the cut-off concentration provides good linear relationships between ozone exposure and plant response for a number of species, thus confirming its biological relevance *(5, 6)*. Furthermore, the ozone concentrations found in most areas of Europe, in the absence of photochemical pollution, are in the range 10–40 ppb, except at very high altitudes. In relation to long-term effects, this sum (referred to as the "Accumulated exposure Over a Threshold of 40 ppb", AOT40), is calculated for a 3-month growing season in the case of crops or herbaceous semi-natural vegetation, or a 6-month growing season for trees. The appropriate months to define the growing season will depend on the vegetation and climate in a specific region or at a specific site. Since uptake of ozone by vegetation occurs primarily during daylight hours when stomata are open, the calculation of the AOT40 considers only those hours when radiation is higher than 50 W/m^2.

To define critical levels, the AOT40 is related to specific effects *(2)*. A reduction in economic yield (such as grain yield in wheat) is considered the most relevant long-term effect of ozone on crop species, and a reduction in biomass is chosen for tree species. For semi-natural vegetation, the effect of ozone is expressed as the change in the species composition. The most important short-term effect of ozone is the appearance of visible leaf injury. The most sensitive species for each vegetation type for which adequate data are available was selected to derive the critical level.

For crops, data on grain yield of spring wheat exposed in open-top field chambers to different ozone concentrations over the growing season were used to set the critical level, since the database is the largest (10 experiments in 6 countries using 10 different cultivars) and most consistent, and wheat is known to be a sensitive species. Statistical analysis of this pooled dataset showed that the least significant deviation in yield that can be estimated with 99% confidence is 4–5%. The critical level determined using this criterion (Table 32) is 3 ppm·h *(5, 7)*.

The critical level for short-term effects of ozone on crops (visible injury) is derived from an extensive database of coordinated European field observations, involving eight countries over two growing seasons using two clover species *(8)*. Using artificial neural network analysis, combinations of ozone exposure and climatic conditions in the five days preceding the onset of visible injury were identified and used to set critical levels (Table 32) of 0.2 ppm·h for humid air conditions (mean vapour pressure deficit below 1.5 kPa) and 0.5 ppm·h for dry air conditions (mean vapour pressure deficit above 1.5 kPa).

For forests, the database available is small. Data sets from three different European studies using open-top field chambers of the effects of ozone on annual biomass increment in beech saplings have been used *(9)*. Statistical analysis of these data showed that the least significant deviation in biomass increment that could be estimated with 95% confidence was about 10%, and this criterion was used to determine a critical level of 10 ppm·h (Table 32).

Table 32. Guidelines for the effects of ozone on vegetation: critical levels

Vegetation type	Guidelines AOT40 (ppm·h)	Time period[a]	Constraints
Crops (yield)	3	3 months	
Crops (visible injury)	0.2	5 days	Humid air conditions (mean daytime VPD[b] below 1.5 kPa)
	0.5	5 days	Dry air conditions (mean daytime VPD[b] above 1.5 kPa)
Forests	10	6 months	
Semi-natural vegetation	3	3 months	

[a] Daylight hours.

[b] VPD = vapour pressure deficit.

Finally, for herbaceous species of semi-natural vegetation, recent studies have reported the effects of ozone in field or laboratory chambers on shoot biomass, seed biomass or relative growth rate of a total of 87 species. All studies showed significant adverse effects, at the 95% confidence level, of exposures in the range 3–5 ppm·h on the most sensitive species studied. Since there is also evidence that the most sensitive of these species are as sensitive as the most sensitive known crop species, a critical level of 3 ppm·h (Table 32), equivalent to that for crops, has been adopted *(10)*.

GUIDELINES

The data used to derive critical levels are almost entirely drawn from experiments in open-top chambers in central and northern Europe, using plants that are adequately supplied with water and nutrients. There are uncertainties in using these data to define air quality guidelines for vegetation throughout Europe. Among the most important of these uncertainties are the following.

- The open-top chamber technique will tend to overestimate the effects because of the higher ozone fluxes within the chambers compared with outside.
- There are a great many species that have not been investigated experimentally in Europe, especially in the Mediterranean region.

- The critical level is likely to be higher when water availability is limited, because ozone flux is reduced. This is a very significant factor in many areas of Europe, especially as periods of water stress often coincide with periods of high ozone concentration.
- There may be physiological, morphological or biochemical changes induced by ozone exposures below the critical level that could be important, for example in altering sensitivity to other abiotic and biotic stresses.
- The data on trees are more variable than those for annual crops and there is uncertainty about the extent and significance of night-time ozone uptake. Furthermore, there are uncertainties in extrapolating from experiments of limited duration with young pot-grown trees to long-term effects on forest ecosystems. For these reasons, there is greater uncertainty attached to the recommended guidelines for trees.
- For changes in species composition, the experiments are also of limited duration, and there is great uncertainty about the long-term effects of ozone exposure.

When determining whether ozone exposures at a specified location exceed the critical levels (Table 32), two points need to be carefully considered.

1. Over short vegetation, but not over forests, there may be significant gradients in AOT40 immediately above the vegetation, and thus AOT40 values determined at the measurement height of most monitoring stations may be larger than at the surface of the vegetation. In contrast, in experimental chambers used to generate the exposure–response data, the air is well mixed and the gradients do not exist.

2. AOT40 values can vary substantially from year to year, because of the variability of the climate. Because the critical level for crop yield was based on analysis of data in several different growing seasons, and because the critical level for forests was based on multi-year experiments, it is recommended that the exceedance of these critical levels, and that for semi-natural vegetation, be evaluated on the basis of mean AOT40 values over a 5-year period. Where visible injury to crops resulting from short-term exposures is of direct economic concern, however, examination of monitoring data for the year with highest ozone exposures is recommended.

REFERENCES

1. Jäger, H.J. et al., ed. *Effects of air pollution on agricultural crops in Europe*. Brussels, European Commission, 1993 (Air Pollution Research Report, No. 46).

2. FUHRER, J. & ACHERMANN, B., ED. *Critical levels for ozone: a UN-ECE workshop report*. Liebefeld-Bern, Swiss Federal Research Station for Agricultural Chemistry and Environmental Hygiene, 1994 (FAC Report, No. 16).
3. DERWENT, R.G. & KAY, P.J.A. Factors influencing the ground level distribution of ozone in Europe. *Environmental pollution*, **55**: 191–219 (1988).
4. RUNECKLES, V.C. Dosage of air pollutants and damage to vegetation. *Environmental conservation*, **1**: 305–308 (1974).
5. ASHMORE, M.R. & WILSON, R.B., ED. *Critical levels for air pollutants in Europe*. London, Department of the Environment, 1994.
6. FUHRER, J. The critical level for ozone to protect agricultural crops. An assessment of data from European open-top chamber experiments. *In*: Fuhrer, J. & Achermann, B., ed. *Critical levels for ozone: a UN-ECE workshop report*. Liebefeld-Bern, Swiss Federal Research Station for Agricultural Chemistry and Environmental Hygiene, 1994 (FAC Report, No. 16), pp. 42–57.
7. SKÄRBY, L. ET AL. Responses of cereals exposed to air pollutants in open-top chambers. *In*: Jäger, H.J. et al., ed. *Effects of air pollution on agricultural crops in Europe.* Brussels, European Commission, 1993 (Air Pollution Research Report, No. 46), pp. 241–259.
8. BENTON, J. ET AL. The critical level of ozone for visible injury on crops and natural vegetation (ICP Crops). *In*: Kärenlampi, L. & Skärby, L., ed. *Critical levels for ozone in Europe: testing and finalizing the concepts. UN-ECE workshop report.* Kuopio, Department of Ecology and Environmental Science, University of Kuopio, 1996, pp. 44–57.
9. BRAUN, S. & FLÜCKIGER, W. Effects of ambient ozone on seedlings of *Fagus silvatica* L. and *Picea abies* (L.) Karst. *New phytologist*, **129**: 33–44 (1995).
10. ASHMORE, M.R. & DAVISON, A.W. Toward a critical level of ozone for natural vegetation. *In*: Kärenlampi, L. & Skärby, L., ed. *Critical levels for ozone in Europe: testing and finalizing the concepts. UN-ECE workshop report.* Kuopio, Department of Ecology and Environmental Science, University of Kuopio, 1996, pp. 58–71.

Indirect effects of acidifying compounds on natural systems: critical loads

ACIDIFYING DEPOSITION AND ECOSYSTEM DAMAGE

Historical data provide evidence of increasing transport of sulfate (SO_4^{2-}), up to a factor of 2 and 3.5 in 1950 and 1980, respectively, compared to pre-industrial levels in Europe. Emissions of sulfur dioxide in the air are transformed to sulfate, which constitutes the major compound of acid deposition *(1, 2)*. The effects and risks of sulfur dioxide emissions and resulting deposition are described for soils in general and for forest soils and surface waters in particular.

Soil acidification is defined as a decrease in the acid neutralizing capacity (ANC) of the inorganic fraction of the soil including the solution phase, and is directly dependent on the net supply of base cations (by weathering and deposition) and the net supply of anions (deposition minus retention) in the mineral soil *(3, 4)*. Deposition of acidifying compounds such as sulfur dioxide, nitrogen oxides and ammonia leads to soil acidification by oxidation to sulfuric and nitric acids and leaching of sulfate and nitrate, respectively.

The dynamics of forest soil acidification is very site-specific and depends on soil characteristics such as weathering rate, sulfate adsorption capacity and cation exchange capacity. The acidification of soils ultimately leads to an increase in the soil solution of the aluminium concentration, which increases the risk of vegetation damage. By defining the relationship between the chemical status (base cation and aluminium concentrations in the soil solution) and vegetation response, the so-called critical load for that particular ecosystem can be derived *(5)*. Damage to forests in Europe, including defoliation, discoloration, growth decrease and tree dieback, have been reported over the last decade, and have to a large extent been attributed to soil acidification, but also to eutrophication and photochemical oxidant effects.

Acidic deposition has caused acidification of surface waters, fish mortality and other ecological changes in large areas of northern Europe and eastern parts of North America.

Sulfate is normally a mobile anion in catchments located in glaciated areas. Increased sulfate concentrations in runoff due to increased acidifying inputs are accompanied by an increase of base cations and a decrease in bicarbonates, resulting in an acidifying effect on surface waters.

For most of the sensitive soils in Europe, the sulfate deposition is directly related to the acidifying load of sulfate in watershed runoff. The deposition/runoff relationship for nitrogen is not as well defined. The nitrate concentration in runoff, and hence the contribution to the total acidity loading, is due to a combination of factors including the amount of deposition, the ability of vegetation to take up nitrogen and the denitrifying processes. Even in cases of substantial nitrogen runoff, models calculate a deposition/runoff ratio greater than 1 due to denitrification. Determining the nitrate runoff response to a change in nitrogen deposition requires site-specific information.

Under natural conditions, most of the nitrogen deposited on terrestrial catchments is taken up by vegetation, leading to low concentrations of ammonia and nitrate in the runoff. In some areas in Europe, however, including Denmark, southern Norway and southern Sweden, nitrogen concentrations in runoff water appear to be above background values. This excess nitrate in runoff is mostly due to a disruption of the nitrogen cycle and not only to increased nitrogen deposition. In such cases, nitrogen deposition exceeds the rate of nitrogen retention mechanisms, i.e. growth uptake, denitrification and immobilization. When nitrate is leached from the soil solution and appears in surface waters, it will contribute to soil and surface water acidification in the same manner as sulfate.

In cases of low soil pH, excess nitrogen deposition leads to acidification of natural vegetation systems other than trees. Plant species from poorly buffered habitats are adapted to nitrate uptake, while plants from acid environments are generally adapted to ammonia uptake. A low pH may thus lead to a shift of these systems from a nitrate-dominated to an ammonia-dominated system. Such a disruption of the nitrogen cycle in combination with low pH ultimately leads to acidification by nitrogen.

CRITICAL LOADS

The critical load of acidity means "the highest deposition of compounds that will not cause chemical changes leading to harmful effects on ecosystem structure and function" *(6)*.

A relationship has been established between increased aluminium concentrations in the soil solution and adverse effects to roots and growth of trees. For example, it has been shown that the tree growth of Norway spruce decreases as the base cation (calcium, magnesium, potassium) to aluminium (BC/Al) ratio is smaller than a critical limit of 1 *(7)*. Other critical limits for forest soils are based on aluminium concentration and pH in soil solution. Laboratory results of aluminium damage indicate that tolerance to aluminium varies among tree species. For example, a growth reduction of 80% has been demonstrated at a BC/Al ratio of 0.1 for the northern white cedar (*Thuja occidentalis*) and of 4 for the masson pine (*Pinus massoniana*). It has been found that a BC/Al ratio exceeding or equal to 1 seems to provide appropriate sustainability for European forests. However, species that grow in non-glaciated old soils rich in aluminium oxide, such as teak, guapira, orange and cotton, seem to be more accustomed to aluminium than trees from the temperate zone. Computations of critical loads in Europe have therefore generally applied a BC/Al ratio of 1 *(7)*.

For surface waters, the ANC has been considered a chemical criterion that is used to explain the increased risk of damage to fish populations. The critical chemical value, ANC limit = 20 μeq/l, has been derived from the information on water chemistry and fish status obtained from the 1000-lake survey carried out in Norway in 1986 *(8, 9)*. The selected ANC limit was assessed by examining the relationship between the critical load exceedance, and damage to fish populations on the basis of data from the Norwegian 1000-lake survey can again be used *(10)*. The probability of damage to fish populations increases clearly as a function of the critical load exceedance. Table 33 gives an overview of average limits that have been established to compute critical loads.

Calculation of critical loads is based on the steady state mass balance method which assumes a time-independent steady state of chemical interaction involving an equilibrium between the production and the consumption of acidic compounds.

Current United Nations Economic Commission for Europe (ECE) protocols concentrate on distinctive acidifying compounds, such as sulfur and nitrogen, rather than on acidity as a whole. It was necessary to subdivide

Table 33. Critical limits for chemical compounds and properties in forest soils and freshwater systems

Compound / property	Unit	Forest soil	Fresh water	Groundwater
Aluminium	mol_c/m^3	0.2	0.003	0.02
BC/Al	mol/mol	1	–	–
pH	–	4.0 [a]	(5.3, 6.0) [b]	6.0
ANC	mol_c/m^3	–	(0.02, 0.08) [b]	0.14
NO_3^-	mol_c/m^3	–	–	0.8

[a] Assuming log K_{gibb} of 8.0 and Al = 0.2 mol_c/m^3.

[b] A pH of 6.0 relates to peak flow situations and is associated with ANC = 0.08 mol_c/m^3.

the critical load of acidity between the acidifying share of sulfur and that of nitrogen. For the purposes of the guidelines no subdivisions are performed.

GUIDELINES

In Europe, critical loads have been established at the EMEP resolution (see page 225) to allow for comparisons between critical loads and sulfur deposition values, and to identify areas where critical loads are exceeded. Critical loads of acidity, as computed by the steady state mass balance method, depend predominantly on the rate of base cation weathering. For terrestrial ecosystems, the weathering rate can be estimated by combining information on soil parent material and texture properties. The critical loads of acidity in relation to combinations of parent material and texture classes range from smaller than 250 eq/ha per year to more than 1500 eq/ha per year (see Table 34).

Additional factors, such as vegetation cover, further modify the value of the critical load. To calculate precise critical loads for a given geographical area, it is recommended that the mass balance equation be used. For surface waters, the weathering rate can be estimated on the basis of water quality and quantity variables, of which base cation concentrations and runoff are the most influential ones.

Table 34. Critical load ranges of acidity used for the various combinations of parent material and texture in terrestrial ecosystems

Guideline range of critical loads of acidity (eq/ha per year)	Parent material[a]	Texture[b]
< 250	acidic	coarse
250–500	acidic	coarse-medium
	intermediate	coarse
	basic	coarse
500–1000	acidic	medium, medium-fine
	intermediate	coarse-medium, medium
	basic	coarse-medium
1000–1500	intermediate	medium-fine
	basic	medium
> 1500	intermediate	fine
	basic	medium-fine

[a] Acidic: sand (stone), gravel, granite, quartzine, gneiss (schist, shale, greywacke, glacial till).
Intermediate: gronodiorite, loess, fluvial and marine sediment (schist, shale, greywacke, glacial till).
Basic: gabbro, basalt, dolomite, volcanic deposits.

[b] Coarse: clay content < 18%.
Medium: clay content 18–35%.
Fine: clay content > 35%.

Table 35 lists the ranges of critical loads in relation to combinations of base cation concentration and runoff classes. For each critical load class, at least 50% of the critical load values computed on the basis of lake data from Finland (1450 lakes), Norway and Sweden fall within the class boundaries, given the ranges for present base cation concentrations and runoff. Only in two cases did the boundaries for the base cation concentration classes overlap between two critical load classes, when the class boundaries were set on the basis of the 25th and 75th percentile base cation concentrations for given runoff classes. For those cases the critical loads are determined more by other factors than base cation levels and runoff, and the guideline value set is therefore more uncertain than those without overlap.

Table 35. Critical load ranges of acidity used for various combinations of base cation concentration and runoff for surface waters

Guideline range of critical loads of acidity (eq/ha per year)	Base cation concentration (meq/m^3)	Runoff (m)
< 250	< 45	> 1.0
	< 100	0.3–1.0
	< 270 [a]	< 0.3
250–500	45–70	> 1.0
	100–190	0.3–1.0
	250–400 [a]	< 0.3
500–1000	70–103	> 1.0
	190–290	0.3–1.0
	400–650	< 0.3
1000–1500	103–170	> 1.0
	290–465 [a]	0.3–1.0
	650–1300	< 0.3
> 1500	> 170	> 1.0
	> 350 [a]	0.3–1.0
	> 1300	< 0.3

[a] The class boundaries overlap.

REFERENCES

1. FINKEL, R.C. ET AL. Changes in precipitation chemistry at Dye 3, Greenland. *Journal of geophysics research*, **910**: 9849–9855 (1986).
2. TUOVINEN, J.-P. ET AL. *Transboundary acidifying pollution in Europe: calculated fields and budgets 1985–93*. Oslo, Norwegian Meteorological Institute, 1994 (Technical Report, No. 129).
3. VAN BREEMEN, N. ET AL. Acidic deposition and internal proton sources in acidification of soils and waters. *Nature*, **307**: 599 (1984).
4. DE VRIES, W. & BREEUWSMA, A. The relation between soil acidification and element cycling, *Water, air and soil pollution*, **35**: 293–310 (1987).

5. DE VRIES, W. *Soil response to acid deposition at different regional scales: field and laboratory data, critical loads and model predictions*. Dissertation, Agricultural University, Wageningen, 1994.
6. NILSSON, J. & GRENNFELT, P., ED. *Critical loads for sulphur and nitrogen.* Copenhagen, Nordic Council of Ministers, 1993 (Miljørapport No. 15).
7. SVERDRUP, H. & WARFVINGE, P. *The effect of soil acidification on the growth of trees, grass and herbs as expressed by the (Ca+Mg+K)/Al ratio*. Lund, University of Lund, 1993 (Report No. 2).
8. HENRIKSEN, A. ET AL. Lake acidification in Norway. Present and predicted chemical status. *Ambio,* **17**: 259–266 (1988).
9. HENRIKSEN, A. ET AL. Lake acidification in Norway. Present and predicted fish status. *Ambio,* **18**: 314–321 (1989).
10. HENRIKSEN, A. & HESTHAGEN, T. *Critical load exceedance and damage to fish populations*. Oslo, Norwegian Institute for Water Research, 1993 (Project Naturens Tålegrenser, Fagrapport No. 43).

Effects of airborne nitrogen pollutants on vegetation: critical loads

Most of earth's biodiversity is found in natural and seminatural ecosystems, both in aquatic and terrestrial habitats. Man's activities pose a number of threats to the structure and functioning of these ecosystems, and thus to the natural variety of plant and animal species. One of the major threats in recent years is the increase in airborne nitrogen pollution, namely NH_y (consisting of ammonia and ammonium ions), and NO_x (consisting of nitrogen dioxide and nitric oxide). Nitrogen is the limiting nutrient for plant growth in many of these ecosystems. Most of the plant species from these habitats are adapted to nutrient-poor conditions, and can only compete successfully on soils with low nitrogen levels *(1)*. Nitrogen is the only nutrient whose cycle through the ecosystem is almost exclusively regulated by biological processes.

To establish reliable critical loads for nitrogen, it is essential to understand the effects of nitrogen on these ecosystem processes. The critical loads for nitrogen depend on:

- the type of ecosystem;
- the land use and management in the past and present; and
- the abiotic conditions, especially those that influence the nitrification potential and immobilization rate in the soil.

The impacts of increased nitrogen deposition on biological systems are diverse, but the most important effects are:

- short-term direct effects of nitrogen gases and aerosols on individual species (see Chapter 11);
- soil-mediated effects;
- increased susceptibility to secondary stress factors; and
- changes in (competitive) relationships between species, resulting in loss of biodiversity.

The empirical approach has been used to establish guidelines for excess nitrogen deposition on natural and seminatural vegetation. It was decided not to include the results of the mass balance approach with nitrogen as a nutrient for non-forest ecosystems, because essential data are missing. The acidifying effects of airborne nitrogen are incorporated in the guidelines for excess acidity based on steady state mass balance models (see Chapter 13).

EVALUATION OF CRITICAL LOADS

The main aim of this evaluation was to update the guideline for airborne nitrogen deposition on vegetation, which was estimated at 30 kg/ha per year for sensitive vegetation *(2)*. Since 1987, significant progress has been made in understanding the ecological effects of nitrogen deposition on several types of vegetation. Critical loads of nitrogen have been formulated on an empirical basis by observing changes in the vegetation, fauna and biodiversity *(3, 4)*. Experiments under controlled and field conditions, and comparisons of vegetation and fauna composition in time and space, are used to detect changes in ecosystem structure *(5–7)*.

Changes in plant development and in species composition or dominance have been used as a "detectable change" for the impacts of excess nitrogen deposition, but in some cases a change in ecosystem function, such as nitrogen leaching or nitrogen accumulation, has been used. The results of dynamic ecosystem models, integrating both biotic and abiotic processes, are also used where available. Based on these data, guidelines for nitrogen deposition (critical loads) have been presented for receptor groups of natural and seminatural ecosystems, namely:

- wetlands, bogs and softwater lakes
- species-rich grasslands
- heathlands
- forest ecosystems (including tree health and biodiversity).

Critical loads have been defined within a range per ecosystem, because of (*a*) real intra-ecosystem variation within and between countries, (*b*) the range of experimental treatment where an effect was observed or not observed, or (*c*) uncertainties in deposition values, where critical loads are based on field observations. The reliability of the figures presented is shown in Table 36.

It is advised, where insufficient national data are available, to use the lower, middle or upper part of the ranges of the nitrogen critical loads for terrestrial

Table 36. Guidelines for nitrogen deposition to natural and seminatural freshwater and terrestrial ecosystems

Ecosystem	Critical load [a] (kg N/ha per year)	Indication of exceedance
Wetlands		
Softwater lakes	5–10 ##	Decline in isoetid aquatic plant species
Ombrotrophic (raised) bogs	5–10 #	Decrease in typical mosses; increase in tall graminoids; nitrogen accumulation
Mesotrophic fens	20–35 #	Increase in tall graminoids; decline in diversity
Species-rich grasslands		
Calcareous grasslands	15–35 #	Increase in tall grasses; decline in diversity [b]
Neutral–acid grasslands	20–30 #	Increase in tall grasses; decline in diversity
Montane–subalpine grassland	10–15 (#)	Increase in tall graminoids; decline in diversity
Heathlands		
Lowland dry heathland	15–20 ##	Transition from heather to grass
Lowland wet heathland	17–22 #	Transition from heather to grass
Species-rich heaths/acid grassland	10–15 #	Decline in sensitive species
Upland *Calluna* heaths	10–20 (#)	Decline in heather, mosses and lichens
Arctic and alpine heaths	5–15 (#)	Decline in lichens, mosses and evergreen dwarf shrubs; increase in grasses
Trees and forest ecosystems		
Coniferous trees (acidic; low nitrification rate)	10–15 ##	Nutrient imbalance
Coniferous trees (acidic; moderate–high nitrification rate)	20–30 #	Nutrient imbalance

Table 36. (contd)

Ecosystem	Critical load [a] (kg N/ha per year)	Indication of exceedance
Trees and forest ecosystems (contd)		
Deciduous trees	15–20 #	Nutrient imbalance; increased shoot/root ratio
Acidic coniferous forests	7–20 ##	Changes in ground flora and mycorrhizas; increased leaching
Acidic deciduous forests	10–20 #	Changes in ground flora
Calcareous forests	15–20 (#)	Changes in ground flora
Acidic forests (unmanaged)	7–15 (#)	Changes in ground flora and leaching
Forests in humid climates	5–10 (#)	Decline in lichens; increase in free-living algae

[a] ## *Reliable*: a number of published papers on various types of study show comparable results.
Fairly reliable: the results of some studies are comparable.
(#) *Expert judgement*: no data are available for this type of ecosystem; the critical load is based on knowledge of ecosystems likely to be more or less comparable with this ecosystem.

[b] Use low end of the range for nitrogen-limited and high end for phosphorus-limited calcareous grasslands.

receptor groups according to the general relationships between abiotic factors and critical loads for nitrogen (Table 37).

At this moment, the critical loads are set in values of total nitrogen inputs. More information is needed in future on the relative effects of oxidized and reduced nitrogen deposition. Critical loads for nitrogen are formulated as reliably as possible. As most research has focused on acidification in forestry, serious gaps in knowledge exist on the effects of enhanced nitrogen deposition on natural and seminatural terrestrial and aquatic ecosystems. The following gaps in knowledge are particularly important:

- more research is needed in Mediterranean, tropical and subtropical vegetation zones;
- quantified effects of enhanced nitrogen deposition on fauna in all types of vegetation reviewed are extremely scarce;
- the critical loads for nitrogen deposition to Arctic and alpine heathlands and forests are largely speculative;

- more research is needed on the effects of nitrogen on forest ground vegetation and (ground) fauna, because most research had focused on the trees only;
- there is a serious gap in knowledge on the effects of nitrogen on neutral/calcareous forests that are not sensitive to acidification;
- more long-term research is needed in montane/subalpine meadows, species-rich grasslands and ombrotrophic bogs;
- the long-term effects of enhanced atmospheric nitrogen in grassland and heathland of great importance for nature conservation under different management regimes are insufficiently known and may affect the critical load value;
- the possible differential effects of the deposited nitrogen species are insufficiently known for the establishment of critical loads; and
- the long-term effects of nitrogen eutrophication in (sensitive) aquatic ecosystems (freshwater and marine) need further research.

GUIDELINES

To establish reliable guidelines, it is crucial to understand the long-term effects of increased nitrogen deposition on ecological processes in a representative range of ecosystems. It is thus very important to quantify the effects of nitrogen loads on natural and seminatural terrestrial and freshwater ecosystems by manipulation of nitrogen inputs in long-term ecosystem studies in unaffected and affected areas. These data are essential to validate the presented critical loads and to develop robust dynamic ecosystem models reliable enough to calculate critical loads for nitrogen deposition in such ecosystems.

Table 37. Suggestions for using the lower, middle or upper part of the set critical loads of terrestrial ecosystems (excluding wetlands) if national data are insufficient

Action	Temperature	Soil wetness	Frost period	Base cation availability
Move to lower part	Cold	Dry	Long	Low
Use middle part	Intermediate	Normal	Short	Intermediate
Move to higher part	Hot	Wet	None	High

Guidelines for nitrogen deposition to natural and seminatural ecosystems are given in Table 36. The most sensitive ecosystems have critical loads of 5–10 kg N/ha per year. An average value for natural and seminatural ecosystems is 15–20 kg N/ha per year.

REFERENCES

1. TAMM, C.O. *Nitrogen in terrestrial ecosystems. Questions of productivity, vegetational changes, and ecosystem stability*. Berlin, Springer-Verlag, 1991.
2. The effects of nitrogen on vegetation. *In*: *Air quality guidelines for Europe*. Copenhagen, WHO Regional Office for Europe, 1987 (WHO Regional Publications, European Series, No. 23), pp. 373–385.
3. BOBBINK, R. ET AL. Critical loads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna. *In*: Grennfelt, P. & Thörnelöf, E., ed. *Critical loads for nitrogen*. Copenhagen, Nordic Council of Ministers, 1992, pp. 111–159.
4. ROSEN, K. ET AL. Nitrogen enrichment of Nordic forest ecosystems – the concept of critical loads. *Ambio*, **21**: 364–368 (1992).
5. HENRIKSEN, A. Critical loads of nitrogen to surface water *In*: Nilsson, J. & Grennfelt, P., ed *Critical loads for sulphur and nitrogen*. Copenhagen, Nordic Council of Ministers, 1993 (Miljørapport No. 15), pp. 385–412.
6. HULTBERG, H. Critical loads for sulphur to lakes and streams. *In*: Nilsson, J. & Grennfelt, P., ed *Critical loads for sulphur and nitrogen*. Copenhagen, Nordic Council of Ministers, 1993 (Miljørapport No. 15), pp.185–200.
7. KÄMÄRI, J. ET AL. Nitrogen critical loads and their exceedance for surface waters. *In*: Grennfelt, P. & Thörnelöf, E., ed. *Critical loads for nitrogen*. Copenhagen, Nordic Council of Ministers, 1993 (Nord Miljörapport No. 41), pp. 161–200.

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WHO published the first edition of these guidelines in 1987. Since then new data have emerged and new developments in risk assessment methodology have taken place, necessitating the updating and revision of the existing guidelines. The Bilthoven Division of the WHO European Centre for Environment and Health has undertaken this process in close cooperation with the International Programme on Chemical Safety and the European Commission.

It is the aim of the guidelines to provide a basis for protecting public health from adverse effects of air pollutants and to eliminate or reduce exposure to those pollutants that are known or likely to be hazardous to human health or wellbeing. The guidelines are intended to provide background information and guidance to international, national and local authorities in making risk assessment and risk management decisions. In establishing pollutant levels below which exposure – for life or for a given period of time – does not constitute a significant public health risk, the guidelines provide a basis for setting standards or limit values for air pollutants.

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Natural England Commissioned Report NECR210

Assessing the effects of small increments of atmospheric nitrogen deposition (above the critical load) on semi-natural habitats of conservation importance

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NATURAL ENGLAND

Foreword

Natural England commission a range of reports from external contractors to provide evidence and advice to assist us in delivering our duties. The views in this report are those of the authors and do not necessarily represent those of Natural England.

Background

The work was commissioned as part of a review of the thresholds used in air quality impact assessments that was required through the Inter-agency Air Quality Technical Advisory Group. The report aimed to analyse existing scientific data to demonstrate and quantify the effect of incremental additions of atmospheric nitrogen deposition (above the critical load) on different semi-natural habitat types.

The report will be used to inform specialist advice on air quality effects on habitat that is used in planning advice, agri-environment schemes and to protect and enhance designated sites. The Environment Agency have planned (subject to approval) to use this science report to review the thresholds they use for controlling ammonia emissions from intensive farming.

This report should be cited as:

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Further information
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Assessing the effects of small increments of atmospheric nitrogen deposition (above the critical load) on semi-natural habitats of conservation importance

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Summary

1. Around two thirds of all Sites of Special Scientific Interest in the UK exceed their critical loads as a result of current atmospheric nitrogen (N) deposition. In regions where the critical load is already exceeded there is a need to understand how further increases in N deposition may affect ecological communities.
2. The objective of this report was to examine recent vegetation survey data to understand the relationships that exist between species (composition and richness) and nitrogen deposition, and to determine the effect of incremental increases in N. Vegetation data were analysed from 226 sites, collected over 8 surveys of 5 UK priority habitats for conservation (sand dune, bog, lowland heath, upland heath, acid grassland). Further evidence was gained from published survey data and the network of UK nitrogen addition experiments.
3. The relationships examined in this report use modelled annual N deposition as the pollutant variables; however, the relationships studied between N deposition and species richness and presence have developed over many years of pollution. The current rate of N deposition is used as a proxy for long-term cumulative N deposition. In some/many cases, sites will have experienced high N deposition for many years and, because of this legacy, it is unlikely that an increase or decrease in nitrogen deposition will immediately cause changes in species richness or composition.
4. Across the habitats and datasets, increasing N deposition (total, reduced or oxidised) was correlated with quantifiable declines in species richness and changes in species composition. Species richness was also correlated to climate, with increasing species richness being a function of increasing precipitation and decreasing temperature. Evidence from the literature review (N addition experiments where climatic drivers are controlled for and other field surveys) supports the findings from the data that N is driving considerable change within the habitats studied.
5. When all the habitats are considered separately, the response of species richness to long-term N deposition is curved, with sharper losses in diversity from well below the habitat-specific critical load range. At levels of N deposition at and above the upper end of each habitat-specific critical load, additional increments of long-term N are associated with further declines in species richness.
6. Not all species responded negatively, nitrogen loving plants such as the graminoids (grasses and sedges) *increased* their cover in response to increasing N deposition in bog, heath and sand dune habitats. This may result in the loss of key habitat species due to increased competition from faster-growing species, and further threaten site integrity. In addition, some species groups responded in some habitats but not in others, for example bryophyte species richness.
7. Gaps in the data mean that there remain many habitat types in the UK for which the responses to N deposition are not fully understood. Ecosystems which share similarities in species and soil type are likely to show similar responses to those found within this report. In these cases it is recommended that the findings in this report, subject to local conditions, be used to predict responses to an incremental increase in N deposition sustained over the long term. Further work should be undertaken to fill the data gaps in these habitats and those that are dissimilar to the ones studied.
8. The atmospheric concentration of NO_x and NH_3 can also influence responses. Over the long-term, changes in pollutant concentration are reflected by changes in deposition, therefore changes in annual mean concentrations could be converted to N deposition and responses predicted using the relationships developed in this report. However, it is important to recognise that the differing effects between concentration and deposition are unclear and high pollutant concentrations, even in the short-term, may be very damaging, especially for lower plants. Dose-response relationships to changes in N concentration are not fully understood and should be further researched experimentally.

9. The findings of this work, in conjunction with other recent studies, have important implications for the way that pollution regulators and the conservation agencies assess new or existing pollution sources and the assessment thresholds applied.

Acknowledgements

We are grateful for all the people and organisations that provided data and professional advice during the writing of this report. Many of the data sets examined in this study were gained as part of research by the UKREATE umbrella consortium funded by Department for Environment, Food and Rural Affairs (DEFRA). In addition, we would like to thank the Botanical Society of the British Isles, British Bryological Society and the British Lichen Society whose data was summarised as part of Task 4; Iain Diack for advice on Fens; Keith Kirby for advice on woodlands; Zoe Russell of Natural England for her expertise and direction throughout the project and the other members of the steering group, including the Countryside Council for Wales and the Environment Agency for guidance during the project and comments on the draft report.

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Contents

1. Introduction

The nitrogen problem

Emissions to the atmosphere of ammonia (NH_3) and nitrogen oxides (NO_x) dramatically increased in the 20th century due to increased combustion of fossil fuels and intensification of agriculture. Ammonia is volatilised from intensive agricultural systems such as dairy farming and intensive animal husbandry, while nitrogen oxides come mainly from burning of fossil fuel by traffic and industry (Asman *et al*., 1988; Galloway, 1995; Bobbink and Hettelingh, 2011). These combined activities result in a more than doubling of the deposition of reactive nitrogen compounds to the earth's surface (Galloway *et al*., 2004). The problems that result from increased aerial deposition of reactive nitrogen compounds have been recognised only in recent decades but are now believed to be widespread in ecological communities on regional and global scales (Emmett, 2007; Phoenix *et al*., 2006; Bobbink *et al*., 2010). There is particular concern over the impacts on natural and semi—natural ecological communities, where the normal low rates of nitrogen supply often provide important limits to ecological processes. For this reason the most obvious potential influence of pollutant nitrogen deposition is as a fertilizer, i.e. eutrophication, threatening the natural composition of those ecological communities that are well adapted to nutrient-poor soils. Another ecological impact of nitrogen deposition results from soil and water acidification which affects some species directly but also causes impacts through release of toxic metals such as aluminium (Stevens *et al*., 2010). A wider range of biogeochemical changes are also likely to occur in impacted sites such as nitrogen leaching and nutrient imbalances in soils and vegetation (see RoTAP, 2012).

The evidence base

The scientific evidence demonstrating that nitrogen pollution can affect ecosystems in the UK and elsewhere has grown substantially in the past decade and has recently been re-evaluated in RoTAP (2012). Much of the early knowledge about nitrogen impacts on ecological communities came from laboratory and field experiments which have demonstrated the potential for change in structure and function of ecosystems and communities. An alternative approach using field-based monitoring and targeted vegetation surveys provides complementary and compelling evidence that the changes seen in nitrogen addition experiments have actually occurred in the field as a result of atmospheric deposition. Various vegetation monitoring schemes such as the UK Countryside Survey (Maskell *et al*., 2010) and specific habitat surveys across deposition gradients (e.g. Stevens *et al*., 2006), supported by experiments (see Emmett *et al*., 2007), indicate that long range nitrogen pollution is or has been responsible for community changes and significant losses of plant diversity across large areas of the UK.

Critical loads

The growing knowledge base from the combined experimental studies and field surveys enable us to generate, for several plant communities, nitrogen dose – ecological response relationships and these can be used to evaluate and position critical load guidelines. The Critical Loads approach is a tool used to judge the risk of harm to the environment from several forms of air pollutants. Critical Loads are defined as: *"a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge"*. The empirical Nutrient nitrogen critical loads were revised in June 2010 (Bobbink and Hettelingh, 2011) and are shown for all EUNIS habitats in Appendix 6 and summarised in Table 1 where they relate to specific habitats studied here. Large areas of the country now exceed the critical loads for nutrient N and are predicted to continue to do so in 2020 despite reductions in emissions of reactive N gases (Hall *et al*., 2006). For an overview of Critical Loads see the UK Air Pollution Information System (http://www.apis.ac.uk/overview/issues/overview_Cloadslevels.htm).

Table 1: Summary of current critical loads relevant to the habitats studied in this report

Ecosystem type	EUNIS code	2011 critical load (kg ha^{-1} yr^{-1})	Indication of exceedance
Upland and lowland heath	F4.2	10-20	Transition from heather to grass dominance, decline in lichens, changes in plant biochemistry, increased sensitivity to abiotic stress
Sand dune grassland	B1.4	8-15	Increase in tall graminoids, decrease in prostrate plants, increased N leaching, soil acidification, loss of typical lichen species
Bog (raised and blanket)	D1	5-10	Increase in vascular plants, altered growth and species composition of bryophytes, increased N in peat and peat water
Acid grassland	E1.7	10-15	Increase in graminoids, decline of typical species, decrease in total species richness

Environmental protection

An application for an Environment Permit (under the Environmental Permitting Regulations 2010) from an industrial installation wishing to commence or expand activities triggers an assessment under the Conservation of Habitats and Species Regulations 2010 (known as the 'Habitats and Species Regulations'), in relation to European protected sites, and the Wildlife & Countryside Act 1981, as amended by the Countryside & Rights of Way (CRoW) Act 2000 in relation to SSSIs. In accordance with the legislation, permits should only[1] be given where it is possible to conclude that the installation will have no adverse effect on the integrity of a European site (SAC, SPA or, by Government Policy, Ramsar site) and is not likely to damage a SSSI. This assessment is considered in context with the thresholds within the Environment Agency (EA) H1 guidance, together with the understanding that around two thirds of all protected sites are already predicted to exceed their nitrogen critical loads as a result of existing levels of air pollution (RoTAP, 2012).

This raises a key question for the Government nature conservation advisors and environmental regulator: if there is already an identified risk of harmful effects from existing air pollution (i.e. predicted critical load or critical level exceedance), what, if any, additional air pollution arising from a new installation is acceptable. Furthermore, what, if any, benefits are likely to be evident due to a reduction of pollutant exposure while the critical load or level remains exceeded?

In order to address these questions of the consequences of changes in nitrogen deposition above and below the critical load, this report will consider in detail the form and the quantitative nature of the relationships between atmospheric nitrogen deposition and ecological response in a number of different important UK ecological communities using

[1] With the exception where Overriding Public Interest is determined by the Secretary of State or Welsh Assembly Government, under the Habitats Regulations.

recent evidence from surveys and experiments. The aim is to quantify the effect of incremental changes in long-term nitrogen deposition both above and below the critical load on important measures of plant community diversity and species composition.
The approach is to use recently available scientific data, including the UKREATE (Terrestrial Umbrella) survey dataset, and apply new statistical analysis such as canonical correspondence analysis, stepwise and LOESS (Locally weighted scatter plot smoothing) regression, and constrained cluster analysis to define and visualise the response variables within a range of habitats. Other published studies and experimental information are also assessed. The nature of the relationships between these response variables and nitrogen deposition are then examined. The results of our analysis are discussed in context with incremental increases in N deposition above and below the critical loads.

Report structure

First the report introduces the data sets and statistical methods used. The report is then structured by Tasks:

Tasks 1 and 2: To collate the relevant scientific information and categorise by habitat type
These tasks collated the available scientific information and categorised these data by habitat. Vegetation data sets from a number of surveys were used for the project, together with responses found at a number of nitrogen-addition experimental sites: details of these are provided here. In addition, key literature reporting survey responses from each habitat is also summarised in this section. In some habitats data were not available; these are identified and discussed separately in Task 6.

Task 3: To identify the relevant response variables for each habitat type
This studied each habitat for which vegetation survey data were available and how species richness or species composition varied across each dataset. The responses of these variables are analysed alongside ecological driver data such as nitrogen and sulphur pollution, temperature and precipitation and response variables strongly associated with nitrogen pollution are identified.

Task 4: To determine the relationship between nitrogen deposition and the key response variables
The nature of the relationship between the response variables identified in Task 3 and nitrogen pollution is considered further in Task 4.

Task 5: To assess the effects of different increments of nitrogen deposition above the critical load
Here the relationships identified in task 4 between N deposition and the response variables within each habitat were further considered and the effect of an incremental increase in long-term N pollution upon each was derived. This is reported as percent change in species richness or cover of selected indicator species for a 1 kg ha^{-1} yr^{-1} rise in long-term N pollution, and the amount of long-term N that would lead to a reduction in species richness of 1 species at different background levels of N pollution. Results for other increments of nitrogen are provided in Appendix 5. Comparable responses from dose-response experiments and the literature are also presented for Tasks 4 and 5.

Task 6:
This task reviews the information presented in the preceding tasks and assesses whether the relationships between the response variables and N can be applied to habitats where survey datasets were not available for analysis. Similarities between habitats in soil type and vegetation are used to complete this task. This task also considers whether the

relationships can be used when considering pollutants concentration i.e. critical levels rather than loads. A final discussion draws together the information presented within the report.

2. Methods

2.1 Vegetation survey data

This project analysed vegetation survey data collected during 8 surveys of 5 key UK semi-natural habitats between 2002 and 2009 encompassing the 2009 Terrestrial Umbrella (TU) multi-habitat survey, the 2006 TU Moorland Regional Survey (MRS), a 2002 Sand dune survey (Jones *et al*., 2004), and the BEGIN UK Acid Grassland dataset (Stevens *et al*., 2010). Mean vegetation data were collected over 5 separate quadrats per site (in most cases these quadrats were 2 x 2 m^2) and vegetation cover of all the species present within the quadrat was estimated. In the case of the Moorland Regional Survey, 0.5 x 0.5 m^2 quadrats were used. Full details of each survey are included under Tasks 1 and 2 and in Tables 2 and 3.

For the quadrat-survey technique to be directly comparable, quadrat size should be identical. It is important to note that this measure of species richness is a probability of finding a species at each site; it does not necessarily mean that fewer species are present at each location, although this may be the case. However, it does imply that the evenness of species is reduced and there is a tendency for the vegetation community to be dominated by fewer species and individual species to be present at lower frequencies. Figure 1 overleaf illustrates this concept.

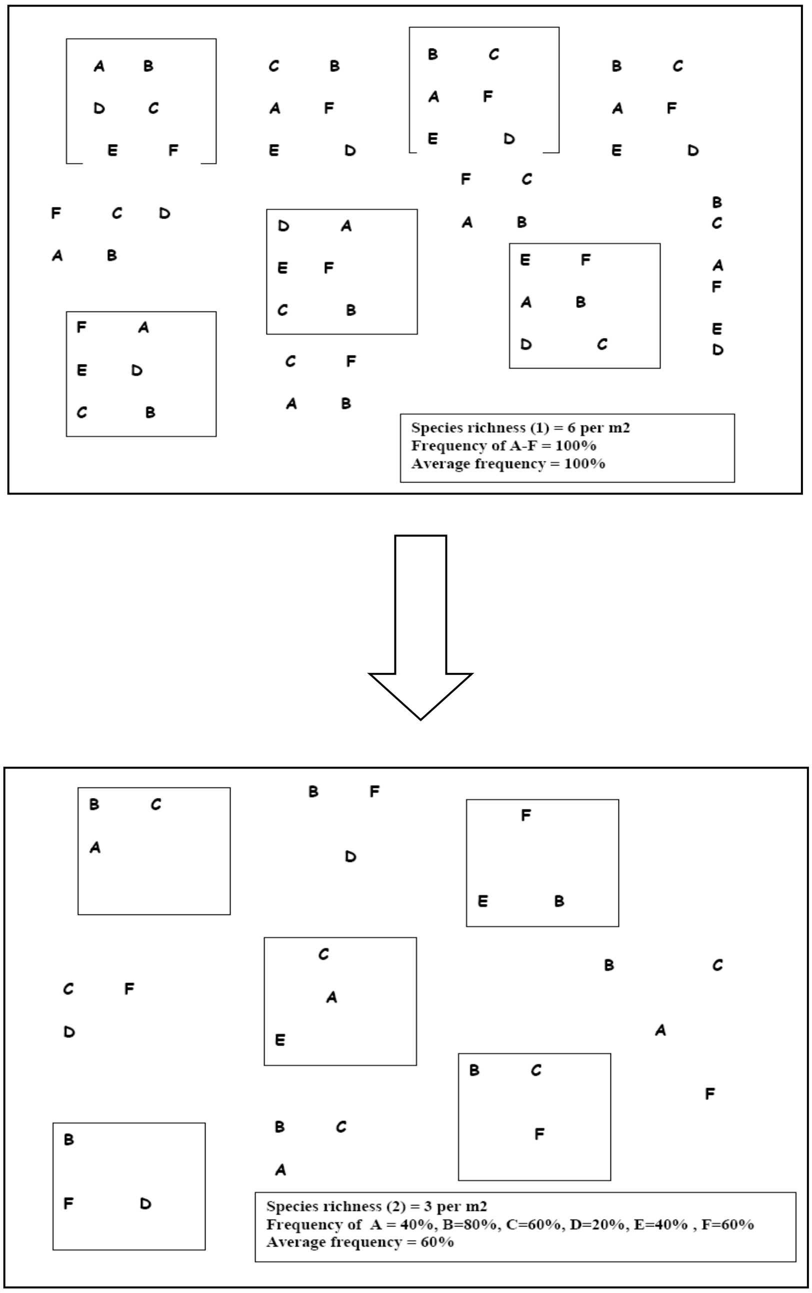


Figure 1: Species richness reduction vs. species loss. In both Figure 1a) and 1b), six species are present in the field. Both fields are surveyed using five 1 m^2 quadrats. In Figure 1 a, each quadrat surveyed contains 6 species and measured species richness is therefore 6. In Figure 1 b, each quadrat only contains 3 species, giving a species richness of 3. The same species are present in both fields, but at a lower frequency in the second: this generates a lower measure of species richness. Figure by N. Dise.

2.2 Environmental and pollutant driver variables

Species richness and composition may be affected by a number of physical and chemical variables. These drivers include variables such as temperature, precipitation, pH etc. Driver data shown in Table 2 were assembled from national datasets for use in the multivariate analyses. The climate data used were based upon UK 5 km^2 gridded data sets provided by the Met Office. Variables representing total annual precipitation and temperature were used, the latter represented by growing degree data (sum of degree days above 5^oC) in the TU and BEGIN survey data and by extreme temperature range in the MRS data. Both precipitation and growing degree data were averaged over the period 1997-2006.

The pollutant deposition data used were the 5 km^2 Concentration Based Estimated Deposition (CBED) values for 2004-2006, provided by the Centre for Ecology and Hydrology (CEH). Variables for total nitrogen deposition (further divided into wet and dry and reduced and oxidised forms), total sulphur deposition (split further into wet and dry) and non-marine calcium + magnesium deposition were included in the data analysis.

Co-correlation of deposition data between pollutant types and the response variable is acknowledged due to the intrinsic link between some pollutants, for example, nitrogen and sulphur are both by-products of fossil fuel combustion and therefore fluctuations in deposition of each follow broadly the same spatial pattern. Therefore in some cases a judgement was made of the ecological significance of a particular type of pollutant deposition. Such judgements were based where possible on the observed effects: for instance if nitrogen and sulphur were closely correlated but the effects were typical of eutrophication rather than acidification it was possible to exclude sulphur as a possible cause. It is also recognised that different forms of a pollutant have different effects on an ecosystem (e.g. oxidised and reduced nitrogen) and that more than one form of a pollutant can be correlated to ecological change. Separating closely correlated environmental variables is difficult in exploratory analyses such as the gradient studies presented here. We consider that it is better to include all possible environmental variables rather than to make *a priori* judgements about which variables are important. In cases where many variables are highly correlated the selection of one highly-correlated variable over another must be interpreted with caution. Although in many of our analyses we present results for the variable with strongest correlation statistics, in general these are best viewed as representing a broader gradient. So, for instance, although we might find strongest correlation statistics with dry deposition of oxidised nitrogen typically this variable is very strongly correlated with other forms of nitrogen deposition and the result is best seen as simply representing ‘nitrogen pollution’.

A further environmental variable included within the bog habitat was a hydrological index based upon field observations on a scale 1-5, with 1 relatively dry (similar to an upland heath) and 5 very wet: a quaking or floating bog. The environmental variables used in the statistical analysis and their acronyms when included on ordination plots are summarised in Table 2. Variables such as radiation index, latitude and longitude were not included as these are correlated with both climate and pollution; variability in these is accounted for by precipitation and growing degree days.

It is recognised that in these semi-natural habitats site management is an important determinant of vegetation structure. The term ‘management’ encompasses a range of human interventions (burning, grazing, drainage) that are difficult to quantify and for which national-scale data is rarely available. We attempt to account for these variables using field-observed indices. For the bog data, the hydrological index largely reflects the history of drainage and peat cutting. For the Moorland Regional Survey we included a ‘habitat’ term which captured the development phase of the *Calluna vulgaris* growth cycle i.e. ‘pioneer’, ‘building’, ‘mature’, ‘degenerate’ (Gimingham, 1972). For the TU heathland and sand dune data no management variables were included. In these studies (indeed in all of the studies) site and quadrat selection was carefully considered to maximise consistency between sites.

Table 2: Summary of driver variables used in the statistical analysis

Driver variables	Acronym	Comment
Growing degree days	growdeg	sum of degree days above 5^{o}C
Precipitation	precip	
Extreme temperature range		Moorland regional survey only
Habitat		Moorland regional survey only
Grazing		Acid grasslands only
Altitude	altimetr	not sand dunes
Hydrology	bog_hydr	Bog habitat only
pH	pH	
Loss on ignition	LOI	
Total acid deposition	aciddepo	
Sulphur deposition	sulpdepo	
Nitrogen deposition	Nitrdepo	
Oxidised nitrogen deposition	oxiNdepo	
Reduced nitrogen deposition	redNdepo	
Calcium + magnesium deposition	Cmgdepo	
Wet sulphur deposition	wet_sulp	
Dry sulphur deposition	dry_sulp	
Wet oxidised nitrogen deposition	wet_oxiN	
Dry oxidised nitrogen deposition	dry_oxiN	
Wet reduced nitrogen deposition	wet_redN	
Dry reduced nitrogen deposition	dry_redN	

2.2.1 Cumulative N versus recent N deposition

The relationships examined in this report use modelled recent annual N deposition as the pollutant variable(s), however, the relationships studied between N deposition and species richness and presence have developed over many years (Dise *et al*., 2011). The recent rate of N deposition is primarily a proxy for longer-term cumulative N deposition. Thus we would not expect that a change in N deposition, either increasing or decreasing, would immediately change species richness or composition, but instead these would be gradually influenced by longer-term changes in N deposition. However, different plant groups respond in different ways: bryophytes and lichens are likely to respond quicker than vascular plants and the responses of both may be affected by management interaction which could alter inter-species composition.

Since cumulative deposition data from the survey locations was not available for use in this study, the current N deposition was instead used as a proxy for cumulative deposition. If cumulative data are estimated from current deposition patterns (as in Dupre *et al*., 2010) we would expect to see very similar results. However, if cumulative deposition data based on emission changes over time were available, this may show different results. An example of this could be in an area where agricultural N emissions have increased markedly in recent years: long term cumulative N deposition based upon current N deposition would over-estimate the total N deposited to the site. Conversely, the use of current N deposition estimates for an area to which N deposition has reduced dramatically over recent years at a rate different to broad-scale trends would under-estimate the cumulative N deposited. Fowler *et al*. (2004) calculated cumulative N deposition for 1900-2000 based upon historic emissions data. A comparison of the cumulative N deposition map for 1900-2000 with modelled nitrogen deposition maps for the year 2000 revealed that broad-scale patterns of N deposition across the country were very similar in these datasets; however, one area of notable difference is apparent. Relatively recent modelled N deposition in East Anglia is much greater than that indicated long-term cumulative N, presumably owing to growing agricultural emissions.

Given the potential impacts of high N deposition both over the long term and currently, a sensible approach would be to consider both N deposition scenarios when judging the vulnerability of a site to raised N and assessing the impact of N deposition at a site. However, given the similarities between the broad-scale spatial patterns of cumulative and contemporary N deposition for much of the UK, the use of current modelled deposition as a proxy for long-term N to predicting responses to increases in N deposition seems reasonable.

2.3 Experimental data

A number of long-term nitrogen addition experiments exist in the UK, many within the DEFRA 'UK Research on The Eutrophication and Acidification of Terrestrial Ecosystems' (UKREATE) project, and data from these were included where relevant to the findings of this project. These sites are summarised in Task 3 and Tables 5 and 6.

2.4 Data analysis

Several analytical techniques have been used to determine the effect of air pollution and environmental variables on key response variables in the survey data. Responses studied included species richness, species composition, changes in species richness of different functional groups and the response of certain individual species. The techniques used include ordination analysis, stepwise and conventional regression, LOESS (Locally weighted scatterplot smoothing) regression and cluster analysis.

2.4.1 Ordination analysis

Ordination analysis is a suite of techniques for the analysis of multivariate data in which the aim is to arrange samples along axes on the basis of their species compositions. At their simplest ordination techniques function as a dimension-reduction technique allowing the representation of difference or similarity in species composition of samples in a simple two-dimensional plot. Axes can be determined simply by the species composition of those samples (unconstrained ordinations, also termed indirect gradient analysis) or can be constrained to be composed of linear combinations of measurable environmental variables (constrained ordinations, also termed direct gradient analysis). In our analyses of gradient studies ordination techniques allowed us to understand and visualise the relationships between overall community composition and the environmental gradients which drive changes in that composition (pollution, climate etc). Furthermore, ordination plots allow us to identify individual species which are particularly responsive to individual environmental variables and which may function as indicator species. These analyses are therefore separate from, but complimentary to, analyses of univariate variables which integrate some aspect of community composition such as species richness or functional group ratios. Environmental controls on overall species composition are separate from, but often overlap with those on species richness.

A Detrended Correspondence Analysis (DCA) was first performed to analyse the length of environmental gradients underlying each dataset. Then either Redundancy Analysis (RDA) or Canonical Correspondence Analysis (CCA) were used as appropriate, assuming linear and unimodal species responses along the environmental gradients respectively (Leps and Smilauer, 2003). We present two key outputs from RDA or CCA, the numerical results namely the % variance explained (analogous to the R^2 of a regression) and P-value (determined by Monte Carlo permutation test) and the ordination plot. Large-scale vegetation datasets will typically contain considerable noise due to non-measured variables and random variability so the proportion of variance explained by environmental variables is often low. However the relationships identified can still be highly significant with small P-values.

Initially, all environmental variables were entered into each analysis and a forward selection procedure using Monte Carlo permutation tests carried out to establish a minimal suite of variables that independently explained significant variance in the data. The variable that explained greatest variance in the data (greatest marginal effect) was first selected in the analysis, and then included as a co-variable in subsequent analysis. The variance explained by all other variables was then tested to identify the next variable that then explained the greatest *additional* variance. This variable could then be tested for significance, and if $P<0.05$ included in the analysis. The selection process continued until no further variables explained significant additional variance. This approach enabled the statistical effect of climate and pollutant variables to be separated, however, it can mean that variables that are correlated with each other (such as different types of air pollution) may be excluded as only the variable with the strongest association is included, and when this variable is included as a covariable, the other variables that are correlated with it may be removed from the analysis as they explain no additional variation.

Subsequently variance partitioning was carried out to test the % variance and significance of each forward selected variable with other selected variables as co-variables. CANOCO software for Windows version 4.53 (ter Braak and Smilauer, 2004) was used for both CCA and RDA analysis.

An example ordination plot from an RDA analysis is provided below (Figure 2). In all ordination plots the values of the axes and the position of species relative to those axes is of less interest in general terms than the position of species relative to other species and environmental variables. The red arrows represent environmental variables and their direction of influence relative to the species shown in the diagram. The strength of each driver variable on species composition is represented by the relative length of arrow, in this example ‘redNdepo’ (reduced nitrogen deposition) has the longest red arrow and as such exerts the most influence on species composition. The correlation between environmental variables can be judged by their similarity of direction, arrows pointing in the same direction represent correlated variables. In the plot below it will be noted that the three red arrows point in different directions, there is little correlation between variables. Similarly, individual species are shown on the plot and positioned relative to the driver variables and the influence of the driver variables shown by relative length of grey arrow presented on the plot. The direction of an arrow relative to the axes does not imply a positive or negative direction of influence. In the example below, most species are ordinated away from redNdepo implying that the cover of these species is negatively affected by deposition of reduced N. These species are also correlated with each other, the vectors for *Cladonia portentosa* (cladport) and *Cladonia gracilis* (cladgrac) are adjacent to each other, most likely the species are found in the same sites. *Cladonia fimbriata* (cladfima) is the only species that appears positively associated to ‘redNdepo’, however, it also appears associated with ‘growdegr’ (growing degree days) suggesting that its relative cover is influenced both by reduced N deposition and temperature.

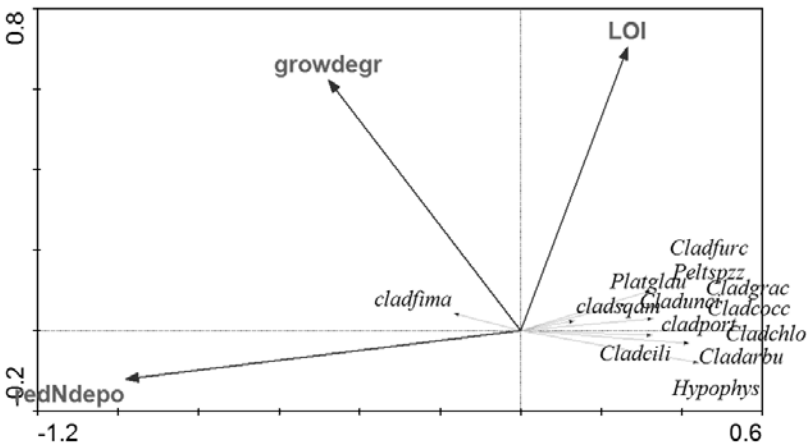


Figure 2: Example ordination plot

2.4.2 Stepwise and conventional regression

Regression techniques were used to model change in species richness with regard to an individual variable (simple regression) or a combination of variables (stepwise regression) that explain the most variation in the independent variable, in this case species richness. Stepwise regression is a form of multiple regression using a combination of forward and backward selection of variables. Variables are included if they explain significant variation in addition to those already in the model, and excluded if their removal does not increase the residual sum of squares. In both forms of regression a test for normality was performed and corrections made as necessary.

2.4.3 LOESS regression

LOESS (locally weighted scatterplot smoothing) regression is a form of non-parametric regression which acts as a ‘smoothing’ tool to aid visualisation of the response of taxa (individual species or functional groups) to a variable such as nitrogen pollution.

Linear regression techniques (including simple, multiple and stepwise) assume that the underlying relationship between dependent and independent variables is linear. LOESS regression enables a curve to be fitted to the data without making any assumptions about the form of the underlying relationship. In doing so it allows more flexibility than classical regression by fitting a smoothing function that varies with the data. However, simple mathematical equations describing the relationships cannot be generated with LOESS as with linear techniques.

PAST software version 2.06 (Hammer *et al.*, 2001) was used for this analysis.

2.4.4 Cluster analysis, sample grouping and ecological thresholds

An important question that is rarely explicitly addressed is whether the response of plant communities to nitrogen deposition is linear, or if there are ecological thresholds. If threshold responses do exist, this may have important implications for regulation of pollution loading, suggesting that there are points above which, or below which, further nitrogen deposition may have a disproportionate impact on the ecosystem. As detailed elsewhere in this report, gradient studies are now available for a large number of semi-natural habitats within the United Kingdom. Results from these studies show a reduction in species richness along the nitrogen pollution gradient and characteristic changes in plant communities.

Here we apply constrained cluster analyses to these datasets. This analysis attempted to identify non-linearities in the community response using a variety of statistical techniques originally developed for time-series data from bio-stratigraphy but theoretically equally applicable to changes along any gradient. These techniques are similar to conventional cluster analysis techniques but with the constraint that clusters be composed of samples with similar levels of nitrogen deposition.

In the context of this report this analysis has two important functions. Firstly it enables us to validate the results of the ordination analyses (discussed below) which show nitrogen pollution to be an important environmental control on the species composition of many habitats. If significantly different groupings can be identified solely on the basis of their nitrogen-loading this provides convincing evidence that nitrogen is an important control on community composition. Secondly, the location of 'break-points' between sample groupings is of interest because these may relate to ecological threshold responses. It is important to note that our approach is subtly different from direct identification of a threshold. If a threshold is abrupt the groups of samples on either side of a break-point will be distinctly different and are likely to be easily separated by clustering, if however the threshold is more gradual there may be variability in the group to which marginal samples are assigned.

We trial three methods derived from two contrasting approaches. We first test an agglomerative approach: with groups built by successively combining samples, as for many conventional cluster analysis techniques. Our approach is based on Ward's method (Ward, 1963) where clusters are built so as to minimize the increase in total within-cluster sum of squares. Conventional cluster analysis produces groups that are difficult to interpret in terms of a single environmental variable. To avoid this problem we introduce a constraint that clusters must be composed of samples with adjacent levels of nitrogen deposition. Essentially, we force the cluster analysis to produce groups which represent differing levels of nitrogen deposition. This method – constrained incremental sum of squares (CONISS) – is widely used for temporally-structured data (Grimm, 1987). CONISS produces a dendrogram, but only the first few splits are likely to be ecologically meaningful. Table 1 presents the first two. A limitation of this technique is that Ward's method has an inherent tendency to produce clusters of similar size (e.g. Morse, 1980).

The other two methods take a contrasting approach: instead of building up groups by successively adding samples in an agglomerative approach, they consider the whole dataset and the reduction in overall variance which may be achieved by the insertion of zone boundaries. As such the methodology is more focused on the sequence as a whole, unlike the agglomerative methodology which is more focused on the individual samples. We treat our samples as a transect along the gradient of total nitrogen deposition (as for CONISS), and test the validity of inserting splits in all alternative positions. Two variants of this divisive methodology are examined, with variance assessed by information content (SPLITINF) or least squares (SPLITLSQ; Gordon and Birks 1972, Birks and Gordon 1985). The SPLITINF and SPLITLSQ techniques are binary approaches that first split the overall dataset in two and then successively split these zones into smaller sub-divisions. As for CONISS, the first two divisions are presented in Table 17. We apply all three of these techniques using ZONE vers.1.2 (Juggins, 1992) with a squared Euclidean distance matrix.

Results of constrained agglomerative techniques can be presented as a dendrogram showing the relationships of samples along the gradient. An example of such a dendrogram is shown in Figure 3 below. The relationships of individual samples are shown by the proximity of their branches. Although such dendrograms present a large amount of information, typically only the first 'branches' are significant and useful. In the results of this analysis we only present the locations of the first two sample divisions.

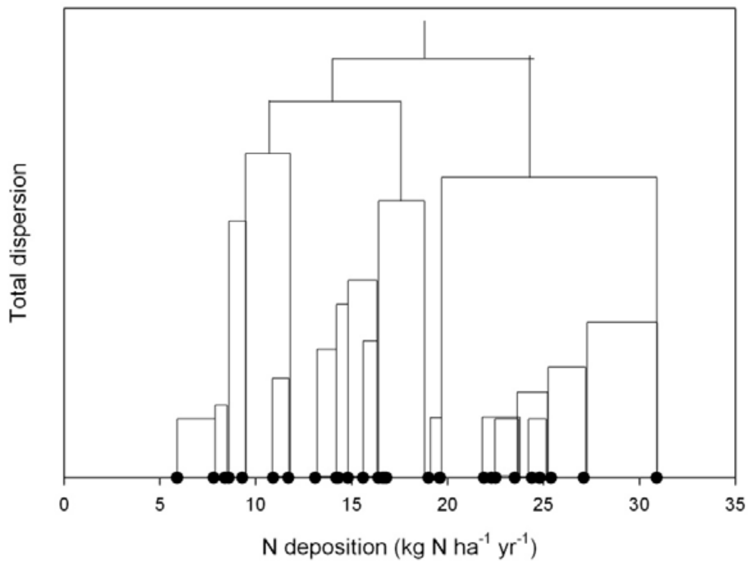


Figure 3: Example of a dendrogram showing the relationships of samples along a gradient

None of these three methods include a test of the validity of the clusters produced. To determine whether the community composition of the different clusters is significantly different, we apply a simple test of community similarity using ANOSIM. ANOSIM is a non-parametric test of similarity between pre-defined groups (Clarke, 1993). The test statistic (R_{ANOSIM}) has a value between -1 and +1 (although negative values are unusual); a value of 0 indicates the null hypothesis, that there is no difference between groups, while a value of 1 indicates that all samples within groups are more similar to one another than to any samples from different groups. Significance testing is achieved using permutation tests.

We applied ANOSIM with a Bray-Curtis distance measure and 10,000 permutations in PAST ver. 1.71 (Hammer *et al*., 2001). The ANOSIM results tell us whether the groups of samples are different, but give no assessment of whether those boundaries are in the optimum position (i.e. the probability of any random division producing a significant result in ANOSIM is relatively high). To give some assessment of the distinctness and validity of the cluster boundaries, we can compare the results of the three methods used. As these techniques rely on different underlying principles, if they identify similar cluster boundaries, this can give us confidence that the groupings are valid and useful. Additionally, where we have replication within an ecosystem type (acid grasslands and upland heaths) we can compare the results between the datasets.

Two general problems occur with all of these three approaches. Firstly there are issues with confounding environmental variables. In all of the datasets, many variables other than total nitrogen deposition affect plant communities, particularly climate. The response of the plant communities to these other variables is likely to complicate the identification of meaningful groups according to nitrogen deposition values. A second general problem is the size of the datasets and inconsistent sampling along the nitrogen gradient. With the exception of the larger Stevens *et al*. (2004, 68 sites) dataset the number of sites in each dataset is small (22-29 sites) and at the lower limit of the sample size for which many multivariate techniques are appropriate. Partly as a result of this limited sample size, the distribution of samples along the N deposition gradient is non-uniform, with gaps apparent in some datasets (for instance, no sites between 30.3 and 40.8 kg N ha^{-1} yr^{-1} in the TU-acid grasslands data, and no sites between 5.9 and 10.6 kg N ha^{-1} yr^{-1} in the TU-lowland heath data). Inconsistent sampling along the gradients both reduces the precision with which cluster boundaries can be located and increases the probability of false identification of a group boundary.

3. Tasks 1 and 2: Collation of scientific information and categorisation by habitat type

The bulk of the data used in this report is from a compilation of vegetation survey data from 8 field surveys encompassing vegetation quadrat data from a total of 226 UK sites surveyed between 2002 and 2009 (see Figure 4). The habitats are: dwarf shrub heathland (upland and lowland), acid grassland, bog and sand dune (fixed-dune grassland). Each represents a UK Biodiversity Action Plan (UK BAP) priority habitat for conservation (Natural England, 2011).

Figure 4: The survey locations of the 226 sites from which data has been used in the analysis for this project

The surveys comprise of a 2002-2003 acid grassland survey, a 2002 sand dune survey, a 2006 upland heath survey, a 2007 acid grassland survey and 2009 surveys of sand dune, bog, upland and lowland heaths. Where data were comparable between surveys, results were combined. Details of the surveys are provided in Table 3.

Within each survey, locations were carefully chosen to enable site comparisons to be made, ensuring vegetation structure between sites was consistent. Locations were identified along a UK nitrogen pollution gradient which was typically cleaner in the north and more polluted in the south. This UK pollution gradient also closely follows a climate gradient, with northern sites being cooler and wetter than their southern counterparts. For this reason an east-west gradient was also maximised within each habitat to provide survey locations that were 'clean and warm', 'polluted and cooler', 'wetter and polluted' and 'drier and less-polluted'. Such an approach aided statistical partitioning of the effects of pollutant and climatic drivers of vegetation change.

Further data were obtained from a network of UK nitrogen addition experiments funded as part of the Terrestrial Umbrella (TU) UKREATE project (http://ukreate.defra.gov.uk/) which include habitats similar to the surveys. Data from these experiments that support or challenge findings from the gradient surveys are presented and details of these experiments are shown, in Tables 5 and 6.

Published data from similar habitats to the survey data have also been drawn upon and reviewed as part of this report: these are summarised in Task 4 and Table 4.

Table 3: Survey data sets by habitat type under new analysis as part of this report

Habitats	New analysis here or Reviewed	NVC	Survey date	Location	Method of data collection/ number of sites	Author /affiliation	Reference
Acid grassland	New New	U4	2002/3 2007	GB	Quadrat survey/68 sites Quadrat survey/22 sites	C. Stevens, Open University/CEH BEGIN grassland survey	Stevens *et al*. (2004) Stevens *et al*. (2010)
Bogs	New	M19, M18	2009	GB	Quadrat survey/ 29 sites	TU consortium led by MMU	UKREATE, 2010 (TU report for 2007-2010)
Lowland heath	New	H2-H13	2009	GB	Quadrat survey/27 sites	TU consortium led by MMU	UKREATE, 2010 (TU report for 2007-2010)
Upland heath	New	H12	2009	GB	Quadrat survey/25 sites	TU consortium led by MMU	UKREATE, 2010 (TU report for 2007-2010)
Sand dunes	New	SD12, SD8 transitional	2009	GB	Quadrat survey/24 sites	TU consortium led by MMU	UKREATE, 2010 (TU report for 2007-2010)
Sand dunes	New	SD8, SD11, SD12 transitional	2002	GB	Quadrat survey/11 sites	L. Jones, CEH Bangor	Jones *et al*. (2004)
Upland heath	New	H12	2006	GB	Quadrat survey/20 sites	J. Carroll & S. Caporn, MMU	TU report, Caporn *et al*., (2007) JNCC report, Stevens *et al*., (2009)

Table 4: Published data by habitat type reviewed in this report

Habitats	New analysis here or Reviewed	Survey date	Location	Method of data collection	Author /affiliation	Reference
Acid grassland	Reviewed	2002-3	GB	Field survey	Stevens *et al.*	Stevens *et al*. (2004, 2006)
Acid grassland	Reviewed	2007	Europe inc. GB	Field survey	BEGIN consortium	Stevens *et al*. (2010)
Calcareous grassland	Reviewed		GB	Field survey	L. Van den Berg	Van den Berg *et al*. (2010)
Acid grassland **Calcareous grassland** **Mesotrophic grassland** **Heathland**	Reviewed	1998	GB	Field survey	CEH Countryside Survey	Maskell *et al*. (2010); JNCC report Stevens *et al*. (2009)
Acid grassland **Calcareous grassland** **Heathland** **Bogs**	Reviewed	Range	-	Collation of archived field survey	Stevens & CEH	Stevens *et al*. (2011)
Upland dry heath	Reviewed	2005	England, Wales	Field survey	J. Edmondson, MMU	Edmondson, *et al*. (2010)

Table 5: Name, location, vegetation type, soil type, and background atmospheric N deposition rates for the 9 TU sites

	Site name	Location in UK	Vegetation type: NVC classification	Soil type	Approx N dep. at site (kg N ha^{-1} yr^{-1})
Heath	Ruabon	North East Wales	Upland heath: H12 *Calluna –Vaccinium*	Peaty podzol	20 20
	Budworth	North west England	Lowland heath: H9 *Calluna –Deschampsia*	Humo ferric podzol	20
	Thursley	Southeast England	Lowland heath: H8 *Calluna*	Podsol, over lower greensand	10-15
	Culardoch	Northeast Scotland	Low Alpine Heath: H13 *Calluna-Cladonia*	Sub-alpine podsol	11
Bog	Whim	Southern Scotland	Ombrotrophic bog: M19, *Calluna-Eriophorum*	*Sphagnum* peat	8-10
Grassland	Pwllperian	Central Wales	Upland acid grassland U4	Shallow ferric stagnopodzol	25
	Wardlow	Central England	Acid grassland: U4e *Festuca-Agrostis-Galium*	Paleo-argillic	20-25
		Central England	Calcareous grassland: CG2d *Festuca –Avenula*	Rendzina	20-25
Sand dune	Newborough	Northwest Wales	Fixed sand dune grassland: SD8 *Festuca – Galium*	Rendzina	11

Table 6: Experimental site name and simulated N deposition treatments for the 9 TU sites. [1]= first experiment (treatments ceased in 1996 to follow recovery); [2]= ongoing experiment; [4] = plots where treatments are no longer ongoing. [5]=includes plots split in half with recovery since August 2005; [6]=includes plots split in half with recovery since spring 2003; [9]= a number of experiments have some plots where treatments have ceased in order to assess recovery.

	Site name	N treatment rates (kg N ha^{-1} yr^{-1})	N form (as solution unless stated)	Year started	Duration of N treatment years to date or until ceased[9]	Key references to experiment
Heath	Ruabon	0,40,80,120 0,10,20,40, 120	NH_4NO_3 solution	1989 1998	22 13[6]	Pilkington *et al*. (2005); Edmondson *et al*. (2010)
	Budworth	0,20,60,120	NH_4NO_3 solution	1996	115	Wilson (2003), Field (2010) Lageard *et al*. (2005)
	Thursley	0, 7.7, 15.4[1] 0, 30[2]	$(NH_4)_2SO_4$	1989-1996[1] 1998[2]	7[1] 12[2]	Power *et al*, 1998; Barker *et al*, 2004; Power *et al*, 2006 Barker, 2001, Green, 2005
	Culardoch	0, 10, 20, 50	NH_4NO_3	2000	11	Britton and Fisher, 2007
Bog	Whim	8,24,56 for wet dep. NH_3 transect 70-4	NH_4Cl $NaNO_3$ $NH_{3(g)}$	2002 2002	8 8	Sheppard *et al*. (2004). Sheppard *et al*. (2008)
Grassland	Pwllperian	10, 20	$NaNO_3$ NH_4SO_4	1996	12	Emmett *et al*. (2007)
	Wardlow	35, 70, 140 35, 140	NH_4NO_3	1990 1995	12[4] 15[5]	Morecroft *et al*. (1994) Horswill *et al*.(2008)
	Wardlow	35, 70, 140 35, 140	NH_4NO_3	1990[4] 1995[5]	12[4] 15[5]	Morecroft *et al*. (1994) Horswill *et al*.(2008)
Sand Dune	Newborough	7.5, 15	NH_4NO_3	2003	7	Plassmann *et al*. (2009)

4. Task 3: Identify the relevant response variables for each habitat

4.1 Introduction

This section of the report uses stepwise regression and ordination analysis to identify the key potential response variables in the survey datasets for each of the habitats studied. Stepwise regression considers overall changes in species richness and, where the data exist, by functional group. Ordination analysis considers changes in the composition of the vegetation community. Both analyses measure these changes relative to climatic and pollutant driver data.

Key potential response variables are also identified from published literature and from the Terrestrial Umbrella experiments.

4.2 Vegetation species richness responses to nitrogen pollution

Across all the datasets studied, increasing nitrogen deposition (total, reduced or oxidised) was correlated with reductions in species richness. This pattern was similar across all habitats. In many cases, climate was also correlated with species richness, with increasing species richness being a function of increasing precipitation and decreasing temperature (expressed as growing degree days or extreme temperature range). An exception to the latter was sand dunes, with pH ≥ 6.5 where increasing temperature was correlated with an increase in overall species richness. The output from the stepwise regressions are summarised in Table 7 and the relationship between nitrogen deposition and species richness presented in more detail in Task 4.

Consistency in survey methods and data collection in habitats visited as part of the TU 2009 survey enabled a direct comparison across all habitats (upland and lowland heath, sand dune, bog and acid grasslands – the latter representing a subset of 23 of the sites visited by Stevens *et al*. (2004)). For this cross habitat comparison, species richness was converted and expressed as a percentage of the maximum number of species recorded in that habitat. From this stepwise regression, nitrogen deposition explained most of the reduction in species richness (expressed as either total nitrogen deposition or dry-oxidised nitrogen deposition), followed by mean annual temperature.

Many plant groups were negatively associated with N pollution: within bogs the relationship was strongest in forbs including *Drosera rotundifolia* and *Narthcium ossifragum* and lichens; in upland heaths mosses and lichens reduced in diversity, although the change in the former was more significantly correlated with sulphur deposition; and in acid grasslands forbs showed a reduction in both richness and diversity (as previously reported in Stevens *et al*., 2006). For lowland heaths, wet-oxidised N deposition was significantly correlated with reductions in overall species richness however, climate explained more of the variation in species richness across the plant groups. This probably reflects the shift in lowland heath soil types as their geographical location changed from acid, base-poor, sandy soils of the Cornish heaths to the moister, more organic soils of the northern lowland heaths. Interestingly, across the ericaceous habitats which are often defined by a competitive balance between shrub, graminoid and moss species groups, graminoid species richness also fell as a function of rising N deposition. However, graminoid cover increased, suggesting a shift toward dominance by fewer species.

In sand dunes, moss species richness showed a strong reduction with increasing N deposition. Forb species richness was more weakly correlated with N pollution. However, when sand dune type was split by pH these responses were only seen in more calcareous sand dunes with pH ≥ 6.5, although limited data were available from sites with pH less than 6.5. In general, sand dune species richness was more strongly correlated to pH and the extent of decalcification. Some responses were, however, seen with soil N indicators in sand dunes (not shown in this report) such as N% and mineralisation at sites with pH lower than 6.5; this could indicate a longer-term response to N deposition.

The different forms of N pollution were also related to responses in species richness. However, it is difficult using modelled data to attribute change to a specific form of N pollution, and specific locations may be more vulnerable to either reduced or oxidised N dependent upon their proximity to a point source. In some cases, for example upland heath moss species richness and bog species richness, sulphur deposition was more strongly correlated with the species richness. It is difficult to be certain if these relationships are ecologically significant as sulphur levels are low across the range of the dataset, or indicative of a legacy effect from earlier years of high sulphur deposition. In general, significant correlations between pollutants exist and in both of these specific cases a form of N was also strongly correlated with the response variable and at levels more likely to elicit an ecological response.

Table 7: Summary of changes in overall species richness and the response of different functional groups (richness and cover where appropriate) using stepwise regression

Habitat /Survey	**Response variable**	**Best fit model parameters from stepwise regression and influence of an increase in the parameter on the response variable (↑↓)**	**Variance explained by model and statistical significance**
All habitats from TU 2009 survey	Overall species richness (% of maximum species recorded in each habitat)	Dry-oxidised nitrogen deposition (↓) Growing degree days (↓)	R^2=0.37, P<0.001
Upland heathland (TU 2009)	Overall species richness (total number of species recorded)	Reduced nitrogen deposition (↓)	R^2=0.39, P=0.002
	Moss species richness	Sulphur deposition (↓) (Wet-oxidised Nitrogen deposition also significant : ↓)	R^2=0.25, P=0.011 (R^2=0.21, P=0.021)
	Lichen species richness	Reduced nitrogen deposition (↓)	R^2=0.26, P<0.01
	Graminoid species richness	Dry-reduced nitrogen deposition (↓) Altitude (↓)	R^2=0.46, P<0.001
	Graminoid cover (%)	Dry-reduced nitrogen deposition (↑)	R^2=0.24, P=0.014
Upland heathland (MRS)	Overall species richness (total number of species recorded)	Dry-reduced nitrogen deposition (↓) Altitude (↑) Temperature extreme range (↑)	R^2=0.87, P=0.001
Lowland heathland (TU 2009)	Overall species richness (total number of species recorded)	Growing degree days (↓) Altitude (↓) Wet-oxidised nitrogen deposition (↓)	R^2=0.64, P<0.001
	Moss species richness	Growing degree days (↓) pH (↑)	R^2=0.42, P=0.005
	Lichen species richness	no combination of variables explain significant variation in the data	-
	Graminoid species richness	Growing degree days (↓)	R^2=0.46, P<0.001
	Graminoid cover (%)	Dry-reduced nitrogen deposition (↑)	R^2=0.35, P=0.001

Habitat /Survey	Response variable	Best fit model parameters from stepwise regression and influence of an increase in the parameter on the response variable (↑↓)	Variance explained by model and statistical significance
Bog (TU 2009)	Overall species richness (total number of species recorded)	Dry-sulphur deposition (↓) (Dry-oxidised Nitrogen deposition also significant : ↓)	R^2=0.56, P=0.01 (R^2=0.50, P=0.01)
	Moss species richness	no combination of variables explain significant variation in the data	-
	Lichen species richness	Dry-oxidised nitrogen deposition (↓)	R^2=0.37, P<0.01
	Forb species richness	Total acid deposition (↓) (Nitrogen deposition also significant : ↓)	R^2=0.39, P=0.002 (R^2=0.38, P=0.002)
	Graminoid cover (%)	Wet-reduced nitrogen deposition (↑) Growing degree days (↑)	R^2=0.68, P<0.001
Sand dunes TU 2009 (all sites)	Overall species richness (total number of species recorded)	pH (↑) Wet-oxidised nitrogen deposition (↓)	R^2=0.57, P<0.005
	Moss species richness	oxidised nitrogen deposition (↓) LOI (↑)	R^2=0.67, P<0.001
	Forb species richness	pH (↑) Wet-oxidised nitrogen deposition (↓) Wet-sulphur deposition (↓)	R^2=0.53, P<0.001
Sand dunes pH <6.5 (TU 2009)	Overall species richness (total number of species recorded)	no significant relationship with N	-
	Moss species richness	wet-sulphur deposition (↓)	
Sand dunes pH ≥6.5 (TU 2009)	Overall species richness (total number of species recorded)	Oxidised nitrogen deposition (↓) Ca + Mg deposition (↑) Growing degree days (↑)	R^2=0.76, P<0.001
	Moss species richness	Oxidised nitrogen deposition (↓)	R^2=0.62, P<0.001
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)	Overall species richness (total number of species recorded)	pH (↑) Nitrogen deposition (↓)	R^2=0.55, P<0.001
	Moss species richness	Total acid deposition (↓)*	R^2=0.31, P=0.001
Acid grasslands (BEGIN UK)	Overall species richness (total number of species recorded)	Nitrogen deposition (↓) Precipitation (↑)	R^2=0.38, P=0.001
	Forb species richness	Nitrogen deposition (↓)	R^2=0.48, P<0.001

*Total acid deposition incorporates both nitrogen and sulphur deposition as a total acid equivalent

Bryophyte species richness reduced in lowland heaths, sand dunes and the upland heath MRS survey – the latter included liverworts whereas the TU upland heath survey did not. Lichen species richness reduced in bogs and upland heaths and forb species richness reduced across acid grasslands, bogs and sand dunes. Graminoid species richness reduced in both heathland types whilst graminoid cover increased in all ecosystems except acid grasslands. However, within sand dunes, whilst this increase was significant, graminoid cover was more strongly associated with soil pH which could reflect either an interaction with precipitation and decalcification or acidification caused by pollutant deposition.

4.3 Vegetation community composition responses to nitrogen pollution

Table 8 presents the results from the ordination analysis. Vegetation community composition was also related to nitrogen (N), with a form N of nitrogen the first variable through the forward selection process in all habitats except sand dunes. In sand dunes, change was more strongly associated with climate (either growing degree days or precipitation) and the effect of rainfall on leaching of base cations, decalcification and acidification. Climate explained significant additional variation in the all the habitats after N pollution. In bogs, hydrology (typically influenced by management and drainage rather than rainfall) also was significantly related to change in species composition. Similarly, in upland heaths the amount of soil organic matter (LOI – loss on ignition) was related to community composition as was pH in lowland heaths: both these responses are indicative of changes in the soil from organic and peaty to calcareous and, at a broad-scale, driven by rainfall and climate.

The percentage of variance explained by these models is often low. Many factors drive variation within different habitats other than the ones chosen for this analysis and there is considerable heterogeneity between sites and also within a site. The fact that the relatively small number of environmental drivers explains as much variance as they do is testament to their strength as drivers of change in species composition.

Table 8: Summary of the analysis of species community composition using ordination (RDA) in CANOCO. ns = not significant

Habitat /Survey	Statistically significant drivers of change in species composition	Variance explained by model and statistical significance	Variance partitioning by driver*	Specific species showing a strong response to Nitrogen with good distribution across dataset (direction ↑↓)
Upland heathland (TU 2009)	Reduced nitrogen deposition	36.3%	15.8% P=0.001	*Cladonia fimbriata* (↑) *Cladonia portentosa* (↓) *Deschampsia flexuosa* (↑) *Brachythecium rutabulum* (↑) *Hylocomium splendens* (↓)
	Growing degree days		7.9% P=0.006	
	Loss on ignition		7.4% P=0.01	
Upland heathland (MRS 2006)	Dry-oxidised nitrogen deposition	87.8%	37.7% P=0.001	*Campylopus introflexus* (↑) *Hylocomium splendens* (↓)
	Reduced nitrogen deposition		22.1% P=0.001	
	Consecutive dry days		15.1% P=0.02	
	Habitat		12.9% P=0.033	
Lowland heathland (TU 2009)	Dry-oxidised nitrogen deposition	32%	8.2% P=0.005	*Cladonia fimbriata* (↑) *Cladonia portentosa* (↓) *Brachythecium rutabulum* (↑) *Campylopus introflexus* (↑) *Hylocomium splendens* (↓)
	Growing degree days		13.3% P=0.001	
	Soil pH		6.5% P=0.019	

Habitat /Survey	Statistically significant drivers of change in species composition	Variance explained by model and statistical significance	Variance partitioning by driver*	Specific species showing a strong response to Nitrogen with good distribution across dataset (direction ↑↓)
Bog (TU 2009)	Dry-reduced nitrogen deposition Hydrological index Dry-oxidised nitrogen deposition	22.6%	7.0% P=0.004 8.3% P=0.001 5.9% P=0.011	*Eriophorum vaginatum* (↑) *Sphagnum fimbriatum* (↑) *Cladonia portentosa* (↓)
Sand dunes TU 2009 (all sites)	Growing degree days Precipitation Dry-reduced nitrogen deposition pH	36.6%	5.6% P=0.03 10.6% P=0.001 5.1% P=0.035 14.6% P=0.001	*Hylocomium splendens* (↓) *Ammophila arenaria* (↓)
Sand dunes pH <6.5 (TU 2009)	Growing degree days Dry-oxidised nitrogen deposition	38.6%	*18.8% P=0.035* *22.0% P=0.008*	*Hylocomium splendens* (↓)
Sand dunes pH ≥6.5 (TU 2009)	Precipitation Dry-reduced nitrogen deposition	29.3%	*17.1% P=0.001* *9.5% P=0.025*	*Hylocomium splendens* (↓)
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)	Precipitation Ca+Mg deposition Dry-sulphur deposition Dry-oxidised nitrogen deposition pH	28.1%	*4.9% P=0.06* *3.2% ns* *3% ns* *3% ns* *8.2% P=0.006*	*Hylocomium splendens* (↓) *Carex arenaria* (↑ but ns). The relationship is significant when the 2002 data is analysed independently.
Acid grasslands (BEGIN UK)	Nitrogen deposition Growing degree days Precipitation Ca+Mg deposition	15.5%	*3.1% P=0.002* *3.8% P=0.001* *2.8% P=0.001* *2.1% P=0.02*	*Deschampsia flexuosa* (↑) *Hypnum cupressiforme (agg.)* (↑) *Nardus stricta* (↑) *Carex panicea* (↑) *Euphrasia officianlis* (↓) *Hylocomium splendens* (↓) *Lotus corniculatus* (↓)

*the sum of the variance explained by individual drivers will not always equal the total variance explained due to the use of covariables in the variance partitioning process

The species richness relationships detailed in the previous section were also largely reflected in community composition, for example, see Figure 5 which illustrates the response of sensitive lichen species in the upland heath habitat to N deposition: in this case reduced N deposition was the most strongly correlated variable (N.B. all species were included in the ordination but only lichen species are shown on the plot, for other ordination plots refer to appendix 1). The ordination process also suggested individual species that appeared strongly associated, either positively or negatively, with N and specific species for each habitat are suggested in Table 8. For example, in Figure 5, *Cladonia fimbriata* (cladfima) is the only species positively associated with N deposition while other lichen species are associated with low N conditions.

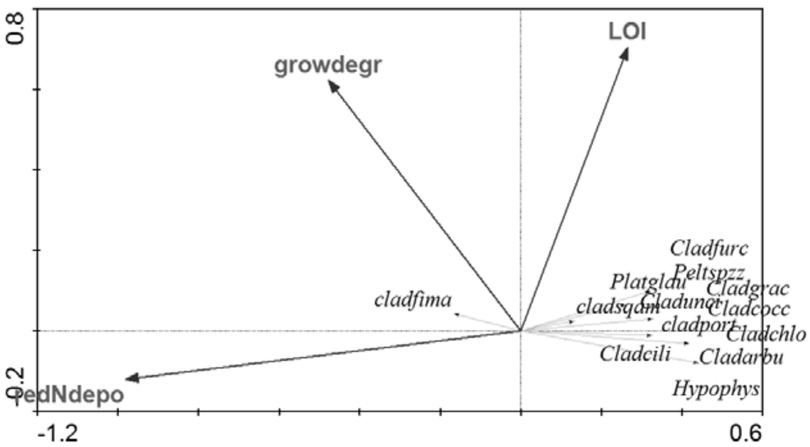


Figure 5: Ordination plot produced from RDA of upland heath data (TU 2009), lichen species only shown

In the example ordination plot above, many lichen species are ordinated away from N. This reflects reductions in lichen species richness as a function of N deposition, however, few lichen species are represented consistently across the dataset to enable a relationship with N to be examined with any statistical confidence. *Cladonia fimbriata* and *Cladonia portentosa* are two widely-represented lichen species, and these relationships alongside the other species shown in Table 8 are examined in greater detail in Task 4.

Typically, the strongest species related to N within the survey were mosses and lichens, showing both positive and negative relationships to N. Across the heathland, acid grassland, and sand dune datasets, the moss *Hylocomium splendens* showed a consistent negative response to N. The MRS survey of upland heaths also recorded presence of liverworts and these too were strongly sensitive to pollution. Within upland heath and bog habitats the graminoid species *Deschampsia flexuosa* and *Eriophorum vaginatum* showed positive responses to N deposition. In the 2009 sand dune survey, the grass *Ammophila arenaria* (marram grass) decreased in cover with increasing N deposition, however, this is at odds with findings from the 2002 survey where an increase in *A. arenaria* cover was found (Jones *et al*., 2004). *Carex arenaria* showed an increase with N, although this was non-signficant. This difference in response may be related to the differing types of sand dune surveyed between the surveys, since the 2009 survey focused on older de-calcified habitats where *Ammophila* persists only at low cover with low vigour, and is termed 'relict *Ammophila'*. Within the acid grassland habitat, forb species were strongly negatively correlated to N deposition, most notably *Lotus corniculatus* and *Euphrasia officianlis*.

Moss, lichen and forb species are an important component of the biodiversity within all the habitats studied, and key to maintaining high species richness and favourable habitat condition. Within the nature conservation agencies Common Standards Monitoring is a tool used to assess the condition of the habitat or feature. In heathlands, bryophytes and lichens play an important role in the overall habitat and are indicators of favourable condition in Common Standards Monitoring (CSM) (JNCC, 2006). The balance between graminoid and shrub cover is important in maintaining the intrinsic diversity in ericaceous habitats. In some cases, high graminoid cover may be detrimental to site integrity, for example, *Deschampsia flexuosa* cover above 25% in lowland heaths (JNCC, 2004). In two of the lowland heaths surveyed graminoid cover exceeded 30% and in several upland heaths graminoid cover was above 25 % suggesting that N deposition posed a long-term threat to site integrity.

4.4 Key response variables in the reviewed literature

Numerous vegetation categories have been described in the literature, such as individual species, botanical groups (grasses, bryophytes etc), functional groups (e.g. Ellenberg score) and plant characteristics (e.g. canopy height). The main ones relevant to N pollution response are listed in Table 9, while their relationships with nitrogen deposition are discussed under Task 4.

Table 9: Response variables frequently reported in the literature on vegetation and N deposition

Habitats	Reported response variable	Reference
Upland dry heath	Bryophytes richness	Edmondson *et al*. (2010)
Upland dry heath	Bryophytes richness, total spp. richness, individual species	Stevens *et al*. (2009) - pooled data of Edmondson *et al*. (2010) and Payne *et al*. 2014)
Acid grassland	Forbs, grass, bryophyte richness and cover, grass/forb ratio Individual species	Stevens *et al*. (2004, 2006, 2009)
Acid grassland (includes European sites)	Forbs, grass, bryophyte richness	Stevens *et al*. (2010)
Calcareous grassland	Species richness & diversity Functional groups species richness, Ellenberg N & R, individual species including (rare & scarce) species	Van den Berg *et al*. (2010)
Acid grassland **Calcareous grassland** **Mesotrophic grassland** **Heathland**	Vascular, bryophyte species richness	Maskell *et al*. (2010)
Acid grassland	Acid preference index Ellenberg N & R Competitive, stress tolerant, ruderal strategy	Stevens *et al*. (2010)
Acid grassland **Calcareous grassland** **Heathland** **Bogs**	Ellenberg N & R, canopy height, specific leaf area, species richness, individual species occurrence	Stevens *et al*. (2011)

4.5 Key response variables in the experimental site data

In the Terrestrial Umbrella (TU) experiments, vegetation and soils have been subjected to detailed study over many years, and several variables, both ecological and biogeochemical, were found to respond to additions of nitrogen. In relation to this report, the key response variables of interest considered were: changes in presence and abundance of individual species, botanical groups (vascular plants, bryophytes and lichens); changes in visible plant injury due to stress (winter damage, heather beetle); and changes in plant and soil chemistry (with potential consequences for nutrient imbalance, soil leaching and pH).

4.6 Conclusion and summary of response variables taken forwards to task 4

The results from the survey datasets strongly support the findings from the literature review. Increasing nitrogen deposition is strongly associated with both detrimental changes in species composition and reductions in species richness. In some cases a specific form of N was more strongly associated with a response however, to enable comparison with critical loads, total N deposition is used in further analysis. This will not affect the overall relationship as modelled total nitrogen deposition was strongly correlated with both modelled reduced and oxidised N deposition over the data used (R^2=0.89 and 0.72 respectively, both $P<0.01$). The nitrogen addition experiments provide data to support the hypothesis that nitrogen pollution, in the absence of change in other environmental variables, has a direct adverse effect on community composition in many different types of vegetation. Important changes in habitats were seen especially regarding the abundance of sensitive species and some of these are described under Task 4.

Table 10 below summarises the response variables that are strongly associated with N deposition for each habitat, these are further analysed in Task 4. The consistency shown between the new research discussed here and the published data is remarkable and reflects the strength of N as a driver of change within the UK's semi-natural ecosystems. The responses to N that are found are in addition to those explained by a climatic gradient and, in many instances, N deposition is statistically the strongest driver of change.

Table 10: Summary of the strongest response variables found during the statistical analysis that will be carried forward to Task 4. '#' indicates found within vegetation datasets analysed as part of this report, '$' indicates found within the literature reviewed as part of this report.

Response variable	Acid grassland	Bog	Upland heath	Lowland heath	Sand dune
Species composition	#$	#$	#$	#$	#
Total species richness	#	#$	#$	#$	#
Bryophyte species richness	$		#$	#	#
Lichen species richness		#	#		
Forb species richness	#$	#			#
Graminoid species richness			#	#	
Graminoid cover	$	#	#	#	#

In some cases, for example lowland heath mosses or lichens, N deposition did not emerge from the stepwise regression as a potentially significant driver of change in species richness. However, in these cases N deposition was associated with changes in species composition in the ordination analysis, with individual moss, lichen, forb and graminoid species strongly associated with changes in a form of N deposition.

Changes in certain individual species were related to changes in N deposition and these are summarised in Table 11 overleaf. Many more species appeared to show some relationship to N but, low frequency in the dataset meant that this was not significant. However, their response does contribute to changes in overall species richness and the species richness of functional groups.

Table 11: Summary of individual species that showed a strong response to N in the ordination analysis. The nature of these relationships with N deposition is examined further under task 4.

Habitat	Species with strong response (direction of response)
Upland heath (TU & MRS)	*Cladonia fimbriata* (↑)
	Cladonia portentosa (↓)
	Deschampsia flexuosa (↑)
	Brachythecium rutabulum (↑)
	Hylocomium splendens (↓)
	Campylopus introflexus (↑)
Lowland heath	*Cladonia fimbriata* (↑)
	Cladonia portentosa (↓)
	Brachythecium rutabulum (↑)
	Campylopus introflexus (↑)
	Hylocomium splendens (↓)
Acid grassland	*Deschampsia flexuosa* (↑)
	Hypnum cupressiforme (agg.)(↑)
	Nardus stricta (↑)
	Carex panicea (↑)
	Euphrasia officianlis (↓)
	Hylocomium splendens (↓)
	Lotus corniculatus (↓)
Bog	*Eriophorum vaginatum* (↑)
	Sphagnum fimbriatum (↑)
	Cladonia portentosa (↓)
Sand dune	*Hylocomium splendens* (↓)
	Ammophila arenaria (↓)

5. Task 4: Determine the relationship between N deposition and the key response variables

5.1 Introduction

The relationships between N deposition and the key response variables determined in Task 3 are examined in more detail here. This task presents results over 6 main sections: 1) those from the gradient surveys analysed in this report; 2) supporting evidence of change from the dose response experiments; 3) evidence from the literature; 4) a review of the relationships between N and the response variables found in the JNCC collation report (Stevens *et al*., 2011); 5) the use of cluster analysis to determine if any relationship exists between species composition and nitrogen deposition in the survey datasets and the possible presence of threshold responses and 6) Concluding comments.

The relationships examined in this chapter use modelled annual N deposition as the pollutant variables, however, the relationships studied between N deposition and species richness and presence have developed over many years. The current rate of N deposition is primarily a proxy for long-term cumulative N deposition. Thus we would not expect that a change in N deposition, either increasing or decreasing, would immediately change species richness or composition, however, long-term changes in N deposition are likely to affect species richness and composition. Furthermore, different species groups will respond in different ways: bryophytes and lichens with no root structure are likely to respond quicker than vascular plants and the responses of both may be affected by management interaction which could alter inter-species composition. Refer to the methods section for further information.

5.2 Relationships determined in the gradient surveys

The nature and strength of the relationships between N deposition and total species richness, functional group richness and plant cover, where significant, are summarised in Table 12 and the relationships between N and individual species cover or presence are presented in Table 13. Regression curves for each of these relationships are provided in Appendix 2 for total and functional group species richness and Appendix 3 for the individual species responses, however, the broad relationship between N and the percentage of the maximum number of species within habitats with comparable survey techniques (TU 2009 upland heath, lowland heath, bog, sand dune and subset of BEGIN/Stevens *et al*. (2004) grasslands) is shown in Figure 6.

Across the 135 survey sites represented in this plot, a highly significant pattern of species richness reduction as a function of increasing atmospheric nitrogen deposition is apparent with a wedge shaped response. The pattern indicates that at low N deposition the species number can be both high and low, but at high N deposition the species number is always low. The large scatter in the data is related to the variation between- as well as within- habitats. Within this overall dataset, a negative-linear relationship best describes the response, however, within each habitat and functional group a negative, curvi-linear relationship is more common, indicating a more rapid loss of species associated with increasing N deposition at

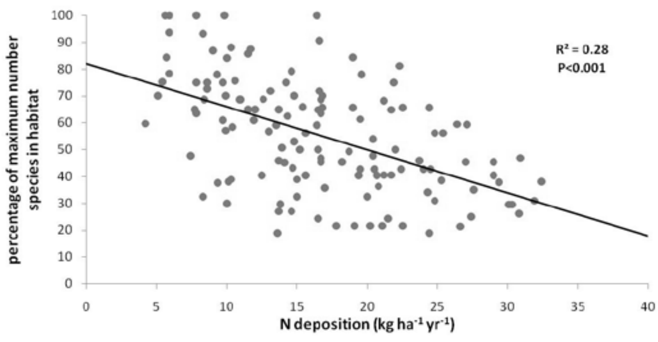


Figure 6: Change in % of maximum species richness as N deposition increases at 135 sites studied in the TU 2009 multi-habitat survey

lower levels of N pollution. Species richness curves from the TU sand dune (all pH) and TU upland heath surveys and are shown in Figure 7 below, and a complete set of response curves provided in Appendix 2. A curvi-linear response suggests that less-polluted sites are more sensitive to increases in N deposition and that at sites already receiving high levels of pollution, much species diversity has already been lost. In all habitats, the magnitude of the response is large, with 50-75% fewer species in the least diverse sites within each habitat, compared with the most diverse.

Loss of species richness related to increases in N deposition is consistent across all the habitats and functional groups, with the exception of sand dunes with a pH less of than 6.5, where limited data and strong climatic effects occurred. Whilst graminoid species richness also declined, graminoid cover increased, and this relationship was also curvilinear, indicating more rapidly increasing cover of fewer graminoid species as N deposition increased. In this respect, the potential for adverse change in each habitat increased at locations with a higher background N deposition.

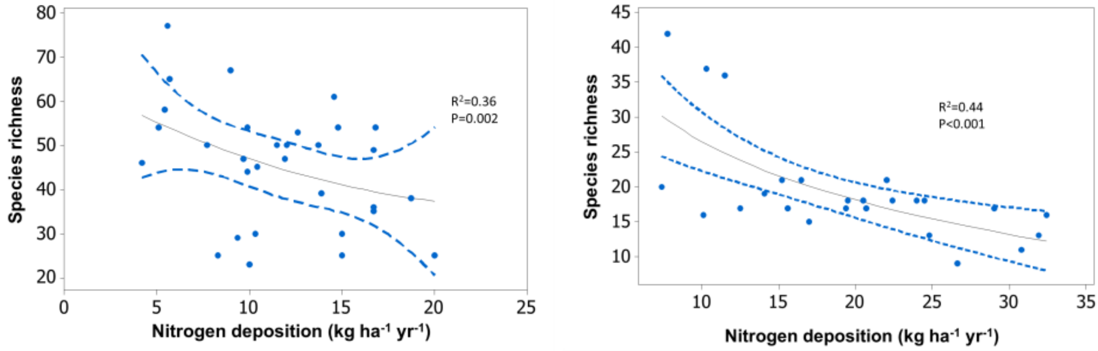


Figure 7: Measured species richness within a) TU 2009 survey sand dunes (all pH) and b) TU survey upland heaths as nitrogen deposition increases

To further examine the range over which the response variables from Task 3 show the most rapid change, each variable was analysed using LOESS regression. Summary data from these analyses are presented in Tables 12 and 13 and example regression curves for TU Sand dune (all pH) and TU Upland heath surveys are shown in Figure 8.

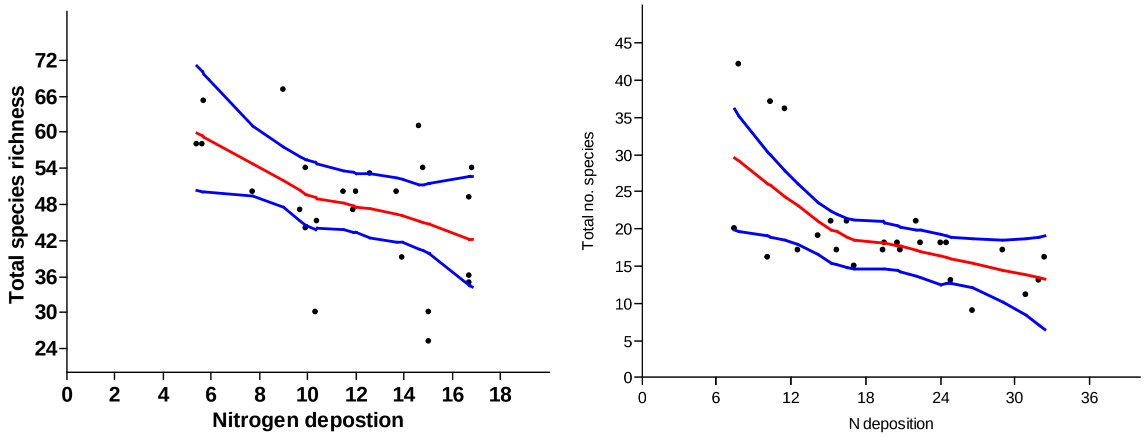


Figure 8: LOESS regression curves showing change in species richness as nitrogen deposition increases for a) TU 2009 survey sand dunes (all pH) and b) TU survey upland heaths. Best fit to data line in red, 95% confidence limits shown in blue and fitted by bootstrapping.

Although data in most habitats is limited to around 25 sites, clear response points were usually found, and these mostly supported the curvilinear relationships from the linear regressions. In many cases the inflection was at an N deposition level typically between 17 and 22 kg N ha^{-1} yr^{-1} for many negative and positive responses to N. The general exception to this was *Cladonia fimbriata* cover which showed a ‘humpback’ uni-modal response in upland (16-25 kg N ha^{-1} yr^{-1}) and lowland heaths (17-28 kg N ha^{-1} yr^{-1}).

Table 12: Summary of relationship type and direction between modelled nitrogen deposition (kg ha^{-1} yr^{-1}) and species richness/cover for each habitat. Relationship equations shown: y=species richness; x=nitrogen deposition (kg ha^{-1} yr^{-1}). LOESS regression range highlights the range over which the species response is the most responsive, a dash indicates no tipping point was apparent.

Habitat/ Survey	Response variable	Direction of response to N	Relationship type	Statistical significance	Relationship equation	LOESS regression range of max. loss or gain
All habitats (from TU 2009)						
Total species richness		negative	linear	R^2=0.28 P<0.001	y=1.56*x + 82.9	n/a
Upland heathland (TU 2009)						
Total species richness		negative	mild curvilinear	R^2=0.44 P<0.001	y=54.37 - 12.11*ln(x)	7-17 kg N
Moss species richness		negative	curvilinear	ns	-	-
Lichen species richness		negative	linear	R^2=0.23 P=0.015	y=11.34 – 3.18*ln(x)	7-16 kg N
Graminoid species richness		negative	mild curvilinear	R^2=0.27 P<0.01	y=9.62-2.28*ln(x)	7-22 kg N
Graminoid cover		positive	mild curvilinear	R^2=0.31 P=0.017	y=0.042x^2 - 0.88x + 9.19	>22 kg N
Upland heathland (MRS)						
Total species richness		negative	mild curvilinear	R^2=0.61 P<0.001	y = 0.011*x^2 - 0.709*x + 19.8	20 kg N
Lowland heathland (TU 2009)						
Total species richness		negative	mild curvilinear	R^2=0.32 P=0.002	y=-11.25*ln(x) + 47.27	< 17 kg N
Moss species richness		negative	mild curvilinear	R^2=0.15 P<0.05	y = -3.29*ln(x) + 14.685	-
Graminoid species richness		negative	mild curvilinear	R^2=0.26 P<0.01	y=0.16+38.99/x	-
Graminoid cover		positive	mild curvilinear	R^2=0.25 P<0.05	y=8.45-1.15*x+0.05x^2	> 23 kg N
Bog (TU 2009)						
Total species richness		negative	linear	R^2=0.23 P=0.009	y=27.9 - 0.30*x	> 19 kg N
Lichen species richness		negative	linear	R^2=0.19 P=0.018	y= 4.79 - 0.13*x	-
Forb species richness		negative	mild curvilinear	R^2=0.46 P<0.001	y = 8.07 - 2.33*ln(x)	-
Graminoid cover		linear	linear	R^2=0.27 P=0.004	y=1.35*x + 17.4	-
Sand dunes TU 2009 (all sites)						
Total species richness		negative	mild curvilinear	R^2=0.36 P=0.002	y=30.4 + 194.6/x	< 10 kg N
Moss species richness		negative	strong curvilinear	R^2=0.81 P<0.001	y= -1.3 + 84.4/x	< 12 kg N
Graminoid cover		positive	mild curvilinear	R^2=0.17 P<0.05	y=75.3 - 214.8/x	< 10 kg N
Forb species richness		negative	mild curvilinear	R^2=0.17 P<0.05	y=12.8 + 84.1/x	-
Sand dunes TU 2009 (pH <6.5)						
Total species richness		too few data points	-	-	-	-

Habitat/ Survey	Response variable	Direction of response to N	Relationship type	Statistical significance	Relationship equation	LOESS regression range of max. loss or gain
Sand dunes TU 2009 (pH ≥6.5)						
Total species richness		negative	mild curvilinear	R^2=0.42 P=0.009	y=94.1-16.8*ln(x)	-
Moss species richness		negative	mild curvilinear	R^2=0.85 P<0.001	y= -1. + 86.1/x	
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)						
Total species richness		negative	mild curvilinear	R^2=0.27 P<0.001	y=104.3-22.6*ln(x)	-
Moss species richness		negative	mild curvilinear	R^2=0.26 P=0.002	y=22.9-6.98*ln(x)	
Acid grasslands (BEGIN)						
Total species richness		negative	mild curvilinear	R^2=0.29 P<0.001	y = 0.0052x^2 - 0.68*x + 34.6	-
Forb species richness		negative	linear	R^2=0.48 P<0.001	Y= 11.8 - 0.35*x	-

Some of the individual species also revealed an apparent threshold level above or below which the response was strong. Most notable were the rapid reduction in the probability of presence (number of quadrats in which a species was found) of *Hylocomium splendens* in both upland heath surveys and the sand dunes at N deposition above 20 kg N ha^{-1} y^{-1} and the rapid increase in *Brachythecium rutabulum* at a similar point. *H. splendens* is thought to be sensitive to N however, its absence should not lead to the assumption that a site is negatively affected by N as the moss generally only occurs at less-polluted sites that are also moist. Similarly, *B. rutabulum* exhibits a preference for moister sites. It is therefore important that other factors be considered when making judgement of a site's N-status by the presence or absence of a single species.

Table 13: Summary of key relationships between nitrogen deposition and the individual species in each habitat identified in Task 3. Type, direction, statistical significance and equation of curve shown.

Habitat/ Survey	Response variable	Direction of response to N	Relationship type	Statistical significance	Relationship equation
Upland heathland (TU 2009)					
	Hylocomium splendens cover (%)	negative	mild curvilinear	R^2=0.16 P<0.05	y = 0.038x^2 - 2.37*x + 36.44
	Hylocomium splendens probability of presence	negative	threshold	ns	-
	Cladonia portentosa cover	negative	curvilinear	ns	-
	Deschampsia flexuosa cover	positive	mild curvilinear	R^2=0.30 P=0.018	y = 0.04x^2 - 0.75*x + 6.52
	Cladonia fimbriata cover	positive	curvilinear	ns	-
	Brachythecium rutabulum cover	positive	curvilinear	ns	-
	Brachythecium rutabulum probability of presence	positive	threshold	ns	-
Upland heathland (MRS)					
	Hylocomium splendens presence (%)	negative	threshold	R^2=0.65 P<0.001	y= -1.33 +39.74/x
	Campylopus introflexus presence (%)	positive	threshold	R^2=0.39 P=0.01	y = 0.003x^2 – 0.05*x + 0.23

Habitat/ Survey	Response variable	Direction of response to N	Relationship type	Statistical significance	Relationship equation
Lowland heathland (TU 2009)					
	Hylocomium splendens cover (%)	negative	curvilinear	R^2=0.44, P<0.001	y=6.21+ 119.51/x
	Hylocomium splendens probability of presence	negative	curvilinear	R^2=0.35, P<0.001	y=6.69-2.14 *ln(x)
	Cladonia portentosa cover	negative	mild curvilinear	R^2=0.35, P<0.001	y=11.08 -3.60 * ln(x)
	Cladonia portentosa presence	negative	strong curvilinear	R^2=0.37, P<0.001	y=8.72 -2.69 *ln(x)
	Cladonia fimbriata cover	positive	linear	ns	
	Cladonia fimbriata presence	positive	linear	R^2=0.14, **P=0.06**	-
	Campylopus introflexus cover	positive	curvilinear	ns	-
	Brachythecium rutabulum cover	positive	curvilinear	ns	-
	Brachythecium rutabulum presence	positive	strong curvilinear	R^2=0.25, P<0.05	y=0.06 -0.014*x + 0.005*x^2
Bogs (TU 2009)					
	Cladonia portentosa cover	negative	linear	R^2=0.13, **P=0.055**	-
	Cladonia uncialis cover	negative	mild curvilinear	R^2=0.25, P=0.006	y=0.89 – 0.29*ln(x)
	Cladonia uncialis presence	negative	mild curvilinear	R^2=0.53, P<0.001	y= -1.17 +24.52/x
	Eriophorum vaginatum cover	positive	linear	R^2=0.41, P<0.001	y=1.48*x + 5.27
	Sphagnum fimbriatum cover	positive	mild curvilinear	R^2=0.20, P=0.015	y= -0.58 + 0.24*ln(x)
	Sphagnum fimbriatum presence	positive	mild curvilinear	R^2=0.21, P=0.013	y= -2.0 + 0.83*ln(x)
Sand dunes (TU 2009 all sites)					
	Hylocomium splendens cover	negative	mild curvilinear	R^2=0.21, P<0.01	y= -5.84 +106.88/x
Acid Grasslands (BEGIN)					
	Hylocomium splendens cover	negative	mild curvilinear	R^2=0.16, P=0.001	y= -1.01 +42.06/x
	Hypnum cupressiforme cover	positive	linear	R^2=0.16, P<0.001	y = 0.19*x - 2.07
	Nardus stricta cover	positive	linear	R^2=0.11, P=0.003	y = 0.30*x - 2.38
	Carex panacea cover	positive	linear	R^2=0.08, P=0.014	y = 0.072*x – 0.88
	Euphrasia officianlis cover	negative	mild curvilinear	R^2=0.11, P=0.005	y= 1.83 - 0.54*ln(x)
	Lotus corniculatus cover	negative	mild curvilinear	R^2=0.09, P=0.009	y= 3.58 – 1.03*ln(x)

5.3 Evidence from Dose–response relationships in the TU experiments

The UKREATE Terrestrial Umbrella (TU) project is funded by the Department for Environment Food and Rural Affairs (Defra) and the Natural Environment Research Council (NERC). The nine UKREATE sites (see Tables 5 and 6) are long term experiments in locations representing a broad range of priority UK habitats. Although established at different times over the past 22 years and involving different levels of nitrogen additions, the common features of their research design and monitoring enable consistent comparisons to be made among the sites. A recent overview is presented in RoTAP (2012), while the published sources of results from individual sites are given in Table 6 as well as in the report section on the UKREATE web site (http://ukreate.defra.gov.uk/publications/reports/index.htm).

5.3.1 Complementary nature of the Field surveys and the UKREATE experiments

The national-scale field surveys and the UKREATE experiments provide different, but complementary information. The changes observed in the spatial field surveys take place in the 'real-world' under normal timescales but may also be driven by climate, management, air pollution, soil chemistry and a host of other factors. Where air pollution, and in particular nitrogen deposition, is separated out as a main driver of change, the timescales of the influence of pollutants are very lengthy, possibly at least over the two centuries since the Industrial Revolution and periods of agricultural expansion but certainly over recent decades (Fowler *et al*., 2004).

The UKREATE experiments are short by comparison and in most cases involve nitrogen additions that are beyond the normal range of current deposition starting from ambient loadings which are already around the critical load. However, the controlled experiments provide evidence for ecosystem responses that result directly from nitrogen addition since other factors (soils, climate management etc) are a constant. They can also reveal the potential for changes that may occur at higher levels of nitrogen deposition but are not yet detectable in the natural landscape. Furthermore, some of the UKREATE experiments include the cessation of treatments along with maintained monitoring in order to investigate the consequences of reduction of nitrogen inputs.

Within this report the results of the UKREATE experiments are used to provide evidence regarding the nature of changes in plant community composition and individual species in response to added nitrogen.

5.3.2 Responses of vegetation to nitrogen addition and recovery

Selected examples of responses of vegetation to nitrogen additions in the UKREATE experiments are presented below. A general outcome from the experiments to date is that bryophytes and lichens are strongly and negatively affected by the nitrogen additions, but that changes in the vascular flora are modest and have appeared more slowly (RoTAP, 2012; Phoenix *et al*, 2012). One important exception is that both vascular species as well as lower plants in the Whim bog experiment have been greatly affected by gaseous ammonia treatments (Figure 9). The data from Whim bog show that responses in the most sensitive plants, in this case the bryophytes, are evident even at the lowest level of N addition i.e. 8 kg N ha^{-1} y^{-1} above a background of the same. At the higher end of the nitrogen addition range there is evidence of increasing damage to bryophytes at additions beyond the critical load range (for bogs 5-10 kg N $ha^{-1}y^{-1}$). In the gaseous NH_3 experiment, increased nitrogen also led to substantial community change with large increases in *Eriophorum* and loss of *Calluna* (Figure 9, top).

The sensitivity of bryophytes and lichens to wet deposited nitrogen treatments was also demonstrated at the original Ruabon upland heath (heather moorland) experiment (Carroll *et al*., 1999). In the newer Ruabon experiment, which used a wider range of treatments, a gradual pattern of change in lichen cover (mainly *Cladonia portentosa*) was observed (Figure 10) which after 5 years showed sensitivity to just 10 kg N $ha^{-1}y^{-1}$ above the ambient input of around 20 kg N $ha^{-1}y^{-1}$. This being the upper end of the critical load range for heathland (Pilkington *et al*., 2007). At Ruabon the same experiment also found that certain liverworts were particularly sensitive to increasing nitrogen additions (Figure 11), and again their abundance continued to decline as loadings were raised above the heathland critical load (Edmondson, 2007).

The sensitivity of lichens to nitrogen pollution, a clear outcome of the JNCC collation report (Stevens *et al*, 2011) was further confirmed in the experiment at Culardoch in the Cairngorms where lichens are an important part of the *Calluna*-*Cladonia* montane heath (Figure 12). Lichen cover was also very sensitive to nitrogen additions of just 7.7 kg N ha^{-1} y^{-1} (above a background deposition of around 8 kg N ha^{-1} y^{-1}) at Thursley Common (Figure 13) and here the effect still persisted at least 8 years after the nitrogen treatments had ceased (Power *et al*., 2006).

The above examples from the UKREATE experiments are consistent with the overall results from the field surveys since they show that small, realistic nitrogen additions can have adverse effects on the abundance of sensitive vegetation, even where the site is already close to the critical load. Increasing nitrogen inputs beyond the critical load can also change the character of the community by increasing the cover of dominant graminoid species.

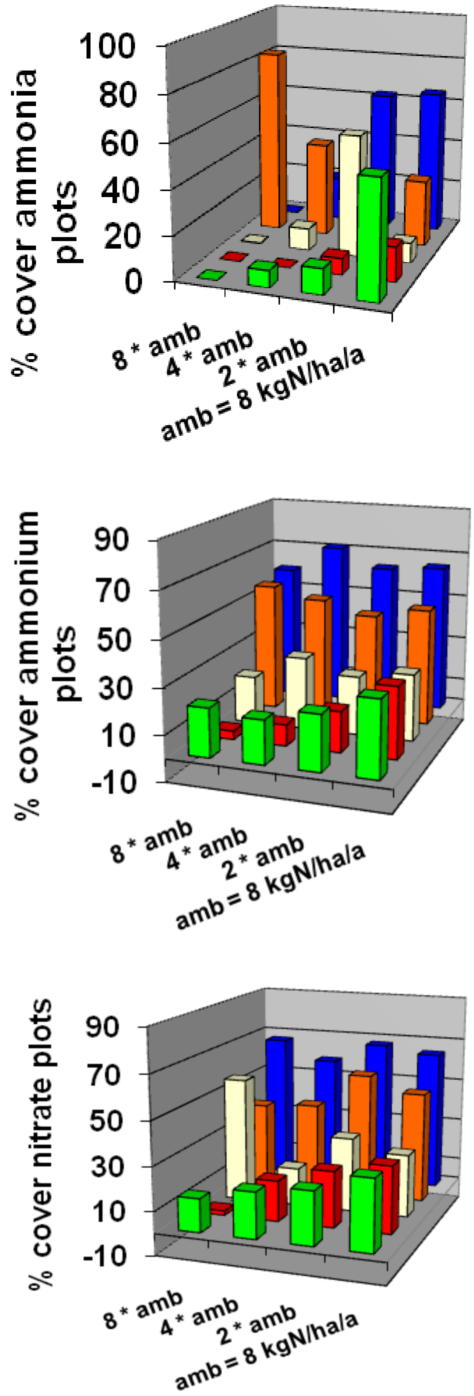


Figure 9: Response to different forms of nitrogen addition on Whim bog in 2009 after seven years treatment: ammonia gas (top), wet ammonium (middle), wet nitrate (lower) in different plants from front to back – *Sphagnum capillifolium, Pleurozium schreberi, Hypnum jutlandicum, Eriophorum vaginatum, Calluna vulgaris* (Sheppard, unpub). Ambient (amb) nitrogen deposition (approx. 8 kg N ha^{-1} yr^{-1}) at the right hand side of each graph, increasing N deposition towards the left.

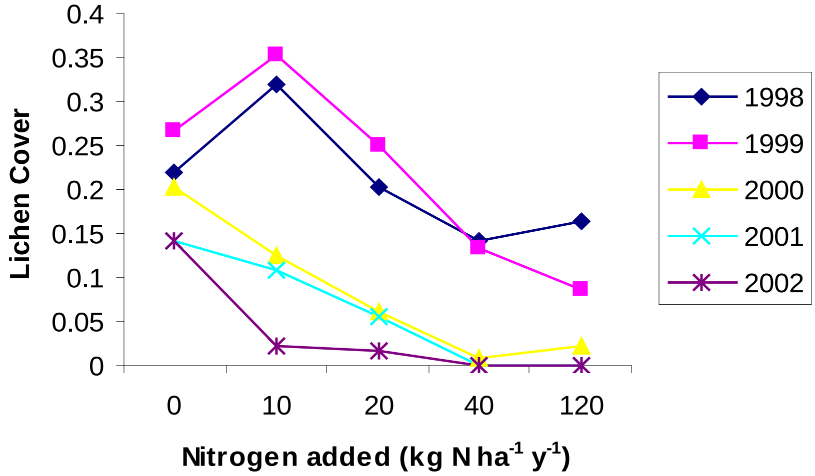


Figure 10: Nitrogen dose-response of lichen cover (mean touches/pin) at the Ruabon upland heath experiment new plots in the first 5 years of treatment. Ambient deposition circa 20 kg N ha^{-1} yr^{-1}. (Pilkington *et al*., 2007).

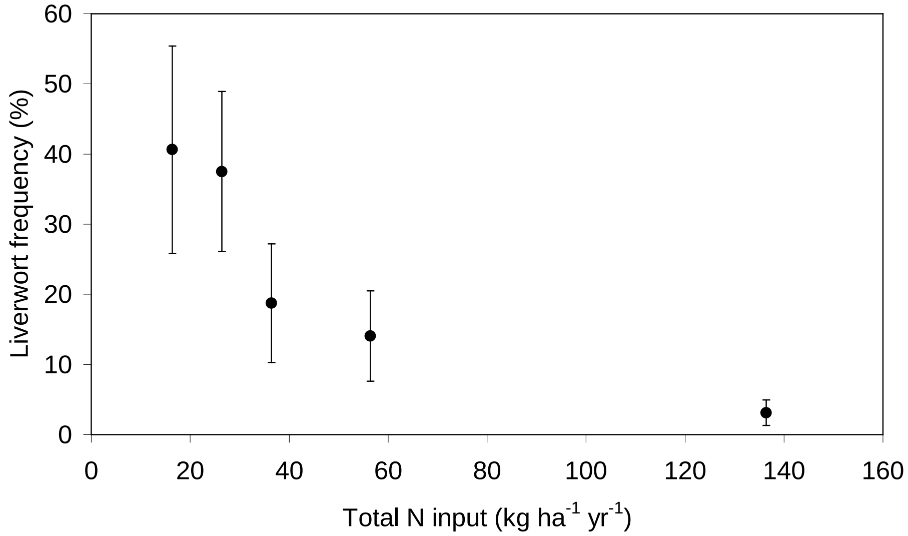


Figure 11: Relationship between total N input (N treatment + ambient deposition) and total liverwort frequency at the Ruabon upland heath experiment new plots. Ambient deposition circa 20 kg N ha^{-1} yr^{-1} (Edmondson, 2007).

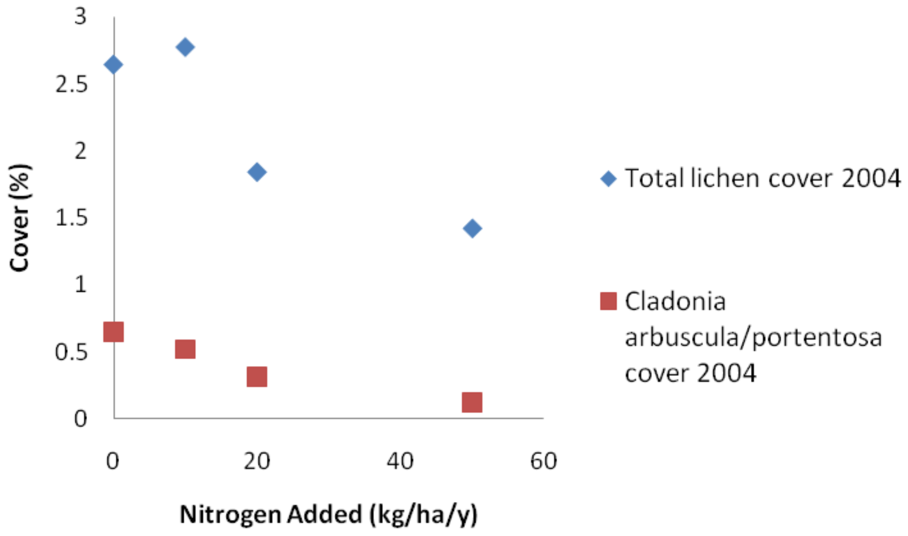


Figure 12: Response of lichens to nitrogen addition in montane heath at Culardoch in the Cairngorms (Britton & Fisher, 2007). Background N deposition circa 10 kg N ha^{-1} yr^{-1}.

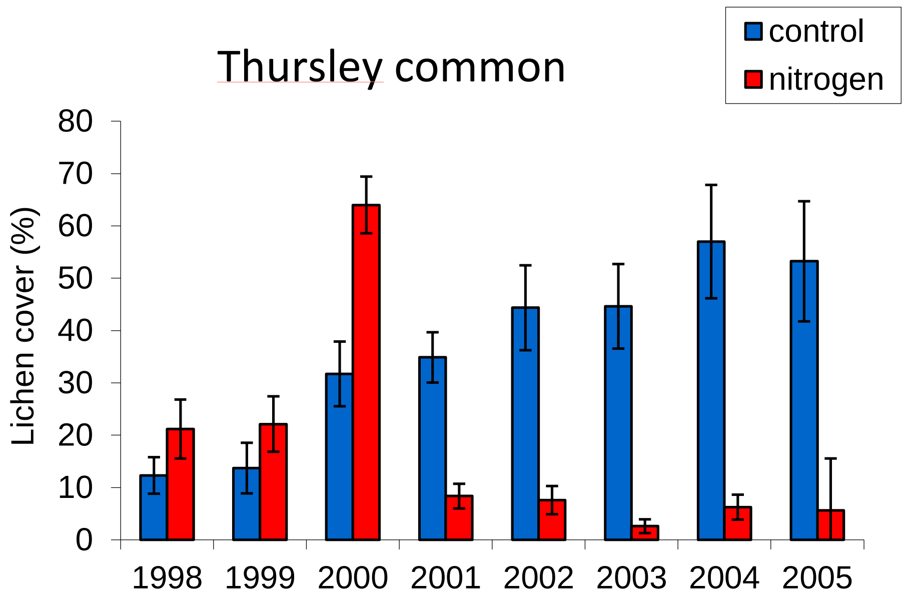


Figure 13: Effect of nitrogen additions (30 kg N ha^{-1} y^{-1}) on lichen cover at Thursley common, lowland heathland in Surrey (Sally Power, pers. comm.). The large decrease between 2000 and 2001 is thought to be related to a closing higher plant canopy following management interaction in 1998.

5.4 Reviewed literature – relationship between N deposition and the key response variables in the Countryside Survey and targeted habitat spatial surveys

The form of the relationship between nitrogen pollution and plant community composition is reviewed here for two types of survey: firstly the Countryside Survey of Great Britain, using a stratified randomised approach, which in this case used data collected in 1998 (Maskell *et al*., 2010). Secondly, several smaller scale surveys that have deliberately targeted particular habitats and enabled separation of air pollution signals from other important influences such as precipitation, temperature and management (Table 14).

The main focus of the Countryside Survey and targeted habitats spatial surveys was change in plant species richness, with further interest in individual sensitive species and functional groups. A range of important UK habitats was addressed, but some significant habitat gaps, notably woodland, remain. Data were gathered using small quadrats (2 x 2 m or 0.5 m^2 for bryophytes only) and analysed by multivariate statistical methods that examined a range of potential drivers including climate, air pollution and in some cases management (in other studies management level was kept a constant).

5.4.1 Habitat differences

The results of these surveys (Table 14) show a large degree of agreement, but also some important differences, between habitats. In upland heath, acid grassland and mesotrophic grassland, the different studies show strong agreement i.e. species richness was negatively correlated with nitrogen deposition. However, the two independent studies examining calcareous grassland found no correlation between nitrogen deposition and species richness. The contrast between base-rich calcareous habitats and the others suggests at least one of the likely mechanisms of change in less buffered soils is a long-term shift in soil pH (Stevens *et al*., 2010 Functional ecology paper) that could result from increased nitrogen deposition. While the calcareous grassland spatial survey in 1990-93 reported by Van den Berg *et al*. (2010) found no significant correlation with nitrogen deposition, the smaller repeat survey of 2006-2009 found an reduction in plant species diversity over the two decades that was greater in high-N deposition regions of the UK, particularly in the 25-35 kg N ha^{-1} y^{-1} areas. These changes comprised a decline in the frequency of characteristic calcareous grassland species and a lower number of rare and scarce species.

5.4.2 Taxonomic groups respond differently

In surveys where a significant drop in overall species richness in response to nitrogen deposition was recorded, different plant groups did not always respond in the same way. In acid grasslands, Stevens *et al*. (2006) found a strong decline in richness of forbs, a much weaker (but significant) reduction in grass species richness and no change in bryophyte species richness although the moss *Hylocomium splendens* was consistently reduced. In the Countryside Survey, both bryophyte and total species richness significantly declined in acid grasslands but not in heathlands and mesotrophic grasslands, where total species richness reduced but, bryophytes showed no change (Maskell *et al*., 2010). Bryophytes declined significantly with increasing nitrogen in the upland heath surveys.

5.4.3 The shape of the relationships between species richness and nitrogen deposition, and the critical load.

The decline in total species richness with increasing nitrogen demonstrated in all surveys apart from calcareous grasslands are described by either linear or curvilinear mathematical relationships. Over the nitrogen range investigated in the UK there is no evidence of a limit at the low end of the range below which negative change does not occur. The linear responses to increasing nitrogen found in a number of the studies (or plant groups within the studies) indicate that the rate of change in richness is constant across the nitrogen deposition range while the curvilinear fit that better describes other relationships indicate that reduction in species richness is greater at low than at high N deposition.

The slope of species richness vs nitrogen deposition is remarkably similar for the studies 1-5 (Table 11) (all except calcareous grassland, study 6) falling in a range of between minus 2.5 – 4.3 fewer species per quadrat per 10 kg $ha^{-1}y^{-1}$. Where curvilinear gradients were described (study 4) the rate of change was greater (minus 4.3) below 10 kg $ha^{-1}y^{-1}$ and minus 2.9 above 20 kg $ha^{-1}y^{-1}$. These figures compare broadly with the survey data analysed in this report: see task 5, Table 19.

Note however, that fewer species in a quadrat, or a reduction in species richness, means neither species 'loss' from a site, nor local species extinction – it means that the frequency of at least one species has been reduced. Also note that it is very likely that any inferred reduction in species richness due to N is the product of many years of N deposition, so that the current rate of N deposition is primarily a proxy for this long-term cumulative N.

5.4.4 Species richness declines in relation to the critical load

For acid grasslands and heathlands (Table 14, studies 1-4 and part of 5) the critical load range is 10-15 kg N $ha^{-1}y^{-1}$ and 10-20 kg N $ha^{-1}y^{-1}$ respectively. These fall towards the lower end (left) of the nitrogen deposition range surveyed. This has the following important implications:

(a) as would be expected, *above* the critical load range there is a substantial reduction in species richness (for both linear and curvilinear responses);
(b) what was more unexpected is that where curvilinear responses in species richness are described, the greatest decline in richness is *below* the critical load.

Table 14: Details of other surveys that have targeted specific habitat types along pollution and climatic gradients

Habitats	Significant Trend in response to increasing N deposition	Critical Load N kg $ha^{-1}y^{-1}$	Nitrogen Range kg $ha^{-1}y^{-1}$	Slope	Type of Relationship of response variable with N deposition	Comment	Reference
Upland dry heath	Bryophytes richness declines	10-20	19.5-30.5	3.1 bryophyte species / 10 kg N	Linear best fit		Edmondson *et al*., 2010
Upland dry heath	Bryophytes richness declines	10-20	8-31	2.4 bryophyte species/ 10 kg N	Linear best fit		Combined data of Edmondson & Carroll & Caporn (Stevens *et al*., 2009)
Acid grassland	Forbs & grass richness declines, Bryophytes no change Plant acid preference index score increases	10-15	6-36	4 species (all) / 10 kg N	Linear best fit to NHy and total N deposition; Exponential curvilinear best fit with NOx deposition Linear best fit of plant acid preference with total N deposition	Greater decline at low N deposition	Stevens *et al.*, 2004, 2006, 2010
Acid grassland	Forbs Grasses Bryophytes All groups richness decline	10-15	2-44	4.3 species (all) / 10 kg N below 20 kg /ha/y 2.9 species (all) / 10 kg above 20 kg N/ha/y	Exponential curvilinear best fit with total N	Greater decline at low N deposition BEGIN project Analysis included GB and European sites	Stevens *et al.*, 2010

Habitats	Significant Trend in response to increasing N deposition	Critical Load N kg $ha^{-1}y^{-1}$	Nitrogen Range kg $ha^{-1}y^{-1}$	Slope	Type of Relationship of response variable with N deposition	Comment	Reference
Acid grassland	(Vascular + bryophyte) richness decline Bryophyte (alone) Decline	10-15	c. 5-40	2.5 (vasc+bryo species) / 10 kg N	Linear best fit	Countryside Survey 1998	Maskell *et al*., 2010
Calcareous grassland	No response in richness	15-25	c. 5-40	No change	No change	Countryside Survey 1998	
Mesotrophic grassland	(Vascular + bryophyte) richness decline	15-25	c. 5-40	Not given	Linear fit	Countryside Survey 1998	
Heathland	(Vascular + bryophyte) richness decline	10-20	c. 5-40	3.2 (vasc+bryo) species / 10 kg N	Linear fit	Countryside Survey 1998	
Calcareous grassland	No response in richness (1990-1993 survey)	15-25	7-41	No change	Above 25 kg N/ha/y there was a lower number of rare and scarce species	1990-3 survey part repeated in 2006-9; Increasing decline in species diversity and evenness over +20 years	Van den Berg *et al*., 2010

5.5 Review of relationships between N deposition and the key response variables from the JNCC Collation report

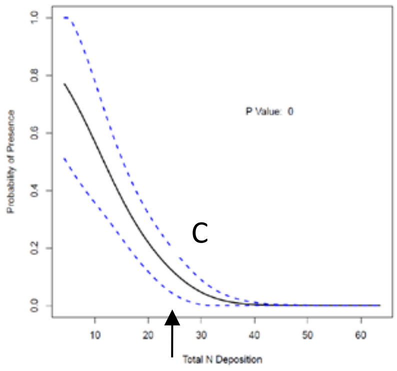
Two recent studies for JNCC have collated and analysed several different vegetation surveillance data sets (within UK or GB) in order to investigate relationships between community composition and nitrogen deposition (Stevens *et al*., 2011) and the impact of this on critical loads and policy (Emmett *et al*., 2011). A summary of relationships in selected data sets from the JNCC report is given here (Tables 16-17). This report will focus on the analysis of the vegetation datasets held within Stevens *et al*. (2011) although for a broader overview of policy implications the reader is directed to Emmett *et al*. (2012).

This JNCC collation study examines in detail the responses to nitrogen in a selection of habitats: heathlands, acid grasslands, calcareous grasslands and bogs. Large scale geographical distributions of plant species in relation to nitrogen deposition were examined using eight different surveillance data sets. Stevens *et al*. (2011) analysed spatial and in addition temporal changes over recent decades where data was available.

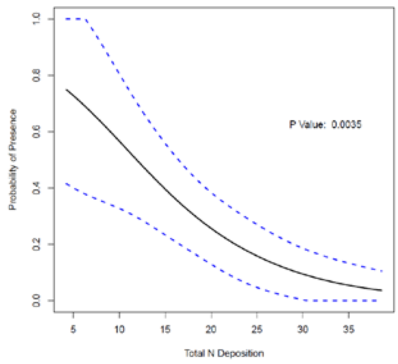
Discussion in this report is limited to examining the nature or shape of the spatial dose response relationships described in the collation report and covers the main databases providing detailed information on individual species over a wide geographical range: Vascular plant database (VPD), Botanical Society of the British Isles (BSBI), British Lichen society (BLS). Comment here is made regarding the British Bryological Society (BBS) data set.

A range of dose-response relationships was mathematically described and plotted in the JNCC Collation report (Stevens *et al*., 2011). Species presence - N relationships were not analysed by Stevens *et al*. (2011) where there were insufficient samples across the nitrogen deposition range. In the current report four types of relationships are proposed, the first three are negative responses of species to nitrogen, while the fourth is a positive response. In the cases of negative responses, an approximate deposition to result in 50% probability of presence is given based on visual assessment of the relationships; this is termed ND_{50}. Examples of these responses taken from Stevens *et al* (2011) are shown below overleaf. The four response types are categorised in the following way and illustrated in Figure 14. The mathematical relationships are represented by the solid line. Dotted lines illustrate the confidence intervals, where narrow our understanding of the response is strong, where the distance between these lines widens this reflects a less understanding of responses usually at extremes of the N deposition range:

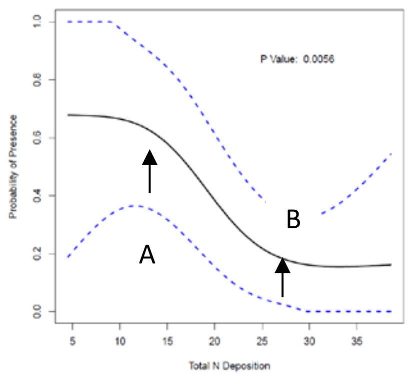
- Type 1: Strong negative curvilinear fall with a turning point ('heel') at point C
- Type 2: Mild negative curvilinear fall, the shape approaching linear within the normal deposition range (up to 30-35 kg N)
- Type 3: Reverse sigmoid fall, indicating a shoulder at point A and a heel at point B
- Type 4: Increase within normal deposition range



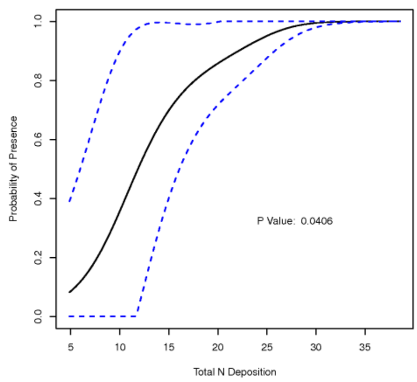
Type 1: strong curvilinear fall
Spatial change in the probability of presence of *Cladonia subulata* in heathland with increasing total current inorganic N deposition (kg N ha^{-1} yr^{-1}). Data from BLS.



Type 2: mild curvilinear fall
Spatial change in the probability of presence of *Peltigera didactyla* in acid grassland with increasing total current inorganic N deposition (kg N ha^{-1} yr^{-1}). Data from BLS.



Type 3: reverse sigmoid fall
Spatial change in the probability of presence of *Cladonia foliacea* in calcareous grassland with increasing total current inorganic N deposition (kg N ha^{-1} yr^{-1}). Data from BLS.



Type 4: Increase
Spatial change in the probability of presence of *Alchemilla xanthochlora* in upland calcareous grassland with increasing total current inorganic N deposition (kg N ha^{-1} yr^{-1}). Data from Vascular Plant Database.

Figure 14: Examples of relationships between individual species probability of presence and nitrogen deposition from JNCC report 447 (Stevens *et al*., 2011) with proposed inflexion points A,B,C which are discussed here in the text

5.5.1 Other types of responses within the VPD, BSBI, BLS and BBS databases

In addition to the cases listed in the Tables 15-17 and shown in the graphs (see Stevens *et al*. 2011), there were also other cases of: no significant relationship; hump-backed responses, U-shaped responses; 'small magnitude changes.' These are not reported in the Tables 15-17 in the current report because they are difficult to interpret and suggest the strong influence of other factors. The tables below show cases where there were clear-cut, significant relationships with nitrogen deposition, both positive and negative.

Where there were sufficient data available for analysis, numerous observations of 'no significant response' were reported. Where significant changes were found the majority of these were negative i.e. decline in presence with increasing nitrogen deposition. There were also a significant number of species showing increasing presence, particularly amongst bryophytes, but the majority of responses within any plant group were negative.

The strongest responses were in the lichen data set (BLS) and these were consistently negative changes. All three decline curve types were seen, but the majority were Type 1 or 2, indicating that there was no minimum threshold and the decline in presence started from a very low dose (around 5 kg N ha^{-1} y^{-1}). The fewer cases found of the reverse sigmoid curve type (Type 3) suggested that a decline commenced above a nitrogen deposition of approximately 15 kg N ha^{-1} y^{-1}. The approximate value for the ND_{50} was, for the lichens, within or around the low end of the critical load range for the habitats, with an average of 14 kg N ha^{-1} y^{-1} for those lichens shown in Table 14.

There were fewer cases of clear relationships with nitrogen in the BSBI higher plant data sets (Table 16), but these were all negative with either Type 1 or Type 3 curves over the normal range of deposition. Sigmoid type curves (Type 3) for three species showed that decline commenced above a nitrogen deposition of approximately 5, 8, 18 kg N ha^{-1} y^{-1} respectively. The mean ND_{50} for these vascular plants was approximately 16 kg N ha^{-1} y^{-1}.

The Vascular Plant Database (VPD) analysis showed a larger number of significant positive and negative relationships between vascular species presence and Nitrogen deposition (Table 17). The most common decline was described by a strong curvilinear response (Type 1) with a 50% drop by around 8-15 kg N ha^{-1} y^{-1}. A few species declined in a sigmoid manner (Type 3) with a shoulder at around 10-15 kg N ha^{-1} y^{-1}.

Bryophyte response curves in the JNCC collation report were not analysed to the same level of detail in the current study. The individual species data for bryophytes are difficult to interpret with some species increasing and other decreasing and the authors could not find any clear and general trends in the data (Stevens *et al*. 2011).

5.5.2 Response patterns above the critical load

Examination of the individual responses from the BLS, BSBI and VPD datasets suggests that at the higher levels of deposition there usually is a turning point or 'heel' to a slower rate of decline (position C in a type 1 curve, and position B in a type 3 curve - see Figure 14). In the significant negative responses this turning point was observed around an average approximate nitrogen deposition of 25 kg N ha^{-1} y^{-1} in lichens (BLS datasets) and 23 kg N ha^{-1} y^{-1} in vascular plants (VPD and BSBI datasets).

5.6 Summary from the review of published dose response relationships between species richness or individual species presence and nitrogen deposition

Evidence from the BLS, BSBI and VPD national datasets (summarised over Tables 15 to 17) which are based on national botanical surveillance data sets or targeted habitat surveys show a remarkable degree of consistency demonstrating evidence for nitrogen enrichment across several UK habitats. All habitats examined here show evidence of either broad scale reductions in species richness, decline in individual species or plant groups or increases in some nitrophilous species such as graminoids. However, it is also clear that species and habitats do not all respond in the same way to nitrogen deposition. For example, individual bryophytes show a range of different responses to nitrogen, some declining and others increasing that probably reflect the importance of other aspects of habitat/micro-habitat over nitrogen deposition in many species of this group. Calcareous habitats are less affected by nitrogen deposition than less well pH buffered systems suggesting a role for acidification in changes in plant communities.

Negative relationships with nitrogen in species richness or individual species presence tend to show a similar response form – namely either linear indicating an equal rate of decline across the nitrogen deposition range or curvilinear suggesting a greater rate of change at low nitrogen than at high deposition rates. The JNCC Collation study revealed a number of plant species that started to decline with increasing nitrogen inputs only after a level of deposition was reached, but in the majority of affected species no level was apparent implying that within the normal UK deposition range the vulnerable species start to decline with any increasing level of nitrogen and that this is typically at or below the critical load range for the habitats.

At the higher end of the nitrogen deposition scale, all the data sets investigated demonstrated clear evidence that species declines continue to occur above the habitat critical loads, so that small increments of nitrogen pollution within the range of 20-35 kg N ha^{-1} y^{-1} still have the potential to cause adverse and continuing change. Even in the less sensitive calcareous habitat, the temporal change study in calcareous grasslands found the greatest decline in individual species was in the areas receiving 25-35 kg N ha^{-1} y^{-1}. The importance of changes in nitrogen deposition at the higher end of the UK range should not be underestimated.

Table 15: Relationship between probability of presence and nitrogen deposition for lichen species from the BLS database spatial analysis for which significant responses were found. The turning point and ND50 (these are defined in section 5.5) values refer to nitrogen deposition (kg N ha^{-1} y^{-1}) and were estimated by eye from published figures of Stevens *et al*. (2011) and Emmett *et al*. (2012). Examples of response types and turning points are shown in Figure 14.

British lichen society database Habitat / Species (Critical load)	Direction of change	Response Type	Turning Points (kg N ha^{-1} y^{-1})	ND_{50}
Acid grassland (10-15 kg N ha^{-1} y^{-1})				
Cetraria aculeata	Negative	1	C=22	15
Peltigera didactyla	Negative	2		12
Calcareous grassland (15-25 kg N ha^{-1} y^{-1})				
Cladonia foliacea	Negative	3	A=15, B=30	17
Bog (5-10 kg N ha^{-1} y^{-1})				
Cladonia portentosa	Negative	3 (between 5-30 kg N)	A=15, B=30	22
Heathland (10-20 kg N ha^{-1} y^{-1})				
Cetraria aculeata	Negative	1	C=20	10
Cetraria muricata	Negative	1	C=20	10
Cladonia cervicornis cervicornis	Negative	1	C=15	8
Cladonia cervicornis verticillata	Negative	2 (between 5-35 kg N)		10
Cladonia portentosa	Negative	2 (between 5-30 kg N)		15
Cladonia subulata	Negative	1	C=27	12
Cladonia uncialis biuncialis	Negative	1	C=18	9
Peltigera hymenina	Negative	3	A=15, B=40	25
				Mean = 14

Table 16: Relationship between probability of presence and nitrogen deposition for vascular plant species from the BSBI database local change spatial analysis for which significant responses were found. The turning point and ND_{50} values refer to nitrogen deposition (kg N ha^{-1} y^{-1}) and were estimated by eye from published figures of Stevens *et al*. (2011) and Emmett *et al*. (2012). Examples of response types and turning points are shown in Figure 14.

BSBI database Habitat/Species (Critical load)	**Direction of change**	**Curve Type**	**Turning Points (kg N ha^{-1} y^{-1})**	**LD50**
Calcareous grassland				
Lowland (15-25 kg N ha^{-1} y^{-1})				
Bromopsis erecta	Negative	1 (over 5-25 kg N range)	C= 12	8
Campanula glomerata	Negative	3	A=18, B=30	27
Carex spicata	Negative	1	C=18	14
Ononis repens	Negative	3	A=8, B=18	15
Upland Heathland (10-20 kg N ha^{-1} y^{-1})				
Vaccinium vitis-idaea	Negative	3	A=5 , B=27	18
				Mean = 16

Table 17: Relationship between probability of presence and nitrogen deposition for vascular plant species for which significant responses were found in the vascular plant database spatial analysis. The turning point and ND_{50} values refer to nitrogen deposition (kg N ha^{-1} y^{-1}) and were estimated by eye from published figures of Stevens *et al*. (2011) and Emmett *et al*. (2012). Examples of response types and turning points are shown in Figure 14.

Vascular plant database Habitat/Species (critical load)	Direction of change	Curve Type	Turning Points	LD_{50}
Acid grassland: Lowland (10-15 kg N ha^{-1} y^{-1})				
Cerastium arvense	Negative	1	C=15	8
Cerastium semidecandrum	Negative	1	C=22	12
Trifolium arvense	Negative	1	C=19	12
Vicia lathyroides	Negative	1	C=19	10
Viola canina	Negative	1	C=24	15
Calcareous grassland (15-25 kg N ha^{-1} y^{-1})				
Lowland				
Allium vineale	Negative	1	C=25	9
Anacamptis pyramidalis	Negative	1	C=30	8
Carlina vulgaris	Negative	2 (between 5-30)		16
Cynoglossum officinale	Negative	3	A=10, B=35	25
Echium vulgare	Negative	3	A=10, B= 30	Outside range
Geranium columbinum	Negative	1	C=25	12
Lathyrus nissolia	Positive	4 (between 15-30)		
Ononis repens	Negative	3	A=10, B= 22	20
Spiranthes spiralis	Negative	1	C=15	8
Stachys officinalis	Positive	4 (between 5-30)		
Upland				
Alchemilla xanthochlora	Positive	4 (between 5-30)		
Melica nutans	Negative	3	A=15, B=27	17
Heathland (10-20 kg N ha^{-1} y^{-1})				
Lowland				
Platanthera bifolia	Positive	4 (between 5-35)		
Viola canina	Negative	3	A=12, B=25	18
Upland				
Arctostaphylos uva-ursi	Negative	1	C=15	9
				Mean = 13

5.7 Cluster analysis, sample grouping and ecological thresholds results

The analysis in this section of Task 4 sought to establish coherent ecological groupings along the N deposition gradient to support the suggestion that N is an important control on community composition and locate putative loads of N deposition where community composition is found to change significantly. Full details of the methodology used are provided in the methods section of this report.

The results are summarised in Table 18, split by survey dataset. In the acid grassland data (BEGIN) there are significant differences between the first two groups identified by both CONISS and SPLITLSQ. The break-points identified by these methods are different but rather similar, falling around 14.1 and 14.4 kg N ha^{-1} yr^{-1}. In the TU sand dune dataset significantly different groups are identified with only one method and the groups identified are only marginally significant. In the TU bog dataset two methods identify first divisions in adjacent positions around 11-12 kg N ha^{-1} yr^{-1}. In the TU upland heath data CONISS and SPLITINF identify adjacent break-points around 17 kg N ha^{-1} yr^{-1}, SPLITLSQ identifies a slightly lower first break at around 14.6 kg N ha^{-1} yr^{-1} and then a second significant break around 25-26 kg N ha^{-1} yr^{-1}. In the TU lowland heath data identical breaks are identified by all methods with only a first break significant at around 14.6 kg N ha^{-1} yr^{-1}.

Table 18: Results of CONISS, SPLITLSQ and SPLITINF for seven nitrogen gradient studies. Results show only the first two break-points listing the samples (e.g. N14.4, N14.5 etc) between which a division falls and the total nitrogen deposition (N) values for those samples in kg N ha^{-1} yr^{-1}. Analyses were based on squared Euclidean distance using proportion data (mean percent cover in most datasets, proportion of total species occurrences in five quadrats in the moorland regional survey). Differences between groups were tested on the same datasets using ANOSIM with Bray-Curtis dissimilarity, significance testing with permutation tests (10,000 permutations). *=P<0.05, **=P<0.01, ***=P<0.001, grey shading shows non-significant results. Second break-points are only counted as significant if all groups are significantly different (maximum P value between groups shown).

Dataset	CONISS		SPLITLSQ		SPLITINF	
	1st division	2nd division	1st division	2nd division	1st division	2nd division
BEGIN- acid grassland	(N14.4/14.5)**	(N22.8/23.8)	(N14.0/14.2)*	(N14.4/14.5)	(N24.8/25.0)	(11.0/13.4)
Moorland regional survey	N26.1/28.1)***	N20.4/20.6)**	(12.9/20.4)***	N26.1/28.1)***	(12.9/20.4)***	(N26.1/28.1)***
TU-sand dune	(N13.9/14.6)	(N15/16.7)	(N13.9/14.6)	(N15/16.7)	(N5.7/7.7)*	(N10.4/11.5)
TU-bogs	N10.9/11.7)***	(N19.6/21.9)	(N19.6/21.9)	(N14.2/14.3)	(N11.7/13.1)**	(16.6/16.6)
TU- Upland heath	(N16.5/17)**	(N24.5/28)	(N14.1/15.2)***	(N24.8/26.6)**	(N17/19.4)***	(N24.8/26.6)
TU-lowland heath	(N14.6/14.7)**	(N13/13.6)	(N14.6/14.7)**	(N13/13.6)	(N14.6/14.7)**	(N13/13.6)

Comparing the results between different surveys of the same or similar habitat types the case for consistent break-points is not overly strong. In the upland heath data from the TU and MRS there is overlap in some of the break-points however given the inconsistent sampling along the gradient it is difficult to place great confidence in the significance of this observation. The 16.5/17, 14.1/15.2 and N17/19.4 break-points in the TU data all lie within the wide gap between the MR15 and MR20 samples (12.9-20.4 kg N ha^{-1} yr^{-1}) in the MRS data. As discussed above, comparison of the MRS results with other results is particularly difficult due to the differing methodologies. Comparing the TU upland and lowland heath data there is similarity in the position of a first break-point in the lowland heath dataset and the first break-point with SPLITLSQ in the upland heath data while the upland heath first splits with SPLITINF and CONISS are slightly higher.

The results show that although break-points are identified in all cases, not all of the identified groups are significantly different. In only two cases are groups identified by a second split in the data significantly different from each other. The most significant results are identified with the moorland regional survey with all methods producing significantly different groups; however these results should be treated with caution particularly in comparison to the other datasets. In contrast to other datasets instead of recording percent cover this survey recorded presence/absence in each of five quadrats; the data we analyse here is a proportion of total species count and therefore although incorporating some measure of abundance is more weighted towards presence/absence.

In general there is some similarity between the results of different methods, but in many cases also substantial differences. Perhaps surprisingly the results of the two divisive methods are not substantially more similar to each other than to the results of the agglomerative method.

In most datasets our analyses succeed in identifying groups of samples with significantly different plant communities according to levels of nitrogen deposition. In itself this is an interesting result, supporting the suggestion from the ordinations that nitrogen deposition modifies plant community structure. The identification of such groups may be a useful way to approach the output of gradient studies. For instance the identification of indicator species for these groups may allow new sites to be categorised according to their levels of nitrogen-loading, which may be a useful approach to bioindication.

In some datasets different statistical techniques suggest the same break-point locations whereas in others there are differences. Where there are similar results by different methods this provides evidence for the validity of these values. However, where results differ between methods it does not necessarily mean the results are invalid. As different methods work on different principles and responded to different elements in the data it is quite possible for different methods to identify different, but equally valid break-points.

This analysis has studied the relationship between species composition and nitrogen deposition. The analysis of the lowland and upland heath and acid grassland datasets suggest that significant changes in composition may occur at 15 – 17 kg N ha^{-1} yr^{-1}. This value is in broad agreement with the conventional and LOESS regressions which looked for 'heels' in the species richness curves. Analysis of the bog vegetation data suggested a breakpoint where change occurs in species composition of 10-13 kg N ha^{-1} yr^{-1} which is lower than the 19-20 kg N ha^{-1} yr^{-1} suggested in the regression analysis of species richness data. Analysis of the sand dune data also suggested that changes in species composition may occur at much lower levels of N deposition than changes in overall species richness, 5-8 kg N ha^{-1} yr^{-1} compared to 10 kg N ha^{-1} yr^{-1}.

These findings broadly support the species richness regressions and the levels of N deposition where the rate of change in species richness altered, however, they do highlight the possibility that a change in species composition may occur before it is measured by a change in overall species richness. The implication of this is that ecosystems may be showing sensitivity to N deposition at much lower levels of N deposition than previously thought and certainly at the lower end of the critical load ranges.

5.8 Summary

The responses to nitrogen (N) across the habitats studied in the survey datasets are remarkably consistent with strong reductions in measured species richness associated with increases in N deposition. The fact that N deposition is so consistently related to changes in species richness (and composition) demonstrates the deleterious effects of N pollution on biodiversity. In many cases, bryophytes and lichens were shown to be most sensitive to N deposition although in grasslands, the intrinsic biodiversity of the habitat was affected by N through reductions in forb species richness. Within the heathland, bog and sand dune survey datasets graminoid cover increased exponentially with rising N deposition.

The results from the survey datasets are consistent with the findings from the dose-response experiments and the literature review. At a number of heathland experiments bryophyte and lichen diversity or cover reduced as N addition increased, notably so at the Culardoch montane heath where background N deposition was at the low end of the critical load range for heathlands (10 kg N ha^{-1} yr^{-1}). Reductions specifically in *Cladonia portentosa* cover at Ruabon and Culardoch supported the reductions seen in the heathland and bog surveys. Vascular plants were not strongly affected by N in the experiments although this perhaps reflects the dose/time compromise made. An exception to this was increase in *Eriophorum vaginatum* cover at Whim Bog along the gaseous ammonia release transect which reflected increases in cover this sedge as N deposition increased in the survey dataset. It should also be noted that the timescales of even the longest N-addition experiments are short by comparison to real-world N driven changes which have occurred over many decades. Furthermore, experiments tend to be limited by plot size which limits the possible response of vascular plants which may be reflected in change over a wider area. For these reasons, many of the experiments fail to show significant loss of vascular species although in some cases declines in cover are observed. Indeed, as has been observed, cover of certain graminoid species increased with N deposition although graminoid species richness tended to fall.

The similarity in responses seen at the experimental sites provides important supporting evidence that N deposition is driving the changes seen in the survey datasets and that change is not solely driven by a climatic gradient. Larger national survey datasets including the Countryside Survey and plant databases from the British Lichen Society (BLS) and the Botanical Society of the British Isles (BSBI) also provide evidence for widespread changes in response to N deposition. However, it is also accepted that climate plays an important role in influencing species richness and diversity with a tendency towards greater species richness in wetter and colder sites.

Much of the data presented and reviewed in this report suggested a curvilinear response to N with steeper responses at lower background levels of N (< 10 kg ha^{-1} yr^{-1}), decreasing at higher background N (>20 kg) with a turning point or heel in the response curves generally occurring between 15 – 22 kg N ha^{-1} yr^{-1}. These N deposition loads were broadly supported by the cluster analysis that studied change using species composition as the variable, however, in sand dunes and bogs species composition appeared sensitive at much lower levels of N highlighting that ecosystem change occurs across the N deposition range. This is reflected by some of the changes in seen in individual species that are poorly distributed across the datasets but typically found at the less polluted sites. The change in the frequency of presence or cover of some species may therefore be good indicators of N deposition even where a change in overall species richness is not observed.

Table 19 below summarises some of the key findings from the analysis of the vegetation datasets and supporting reviews of literature and experimental site work. The relationships found in the species richness, cover and individual species data will be carried forward to Task 5 where the effect of incremental increases in N deposition on the variables will be examined further. Other relationship exist with the review literature and the experiments as discussed in this chapter, however, where these were not found to be significant in the data analysed as part of this project they have not been carried forward.

Table 19: Summary of the key findings from the analysis of the vegetation datasets and supporting reviews of literature and experimental site work. [1]Details of the UKREATE experiments in this report is presented in Tasks 2, 3 and 4 summarised in Phoenix *et al* (2012). [2]For details of studies included in the literature review see Tasks 2 and 3.

Habitat	Response curve shape from data analysis	Supported by UKREATE experiments[1]	Literature[2]	Spp. richness LOESS max. sensitivity	Spp. composition Cluster analysis change range	Individual species
All habitats						
Species richness (SR)	linear	#	#	n/a	n/a	*Hylocomium splendens*
Upland heath					14-20 kg N	*Hylocomium splendens Deschampsia flexuosa*
Total SR	mild curvilinear		#	7-20 kg N		
Lichen SR	curvi-linear	#	#	7-16 kg N		
Graminoid SR	mild curvilinear			7-22 kg N		
Graminoid cover	mild curvilinear			>22 kg N		
Lowland heath					14-15 kg N	*Hylocomium splendens Cladonia portentosa Brachythecium rutabulum*
Total SR	mild curvilinear	#	#	< 17 kg N		
Moss SR	mild curvilinear	#	#	none		
Graminoid SR	mild curvilinear			none		
Graminoid cover	mild curvilinear			> 23 kg N		
Bog					10-13 kg N	*Cladonia uncialis Eriophorum vaginatum Sphagnum fimbriatum*
Total SR	linear			> 19 kg N		
Lichen SR	linear			none		
Forb SR	mild curvilinear			none		
Graminoid cover	linear	#		none		
Sand dune					5-8 kg N	*Hylocomium splendens*
Total SR	mild curvilinear					
Moss SR	strong curvilinear					
Forb SR	mild curvilinear					
Graminoid cover	mild curvilinear		#			

Habitat	Response curve shape from data analysis	Supported by UKREATE experiments[1]	Literature[2]	Spp. richness LOESS max. sensitivity	Spp. composition Cluster analysis change range	Individual species
Acid grassland					14-15 kg N	*Hylocomium splendens* *Hypnum cupressiforme* *Europhrasia officianalis* *Lotus corniculatus* *Carex panacea* *Nardus stricta*
Total SR	mild curvilinear		#			
Forb SR	linear		#			

6. Task 5: Determine the relative effect of incremental N

6.1 Introduction

The implications of the relationships between the response variables and nitrogen (N) deposition that were detailed within Task 4 are explored further within this task and the effect of incremental increases in long-term N deposition upon these response variables is considered.

6.2 Habitat specific difference in species richness along the survey gradients associated with different levels of N deposition

As described previously, the relationships between species richness and N deposition described by the spatial surveys have developed over many years, and the current rate of N deposition is primarily a proxy for this long-term cumulative N. Thus we would not expect that a change in N deposition, either increasing or decreasing, would immediately change species richness or composition. However, the spatial relationships described above can be examined to estimate how the species richness of different habitats has responded to different levels of long-term N deposition, as represented by differences in current N deposition. Recall from previously that a reduction in species richness does not necessarily mean that any species are 'lost' (see Figure 1), but that the frequency of some species is reduced.

This section 1) compares the difference in species richness for a 1 kg N ha^{-1} y^{-1} increment of long-term N deposition along the gradient studies, expressed as a reduction of percentage of the maximum number of species recorded in each habitat (Table 20**,** other increments are shown in appendix 5), 2) expresses the response relationship as the difference in long-term N deposition associated with a species richness reduction of one species along the gradient (Table 21). For reference, the current (2011) critical load for each habitat is included in the results tables and full details of critical loads for all habitats are provided in appendix 6 with a summary in the introduction to this report).

When the linearly related TU survey data are combined, 1.6% of the maximum number of species within each habitat is reduced with every incremental increases of 1 kg ha^{-1} yr^{-1} of long-term N deposition. When all the habitats are considered separately, the typically curvilinear response of species richness to N deposition produces sharp losses in diversity from well below the habitat-specific critical load ranges. However, even at levels of N deposition at and above the upper end of each habitat-specific critical load, the effect of a 1 kg increase of N is considerable and at the mid-point of the critical load range the losses and subsequent threat to habitat integrity from loss of sensitive species and increases in graminoid cover are considerable. For example, within the upland and lowland heath habitats in the TU 2009 survey, species richness is reduced by nearly 1.0 (around 2 % of the maximum species richness) for each 1 kg $ha^{-1}yr^{-1}$ increase in long-term N above the mid-point of the critical load range (15 kg N $ha^{-1}yr^{-1}$) and by 1.0 for each 2 kg increase (above 1 % of maximum) increase in N deposition well above the critical load: 25 kg N $ha^{-1}yr^{-1}$. The magnitude of change is less in the MRS Upland heath survey probably due to a different recording technique and the use of smaller survey quadrats due to the focus towards the lower plant species. These results from the heathland surveys reveal remarkably high losses in diversity in what are naturally low-diversity systems, typified by specialist low-nutrient plants and these losses highlight the vulnerability of this habitat to eutrophication by N enrichment.

The results from the other habitats follow a similar pattern of reductions in species richness as long-term N deposition increases. Sand dune ecosystems in particular appear to be strongly sensitive to N deposition with a very rapid loss of species diversity as N increases from below the lower end of the critical load range. Even when sand dune type is split between decalcified and calcareous, species richness reduces by 1.2 % species for every 1 kg increase in long-term N deposition above the upper end of the habitat specific critical load (15 kg N $ha^{-1}yr^{-1}$), equivalent to 1 a fall of 1 species for every 1.1 kg N $ha^{-1}yr^{-1}$. Moss diversity is particularly negatively related to N. Within the bog habitat, losses are less severe with species richness reducing by around 1 % for approximately every 3 kg increase in long-term N deposition across the range studied. This is likely due to the hydrology regime limiting species responses to N. The much larger acid grassland dataset (BEGIN) also showed reductions in richness to N and, due to a flatter response curve, these remain high with a 1 species drop in richness for approximately every 2 kg N increase above the upper end of the critical load range. Interestingly, when non-UK European grasslands are included (not included in this report) a curvilinear response is also apparent (Stevens *et al*., 2010).

Table 20: Summary of relationships between nitrogen deposition and species richness/cover by habitat expressed as a percentage of the maximum in a habitat. Change in species richness associated with a 1 kg ha^{-1} y^{-1} difference in long-term N deposition along the survey sites is shown. Modelled relationship only applied over N deposition range in which survey sites fell, where no sites were surveyed at a given N deposition level '-' is shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load (kg N ha^{-1} yr^{-1})	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 1 kg increase in long-term N deposition at different background N deposition levels					
			5 kg N	**10 kg N**	**15 kg N**	**20 kg N**	**25 kg N**	**30 kg N**
All habitats (TU 2009)								
Total species richness	77 spp.	10-20	-1.6 % of maximum number of species/kg N increase					
Upland heath (TU 2009)								
Total species richness	42 spp.	10-20	-5.7 %	-2.9 %	-2.0 %	-1.4 %	-1.2 %	-1.0 %
Lichen species richness	11 spp.	10-20	-5.4 %	-2.7 %	-1.8 %	-1.8 %	-1.0 %	-1.0 %
Graminoid species richness	7 spp.	10-20	-7.0 %	-2.9 %	-2.9 %	-1.4 %	-1.4 %	-1.0 %
Graminoid cover	n/a	10-20	-0.5 %	no change	+0.4 %	+0.8 %	+1.2 %	+1.6 %
Upland heath (MRS)*								
Total species richness	16 spp.	10-20	-3.4 %	-3.1 %	-2.5 %	-1.9 %	-1.3 %	-0.3 %
Lowland heath (TU 2009)								
Total species richness	37 spp.	10-20	-6.2 %	-3.5 %	-2.2 %	-1.6 %	-1.4 %	-1.0 %
Moss species richness	12 spp.	10-20	-5.8 %	-2.5 %	-1.7 %	-1.7 %	-1.7 %	-0.9 %
Graminoid species richness	9 spp.	10-20	-17.8%	-4.4 %	-2.2 %	-1.1 %	-1.1 %	-0.5 %
Graminoid cover	n/a	10-20	-0.6 %	no change	+0.5 %	+1.05 %	+1.6 %	+2.2 %
Bog (TU 2009)								
Total species richness	32 spp.	5-10	-0.9 % of maximum number of species/kg N increase					
Lichen species richness	6 spp.	5-10	-1.7 %					
Forb species richness	6 spp.	5-10	-7.7 %	-3.9 %	-2.6 %	-1.9 %	-1.6 %	-1.3%
Graminoid cover	-	5-10	+1.5 % cover/kg N increase					
Sand dunes (TU 2009, all sites)								
Total species richness	77 spp.	8-15	-10.1%	-2.6 %	-1.2 %	-0.6 %	-	-
Moss species richness	16 spp.	8-15	-21.3%	-5.0 %	-2.5 %	-1.3 %	-	-
Graminoid cover	n/a	8-15	+8.6 %	+ 2.2 %	+ 1.0 %	+ 0.5 %	-	-
Forb species richness	33 spp.	8-15	-10.3%	-2.4 %	-1.2 %	-0.6 %	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total species richness	77 spp.	8-15	-4.4 %	-2.2 %	-1.4 %	-1.0 %	-	-
Moss species richness	16 spp.	8-15	-21.3%	-5.6 %	-2.5 %	-1.3 %	-	-
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)								
Total species richness	77 spp.	8-15	-4.4 %	-2.2 %	-1.4 %	-1.0 %	-	-
Moss species richness	16 spp.	8-15	-8.9 %	-4.4 %	-3.1 %	-2.5 %	-	-

Survey/ Habitat/	Max. species richness	Habitat/species critical load (kg N ha^{-1} yr^{-1})	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 1 kg increase in long-term N deposition at different background N deposition levels					
Acid grasslands (BEGIN)								
Total species richness	42 spp.	10-15	-1.5 %	-1.4 %	-1.2 %	-1.1 %	-1.0%	-0.9%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

The incremental effect of long-term N on species richness reduces as deposition levels increase above the upper end of the critical load for each habitat due to the curvilinear nature of the relationship between N and species richness. However, the positive, curvilinear relationship between graminoid cover in the heathlands means that graminoid cover increases dramatically above the critical load. This outcome is of key importance to site integrity, particularly within heathlands which have been shown to be vulnerable to conversion to grassland, most notably in the highly N-polluted areas of the Netherlands. Within the bog habitat, graminoid cover (principally the sedge, *Eriophorum vaginatum*) was found to increase by 1.5% per additional kg N across the deposition range studied suggesting that the balance between shrubs, graminoid and moss (mainly *Sphagnum* spp.) is at risk of moving towards dominance by sedge species. A similar result was obtained from the sand dune survey data although the relationship with N was weaker and more strongly associated with pH. Nevertheless, other studies have found sand dune integrity vulnerable to increases in graminoid cover (Remke *et al*., 2009).

It is also important to highlight the differing results between surveys, particularly the TU 2009 Upland Heath Survey and the Moorland Regional Survey (MRS). The reason behind this is the different quadrat sizes used to measure species richness: the TU survey used 2 x 2 m quadrats whilst the MRS used 0.5 x 0.5 m quadrats. Both produce valid measures of species richness although the MRS was more focussed on lower plants. For this reason the TU 2009 survey provides the strongest data as the bigger quadrats capture more of a site's biodiversity and therefore are more representative of the changes that occur across the dataset.

Table 21: Summary of relationships between long-term nitrogen deposition and species richness by habitat expressed as the amount of incremental N deposition (in kg N ha^{-1} yr^{-1}) associated with a reduction in species richness of one species along the survey gradient sites. Modelled relationship only applied over N deposition range in which survey sites occurred; where no sites were surveyed at a given N deposition level '-' is shown.

Survey/ Habitat/	Max. species richness	Habitat/ species critical load kg N ha^{-1} yr^{-1}	Increase in N deposition (in kg N ha^{-1} yr^{-1}) required to reduce measured species richness by 1 at different background long-term N deposition levels					
			5 kg N	10 kg N	15 kg N	20 kg N	25 kg N	30 kg N
Upland heath (TU 2009)								
Total species richness	42 spp.	10-20	0.4 kg	0.8 kg	1.3 kg	1.7 kg	2.0 kg	2.4 kg
Upland heath (MRS)*								
Total species richness	16 spp.	10-20	1.7 kg	2.0 kg	2.5 kg	3.3 kg	5.0 kg	20.0 kg
Lowland heath (TU 2009)								
Total species richness	37 spp.	10-20	0.4 kg	0.8 kg	1.3 kg	1.7 kg	2.0 kg	2.4 kg
Bog (TU 2009)								
Total species richness	32 spp.	5-10			3.3 kg			
Sand dunes (TU 2009, all sites)								
Total species richness	77 spp.	8-15	0.1 kg	0.5 kg	1.1 kg	2.0 kg	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total species richness	77 spp.	8-15	0.3 kg	0.6 kg	0.9 kg	1.3 kg	-	-
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)								
Total species richness	77 spp.	8-15	0.3 kg	0.6 kg	0.9 kg	1.3 kg	-	-
Acid grasslands (BEGIN)								
Total species richness	42 spp.	10-15	1.7 kg	1.7 kg	2.0 kg	2.0 kg	2.5 kg	2.5 kg

*in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Table 22: Summary of relationships between long-term nitrogen deposition and species cover (C) or probability of presence (P) by habitat expressed as a percentage of the maximum in a habitat. Difference in species richness associated with a 1 kg ha^{-1} y^{-1} difference in long-term N deposition along the survey sites is shown. Modelled relationship only applied over N deposition range in which survey sites fell, where no sites were surveyed at a given N deposition level '-' is shown. When the relationship between N and species richness was not significant 'ns' is shown.

Habitat (Survey) /Species	**Max cover/ presence (no. of quadrats)**	**Change in species cover expressed as a % of maximum species cover recorded in habitat with a 1 kg increase in long-term N deposition at different background N deposition levels**					
		5 kg N	**10 kg N**	**15 kg N**	**20 kg N**	**25 kg N**	**30 kg N**
Upland heath (TU 2009)							
Hylocomium splendens cover (C)	73	-2.0 %	-1.6 %	-1.2 %	-0.8 %	-0.4 %	-0.1
Deschampsia flexuosa (C)	37	-0.3 %	0.1 %	0.5 %	0.9 %	1.3 %	1.7
Upland heath (MRS)*							
Hylocomium splendens presence (P)	5	-1.3 %	-0.4 %	-0.2 %	-0.1 %	-0.1 %	-0.04
Campylopus introflexus (P)	2	0.0 %	0.02 %	0.04 %	0.07 %	0.10 %	0.13
Lowland heath (TU 2009)							
Hylocomium splendens (C)	31	-4.0 %	-1.1 %	-0.5 %	-0.3 %	-0.2 %	-0.1
Hylocomium splendens (P)	5	-0.4 %	-0.2 %	-0.1 %	-0.1 %	-0.1 %	-0.1
Cladonia portentosa (C)	11	-0.7 %	-0.3 %	-0.2 %	-0.2 %	-0.1 %	-0.1
Cladonia portentosa (P)	5	-0.5 %	-0.3 %	-0.2 %	-0.1 %	-0.1 %	-0.1
Brachythecium rutabulum (P)	5	0.0 %	0.1 %	0.1 %	0.2 %	0.2 %	0.3
Bog (TU 2009)							
Cladonia uncialis (C)	1	-0.1 %	0.0 %	0.0 %	-0.01 %	-0.01 %	-0.01
Cladonia uncialis (P)	4	-0.8 %	-0.2 %	-0.1 %	-0.06 %	-0.04 %	-0.03
Eriophorum vaginatum(C)	65	1.5 %					
Sphagnum fimbriatum (C)	1	0.0 %	0.0 %	0.0 %	0.01 %	0.01 %	0.01
Sphagnum fimbriatum (P)	3	0.2 %	0.1 %	0.1 %	0.04 %	0.03 %	0.03
Sand dunes (TU 2009 all sites)							
Hylocomium splendens (C)	30	-3.6 %	-1.0 %	-0.4 %	-0.3 %	-	-
Acid Grasslands (BEGIN)							
Hylocomium splendens (C)	11	-1.4 %	-0.4 %	-0.2 %	-0.1 %	-0.1 %	0.0
Hypnum cupressiforme (C)	19	0.2 %					
Nardus stricta cover (C)	42	0.3 %					
Carex panacea cover (C)	13	0.1 %					
Euphrasia officianlis cover (C)	1	-0.1 %	-0.1 %	0.0 %	0.0 %	0.0 %	0.0
Lotus corniculatus cover (C)	11	-0.2 %	-0.1 %	-0.1 %	-0.1 %	0.0 %	0.0

6.3 Applicability of this work to pollutant (NO_x and NH_3) concentrations and critical levels

The work in this chapter has focused upon the relationship between nitrogen deposition, critical loads and species richness, however, it is recognised that the concentration of a pollutant and its critical level may also influence species responses. Critical levels were identified in order to protect particular species or groups of plants, for example the critical level for NH_3 is 3 μgm^{-3} for vascular plants but 1 μgm^{-3} for lower plants such as lichens. By contrast critical loads are habitat specific.

Nitrogen deposition data is based on measured concentration data (for wet deposition, ion concentrations in precipitation and the amount of rainfall, and for dry deposition, gas concentrations in air, measured at a specified height, usually 1.5 m from the ground surface, which represent an average over a defined period). Thus deposition reflects pollutant concentration and over the longer-term, vegetation responses to changes in deposition would be comparable to changes in pollutant concentrations of a similar magnitude. It would therefore be reasonable to use the data in this chapter to approximate the response of vegetation to a percentage increase in concentration and to compare responses between changes in long-term deposition and long-term mean concentrations. This could be done by converting an increment in concentration to an increment in N deposition. However, this would only be relevant over the longer-term, and it is important to understand that the differing effects between concentration and deposition over time are unclear. High pollutant concentrations in particular may be very damaging, especially for lower plants (Pearce and van der Waal, 2008).

At present, air concentrations of dry deposited gases are regulated through critical levels. Gaseous NH_3 and NO_x concentrations can be measured relatively easily using passive samplers, normally exposed for one month, so that routine monthly measurements can be made to estimate an annual mean. However, the conversion of a gas concentration to an N load is not straightforward, as deposition velocity varies by species group, and meteorological conditions also affect deposition. In addition, the precise positioning of a new or expanding installation would also influence the frequency of high concentrations.

Much experimental evidence concerning the responses of a number of ecosystem types to changes in nitrogen deposition, mainly in form of wet NH_4NO_3, is available and we are confident that nitrogen deposition is in many cases the strongest driver of the changes in species richness and composition found in the survey data. However, the interactive effects of pollutant concentration and deposition are unclear and within the current report it was not the intention to attribute responses to specific forms of pollutant or changes in modelled concentration owing to co-correlation between these variables and the difficulty in separating out these across the survey sites.

Field experiments investigating the response of ecosystems to changing concentrations in wet deposition are rare and no contemporary experiments have studied the response of semi-natural ecosystems to changes in dry deposition of NO_x at realistic concentrations, however, one notable UK experiment has studied the response of bog vegetation to gaseous NH_3. The CEH Edinburgh Whim Bog experiment adds wet nitrogen deposition in oxidised and reduced N forms and dry-gaseous N deposition in the reduced form as NH_3 to a raised bog consisting mainly of ericoids, *Sphagnum* and cotton grass vegetation (Leith *et al*., 2004; Sheppard *et al*., 2004). Per unit N deposited, gaseous NH_3 has a much greater effect on soils and vegetation in comparison to similar deposition of the wet forms of N (Sheppard *et al*., 2011). The stronger responses to gaseous NH_3 are thought to be linked to effects on the foliage which may in part reflect the intermittent high concentrations, although deposition is not linearly linked to concentration (Jones, 2006) and applying the critical level

of ammonia (1 µg m^{-3}) to the bog provides greater protection than the critical load (5-10 kg N ha^{-1} y^{-1}).
In conclusion, caution should be applied when using the approach developed in this report to legislate for small incremental increases in pollutant concentration as the incremental responses developed within this report are not directly comparable between deposition and concentration and the level of uncertainty from a direct conversion of load to level would be quite large. Projected increases in deposition are more appropriate in understanding the likely long-term change and mean levels of concentration could be converted to deposition to give an indication of the changes that would occur over many decades. However, particular care should be taken where installations are likely to produce high levels of a pollutant over short-timescales as these may be very damaging to vegetation within close proximity to the source.

With the exception of the Whim Bog facility, dose response relationships to changes in N concentration are not fully understood and should be further researched experimentally. Further work on the datasets used in this report could also be carried out to study responses to changes in NH_3 concentration and deposition based on forthcoming 1 km gridded modelled pollutant datasets and the effect on bog vegetation based upon current experimental evidence. Given the commonalities between soil type and vegetation, this could also be used to experimentally support responses seen in some heathland systems and it may be possible to produce similar incremental relationships to those found with N deposition.

6.4 Summary

Even relatively small increases in long-term N deposition of 1 - 2 kg N $ha^{-1}yr^{-1}$ were found to have an impact on species richness across all habitats with the sand dune habitat appearing the most sensitive at low levels of N and the bog habitat the least. As N deposition increased upwards through the critical load range the rate of fall in species richness lessened but still existed and within the gradient of UK N deposition responses were still marked. These changes in rate of response reflected the curvi-linear relationships that were developed in Task 4.

Contrasting the falls in species richness, graminoid cover increased markedly with long-term N deposition and highlighted a potential threat to site integrity in the sand dunes, bogs and heaths. In the latter, cover increased exponentially and at some sites was above a level likely to cause concern and tip the balance between shrubs, lower plants and grasses.

The impacts illustrated in this chapter are calculated from the modelled relationships found in chapter 4. Only the relationships that were statistically significant were used to consider the impact of incremental N and in the most part these relationships were strong, however, with the exception of the acid grassland habitat, only 25-30 sites were visited within each habitat and this has produced some scatter within the data which is to be expected given the heterogeneous nature of site vegetation. The fact that the relationships found *were* so strong demonstrates the strength of the nitrogen signal in the data and if more sites were visited it is likely that the strength of the relationship would increase further.

The measures of species richness used in calculating the incremental effect of long-term N were obtained from a specific survey methodology that was consistently applied across all sites. The relationships produced are not rigid and they are not intended to be a precise prediction of the impact of incremental N although the direction of the impact of N (usually negative), the relative magnitude of the response and the shape of the response (often curvi-linear) would not be expected to change across an alternative selection of sites. In addition, many factors that impact biodiversity vary across the sites surveys including climate and other pollutants such as sulphur and these may also be correlated with changes in species richness. It is important to remember that these, and the relationships used in this report are based upon correlations and which does not imply causation, however, the findings of this work are also supported by significant literature on the subject and work from experimental sites which control for N addition.

The data used in this chapter illustrates what the effect of long-term N deposition has been, and could be used in the assessment of the relative effects of incremental increases in N over the long-term from new sources at different background levels of N deposition. The impact of N on individual species should be more cautiously used as many factors may influence the response of a species at a particular site and the data on individual species within the surveys was limited.

7. Task 6: Applicability of results to other habitats with limited dose-response information

7.1 Introduction

This report has studied the responses of vegetation to long-term N deposition within data from five semi-natural UK habitats: acid grasslands, upland heath, lowland heath, bog and sand dunes, however, evidence suggests that other important UK habitats are also negatively affected by N. Task 6 will consider whether the results and relationships presented within this report can be reasonably applied to other habitats given similarities in soil type and vegetation. Focus will be given to Calcareous grasslands, vegetated shingle, fens and deciduous woodlands.

7.2 Comparisons of responses between habitats

Reductions in total species diversity were found in all the habitats studied in this report. Figure 15 below compares the response curves within each based upon the percentage of the maximum number of species at a given N deposition. Both heaths showed similar response gradients, and this reflects the changes in species richness illustrated in Task 5.

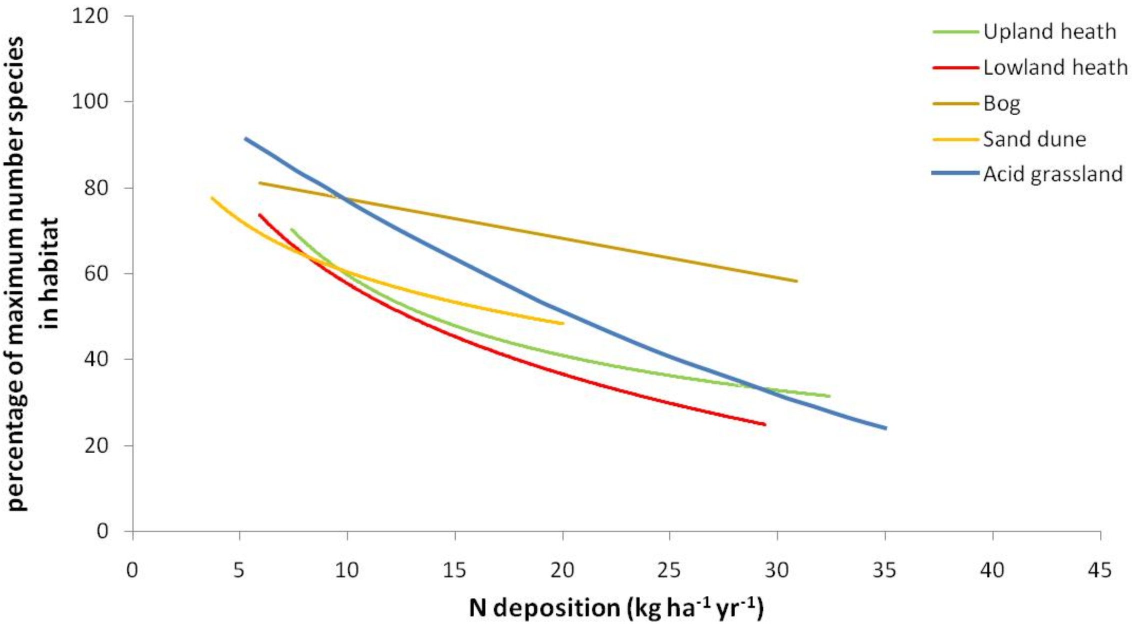


Figure 15: Modelled response curves showing the rate of change in species richness across the habitats studied as part of the TU 2009 multi-habitat survey.

Acid grasslands show a similar magnitude of response as the heaths, albeit less curvilinear, perhaps reflecting similarities in acidic, humus soil types. Changes in species richness in the bog habitat are shallower, indicative of the strong effects of hydrology limiting the response to N and a slightly less diverse habitat than the others. Sand dune response occurs more rapidly initially, then shallow over a much smaller range than the other habitats.

Whilst there is variability between the responses of the different habitats, the general magnitude of response is similar and when all the above data is considered together, as in Tasks 4 and 5, species richness reduces by 1.6% per 1 kg increase in long-term N. This broad approach could be used in similar habitats to those studied in this report, however, would be unsuitable for habitats which do not show change in species richness. An example of the latter would be calcareous grasslands where survey work has found that species richness does not appear affected by N deposition but species composition does change and the frequency of rare or scare plants reduces.

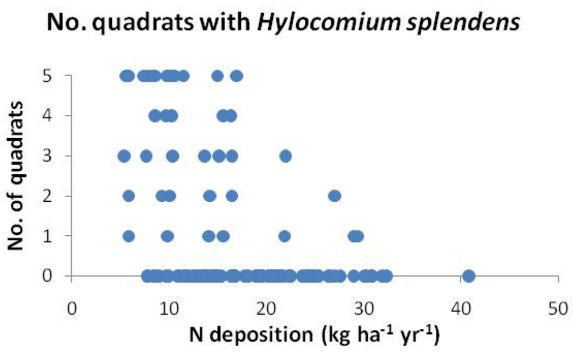


Figure 16. The presence of *Hylocomium splendens* across the habitats studied as part of the TU 2009 multi-habitat survey.

Task 3 showed that changes in overall diversity were often driven by changes in specific functional groups. Within upland heaths the reductions were apparent in total species richness, moss and lichens; in lowland heaths total species richness only; sand dunes moss species richness was strongly affected; in bogs forbs and lichens reduced and in acid grasslands forbs showed the strongest response. The lower plants, where present in significant numbers, are often the most sensitive to N. Of these, the moss *Hylocomium splendens* showed a remarkably consistent negative response to N across all the habitats except bogs, see Figure 16**,** with an abrupt reduction in probability of presence (number of quadrats the species was found in) above 17 kg N ha^{-1} yr^{-1}. The lichen *Cladonia portentosa* was also consistently negatively associated with N deposition in both upland and lowland heath sites and the bog survey.

7.3 Deciduous broadleaf woodland

Elevated nitrogen deposition has driven strong biogeochemical responses in woodlands with many authors documenting reductions in soil CN, acidification and increased nitrate leaching (Dise and Wright, 1995; Emmett *et al*., 1998; Dise *et al*., 2009). However, the impact of N deposition on vegetation composition is poorly understood partly due to the strong influence that tree canopy structure places on ground flora through inception of light, rainfall and pollution and the effect of woodland management and nitrogen deposition upon this structure.

Nevertheless, work has demonstrated that understory plants such as bryophytes, lichens and forbs can be negatively affected by N. Studies of mixed woodlands around four Scottish intensive livestock units showed marked changes in species composition within 300 m downwind of the units (Pitcairn *et al*., 1998; Pitcairn *et al*., 2009), the grasses *Deschampsia flexuosa* and *Holcus lanatus* increased in abundance close to the units as did the shrub *Rubus idaeus* and the forb *Urtica dioica*. Mosses in general were found to decrease in abundance downwind of the farm as did the forbs *Oxalis acetosella*, *Galium odoratum*, *Potentilla erecta* and *Dactylorhiza fuchsii*. A much broader scale survey of 103 woodlands in 1971 and revisited in 2001 found that overall species richness was unaffected by N but changes in composition were found with some species responding positively to N (*Poa nemoralis/trivialis, Galium aparine, Allium ursinum, Athyrium filix-femina, Carex pendula, Urtica dioica*) and others negatively (*Deschampsia flexuosa, Agrostis capillaris, Ajuga reptans, Holcus lanatus, Pteridium aquilinum, Vaccinium myrtillus*). The lack of an overall response in species richness was attributed to three main reasons: 1) much woodland ground flora tends towards the upper and middle of the Ellenberg spectrum; 2) impacts on woodlands may be from localised ammonia sources and unapparent over a national N gradient and 3) the interaction of canopy shading damping responses of the lower plants to N (Kirby *et al*., 2005).

Experimental work in Sweden well summarised by Cunha *et al*. (2002) found that N addition altered the composition of species towards *Deschampsia flexuosa* and ruderal species. In addition bryophyte abundance changed with increases in some *Brachythecium* species and reductions in others including *Hylocomium splendens*. These effects are similar to those found in the datasets studied in this report. Other studies have shown detrimental effects of N deposition on growth of bryophyte species *Isothecium myosuroides, Dicranum scoparium, Frullania tamarisci* in transplant experiments between Atlantic Oak woods (Mitchell *et al*., 2004) and shifts in species composition of epiphytic lichens between nitrophytes and acidophytes (Sutton *et al*., 2009).

A recent study has attempted identify the contribution of N deposition to vegetative change in addition to changes in woodland structure and age. Verheyen *et al*. (2012) studied data from over 1200 vegetation plots and found a shift towards shade tolerant and nutrient demanding species, however, N deposition did not explain these responses. Verheyen *et al* (2012) concluded that the effects of N deposition could be obscured by changes in the tree canopy and highlighted a potential N 'time bomb' which could explode when the canopy opens up again.

Some of the species responses highlighted by the above studies showed a significant response in the survey datasets in this report including the grass *Deschampsia flexuosa* which increased in cover in upland heaths supporting the findings by Pitcairn *et al*. (1998) but contradictory to Kirby *et al*., (2005), and the mosses *H. splendens* and *Brachythecium* species which showed respective decreased and increased in presence. This lack of an overall relationship between species richness and N deposition makes it difficult to assume a dose-response relationship to broad-scale N deposition in woodlands over a national gradient, however, it seems likely that the edges of the woodlands are likely to be more strongly affected by a nearby pollutant source such as an intensive livestock farm (Kirby *et al*., 2005).

7.4 Vegetated shingle

Vegetated shingle is an important habitat for conservation in the UK (Natural England, 2011) yet there is limited knowledge of its responses to N deposition and no experimental data from this community is available for analysis. Vegetated shingle community species composition shifts from pioneer species close to the sea shore able to tolerate high salinity and sea spray to more stable gravel communities consisting of grassland with important moss and lichen assemblages (UK BAP, 2008). In the survey datasets studied in this report the closest analogue are the acidic dune grasslands (sites with pH <6.5) which share their dry, acidic nature with the gravel communities of vegetated shingle.

Results from analysis of the acidic dune grasslands in this report suggests that nitrogen deposition (in the form dry-oxidised N) alongside climate plays an important role in determining species composition but no overall relationship between species richness and N deposition was found. This lack of a relationship between N and species richness is likely due to the limited number of acidic-dune sites that were surveyed (only 9). However, a recent study of 19 coastal dunes around the Baltic Sea on acid soils found that high N deposition increased growth of the sedge *Carex arenaria* and reduced the species richness of lichen, grass and forb species (Remke *et al*., 2009). These responses were strongest in the sites with the lowest pH suggesting the influence of acidification on species richness and the ability for *C. arenaria* to dominate under these conditions. The acidic-dune grassland survey data within this report showed that cover of *C. arenaria* appears to increase with N deposition although the number of data points is limited and the relationship non-significant. Moss species richness was most strongly correlated with Sulphur (S) deposition (in dunes with pH <6.5) and a correlation with N deposition was almost significant; in general a greater number of moss species were present at lower levels of N deposition. As discussed elsewhere in this report it is difficult to ascertain if this relationship with S is ecologically significant, however, both N and S deposition are likely to increase acidity and may be responsible for the decline in moss species richness and concurrent increases in *C. arenaria*.

Whilst caution should therefore be applied in attempting to extrapolate relationships between species richness and N deposition from the sand dune survey data presented it this report, it would seem reasonable to assume that the widespread reductions in species richness observed across all habitats, particularly in lower plant species, would also occur in vegetated shingle, especially above the recommended critical load range of 8-15 kg ha^{-1} yr^{-1}.

7.5 Fens

Fen nutrient budgets are characterized by inputs and outputs of nutrients via groundwater and surface water, and are tightly linked with local hydrology. The extent to which these systems receive and lose nutrients with in- and out-flowing water determines for a large part their sensitivity to excess N (ECE, 2010). Open wetland ecosystems such as floodplain fens would be expected to show little sensitivity to deposition of atmospheric N, whereas the impact on fens with a closed N cycle is likely to be much more significant. Fens with lowered water tables, as a result of drainage or over-abstraction, may also exhibit increased nutrient availability through decomposition of surface layers following drying, so knowledge of hydrological status of sites is important in assessing impact and risk.

Fens are found across a wide range of base cation and nutrient levels, from acid to strongly alkaline, and from oligotrophic to eutrophic. In all of these fen types, elevated N deposition has been found to increase cover of vascular plants including graminoids and dwarf shrubs and reduce cover of bryophytes (ECE, 2010). The vegetation of acidic, oligotrophic fens closely resembles the bog habitat studied in this report however; the critical load for this habitat has been estimated to be higher at 10-15 kg N ha^{-1} yr^{-1} compared with 5-10 kg N ha^{-1} yr^{-1} for bogs, due to the assumed higher buffering capacity of these minerotrophic fens. It has been recommended that the upper end of this range is used for soligenous, acidic valley mires, while the lower end of this range is used for quaking bogs and transition mires (ECE, 2010), which are in many cases functionally ombrotrophic systems.

Experimental work on a mesotrophic fen in Ireland (Verhoeven *et al*., 2011) has demonstrated significant responses to N additions of 35 and 70 kg ha^{-1} yr-^{1} on top of a low background N of 4-8 kg N ha^{-1} yr-^{1} and particularly to reduced-nitrogen. In these plots, vascular plant biomass increased strongly (from 170 g m^{-2} to 340 gm^{-2}) and bryophyte cover reduced considerably (from 350 g m^{-2} to 60 g m^{-2}) as did bryophyte species richness (from an average of 7 to 4). The authors attributed these changes in bryophytes to a combination of increased vascular plant cover and the direct toxic effect of ammonia. Other studies have also found N related increases in vascular plant biomass and shifts in vegetation composition in both mesotrophic and oligotrophic base-rich Dutch fens (Verhoeven and Schmitz, 1991; Verhoeven *et al*., 1996; Paulissen *et al*., 2004). Only limited evidence is available for the impacts of N on base-rich fens and the recommended critical load spans a wide range (15-30 kg N ha^{-1} yr-^{1}). The lower end of this range is recommended for oligotrophic types, such as montane fens,

These responses found in fens closely mirror those found in the bogs habitat survey and by the Whim Bog experiment discussed earlier in this report. It would seem reasonable therefore, given strong similarities in vegetation composition, soil type and hydrological regime to apply the relationships found in the bog survey data also to oligotrophic and mesotrophic fens, particularly in reference to changes in bryophyte, lichen and sedge cover and species richness around intensive livestock units with the caveat that the slightly higher pH may offer marginally greater protection from acidification, however, not from eutrophication.

8. Discussion and overall conclusion

The objective of this report was to determine the effects of incremental increases in long-term nitrogen deposition on species composition and richness for a range of different habitat types. This was carried out by examining recent vegetation survey data in order to understand the relationships that exist between species composition and richness and nitrogen (N) deposition.

Tasks 1 and 2 studied the available datasets and sought other supporting data that existed from N addition experiments and in recent published literature. The survey data chosen consisted of data from 226 sites, across 8 surveys and 5 UK priority habitats encompassing the Terrestrial Umbrella (TU) 2009 multi-habitat survey, the BEGIN UK Acid Grassland dataset, the 2006 TU Moorland Regional Survey (MRS) and a 2002 Sand dune survey.

Task 3 then used statistical techniques to understand the ecological responses within the survey data for each habitat, be it changes in overall species composition, species richness, functional groups or individual species abundance, and related these to key environmental variables including climate and pollutant deposition data. Table 23 below summarises the responses variables that were significantly related to nitrogen deposition resulting from this task.

Table 23: Summary of the response variables found during the statistical analysis of vegetation datasets in Task 3 that were significantly related to atmospheric nitrogen deposition. MRS = Moorland Regional Survey.

Response variable	Acid grassland	Bog	Upland heath	Lowland heath	Sand dune
Species composition	#	#	#	#	#
Total species richness	#	#	#	#	#
Bryophyte species richness			# (MRS only)	#	#
Lichen species richness		#	#		
Forb species richness	#	#			#
Graminoid species richness			#	#	
Graminoid cover		#	#	#	#

Task 4 examined the nature of the relationship between total species richness, the relevant functional groups and key individual species and N deposition. In many cases a strong, curvi-linear relationship existed between N and the response variables. This means that sites located in areas of low background N deposition responded to additional nitrogen with a greater fall in species richness than sites located in more polluted parts of the country. However, contrasting this, graminoid[2] cover *increased* with increasing N deposition in bog, heath and sand dune habitats. Depending on the specific graminoid species affected, and the balance between graminoids and other functional groups, this could have a negative effect on the condition of the site and prevent the site achieving its conservation objectives. This task also examined the data for levels or ranges of pollution over which an ecosystem appeared to be more sensitive and whilst these results are difficult to interpret it was

[2] Graminoid functional group includes species from the grass, sedge and rush families.

apparent that much ecological change occurred between approximately 14 to 23 kg N ha^{-1} yr^{-1}.

Task 5 used the relationships developed within Task 4 and quantified them in terms of change per *x* kg long-term N. This information could be used to understand the likely effect of additional increments in long-term nitrogen deposition (e.g. from a new pollution source) and assist with decisions making under the planning and/or environmental permitting regimes. It was apparent that even though a curvi-linear response dominates the relationship between the response variable and N deposition, significant falls in species richness were modelled for increases in long-term N deposition levels above the upper end of the critical load. Similarly, the positive relationship between graminoid cover and N in the sand dune, heath and bog habitats was reflected by marked increases in cover of grasses and sedges at the higher levels of long-term N. These simultaneous events may be interactive, with graminoids (and shrubs to some degree) enjoying a competitive advantage as N increases and shading out forbs and lower plants, or be a direct toxic effect of N on the lower and less nutrient-tolerant plants.

Consequently, the data suggests that habitat quality is affected across the UK N deposition range. The decline in species richness commences at the low end of the N deposition range and by upper end of the critical load range a substantial loss has already occurred. This reduces the inherent biodiversity of habitats through the loss of more sensitive and rare species. At higher loads of long-term N deposition beyond the critical load range, the integrity of sites may be threatened by graminoid domination and structural change to the habitat. Furthermore, all of the data sets examined as well as the reviewed published studies, show that at high rates of long-term N deposition, above for example 25 kg N ha^{-1} yr^{-1} (i.e. beyond the upper range of all critical loads of the communities addressed in this report) there is significant decline in species richness. Table 24 below summarises the relationships found in the vegetation datasets in Task 4 and indicates where these findings are supported by N addition experiments and the literature reviewed in this report. It should be highlighted that many of these responses are also supported by the broader scientific literature.

Table 24: Summary of the key findings from the analysis of the vegetation datasets and supporting reviews of literature and experimental site work. [1]Details of the UKREATE experiments in this report is presented in Tasks 2, 3 and 4, summarised in Phoenix *et al* (2012) and additionally in the literature cited in the table below. [2]For details of studies included in the literature review see Tasks 2 and 3 and the references in the table below.

Habitat	**Response curve shape from data analysis in this report**	**Direction of change**	**Supported by** UKREATE experiments[1]	Literature[2]
All habitats				
Total species richness (SR)	linear	↓	#	#
Upland heath				
Total SR	mild curvilinear	↓	*lichens and bryophytes only*	Edmondson *et al*, 2010; Stevens *et al*, 2009; Maskell *et al*, 2010; Payne *et al*, 2014
Lichen SR	mild curvilinear	↓	Carroll *et al*, 1999; Pilkington *et al*, 2007	Southon *et al*, 2013; Field *et al*, 2014
Graminoid SR	mild curvilinear	↓	*no*	Southon *et al*, 2013; Field *et al*, 2014
Graminoid cover	mild curvilinear	↑	*no*	Southon *et al*, 2013; Field *et al*, 2014

Habitat	Response curve shape from data analysis in this report	Direction of change	Supported by UKREATE experiments[1]	Literature[2]
Lowland heath				
Total SR	mild curvilinear	↓	*no*	Southon *et al*, 2013; Field *et al*, 2014
Moss SR	mild curvilinear	↓	*no (but lichen cover reduced)*	
Graminoid SR	mild curvilinear	↓	*no*	Southon *et al*, 2013; Field *et al*, 2014
Graminoid cover	mild curvilinear	↑	Barker *et al*, 2004	Southon *et al*, 2013; Field *et al*, 2014
Bog				
Total SR	linear			
Lichen SR	mild curvilinear	↓	*cover only* (Sheppard *et al*, 2011)	Field *et al*, 2014
Forb SR	mild curvilinear	↓		
Graminoid cover	linear	↑	(Sheppard *et al*, 2011)	Field *et al*, 2014
Sand dune				
Total SR	mild curvilinear	↓	*no*	Jones *et al*, 2004; Field *et al*, 2014
Moss SR	strong curvilinear	↓	*biomass only* (Plassman *et al*, 2009)	Remke *et al*, 2009; Field *et al*, 2014
Forb SR	mild curvilinear	↓	*no*	Remke *et al*, 2009; Field *et al*, 2014
Graminoid cover	mild curvilinear	↑	*no*	Jones *et al*, 2004; Remke *et al*, 2009
Acid grassland				
Total SR	curvi-linear	↓	*no*	Stevens *et al*, 2004; Maskell *et al*, 2010; Stevens *et al*, 2010
Forb SR	linear	↓	*no*	Stevens *et al*, 2006

Some individual species appeared to show a response at a specific point in the N deposition range, for example presence of *Hylocomium splendens* declined sharply above 17 kg N ha^{-1} yr^{-1}, whilst overall species richness changed more gradually. It is possible that many more species have a range of N pollution over which they can thrive up to a particular deposition load, however, the relatively small datasets used in this analysis has meant that some of the rarer species within each habitat are present at only a small number of locations. In these cases it was not possible to develop a mathematical relationship with N but each individual species contributed to an overall change in species composition and gradual fall in species richness. The relatively small datasets mean that caution should be applied when drawing conclusions on site integrity based on the presence or absence of individual species and that this information be used in conjunction with changes in species richness and composition.

Equally, some species groups responded in certain habitats (see Table 21) but not others. The reasons for these differences are not immediately obvious as many factors interact to govern how a habitat or species responds. In some cases the effect of a pollutant may be direct, for example a particular plant may be intolerant of N, and in others it could be a competitive interaction as discussed above. In addition to eutrophication and its fertilisation effect, N deposition can also acidify the soil. This could cause a response in rooted plants (and less in bryophytes and lichens), particularly in the poorly-buffered acidic habitats and less so in well-buffered calcareous habitats.

Due to a lack of root structure the lower plants are more directly responsive to current N and are likely to respond quickly to increases in pollution, whilst changes in the dominant species present within the vascular plant community are likely to occur more slowly in response to changes in soil N.   Management interaction such as grazing can also alter the competitive interaction between lower plants and faster growing higher plants through competition for light. The relationships determined in this report were based upon modelled recent annual nitrogen deposition, and no attempt was made to consider long-term cumulative N deposition which occurs over many years. Whilst current day N deposition can be used as a good proxy for long-term deposition and gives a strong indication of changes with regard to N deposition, it would be advised to further consider pollution over the longer-term. However, the overall findings of this report would not be expected to change dramatically if cumulative N deposition is used instead of present day N deposition.

These alternative mechanisms for change mean that species richness is a better indicator of N deposition in some habitats and changes in species composition more suitable in others. An example of the latter is the Calcareous Grassland habitat where existing survey work (Van den Berg *et al*., 2010) has suggested that species richness is not affected by increasing N but found changes in species composition and the less presence of rare or scarce species as N increases. New data studying the Calcareous Grassland habitat has been obtained as part of recent survey work by the BEGIN project and this should be reviewed to develop understanding further.

Whilst the general trend in reducing species richness as N increases holds true across the habitats that were studied in this report, not all of the habitats behaved in the same way. The bog habitat is probably affected more strongly by site hydrology whilst in sand dunes, precipitation and decalcification were the more dominant drivers. For bogs, this means that the species richness response to N is buffered by the hydrological status and the response curve is shallower per unit N than the habitats that are more freely drained. The approach used to predict the response to incremental N can still be used, however, sites with lower rainfall may be more sensitive to N and the relationships expressed in this report for the bog habitat should be regarded as conservative. For sand dunes, site location and rainfall drive acidity with the more acidic sites being less biodiverse. This situation is complicated by the fact that N (and sulphur) deposition can also acidify a habitat. The relationships found in Task 5 can still be used to understand the likely impact, however, limited data is available for sites with a pH less than 6.5 and a conservative approach would be to use the relationships obtained for the largest datasets. These are the TU 2009 all site dataset and the combined TU 2009 and 2002 surveys.

Task 6 used the relationships developed from the survey data and considered if they may be reasonably applied to habitats where significant data is not currently available. Many UK ecosystems share similarities in species and soil type and it seems likely that where this is the case similar responses to those found within this report would occur. However, local site conditions, management interactions affecting canopy structure, and natural variation within a habitat should be carefully considered when applying these relationships to a new habitat. Gaps in data do exist and mean that there remain large habitat types in the UK where the impact of N deposition is not fully understood, for example, woodlands, fens, vegetated shingle and mesotrophic grasslands and further work. Experimentally work and gradient studies should be performed to understand the changes that occur with increases in N. The incremental effect of changes in pollutant concentration is also not fully understood as the differing effects between concentration and deposition over time are unclear. High pollutant concentrations in particular may be very damaging, especially for lower plants and these should be further researched experimentally. Further work on the datasets used in this report could also be carried out to study the evidence of response to changes in NH_3 concentration and deposition based upon current experimental work.

The overall goal in the management of pollution deposition levels should be one of reduction but it should be stressed that lowering current N deposition may not reverse the declining trend in species richness. Indeed it is likely that lowering current N deposition may only slow the decline in species richness. Considerable N remains stored within the soil and management such as cutting or burning may only remove small amounts of the N pool. However, as the rate of decline may be slowed by a reduction in N, management may help tip the competitive balance in favour of lower plants that are not directly affected by the pollutant.

The effects of N deposition extend beyond its impact on biodiversity and species composition. Many experiments have demonstrated an impact on soil biodiversity, ecosystem services and the onset of leaching. Depending upon the ecosystem studied and the timescale of deposition, these responses can have broader implications upon water quality and aquatic systems. The focus of this report has been the study of botanical datasets, further work should be carried out to quantify the effects on incremental N on soils and biogeochemistry. The relationship between N concentration and ecosystem response is poorly understood and further work should be carried out experimentally to develop our knowledge.

The main weaknesses of the analyses performed in this report fall in two main areas. Firstly, due to the time-consuming survey methods used to obtain the data, and with the possible exception of the acid grassland habitat, the size of the datasets is inevitably limited. However, the fact that N deposition still comes through the analysis reflects its strength as a driver of change. Secondly, across the large north-south pollution gradient that exists in the UK a similar climatic gradient also occurs, with cooler, wetter sites in the north and warmer, drier sites in the south. The design of the surveys and choice of site locations attempted to cross these climatic boundaries as much as possible within a given habitat type by also choosing cooler, dryer sites and warmer, wetter sites. However, in a gradient survey it is difficult to completely separate the simultaneous and interactive effects of climate and pollutant.

Overall, in all of the analysis of species composition and species richness, N comes through as a strong driver of change and in many cases is the strongest driver. The fact that this does happen adds considerable assurance to the belief that N is driving considerable change within our semi-natural habitats. This is supported with many years of experimental manipulation of N with climatic change controlled for within the main habitats studied. This provides confidence in using the data presented in Task 5 of this report to understand the relative effects of incremental increases in N over the long-term from new or existing sources at different background levels of N deposition.

References

Asman, W. A. H., B. Drukker and A. J. Janssen (1988). "Modelled historil concentrations and depositions of ammonia and ammonium in Europe." Atmospheric Environment 22: 725-735.

Barker, C.G., 2001. The impact of management on heathland response to increased nitrogen deposition. PhD Thesis, Imperial College, London.

Barker, C. G., S. A. Power, J. N. B. Bell and C. D. L. Orme (2004). "Effects of habitat management on heathland response to atmospheric nitrogen deposition." Biological Conservation 120(1): 41-52.

Birks, H. J. B., and Gordon, A. D. 1985. Numerical methods in Quaternary pollen analysis. Academic Press, New York.

Bobbink, R., K. Hicks, J. Galloway, T. Spranger, R. Alkemade, M. Ashmore, M. Bustamante, S. Cinderby, E. Davidson, F. Dentener, B. Emmett, J. W. Erisman, M. Fenn, F. Gilliam, A. Nordin, L. Pardo and W. De Vries (2010). "Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis." Ecological Applications 20(1): 30-59.

Bobbink, R. and Hettelinghl, J-P. Eds. (2011) Review and revision of empirical critical loads and dose response relationships. Proceedings of an expert workshop, Noordwijkerhout, 23-25th June 2010.

Britton, A.J. and Fisher, J.M. (2007) Interactive effects of nitrogen deposition, fire and grazing on diversity and composition of low-alpine prostrate Calluna vulgaris heathland. Journal of Applied Ecology, 44, 125-135.

Caporn, S.J.M., Edmondson, J., Carroll, J.A., Pilkington, M., and Ray, N. (2007). Long term impacts of enhanced and reduced nitrogen deposition on semi-natural vegetation. UKREATE (2007) Terrestrial Umbrella: Effects of Eutrophication and Acidification on Terrestrial Ecosystems. CEH Contract Report. Defra Contract No. CPEA 18.

Carroll, J. A., S. J. M. Caporn, L. Cawley, D. J. Read and J. A. Lee (1999). "The effect of increased deposition of atmospheric nitrogen on Calluna vulgaris in upland Britain." New Phytologist 141: 423-431.

Clarke, K.R. (1993) Non-parametric multivariate analyses of changes in community structure. Australian Journal of Ecology, 18, 117-143.

Cunha, A., Power, S. A., Ashmore, M. R., Green, P. R. S., Haworth, B. J. and Bobbink, R. (2002) Whole ecosystem nitrogen manipulation: an updated review. JNCC: 126.

Dise, N. B. and R. F. Wright (1995). "Nitrogen leaching from European forests in relation to nitrogen deposition." Forest Ecology and Management 71(1-2): 153-161.

Dise, N. B., J. J. Rothwell, V. Gauci, C. van der Salm and W. de Vries (2009). "Predicting dissolved inorganic nitrogen leaching in European forests using two independent databases." Science of the Total Environment 407(5): 1798-1808.

Dise, N. B., Ashmore, M., Belyazid, S. Aleeker, A., Bobbink, R., de Vries, W., Erisman, J.W., Spranger, T., Stevens, C.J. and van den Berg, L. (2011) Nitrogen as a threat to European terrestrial biodiversity. In: Th e European Nitrogen Assessment, eds M. A. Sutton, C. M. Howard, J. W. Erisman *et al*. Cambridge University Press.

Dupre, C., C. J. Stevens, T. Ranke, A. Bleeker, C. Peppler-Lisbach, D. J. G. Gowing, N. B. Dise, E. Dorland, R. Bobbink and M. Diekmann (2010). "Changes in species richness and composition in European acidic grasslands over the past 70 years: the contribution of cumulative atmospheric nitrogen deposition." Global Change Biology 16(1): 344-357.

ECE/EB.AIR/WG.1/2010/14, 2010. Empirical critical loads and dose-response relationships. Prepared by the Coordination Centre for Effects of the International Cooperative Programme on Modelling and Mapping Critical Levels and Loads and Air Pollution Effects, Risks and Trends, Report to the Working Group on Effects; http://www.unece.org/env/lrtap/WorkingGroups/wge/29meeting_Rev.htm

Edmondson, J.L. (2007) Nitrogen pollution and the ecology of heather moorland. PhD. thesis. Manchester Metropolitan University.

Edmondson, J. L., Carroll, J. A., Price, E. A. C. and Caporn, S. J. M. (2010) "Bio-indicators of nitrogen pollution in heather moorland." Science of the Total Environment, 408(24), 6202-6209.

Emmett, B. A., D. Boxman, M. Bredemeier, P. Gundersen, O. J. Kjønaas, F. Moldan, P. Schleppi, A. Tietema and R. F. Wright (1998). "Predicting the Effects of Atmospheric Nitrogen Deposition in Conifer Stands: Evidence from the NITREX Ecosystem-Scale Experiments." Ecosystems 1(4): 352-360.

Emmett, B. A. (2007) Nitrogen saturation of terrestrial ecosystems: Some recent findings and their implications for our conceptual framework. Water Air and Soil Pollution, Focus 7, 99-109.

Emmett, B.A., Griffiths, B., Williams, D., and Williams, B. (2007) Task 6: Interactions between grazing and nitrogen deposition at Pwllpeiran: Effects of eutrophication and acidification on terrestrial ecosystems. Final Report (July 2007) NERC-Defra Terrestrial Umbrella (Contract No CPEA18).

Emmett, B.A, Rowe, E.C, Stevens, C.J, Gowing, D.J, Henrys, P.A, Maskell, L.C. and Smart, S.M. (2011) Interpretation of evidence of nitrogen impacts on vegetation in relation to UK biodiversity objectives. JNCC Report, No. 449.

Field, C.D. (2010). The effect of nitrogen deposition on carbon sequestration in semi-natural ericaceous dominated ecosystems. PhD Thesis, Manchester Metropolitan University, Manchester, UK.

Field, C., Dise, N., Payne, R., Britton, A., Emmett, B., Helliwell, R., Hughes, S., Jones, L., Lees, S., Leake, J., Leith, I., Phoenix, G., Power, S., Sheppard, L., Southon, G., Stevens, C. and Caporn, S. M. (2014). "The Role of Nitrogen Deposition in Widespread Plant Community Change Across Semi-natural Habitats." Ecosystems: 1-14.

Fowler, D., O'Donoghue, M., Muller, J.B.A., Smith, R.J., Dragosits, U., Skiba, U., Sutton, M.A. and Brimblecombe, P. (2004) A chronology of nitrogen deposition in the UK between 1900 and 2000. Water, Air and Soil Pollution Focus, 4, 9-23.

Galloway, J. N. (1995). "Acid deposition: Perspectives in time and space." Water, Air, & Soil Pollution 85(1): 15-24.

Galloway, J.N., F.J. Dentener, D.G. Capone, E.W. Boyer, R.W. Howarth, S.P. Seitzinger, G.P. Asner, C.C. , Cleveland, P.A. Green, E.A. Holland, D.M. Karl, A.F. Michaels, J.H. Porter, A.R. Townsend, and C.J. Vorosmarty. (2004) Nitrogen cycles: Past, present and future. Biogeochemistry, 70, 153-226.

Gimingham, C. H. (1972) Ecology of Heathlands. London, Chapman and Hall Limited, 242.

Gordon, A.D. and Birks, H.J.B. (1972) Numerical methods in Quaternary palaeoecology. I. Zonation of pollen diagrams. New Phytologist, 71, 961.

Green ER (2005) The effect of N deposition on lowland heath ecosystems. PhD thesis, Imperial College London, London, UK.

Grimm, E.C. (1987) CONISS: A Fortran 77 program for stratigraphically constrained cluster analysis by the method of incremental sum of squares. Computers and Geosciences, 13, 13-35.

Hall, J., Dore, A., Heywood, E., Broughton, R., Stedman, J., Smith, R., and O'Hanlon, S. (2006) Assessment of the Environmental impacts Associated with the UK Air Quality Strategy: London, DEFRA.

Hall, J., Emmett, B., Garbutt, A., Jones, L., Rowe, E., Sheppard, L., Vanguelova, E., Pitman, R., Britton, A., Hester, A., Ashmore, M., Power, S. and Caporn, S. (2011) UK Status Report March 2011: Update to empirical critical loads of nitrogen. Report to Defra under contract AQ801 Critical Loads and Dynamic Modelling. http://cldm.defra.gov.uk/Status_Reports.htm

Hammer, O., Harper, D.A.T. and Ryan, P.D. (2001) Paleontological Statistics software package for education and data analysis. Paleontologica Electronica, 4(1), 9 pp.

Horswill, P., O'Sullivan, O., Phoenix, G. K., Lee, J. A. and Leake, J. R. (2008) Base cation depletion, eutrophication and acidification of species-rich grasslands in response to long-term simulated nitrogen deposition. Environmental Pollution, 155(2), 336-349.

JNCC (2004). Common Standards Monitoring Guidance for Lowland Heathland. JNCC. ISSN: 1743-8160. http://jncc.defra.gov.uk/pdf/CSM_lowland_heathland.pdf

JNCC (2006). Common Standards Monitoring Guidance for Upland Habitats. JNCC. ISSN: 1743-8160. http://jncc.defra.gov.uk/pdf/CSM_Upland_Oct_06.pdf

Jones, M.L.M., Wallace, H.L., Norris, D., Brittain, S.A., Haria, S., Jones, R.E., Rhind, P.M., Reynolds, B.R., and Emmett, B.A. (2004) Changes in vegetation and soil characteristics in coastal sand dunes along a gradient of atmospheric nitrogen deposition. Plant Biology, 6(5), 598-605.

Jones, M.R. (2006). Ammonia deposition to semi-natural vegetation. PhD Thesis. University of Dundee.

Juggins, S. (1992). The ZONE program, version 1.2, Unpublished program, University of Newcastle.

Kirby, K.J., Smart, S.M., Black, H.I.J., Bunce, R.G.H.,Corney,P.M.and Smithers,R.J. (2005). *Long term ecological change in British woodland* (1971-2001). Peterborough: English Nature (Research Report 653).

Leith, I., L. Sheppard, D. Fowler, J. N. Cape, M. Jones, A. Crossley, K. Hargreaves, Y. S. Tang, M. Theobald and M. Sutton (2004). "Quantifying Dry NH3 Deposition to an Ombrotrophic Bog from an Automated NH3 Field Release System." Water, Air, & Soil Pollution: Focus 4(6): 207-218.

Leps, J. and Smilauer, P. (2003) Multivariate analysis of ecological data using CANOCO. Cambridge University Press, Cambridge, 260pp.

Maskell, L. C., S. M. Smart, J. M. Bullock, K. E. N. Thompson and C. J. Stevens (2010). "Nitrogen deposition causes widespread loss of species richness in British habitats." Global Change Biology 16(2): 671-679.

Mitchell, R.J., Sutton, M.A., Truscott, A.M., Leith, I.D., Cape, J.N., Pitcairn, C.E.R. and van Dijk, N. (2004) Growth and tissue nitrogen of epiphytic Atlantic bryophytes: effects of increased and decreased atmospheric N deposition. Functional Ecology, 18, 322-329.

Morecroft, M.D.; Skellers, E.K.; Lee, J.A. 1994 An experimental investigation into the effects of atmospheric nitrogen deposition on two semi-natural grasslands Journal of Ecology 82 475-483.

Morse, J.N. (1980) Reducing the size of the non-dominated set: Pruning by clustering, Computers and Operations Research, 7, 55-66.

Natural England (2011) UK list of priority habitats and species. http://www.naturalengland.org.uk/ourwork/conservation/biodiversity/protectandmanage/prioritylist.aspx

Paulissen, M. P. C. P., van der Ven, P. J. M., Dees, A. J. and Bobbink, R. (2004) Differential Effects of Nitrate and Ammonium on Three Fen Bryophyte Species in Relation to Pollutant Nitrogen Input. New Phytologist, 164(3), 451-458.

Payne, R., Caporn, S.J.M., Field, C., Carroll, J., Edmondson, J., Britton, A. and Dise, N. (2014). "Heather Moorland Vegetation and Air Pollution: A Comparison and Synthesis of Three National Gradient Studies." Water, Air, & Soil Pollution 225(7): 1-13.

Pearce, I. S. K. and R. Van der Wal (2008). "Interpreting nitrogen pollution thresholds for sensitive habitats: The importance of concentration versus dose." Environmental Pollution 152(1): 253-256.

Phoenix, G.K., Hicks, W.K., Cinderby, S., Kuylenstierna, J.C.I, Stock, W.D., Dentener, F.J., Giller, K.E., Austin, A.T., Lefroy, R.D.B., Gimeno, B.S., Ashmore, M.R. and Ineson, P. (2006) Atmospheric Nitrogen Deposition in World Biodiversity Hotspots: the need for a greater global perspective in assessing N deposition impacts. Global Change Biology, 12, 470-476.

Phoenix, G. K., Emmett, B. A., Britton, A. J., Caporn, S. J. M., Dise, N. B., Helliwell, R., Jones, L., Leake, J. R., Leith, I. D., Sheppard, L. J., Sowerby, A., Pilkington, M. G., Rowe, E. C., Ashmorek, M. R. and Power, S. A. (2012). "Impacts of atmospheric nitrogen deposition: responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments." Global Change Biology 18(4): 1197-1215.

Pilkington, M. G., Caporn, S. J. M., Carroll, J. A., Cresswell, N., Lee, J. A., Emmett, B. A. and Johnson, D. (2005). "Effects of increased deposition of atmospheric nitrogen on an upland Calluna moor: N and P transformations." Environmental Pollution 135(3): 469.

Pilkington, M. G., Caporn, S. J. M., Carroll, J. A., Cresswell, N., Lee, J. A., Emmett, B. A. and Bagchi, r. (2007). "Phosphorus supply influences heathland responses to atmospheric nitrogen deposition." Environmental Pollution 148(1): 191-200.

Pitcairn, C. E. R., I. D. Leith, L. J. Sheppard, M. A. Sutton, D. Fowler, R. C. Munro, S. Tang and D. Wilson (1998). "The relationship between nitrogen deposition, species composition and foliar nitrogen concentrations in woodland flora in the vicinity of livestock farms." Environmental Pollution 102(1, Supplement 1): 41-48.

Pitcairn, Carole E.R.; Leith, Ian D.; van Dijk, Netty; Sheppard, Lucy J.; Sutton, Mark A.; Fowler, David. 2009 The application of transects to assess the effects of ammonia on woodland groundflora. In: Sutton, Mark A.; Reis, Stefan; Baker, Samantha M.H., (eds.) Atmospheric Ammonia: Detecting emission changes and environmental impacts. Results of an Expert Workshop under the Convention on Long-range Transboundary Air Pollution. Springer, 59-69.

Plassmann, K., Edwards-Jones, G., and Jones, M.L.M. (2009) The effects of low levels of nitrogen deposition and grazing on dune grassland. Science of the Total Environment, *407(4), 1391-1404.*

Power, S. A., M. R. Ashmore and D. A. Cousins (1998). "Impacts and fate of experimentally enhanced nitrogen deposition on a British lowland heath." Environmental Pollution 102(1, Supplement 1): 27-34.

Power, S.A., Green, E.R., Barker, C.G. & J.N.B. and Ashmore, M.R. (2006) Ecosystem recovery: heathland response to a reduction in nitrogen deposition. Global Change Biology, 12, 1241-1252.

Remke, E., Brouwer, E., Kooijman, A., Blindow, I. and Roelofs, J. G. M. (2009) Low atmospheric nitrogen enrichment loads leads to grass encroachment in coastal dunes, but only on acid soils. Ecosystems, 12, 1173-1188.

RoTAP (2012). Review of Transboundary Air Pollution: Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Contract Report to the Department for Environment, Food and Rural Affairs. Centre for Ecology & Hydrology.

Sheppard, L. J., Crossley, A., Leith, I. D., Hargreaves, K. J., Carfrae, J. A., Dijk, N. v., Cape, J. N., Sleep, D., Fowler, D. and Raven, J. A. (2004) An automated wet deposition system to compare the effects of rediced and oxidised N on ombrotrophic bog species: practical considerations. Water, Air, and Soil Pollution, 4, 197-205.

Sheppard, L. J., Leith, I. D., Crossley, A., Van Dijk, N., Fowler, D., Sutton, M. A. and Woods, C. (2008) Stress responses of Calluna vulgaris to reduced and oxidised N applied under `real world conditions'. Environmental Pollution, 154(3), 404-413.

Sheppard, L.J., Leith, I.D., Kivimaki, S.K., Gaiawyn, J. (2011) The form of reactive nitrogen deposition is important for the provision of ecosystem services. In: Nitrogen Deposition, Critical Loads and Biodiversity (Eds. Sutton M.A. *et al*.) (Proceedings of the INI/CLRTAP/CBD Expert Workshop, 16-18 November 2009).

Southon, G. E., Field, C., Caporn, S. J. M., Britton, A. J. and Power, S. A. (2013). "Nitrogen Deposition Reduces Plant Diversity and Alters Ecosystem Functioning: Field-Scale Evidence from a Nationwide Survey of UK Heathlands." PLoS ONE 8(4): e59031.

Stevens, C. J., Dise, N. B., Mountford, J. O. and Gowing, D. J. (2004) Impact of nitrogen deposition on the species richness of grasslands. Science, 303, 1876-1879.

Stevens, C. J., N. Dise, D. J. Gowing and J. O. Mountford (2006). "Loss of forb diversity in relation to nitrogen deposition in the UK: regional trends and potential controls." Global Change Biology 12: 1823-1833.

Stevens, C. J., Caporn. S.J.M., Maskell, L. C., Smart, S. M. Dise, N. and Gowing, D. J. (2009). Detecting and attributing air pollution impacts during SSSI condition assessment. , JNCC report 426.

Stevens, C. J., Thompson, K., Grime, J. P., Long, C. J. and Gowing, D. J. (2010a) Contribution of acidification and eutrophication to declines in species richness of calcifuge grasslands along a gradient of atmospheric nitrogen deposition. Functional Ecology, 24, 478-484.

Stevens, C.J., Dupre, C., Dorland, E., Gaudnik, C., Gowing, D., Bleeker, A., Diekmann, M., Alard, D., Bobbink, R., Fowler, D., Corcket, E., Mountford, J.O., Vandvik, V., Aarrestad, P.A., Muller, S. and Dise, N.B. (2010b). Nitrogen deposition threatens species richness of grasslands across Europe. Environmental Pollution, 158, 2940-2945.

Stevens, C.J., Smart, S.M., Henrys, P., Maskell, L.C., Walker, K.J., Preston, C.D., Crowe, A., Rowe, E., Gowing, D.J. and Emmett, B.A. (2011). Collation of evidence of nitrogen impacts on vegetation in relation to UK biodiversity objectives. JNCC report 447.

Sutton, M.A., Wolseley, P.A., Leith, I.D., van Dijk, N., Tang, Y.S., James, P.W., Theobald, M.R., and Whitfield, C. (2009) Estimation of the Ammonia critical levels for epiphytic lichens based on observations at farm, landscape and national scales. In Sutton, M.A., Reis, S. and Baker, S.M.H. (Eds.) (2009). Atmospheric Ammonia. Springer Press. 464pp.

ter Braak, C.J.F. and Smilauer, P. (2004) Canoco Software for Windows 4.53. Biometris Plant Research International, Wageningen, The Netherlands.

UKBAP (2008). Coastal vegetated shingle. UK Biodiversity Action Plan; Priority Habitat Descriptions.

UKREATE (2010) Terrestrial Umbrella: Effects of Eutrophication and Acidification on Terrestrial Ecosystems. CEH Contract Report NEC03425. Defra Contract No. AQ0802.

Van Den Berg, L. J. L., Vergeer, P., Rich, T. C. G., Smart, S. M., Guest, D. A. N. and Ashmore, M. R. (2010) Direct and indirect effects of nitrogen deposition on species composition change in calcareous grasslands. Global Change Biology.

Verheyen, K., Baeten, L., De Frenne, P., Bernhardt-Römermann, M., Brunet, J., Cornelis, J., Decocq, G., Dierschke, H., Eriksson, O., Hédl, R., Heinken, T., Hermy, M., Hommel, P., Kirby, K., Naaf, T., Peterken, G., Petřík, P., Pfadenhauer, J., Van Calster, H., Walther, G.-R., Wulf, M. and Verstraeten, G. (2012). "Driving factors behind the eutrophication signal in understorey plant communities of deciduous temperate forests." Journal of Ecology 100(2): 352-365.

Verhoeven, J. T. A. and Schmitz, M. B. (1991) Control of plant growth by nitrogen and phosphorus in mesotrophic fens. Biogeochemistry, 12(2), 135-148.

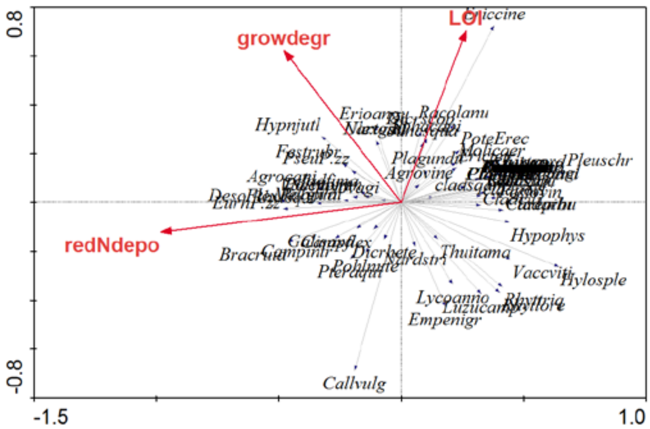
Verhoeven, J., Beltman, B. and De Caluwe, H. (1996) Changes in plant biomass in fens in the vechtplassen area, as related to nutrient enrichment. Aquatic Ecology, 30(2), 227-237.

Verhoeven, J. T. A., B. Beltman, E. Dorland, S. A. Robat and R. Bobbink (2011). "Differential effects of ammonium and nitrate deposition on fen phanerogams and bryophytes." Applied Vegetation Science 14(2): 149-157.

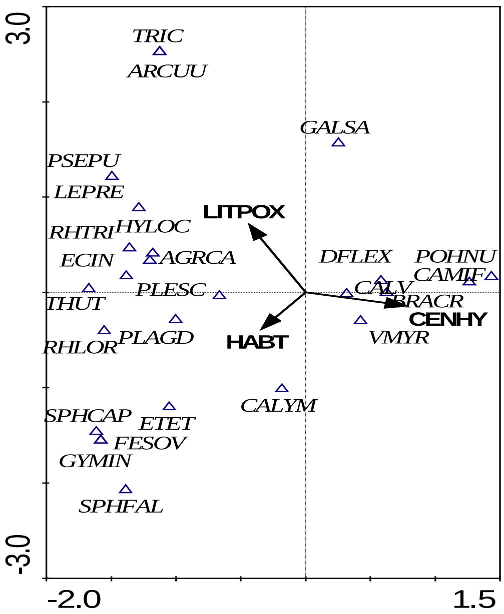
Ward, J.H. (1963) Hierarchical grouping to optimize an objective function. Journal of the American Statistical Association, 58, 236-244.

Appendix 1: Key ordination plots

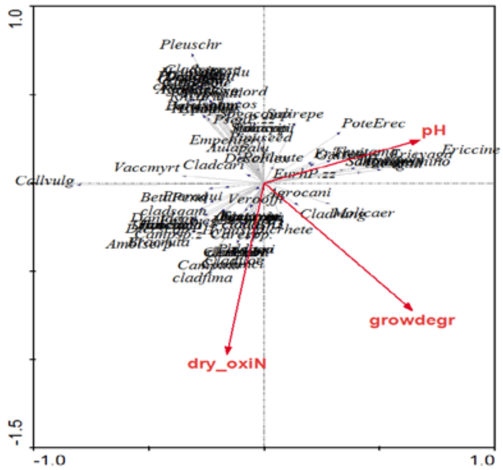
Upland heaths (TU 2009): RDA ordination plot



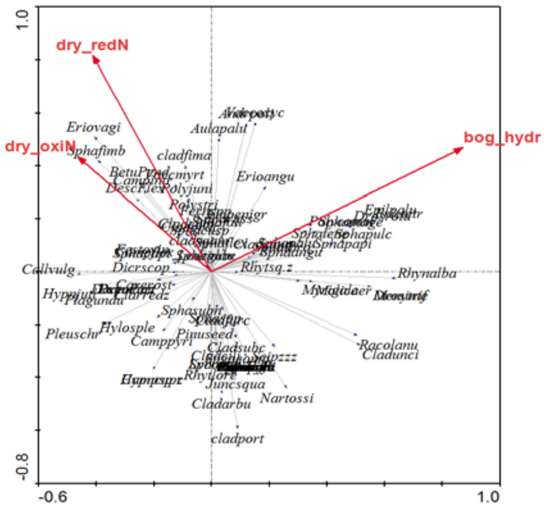
Upland heaths (MRS): CCA ordination plot



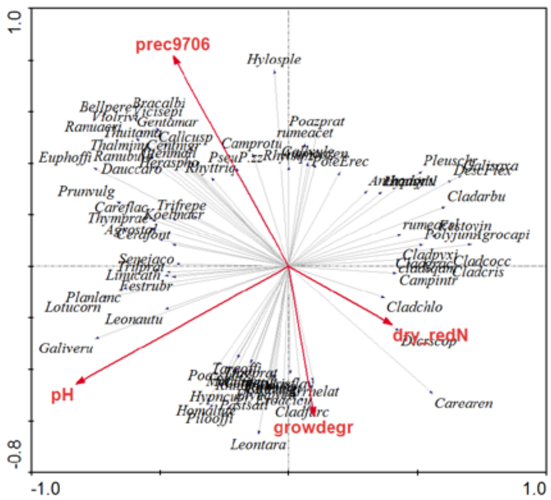
Lowland heaths (TU 2009): RDA ordination plot



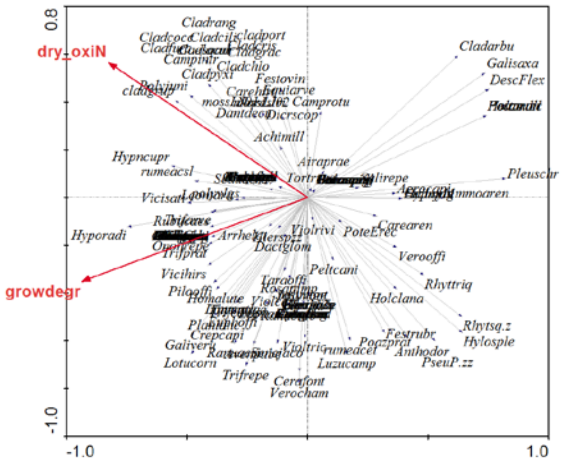
Bog (TU 2009): RDA ordination plot



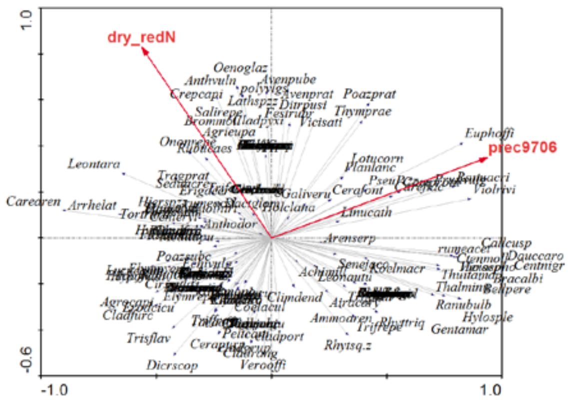
Sand dune (TU 2009 all pH): RDA ordination plot showing response of species with 15% minimum fit to axes



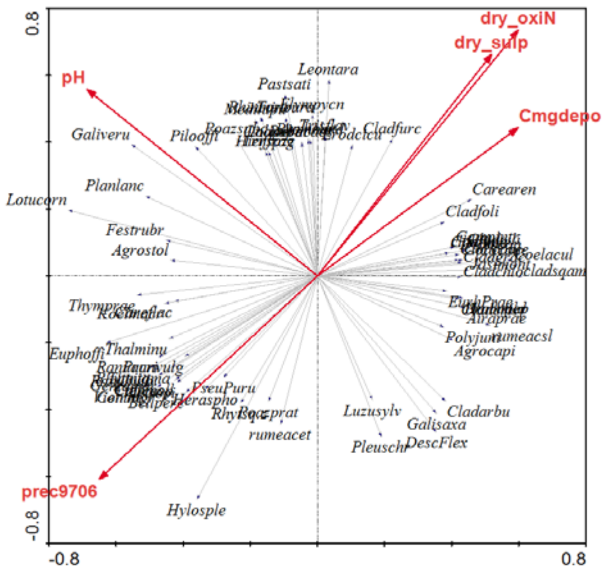
Sand dune (TU 2009 pH< 6.5): RDA ordination plot



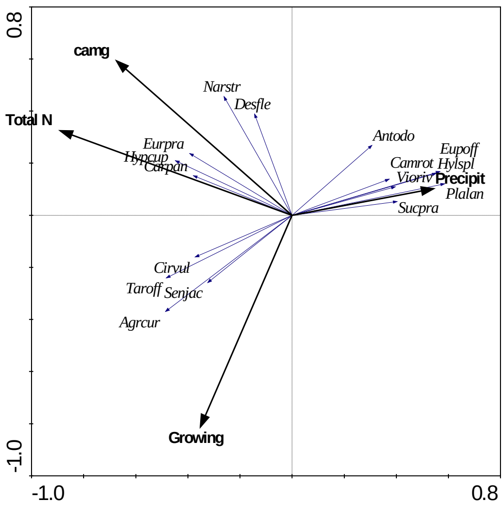
Sand dune (TU 2009 pH≥ 6.5): RDA ordination plot



Sand dune (TU 2009 all pH plus 2002 fixed-dune grasslands): RDA ordination plot showing response of species with 15% minimum fit to axes



Acid grasslands (BEGIN): RDA ordination plot

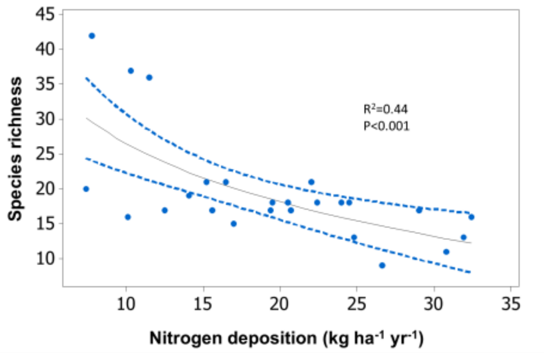


Appendix 2: Species richness nitrogen response curves.

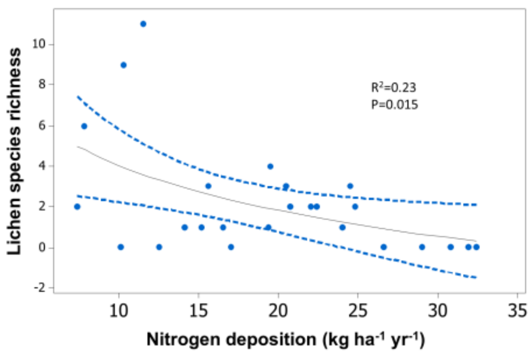
Only those with a significant relationship to nitrogen deposition are shown.

Upland heath: TU Survey 2009

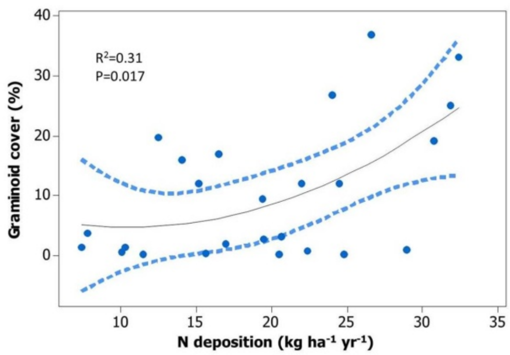
Upland heath: total species richness



Upland heath: lichen species richness

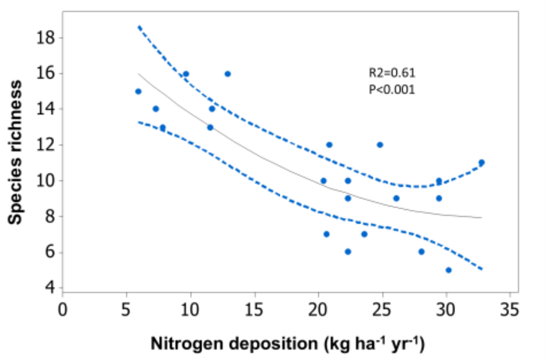


Upland heath: graminoid cover



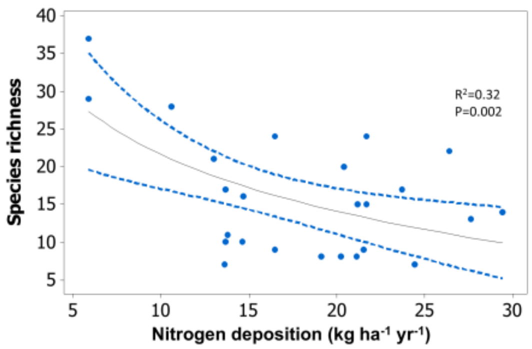
Upland heath: MRS

Upland heath: total species richness

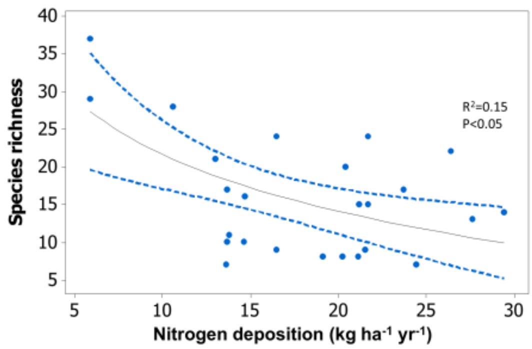


Lowland heath: TU Survey 2009

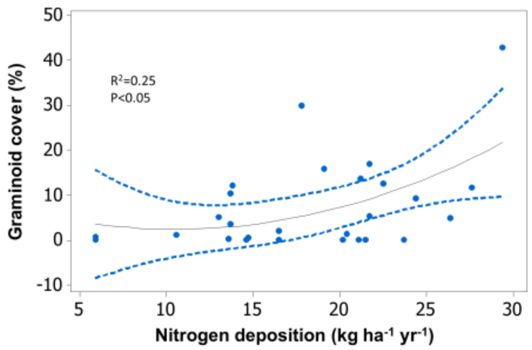
Lowland heath: total species richness



Lowland heath: moss species richness

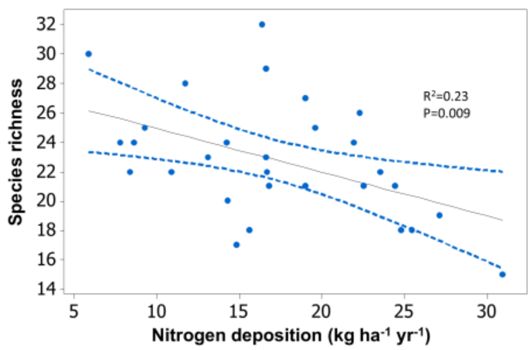


Lowland heath: graminoid cover

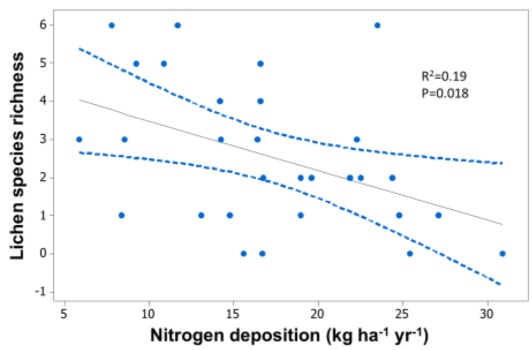


Bog: TU Survey 2009

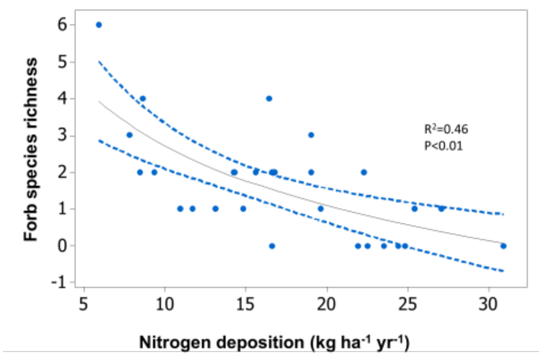
Bogs: total species richness



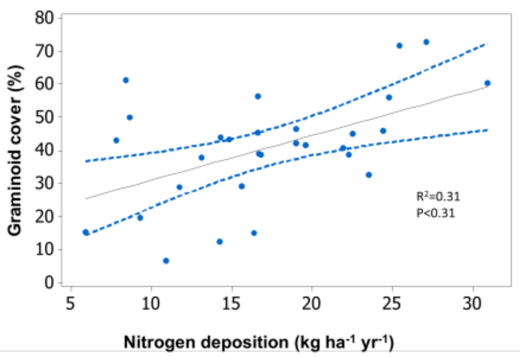
Bogs: lichen species richness



Bogs: forb species richness

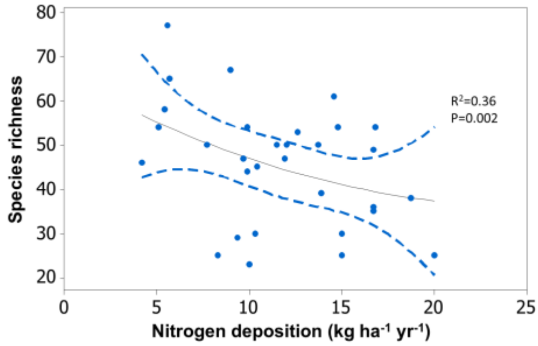


Bogs: graminoid cover

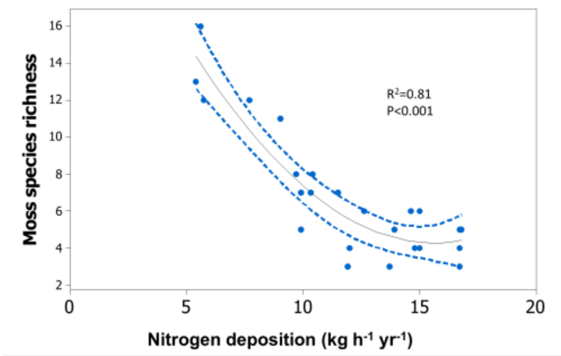


Sand dunes (TU 2009 all pH)

Sand dunes: total species richness

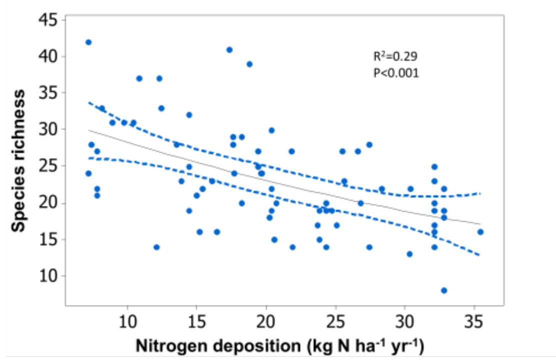


Sand dunes: moss species richness



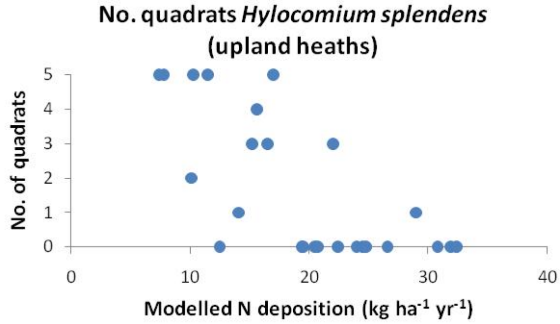
Acid grasslands (BEGIN)

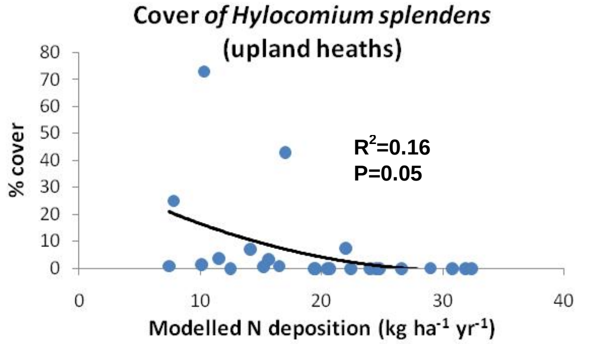
Acid grassland: total species richness

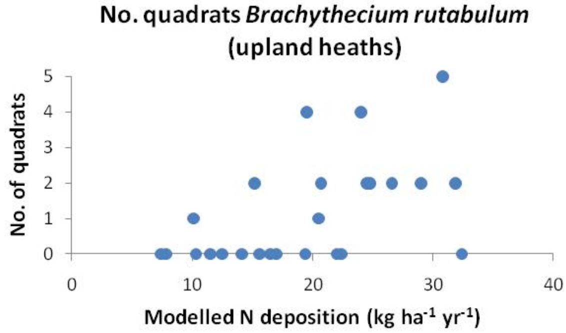


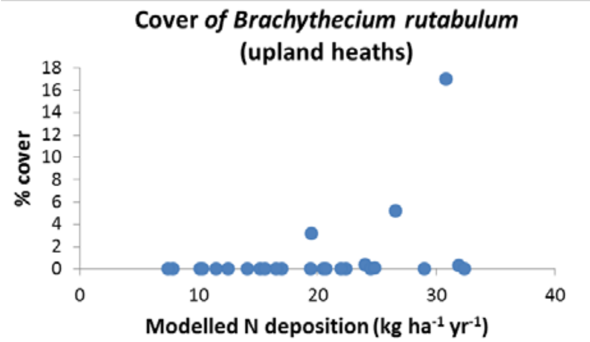
Appendix 3: Individual species N response curves

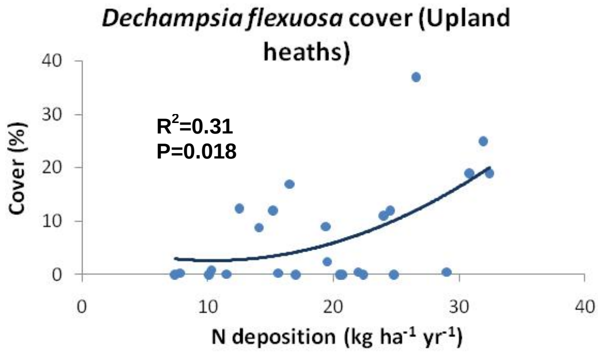
Upland heaths: TU 2009



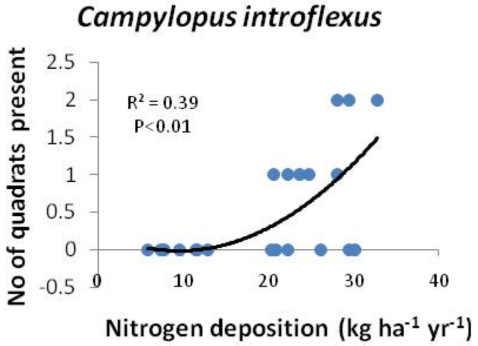


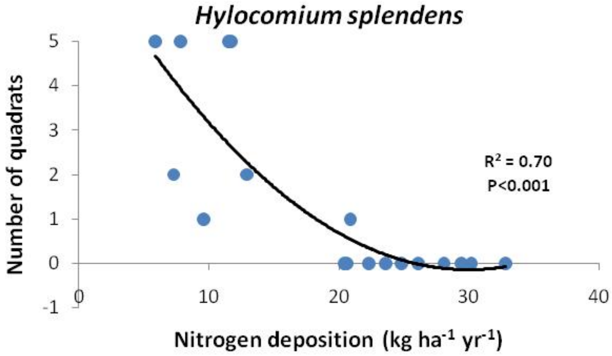




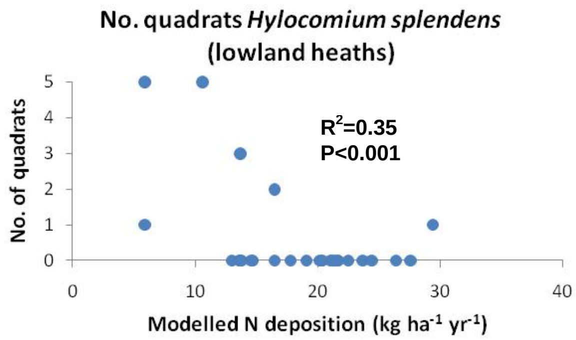


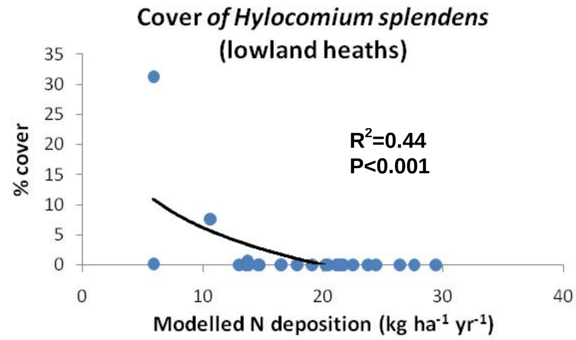
Upland heaths (MRS)

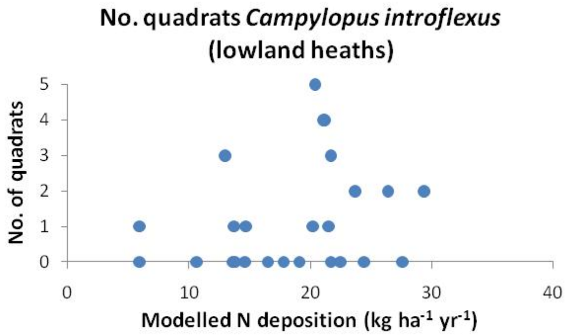


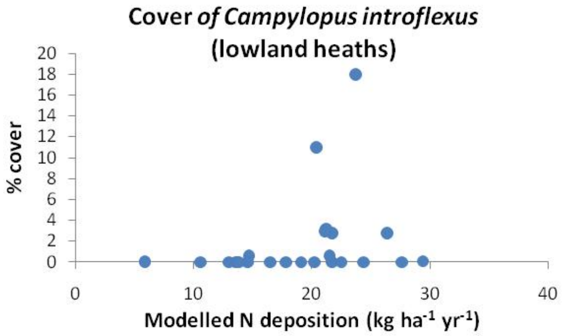


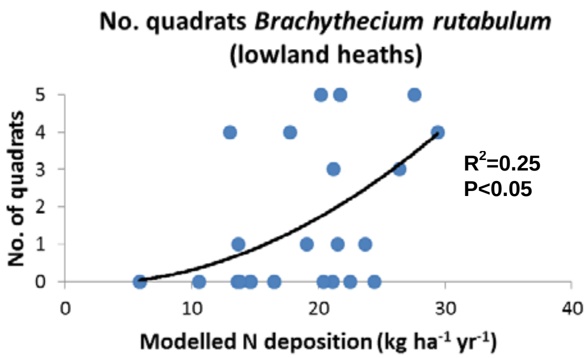
Lowland heaths: TU 2009

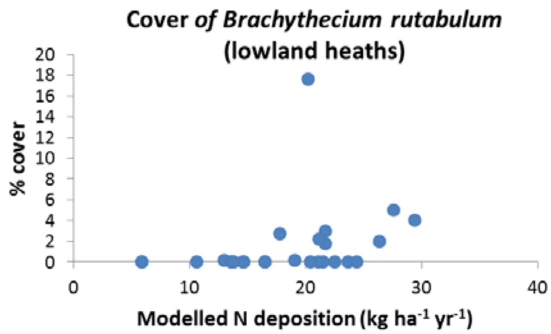


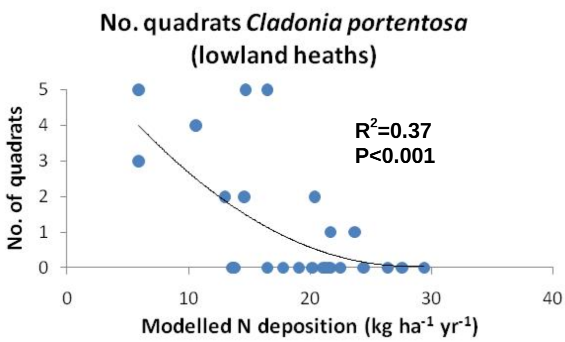


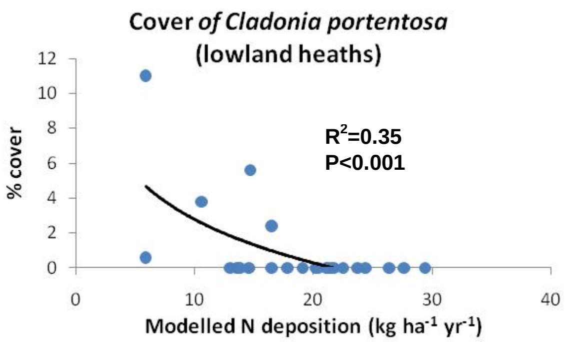


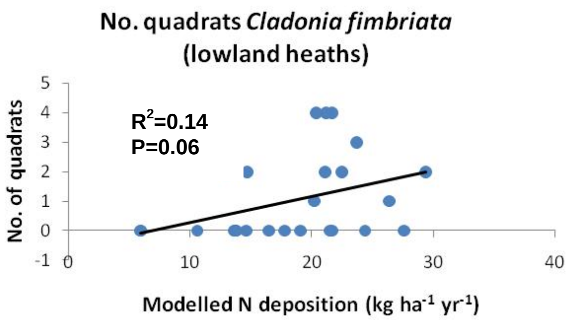


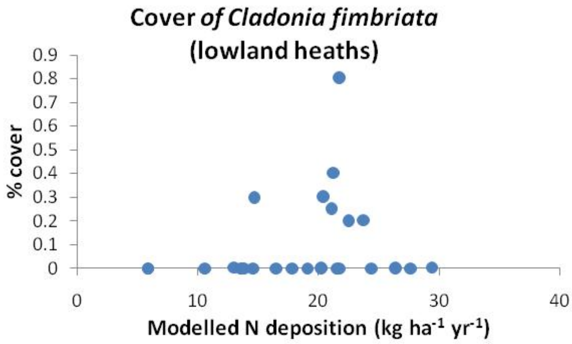




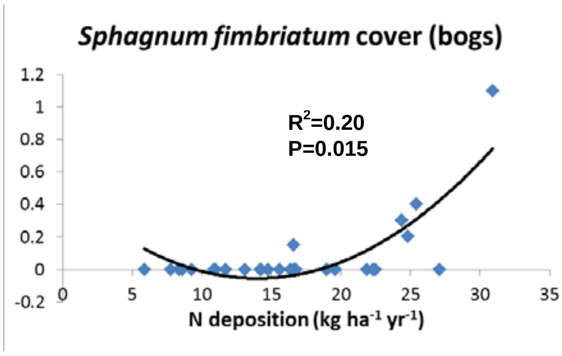


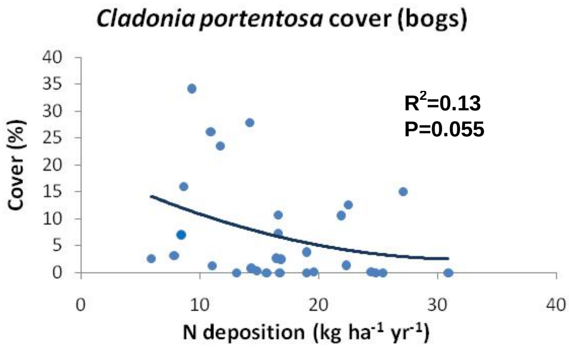




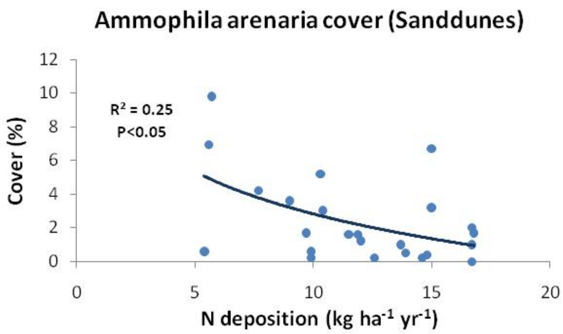


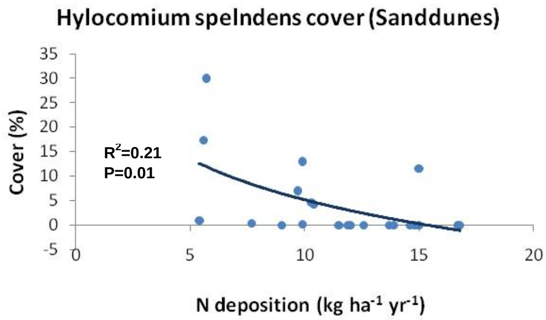
Bogs: TU survey 2009

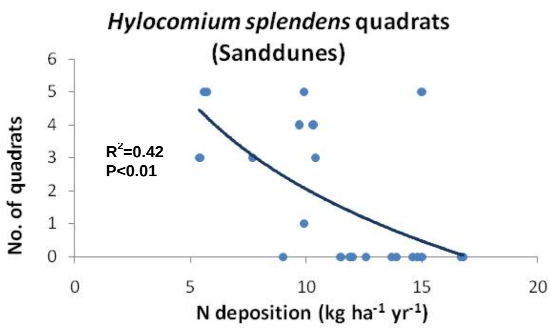




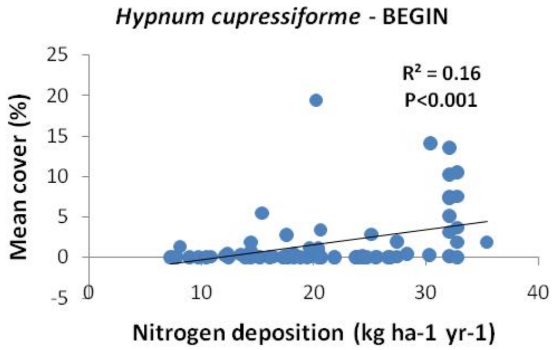
Sand dunes (TU 2009 all sites)

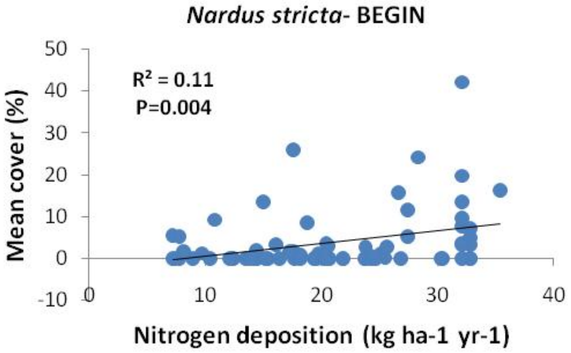


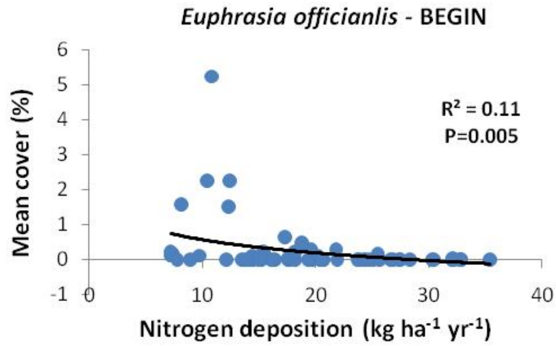


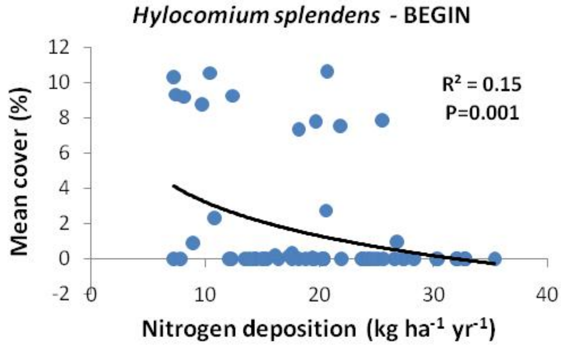


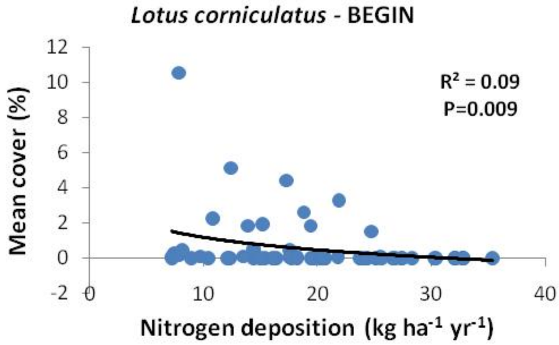
Acid grasslands (BEGIN)

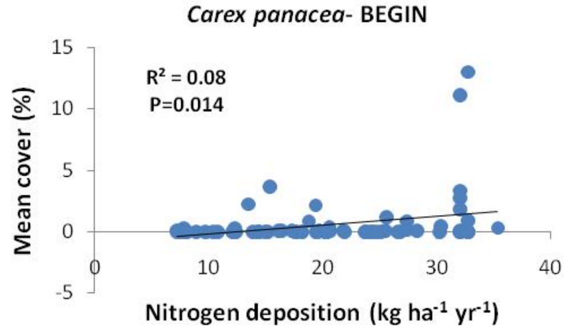








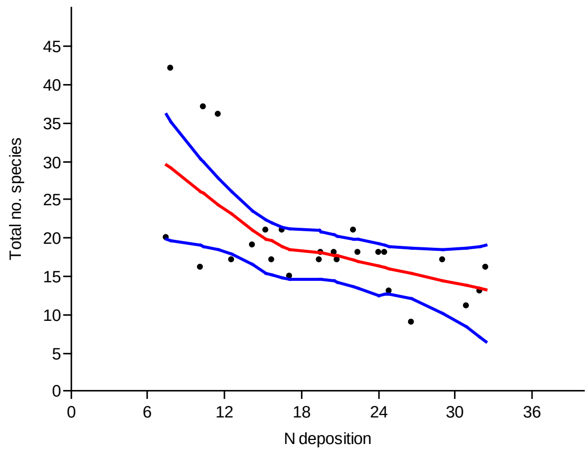


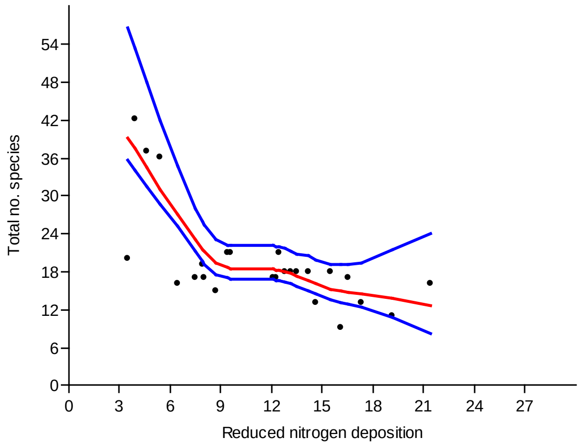


Appendix 4: LOESS regression curves

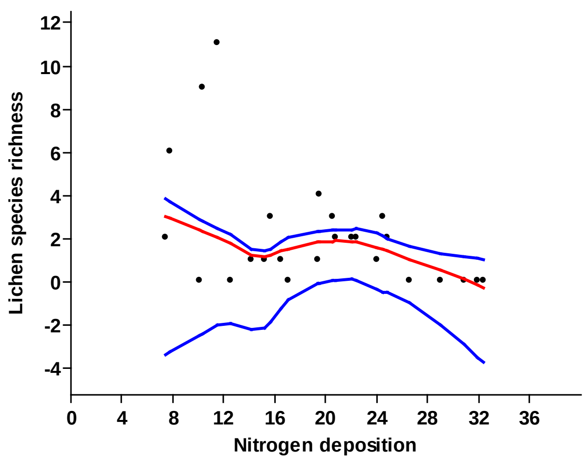
Best fit to data line in red, 95% confidence limits shown in blue and fitted by bootstrapping.

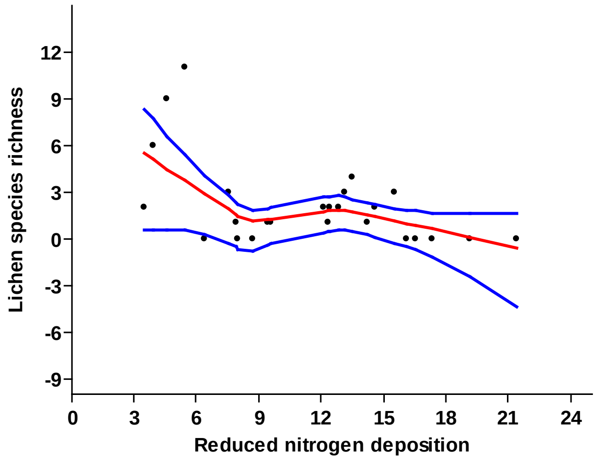
Upland heaths (TU): total species richness



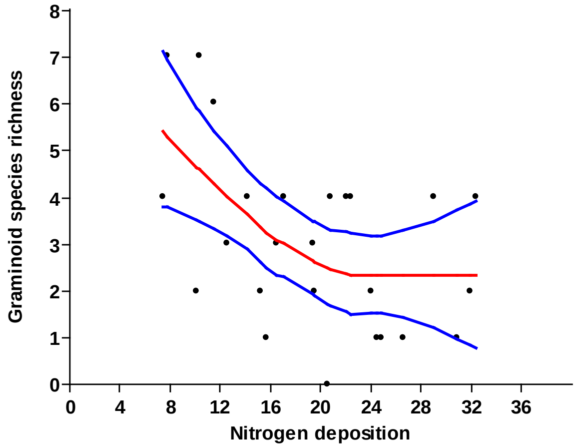


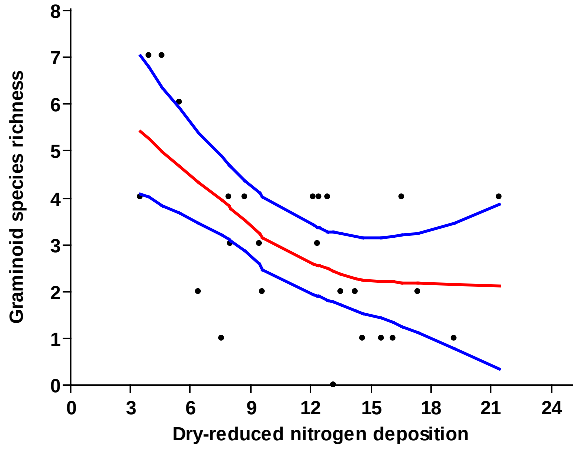
Upland heaths (TU): lichen species richness



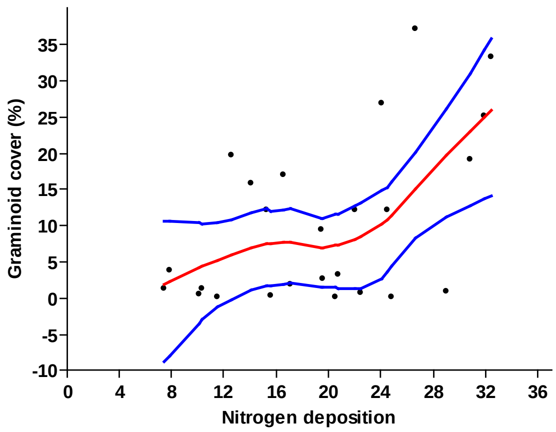


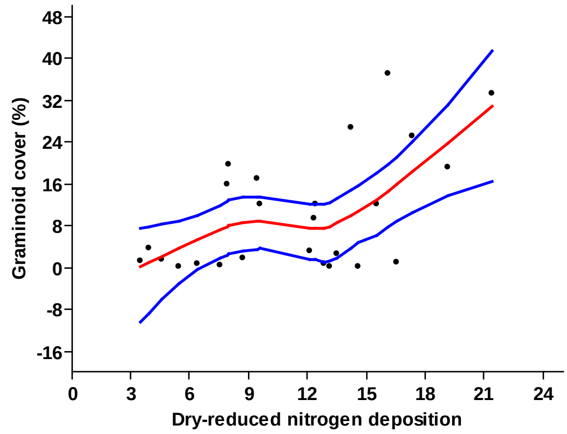
Upland heaths (TU): graminoid species richness



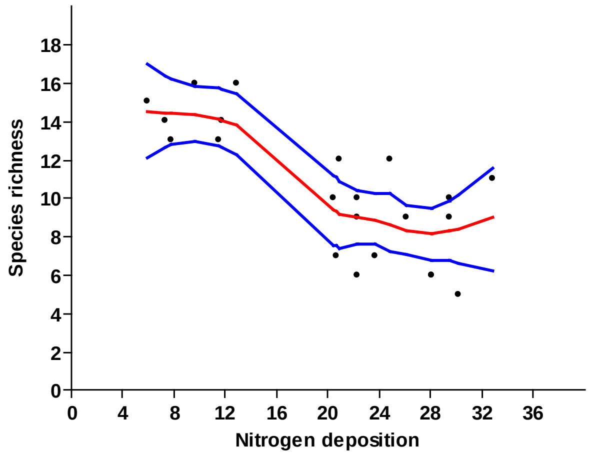


Upland heaths (TU): graminoid cover

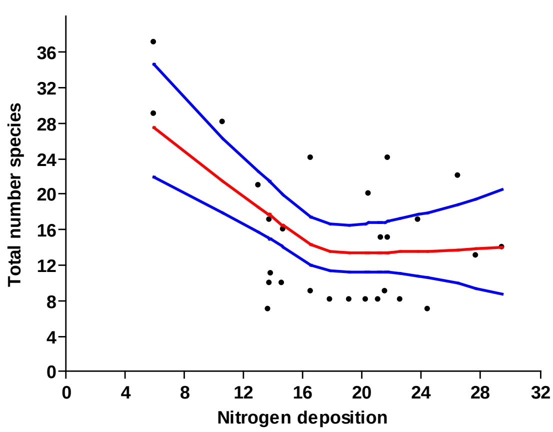


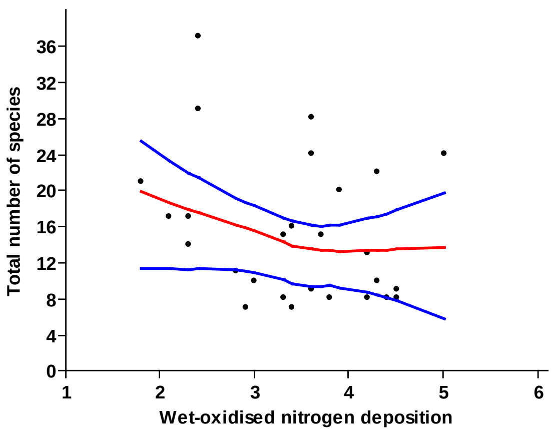


Upland heaths (MRS): total species richness

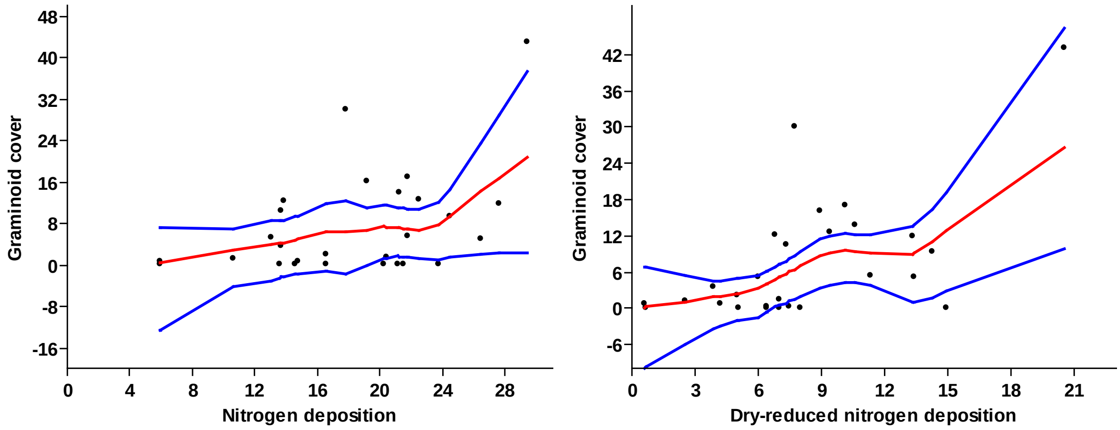


Lowland heaths: total number species

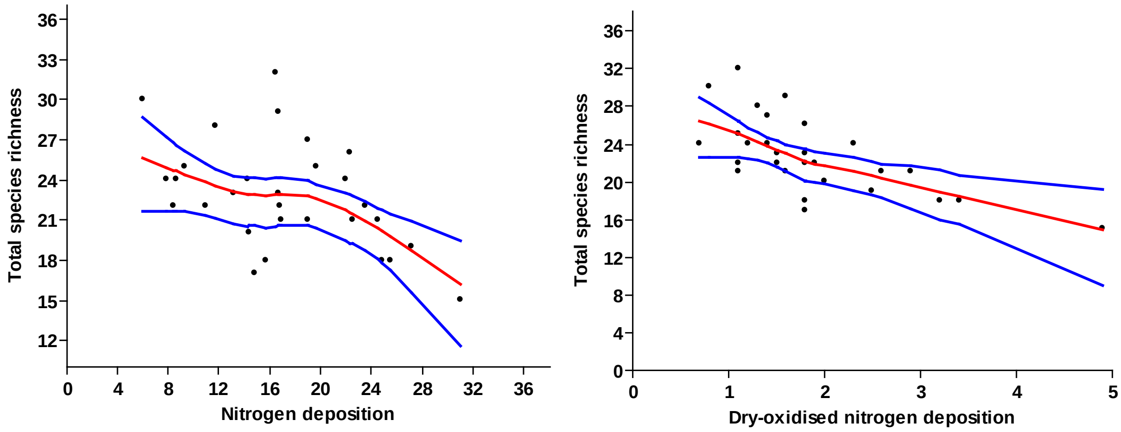




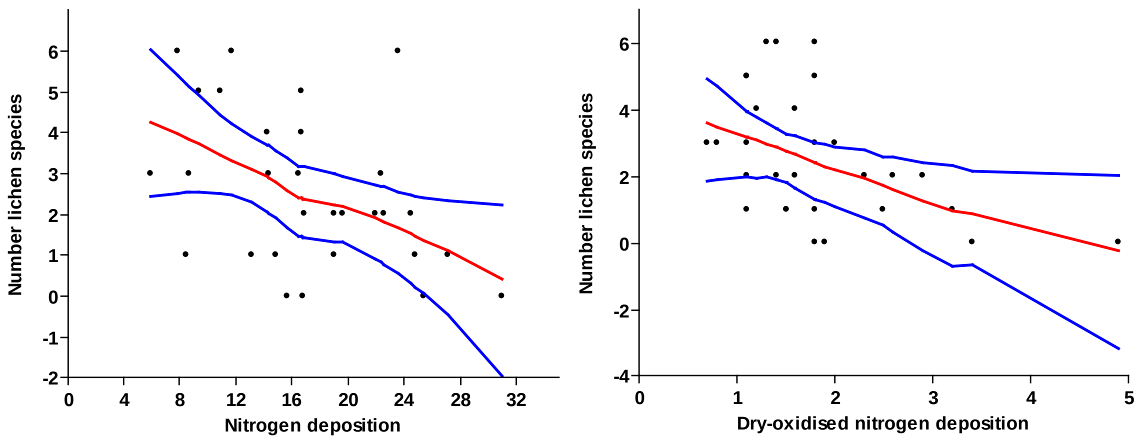
Lowland heaths: Graminoid cover



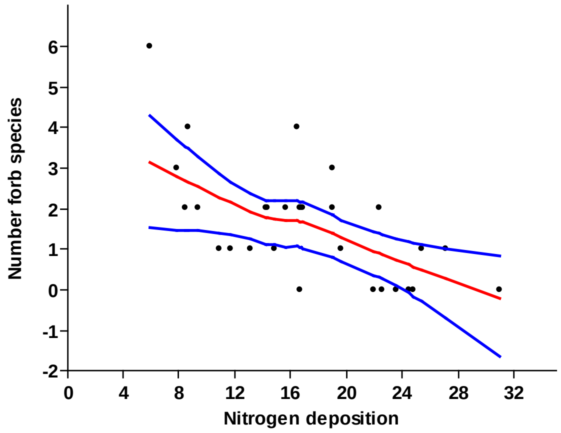
Bogs: total species richness



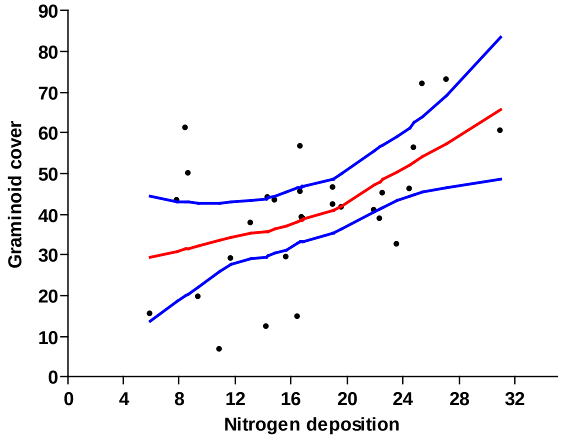
Bogs: lichen species richness

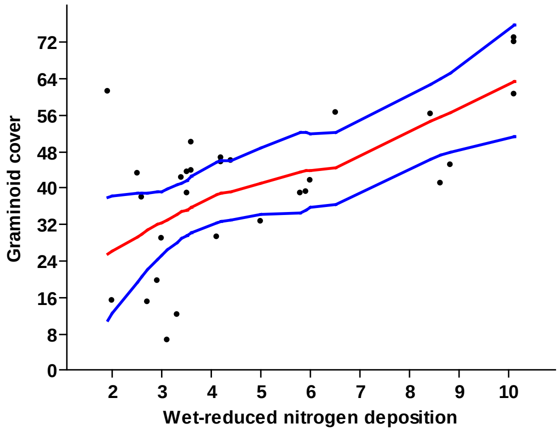


Bogs: Forb species richness

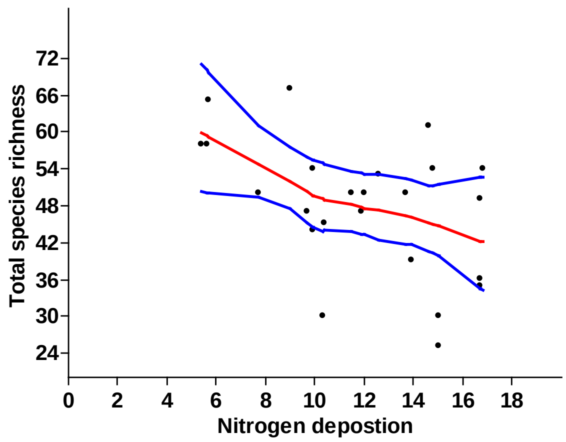


Bogs: Graminoid cover



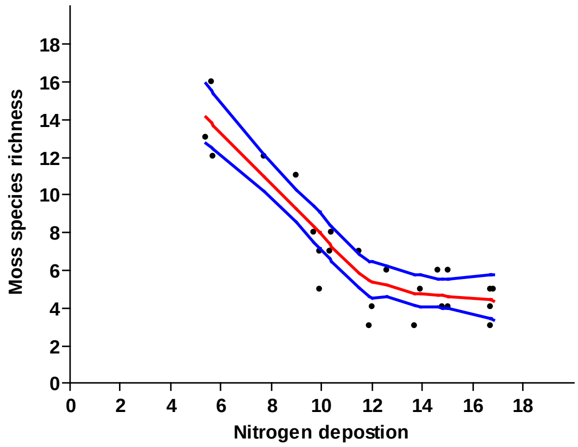


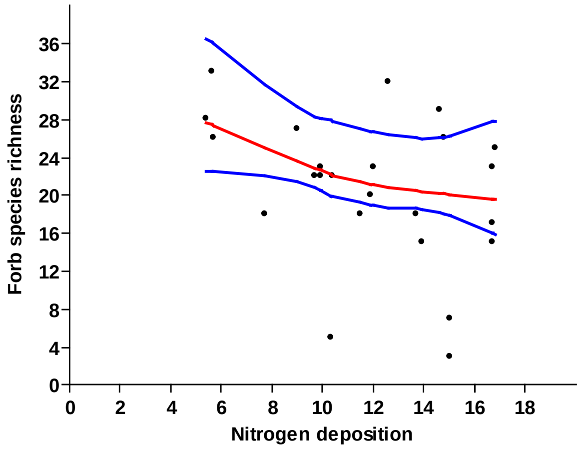
Sand dunes (all TU 2009): Total species richness



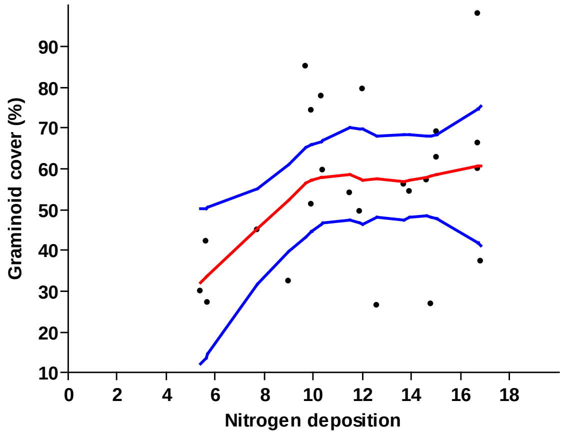
Sand dunes (all TU 2009): Moss species richness

Forb species richness

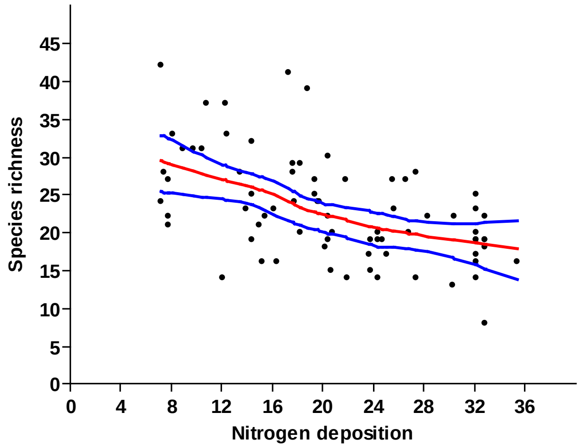




Sand dunes (all TU 2009): Graminoid cover



Acid Grasslands (BEGIN): Total species richness



Appendix 5: Effect of incremental increases in N deposition upon species richness

Summary of relationships between nitrogen deposition and species richness by habitat expressed as a percentage of the maximum in a habitat. Change in cover expresses as an absolute percentage. Incremental effect of a 0.3 kg increase in N deposition shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 0.3 kg increase in N deposition at different background N deposition levels					
			5 kg N	10 kg N	15 kg N	20 kg N	25 kg N	30 kg N
All habitats (TU 2009)								
Total Species Richness (SR)	77 spp.	10-20 kg N ha^{-1} yr^{-1}	-0.5 % of maximum number of species					
Upland heath (TU 2009)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.7%	-0.9%	-0.6%	-0.4%	-0.3%	-0.3%
Lichen SR	11 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.7%	-0.9%	-0.6%	-0.4%	-0.3%	-0.3%
Graminoid SR	7 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.9%	-1.0%	-0.6%	-0.5%	-0.4%	-0.3%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.13%	-0.01%	+0.11%	+0.24%	+0.37%	+0.5%
Upland heath (MRS)*								
Total SR	16 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.1%	-0.9%	-0.7%	-0.5%	-0.3%	-0.1%
Lowland heath (TU 2009)								
Total SR	37 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.8%	-0.9%	-0.6%	-0.5%	-0.4%	-0.3%
Moss SR	12 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.6%	-0.8%	-0.5%	-0.4%	-0.3%	-0.3%
Graminoid SR	9 spp.	10-20 kg N ha^{-1} yr^{-1}	-4.9%	-1.3%	-0.6%	-0.3%	-0.2%	-0.1%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	n/a	-0.04%	+0.11%	+0.26%	+0.41%	+0.56%
Bog (TU 2009)								
Total SR	32 spp.	5-10 kg N ha^{-1} yr^{-1}	-0.3%					
Lichen SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-0.6%					
Forb SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-2.3%	-1.1%	-0.8%	-0.6%	-0.5%	-0.4%
Graminoid cover	-	5-10 kg N ha^{-1} yr^{-1}	+0.41%					
Sand dunes (TU 2009, all sites)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-2.9%	-0.7%	-0.3%	-0.2%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-6.0%	-1.5%	-0.7%	-0.4%	-	-
Graminoid cover	n/a	8-15 kg N ha^{-1} yr^{-1}	+2.43%	+0.63%	+0.28%	-0.16%	-	-
Forb SR	33 spp.	8-15 kg N ha^{-1} yr^{-1}	-2.9%	-0.7%	-0.3%	-0.2%	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-1.7%	-0.9%	-0.6%	-0.4%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-2.5%	-1.3%	-0.9%	-0.6%	-	-
Acid grasslands (BEGIN)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-0.4%	-0.4%	-0.4%	-0.3%	-0.3%	-0.3%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Summary of relationships between nitrogen deposition and species richness by habitat expressed as a percentage of the maximum in a habitat. Change in cover expresses as an absolute percentage. Incremental effect of a 0.5 kg increase in N deposition shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 0.5 kg increase in N deposition at different background N deposition levels					
			5 kg N	**10 kg N**	**15 kg N**	**20 kg N**	**25 kg N**	**30 kg N**
All habitats (TU 2009)								
Total SR	77 spp.	10-20 kg N ha^{-1} yr^{-1}	-0.8 % of maximum number of species					
Upland heath (TU 2009)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-2.7%	-1.4%	-0.9%	-0.7%	-0.6%	-0.5%
Lichen SR	11 spp.	10-20 kg N ha^{-1} yr^{-1}	-2.8%	-1.4%	-0.9%	-0.7%	-0.6%	-0.5%
Graminoid SR	7 spp.	10-20 kg N ha^{-1} yr^{-1}	-3.1%	-1.6%	-1.1%	-0.8%	-0.6%	-0.5%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.22%	-0.01%	+0.20%	+0.41%	+0.62%	+0.83%
Upland heath (MRS)*								
Total SR	16 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.9%	-1.5%	-1.2%	-0.8%	-0.5%	-0.1%
Lowland heath (TU 2009)								
Total SR	37 spp.	10-20 kg N ha^{-1} yr^{-1}	-2.9%	-1.5%	-1.0%	-0.8%	-0.6%	-0.2%
Moss SR	12 spp.	10-20 kg N ha^{-1} yr^{-1}	-2.6%	-1.3%	-0.9%	-0.7%	-0.5%	-0.2%
Graminoid SR	9 spp.	10-20 kg N ha^{-1} yr^{-1}	-7.9%	-2.1%	-0.9%	-0.5%	-0.3%	-0.2%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.31	-0.06%	+0.19%	+0.44%	+0.69%	+0.94%
Bog (TU 2009)								
Total SR	32 spp.	5-10 kg N ha^{-1} yr^{-1}	-0.5%					
Lichen SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-1.1%					
Forb SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-3.7%	-1.9%	-1.3%	-1.0%	-0.8%	-0.6%
Graminoid cover	-	5-10 kg N ha^{-1} yr^{-1}	+0.68%					
Sand dunes (TU 2009, all sites)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-4.6%	-1.2%	-0.5%	-0.3%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-9.6%	-2.5%	-1.1%	-0.6%	-	-
Graminoid cover	n/a	8-15 kg N ha^{-1} yr^{-1}	+3.91%	+1.02%	+0.46%	+0.26%	-	-
Forb SR	33 spp.	8-15 kg N ha^{-1} yr^{-1}	-4.6%	-1.2%	-0.5%	-0.3%	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-2.8%	-1.4%	-1.0%	-0.7%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-4.2%	-2.1%	-1.4%	-1.1%	-	-
Acid grasslands (BEGIN)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-0.7%	-0.7%	-0.6%	-0.6%	-0.5%	-0.4%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Summary of relationships between nitrogen deposition and species richness/cover by habitat expressed as a percentage of the maximum in a habitat. Incremental effect of 1 kg increase in N deposition shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 1 kg increase in N deposition at different background N deposition levels					
			5 kg N	**10 kg N**	**15 kg N**	**20 kg N**	**25 kg N**	**30 kg N**
All habitats (TU 2009)								
Total species richness (SR)	77 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.6 % of maximum number of species/kg N increase					
Upland heath (TU 2009)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-5.7 %	-2.9 %	-2.0 %	-1.4 %	-1.2 %	-1.0 %
Lichen SR	11 spp.	10-20 kg N ha^{-1} yr^{-1}	-5.4 %	-2.7 %	-1.8 %	-1.8 %	-1.0 %	-1.0 %
Graminoid SR	7 spp.	10-20 kg N ha^{-1} yr^{-1}	-7.0 %	-2.9 %	-2.9 %	-1.4 %	-1.4 %	-1.0 %
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.5 %	no change	+0.4 %	+0.8 %	+1.2 %	+1.6 %
Upland heath (MRS)*								
Total SR	16 spp.	10-20 kg N ha^{-1} yr^{-1}	-3.4 %	-3.1 %	-2.5 %	-1.9 %	-1.3 %	-0.3 %
Lowland heath (TU 2009)								
Total SR	37 spp.	10-20 kg N ha^{-1} yr^{-1}	-6.2 %	-3.5 %	-2.2 %	-1.6 %	-1.4 %	-1.0 %
Moss SR	12 spp.	10-20 kg N ha^{-1} yr^{-1}	-5.8 %	-2.5 %	-1.7 %	-1.7 %	-1.7 %	-0.9 %
Graminoid SR	9 spp.	10-20 kg N ha^{-1} yr^{-1}	-17.8%	-4.4 %	-2.2 %	-1.1 %	-1.1 %	-0.5 %
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.6 %	no change	+0.5 %	+1.05 %	+1.6 %	+2.2 %
Bog (TU 2009)								
Total SR	32 spp.	5-10 kg N ha^{-1} yr^{-1}	-0.9 % of maximum number of species/kg N increase					
Lichen SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-1.7 %					
Forb SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-7.7 %	-3.9 %	-2.6 %	-1.9 %	-1.6 %	-1.3%
Graminoid cover	-	5-10 kg N ha^{-1} yr^{-1}	+1.5 % cover/kg N increase					
Sand dunes (TU 2009, all sites)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-10.1%	-2.6 %	-1.2 %	-0.6 %	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-21.3%	-5.0 %	-2.5 %	-1.3 %	-	-
Graminoid cover	n/a	8-15 kg N ha^{-1} yr^{-1}	+ 8.6 %	+ 2.2 %	+ 1.0 %	+ 0.5 %	-	-
Forb SR	33 spp.	8-15 kg N ha^{-1} yr^{-1}	-10.3%	-2.4 %	-1.2 %	-0.6 %	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-4.4 %	-2.2 %	-1.4 %	-1.0 %	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-21.3%	-5.6 %	-2.5 %	-1.3 %	-	-
Sand dunes TU 2009 + 2002 (Fixed dune grasslands)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-4.4 %	-2.2 %	-1.4 %	-1.0 %	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-8.9 %	-4.4 %	-3.1 %	-2.5 %	-	-
Acid grasslands (BEGIN)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-1.5 %	-1.4 %	-1.2 %	-1.1 %	-1.0%	-0.9%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Summary of relationships between nitrogen deposition and species richness by habitat expressed as a percentage of the maximum in a habitat. Change in cover expresses as an absolute percentage. Incremental effect of a 2 kg increase in N deposition shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 2.0 kg increase in N deposition at different background N deposition levels					
			5 kg N	10 kg N	15 kg N	20 kg N	25 kg N	30 kg N
All habitats (TU 2009)								
Total Species Richness (SR)	77 spp.	10-20 kg N ha^{-1} yr^{-1}	-3.2 % of maximum number of species					
Upland heath (TU 2009)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-9.7%	-5.3%	-3.6%	-2.7%	-2.2%	-1.9%
Lichen SR	11 spp.	10-20 kg N ha^{-1} yr^{-1}	-9.7%	-5.3%	-3.6%	-2.8%	-2.2%	-1.9%
Graminoid SR	7 spp.	10-20 kg N ha^{-1} yr^{-1}	-11.0%	-5.9%	-4.1%	-3.1%	-2.5%	-2.1%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-0.8%	+0.1%	+0.9%	+1.8%	+2.6%	+3.5%
Upland heath (MRS)*								
Total SR	16 spp.	10-20 kg N ha^{-1} yr^{-1}	-7.2%	-5.8%	-4.5%	-3.1%	-1.7%	-0.3%
Lowland heath (TU 2009)								
Total SR	37 spp.	10-20 kg N ha^{-1} yr^{-1}	-10.2%	-5.5%	-3.8%	-2.9%	-2.3%	-2.0%
Moss SR	12 spp.	10-20 kg N ha^{-1} yr^{-1}	-9.2%	-5.0%	-3.4%	-2.6%	-2.1%	-1.8%
Graminoid SR	9 spp.	10-20 kg N ha^{-1} yr^{-1}	-24.8%	-7.2%	-3.4%	-2.0%	-1.3%	-0.9%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-1.1%	-0.1%	+0.9%	+1.9%	+2.9%	+3.9%
Bog (TU 2009)								
Total SR	32 spp.	5-10 kg N ha^{-1} yr^{-1}	-1.9%					
Lichen SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-4.3%					
Forb SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-13.1%	-7.1%	-4.9%	-3.7%	-3.0%	-2.5%
Graminoid cover	-	5-10 kg N ha^{-1} yr^{-1}	+2.7%					
Sand dunes (TU 2009, all sites)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-14.4%	-4.2%	-2.0%	-1.1%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-30.1%	-8.8%	-4.1%	-2.4%	-	-
Graminoid cover	n/a	8-15 kg N ha^{-1} yr^{-1}	+12.3%	+3.6%	+1.7%	+1.0%	-	-
Forb SR	33 spp.	8-15 kg N ha^{-1} yr^{-1}	-14.6%	-4.2%	-2.0%	-1.2%	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-9.9%	-5.4%	-3.7%	-2.8%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-14.7%	-8.0%	-5.5%	-4.2%	-	-
Acid grasslands (BEGIN)								
Total species richness	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-2.9%	-2.7%	-2.4%	-2.2%	-2.0%	-1.7%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Summary of relationships between nitrogen deposition and species richness by habitat expressed as a percentage of the maximum in a habitat. Change in cover expresses as an absolute percentage. Incremental effect of a 5.0 kg increase in N deposition shown.

Survey/ Habitat/	Max. species richness	Habitat/species critical load	Change in species richness expressed as a % of maximum species richness recorded in habitat with a 5.0 kg increase in N deposition at different background N deposition levels					
			5 kg N	**10 kg N**	**15 kg N**	**20 kg N**	**25 kg N**	**30 kg N**
All habitats (TU 2009)								
Total species richness (SR)	77 spp.	10-20 kg N ha^{-1} yr^{-1}	-8 % of maximum number of species					
Upland heath (TU 2009)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-20.0%	-11.7%	-8.3%	-6.4%	-5.3%	-4.4%
Lichen SR	11 spp.	10-20 kg N ha^{-1} yr^{-1}	-20.0%	-11.7%	-8.3%	-6.5%	-5.3%	-4.5%
Graminoid SR	7 spp.	10-20 kg N ha^{-1} yr^{-1}	-22.6%	-13.2%	-9.4%	-7.3%	-5.9%	-5.0%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-1.3%	+0.9%	+3.0%	+5.1%	+7.2%	+9.3%
Upland heath (MRS)*								
Total SR	16 spp.	10-20 kg N ha^{-1} yr^{-1}	-17.0%	-13.6%	-10.1%	-6.7%	-3.3%	-
Lowland heath (TU 2009)								
Total SR	37 spp.	10-20 kg N ha^{-1} yr^{-1}	-21.1%	-12.3%	-8.7%	-6.8%	-5.5%	-4.7%
Moss SR	12 spp.	10-20 kg N ha^{-1} yr^{-1}	-19.0%	-11.1%	-7.9%	-6.1%	-5.0%	-4.2%
Graminoid SR	9 spp.	10-20 kg N ha^{-1} yr^{-1}	-43.3%	-14.4%	-7.2%	-4.3%	-2.9%	-2.1%
Graminoid cover	n/a	10-20 kg N ha^{-1} yr^{-1}	-2.0%	+0.5%	+3.0%	+5.5%	+8.0%	+10.5%
Bog (TU 2009)								
Total SR	32 spp.	5-10 kg N ha^{-1} yr^{-1}	-4.7%					
Lichen SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-10.8%					
Forb SR	6 spp.	5-10 kg N ha^{-1} yr^{-1}	-26.9%	-15.7%	-11.2%	-8.7%	-7.1%	-6.0%
Graminoid cover	-	5-10 kg N ha^{-1} yr^{-1}	+6.8%					
Sand dunes (TU 2009, all sites)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-25.3%	-8.4%	-4.2%	-2.5%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-52.8%	-17.6%	-8.8%	-5.3%	-	-
Graminoid cover	n/a	8-15 kg N ha^{-1} yr^{-1}	+21.5%	+7.2%	+3.6%	+2.2%	-	-
Forb SR	33 spp.	8-15 kg N ha^{-1} yr^{-1}	-25.5%	-8.5%	-4.2%	-2.5%	-	-
Sand dunes TU 2009 (pH ≥6.5)								
Total SR	77 spp.	8-15 kg N ha^{-1} yr^{-1}	-20.3%	-11.9%	-8.4%	-6.5%	-	-
Moss SR	16 spp.	8-15 kg N ha^{-1} yr^{-1}	-30.2%	-17.7%	-12.6%	-9.7%	-	-
Acid grasslands (BEGIN)								
Total SR	42 spp.	10-20 kg N ha^{-1} yr^{-1}	-7.2%	-6.5%	-5.9%	-5.3%	-4.7%	-4.1%

* in the upland heath MRS survey quadrat size was 0.5 x 0.5 m. This produced different results than the other surveys which used 2 x 2 m quadrats.

Appendix 6: Current critical loads for all habitats taken from ECE Empirical critical loads and dose-response relationships

Overview of empirical critical loads for nitrogen deposition (kg N ha^{-1} $year^{-1}$) to natural and seminatural ecosystems (column 1), arranged according to EUNIS class and level (column 2), as originally established in 2002 and reported in 2003 (column 3) and as revised in 2010 (column 4). The reliability is expressed in qualitative terms: ## reliable; # quite reliable; and (#) expert judgement (column 5). Column 6 provides a selection of effects that can occur when critical load are exceeded. Changes with respect to values of 2003 are indicated in bold.) For more information see Defra Report AQ801 Hall *et al*. (2011).

Ecosystem type	*EUNIS code*	*2003 kg N ha^{-1} $year^{-1}$ and reliability*	*2010 kg N ha^{-1} $year^{-1}$*	*2010 reliability*	*Indication of exceedance*
Marine habitats (A)					
Mid-upper salt-marshes	A2.53		20–30	(#)	Increase in dominance of graminoids
Pioneer and low-mid salt-marshes	A2.54 and A2.55	30–40 (#)	20–30	(#)	Increase in late-successional species, increase in productivity
Coastal habitat (B)					
Shifting coastal dunes	B1.3	10–20 (#)	10–20	(#)	Biomass increase, increase N leaching
Coastal stable dune grasslands (grey dunes)	B1.4[a]	10–20 #	8–15	#	Increase in tall graminoids, decrease in prostrate plants, increased N leaching, soil acidification, loss of typical lichen species
Coastal dune heaths	B1.5	10–20 (#)	10–20	(#)	Increase in plant production, increase in N leaching, accelerated succession
Moist-to-wet dune slacks	B1.8[b]	10–25 (#)	10–20	(#)	Increased biomass and tall graminoids
Inland surface water habitats (C)					
Soft-water lakes (permanent oligotrophic waters)	C1.1[c]	5–10 ##	3–10	##	Change in the species composition of macrophyte communities, increased algal productivity and a shift in nutrient limitation of phytoplankton from N to phosphorous (P)
Dune slack pools (permanent oligotrophic waters)	C1.16	10–20 (#)	10–20	(#)	Increased biomass and rate of succession
Permanent dystrophic lakes, ponds and pools	C1.4[d]		3–10	(#)	Increased algal productivity and a shift in nutrient limitation of phytoplankton from N to P

Ecosystem type	*EUNIS code*	*2003 kg N ha^{-1} year^{-1} and reliability*	*2010 kg N ha^{-1} year^{-1}*	*2010 reliability*	*Indication of exceedance*
Mire, bog and fen habitats (D)					
Raised and blanket bogs	D1[e]	5–10 ##	5–10	##	Increase in vascular plants, altered growth and species composition of bryophytes, increased N in peat and peat water
Valley mires, poor fens and transition mires	D2[f]	10–20 #	10–15	#	Increase in sedges and vascular plants, negative effects on bryophytes
Rich fens	D4.1[g]	15–35 (#)	15–30	(#)	Increase in tall graminoids, decrease in bryophytes
Montane rich fens	D4.2[g]	15–25 (#)	15–25	(#)	Increase in vascular plants, decrease in bryophytes
Grasslands and tall forb habitats (E)					
Subatlantic semi-dry calcareous grassland	E1.26	15–25 ##	15–25	##	Increase in tall grasses, decline in diversity, increased mineralization, N leaching; surface acidification
Mediterranean xeric grasslands	E1.3		15–25	(#)	Increased production, dominance by graminoids
Non-Mediterranean dry acid and neutral closed grassland	E1.7[b]	10–20 #	10–15	##	Increase in graminoids, decline of typical species, decrease in total species richness
Inland dune pioneer grasslands	E1.94[b]	10–20 (#)	8–15	(#)	Decrease in lichens, increase in biomass
Inland dune siliceous grasslands	E1.95[b]	10–20 (#)	8–15	(#)	Decrease in lichens, increase in biomass, increased succession
Low and medium altitude hay meadows	E2.2	20–30 (#)	20–30	(#)	Increase in tall grasses, decrease in diversity
Mountain hay meadows	E2.3	10–20 (#)	10–20	(#)	Increase in nitrophilous graminoids, changes in diversity
Moist and wet oligotrophic grasslands					
• Molinia caerulea meadows	E3.51	15–25 (#)	15–25	(#)	Increase in tall graminoids, decreased diversity, decrease of bryophytes
• Heath (Juncus) meadows and humid (Nardus stricta) swards	E3.52	10–20 #	10–20	#	Increase in tall graminoids, decreased diversity, decrease of bryophytes
Moss- and lichen-dominated mountain summits	E4.2	5–10 #	5–10	#	Effects upon bryophytes or lichens
Alpine and subalpine acid grasslands	E4.3		5–10	#	Changes in species composition; increase in plant production
Alpine and subalpine calcareous grasslands	E4.4		5–10	#	Changes in species composition; increase in plant production

Ecosystem type	*EUNIS code*	*2003 kg N ha^{-1} year^{-1} and reliability*	*2010 kg N ha^{-1} year^{-1}*	*2010 reliability*	*Indication of exceedance*
Heathland, scrub and tundra habitats (F)					
Tundra	F1	5–10 #	3–5	#	Changes in biomass, physiological effects, changes in species composition in bryophyte layer, decrease in lichens
Arctic, alpine and subalpine scrub habitats	F2	5–15 (#)	5–15	#	Decline in lichens, bryophytes and evergreen shrubs
Northern wet heath	F4.11				
• "U" Calluna-dominated wet heath (upland moorland)	F4.11[e,h]	10–20 (#)	10–20	#	Decreased heather dominance, decline in lichens and mosses, increased N leaching
• "L" Erica tetralix-dominated wet heath (lowland)	F4.11[e,h]	10–25 (#)	10–20	(#)	Transition from heather to grass dominance
Dry heaths	F4.2[e,h]	10–20 ##	10–20	##	Transition from heather to grass dominance, decline in lichens, changes in plant biochemistry, increased sensitivity to abiotic stress
Mediterranean scrub	F5		20–30	(#)	Change in plant species richness and community composition

Ecosystem type	*EUNIS code*	*2003 kg N ha^{-1} $year^{-1}$ and reliability*	*2010 kg N ha^{-1} $year^{-1}$*	*2010 reliability*	*Indication of exceedance*
Forest habitats (G)					
Fagus woodland	G1.6		10–20	(#)	Changes in ground vegetation and mycorrhiza, nutrient imbalance, changes soil fauna
Acidophilous Quercus-dominated woodland	G1.8		10–15	(#)	Decrease in mycorrhiza, loss of epiphytic lichens and bryophytes, changes in ground vegetation
Meso- and eutrophic Quercus woodland	G1.A		15–20	(#)	Changes in ground vegetation
Mediterranean evergreen (Quercus) woodland	G2.1		3–7	(#)	Changes in epiphytic lichens
Abies and Picea woodland	G3.1		10–15	(#)	Decreased biomass of fine roots, nutrient imbalance, decrease in mycorrhiza, changed soil fauna
Pinus sylvestris woodland south of the taiga	G3.4		5–15	#	Changes in ground vegetation and mycorrhiza, nutrient imbalances, increased N_2O and NO emissions
Pinus nigra woodland	G3.5		15	(#)	Ammonium accumulation
Mediterranean Pinus woodland	G3.7		3–15	(#)	Reduction in fine root biomass, shift in lichen community
Spruce taiga woodland	G3.A[i]	10–20 #	5–10	##	Changes in ground vegetation, decrease in mycorrhiza, increase in free algae
Pine taiga woodland	G3.B[i]	10–20 #	5–10	#	Changes in ground vegetation and in mycorrhiza, increase occurrence of free algae
Mixed taiga woodland with Betula	G4.2		5–8	(#)	Increased algal cover
Mixed Abies-Picea Fagus woodland	G4.[j]		10–20	(#)	
Overall					
Broadleaved deciduous woodland	G1[k)]	10–20 #	10–20	##	Changes in soil processes, nutrient imbalance, altered composition mycorrhiza and ground vegetation
Coniferous woodland	G3[k,l]	10–20 #	5–15	##	Changes in soil processes, nutrient imbalance, altered composition mycorrhiza and ground vegetation

[a] For acid dunes, use the 8–10 kg N ha-1 year-1 range, for calcareous dunes use the 10–15 kg ha-1 year-1 range.

[b] Use the lower end of the range with low base cation availability. Use the higher end of the range with high base cation availability.

[c] This critical load should only be applied to oligotrophic waters with low alkalinity with no significant agricultural or other human inputs. Use the lower end of the range for boreal and alpine lakes, use the higher end of the range for Atlantic softwaters.

[d] This critical load should only be applied top waters with low alkalinity with no significant agricultural or other direct human inputs. Use the lower end of the range for boreal and alpine dystrophic lakes.

[e] Use the high end of the range with high precipitation and the low end of the range with low precipitation. Use the low end of the range for systems with a low water table, and the high end of the range for systems with a high water table. Note, that water table can be modified by management.

[f] For D2.1 (quaking fens and transition mires) use lower end of the range (#).

[g] For high latitude systems use lower end of the range.

[h] Use the high end of the range when sod cutting has been practiced; use the lower end of the range with low intensity management.

[i] In 2003 presented as overall value for boreal forests.

[j] Included in studies which were classified into G1.6 and G3.1.

[k] In 2003 presented as overall value for temperate forests.

[l] For application at broad geographical scales.

A.5 Institute of Air Quality Management (2020). A guide to the assessment of air quality impacts on designated nature conservation sites

IAQM Institute of Air Quality Management

A guide to the assessment of air quality impacts on designated nature conservation sites

Version 1.1

May 2020

www.iaqm.co.uk

Contents

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Record of substantive amendments

v1.1. May 2020

Original location	Revised location	Amendment made
Thoughout	---	Reference to Highways Agency Design Manual for Roads and Bridges (DMRB) updated to reflect new guidance published in November 2019.
1.2.1	---	*Deleted text:* The need to take account of complex new case law relating to European sites, coupled with the voluntary nature of producing this guidance has meant that the final joint document has been unavoidably delayed.
1.2.2	1.2.2	*Replacement paragraph as follows*: There were a number of unavoidable delays in producing the document and IAQM made the decision to publish the air quality sections as a standalone document in 2019.
---	1.2.4	*Additional paragraph*: CIEEM intend to publish the ecological sections in 2020. Both documents should be considered together.
Box 3.1	Box 3.1	*Footnote added:* The relevant section of the DMRB has been replaced by LA 105 Air Quality (see reference 23).
3.12	---	*Deleted text:* The 2019 Clean Air Strategy includes a commitment that the EU Withdrawal Bill will ensure existing EU environmental law continues to have effect in law after the UK leaves the EU. Therefore, the above rulings of the CJEU are likely to remain relevant for the foreseeable future.
5.3.14	5.3.2	*Paragraphs 5.3.2 and 5.3.14 merged.* *Paragraph 5.3.2 amended as follows:* The locations and boundaries of international and national designated sites can be found online, e.g. on the MAGIC website or similar online resources from the relevant SNCO. If local sites are to be assessed, details can be obtained by consulting the Environment Agency or local biodiversity records office who may charge a nominal fee for this service. Sufficient time should be allowed to obtain this data.
---	5.4.1.3	*Reference added:* iaqm.co.uk/text/position_statements/screening_tools_interim.pdf.
---	5.4.1.10	*Additional text added:* Although this may be offset to some extent if NH_3 emissions per vehicle km increase in the future.
---	5.4.1.11	*New footnote:* An updated version of the spreadsheet is available from the Overseeing Organisations (e.g. Highways England) for use on their road schemes.
---	5.4.1.17	*Reference added:* laqm.defra.gov.uk/documents/LAQM-TG16-February-18-v1.pdf.
5.4.1.21	---	*Deleted text:* It should be noted that the current DMRB guidance only provides a deposition velocity for NO_2 only and that it is different from the AQTAG NO_2 deposition velocity. IAQM recommends that the AQTAG value is used in preference to the DMRB value. It should also be noted that the deposition velocity for NO is extremely small and assuming that all NO_x is in the form of NO_2 is therefore highly conservative.
5.4.2.5	---	*Reference removed:* Development of the CURED V3A Emissions Model, Air Quality Consultants.
5.4.2.9	---	*Deleted paragraph:* The 2007 DMRB guidance for ecological assessment suggests reducing the background deposition rates by 2% each year. This approach is now considered to be inappropriate as it is not supported by monitoring data.
5.5.4.1	5.5.4.1	*Replacement paragraph as follows:* There is evidence that ammonia emissions from road vehicles may contribute more than half the local traffic related increment to nitrogen deposition.
5.5.4.2	5.5.4.2	*Replacement paragraph as follows:* The DMRB methodology only requires the assessment of NO_x emissions and nitrogen deposition. It does not consider NH_3 or its contribution to nitrogen deposition. As road transport is a source of ammonia, albeit a small source compared to agriculture at a national level, consideration should be given to including it and its contribution to local nitrogen deposition.

Original location	Revised location	Amendment made
5.5.4.3	---	*Deleted paragraph:* Where internationally important sites are involved this should be discussed with the project ecologist (or the HRA co-ordinator) to ensure that the potential for 'in-combination' effects is treated appropriately.
5.5.4.4	---	*Deleted paragraph:* If a formal assessment of 'in-combination' impacts is required, it must take place before applying the 1% criterion. Within this context, it may be possible to screen for effects of nitrogen deposition without specifically calculating the nitrogen deposition rate and identifying relevant critical loads and baseline concentrations at all sensitive sites. This assumes a linear relationship between concentration and deposition of NO_x (through the application of simple conversion factors to calculate deposition from concentration – there may be cases when a more complex relationship is applied) means that where the change in NO_x concentrations is less than 0.4 µg/m3, it is unlikely that it would exceed 1% of the most stringent critical loads for nitrogen and acid deposition for a sensitive habitat. This, however, may not be true for all habitats[66], and depends on the deposition velocity used.

1. Introduction

1.1 Purpose

1.1.1 This document has been produced by the Institute of Air Quality Management (IAQM) to assist its members in the assessment of the air quality impacts of development on designated nature conservation sites. It may also be useful for ecologists, who use the results of air quality assessments (AQAs) to evaluate the effects of air pollution on habitats and species, by increasing their understanding of the information provided by air quality specialists. This subsequent stage of the overall process, i.e. the assessment of the *effects* that air quality impacts may have on habitats and species, is generally outside the expertise of IAQM members and no specific detail on this stage is provided in this guidance.

1.1.2 This document focuses on air quality assessments in support of Habitats Regulations Assessments (HRA)[1], but it will also be useful when assessing the air quality impact on national or local designated nature conservation sites.

1.2 Producing this guidance

1.2.1 The IAQM and the Chartered Institute of Ecology and Environmental Management (CIEEM) originally intended to produce a joint document on the assessment of the ecological effects of air pollution. The members of both organisations, some regulatory bodies and nature conservation agencies were consulted on a draft document in October 2017.

1.2.2 There were a number of unavoidable delays in producing the document and IAQM made the decision to publish the air quality sections as a standalone document in 2019.

1.2.3 A second round of consultation of IAQM members was undertaken for the draft of this document in Spring 2019. The comments received from both consultations have been taken into account in the production of the final document.

1.2.4. CIEEM intend to publish the ecological sections in 2020. Both documents should be considered together.

1.2.5 It is recognised that there may be useful learning points and amendments to be made to this guidance once it has been applied in practice. IAQM, therefore, welcomes comments and feedback on the guidance and will endeavour to produce, if necessary, a revised version at an appropriate time.

1.2.6 The publication of this document replaces the IAQM

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Position Statement on 'Use of a Criterion for the Determination of an Insignificant Effect of Air Quality Impacts on Sensitive Habitats' issued in January 2016.

1.2.7 A glossary of terms is provided in **Appendix A**.

1.3 Sites covered by this guidance

1.3.1 This IAQM guidance is applicable to the assessment of European, national and local designated sites where such assessments are required by the decision maker. This guidance, therefore, applies to the assessment of Special Areas of Conservation (SACs) and Special Protection Areas (SPAs) (known as European sites) and Ramsar sites[2] which are covered by the Habitats Regulations. It also applies to Sites of Special Scientific Interest (SSSIs), Areas of Special Scientific Interest (ASSIs), National Nature Reserves (NNRs), local nature reserves (LNRs), local wildlife sites (LWSs) and areas of ancient woodland (AW)[3]. All these sites may require assessment depending on the type of project and/or the regulatory system under which the application is made. In this document, these are referred to as 'designated sites'.

1.3.2 The Habitats Regulations[4], which transpose the Habitats Directive[5] into legislation in the UK[6], require that a development proposal, or a project or plan, will not cause a likely significant effect or, where likely significant effects cannot be discounted, no adverse effect on the integrity of European sites[7]. Proving the absence of significant effects is more difficult than proving that a significant effect will occur. The air quality practitioner has an important role to play in ensuring the right information is provided to the ecologist to allow them to make that judgement.

1.3.3 Different requirements apply to national and local designated sites. In England, for example, the National Planning Policy Framework (NPPF)[8] states that planning permission should be refused if significant harm to biodiversity cannot be avoided, mitigated or, as a last resort, compensated[8]. The same basic assessment methodology can be used although the final determination of the significance of effect may be different.

1.4 The Habitats Regulations

1.4.1 The European Commission (EC) provides guidance on managing internationally designated nature conservation sites[9]. The flow chart in **Appendix B**, reproduced from this EC guidance, illustrates the stages of the assessment process. It should be noted that the process is iterative, i.e. it is possible to return to earlier stages during the assessment of a project or plan. Air quality assessment may be required at any stage.

1.4.2 The requirement to produce an HRA is driven in England and Wales by the Conservation of Habitats and Species Regulations 2017 (as amended), which states that: 'A competent authority, before deciding to undertake, or give any consent, permission or other authorisation for, a plan or project which

- Is likely to have a significant effect on a European site or a European offshore marine site (either alone or in-combination with other plans or projects); and
- Is not directly connected with or necessary for the management of that site, must make an appropriate assessment of the implications for that site in view of that site's conservation objectives.'

1.4.3 Similar requirements apply in Scotland and Northern Ireland.

1.4.4 The contents of an appropriate assessment are not defined in the legislation, and there is no current Government guidance providing clarification. The competent authority[10] varies depending on the type of the project or plan, but for planning applications is primarily the local planning authority. For appeals, for example, in England, the planning inspector or the Secretary of State can also be the competent authority. For environmental permits the UK competent authorities include the Environment Agency in England (EA), Scottish Environmental Protection Agency (SEPA) in Scotland, Industrial Pollution and Radiochemical Inspectorate (IPRI) in Northern Ireland, Natural Resources Wales (NRW) in Wales, and local authorities. It should be noted that the regulators of industrial installations in the devolved authorities generally, but not exclusively, rely on EA advice[11].

1.4.5 Typically, consultants working for the applicant produce an HRA, which is used by the competent authority to inform the 'appropriate assessment'. The role of the air quality specialist is to assess the potential impacts so as to either demonstrate that a project or plan will not have a likely significant effect (alone or in-combination) or, if this is not possible, to provide an ecologist with an estimate of the air quality impacts. If there is a need for an HRA covering the air quality impacts, the ecologist should produce it in liaison with the air quality specialist.

1.4.6 Where the appropriate assessment concludes that the project or plan will not result in an adverse effect on the integrity of the European site(s) being considered, consent may be granted. If adverse effects on the integrity of the European site(s) cannot be ruled out, consent cannot be granted without further work. This may include the identification of further measures to address the predicted adverse effect(s).

1.4.7 Where no further measures are available, and the project or plan is needed for 'imperative reasons of overriding public interest' (IROPI), the competent authority may authorise the project or plan despite the potential for adverse effects provided there are no reasonable alternatives to achieving the objectives of the plan or project which would have less effect on European sites. Under these circumstances, consent can only be granted if suitable compensatory measures are identified.

1.5 Scope of this document

1.5.1 This IAQM guidance document is not intended to be a primer on how to model air quality impacts[12] but instead is intended to provide practical guidance for those air quality specialists who undertake air quality impact assessments and are already familiar with modelling techniques. It also aims to encourage greater communication and co-operation between air quality and ecological specialists.

1.5.2 The planning and environmental permitting systems are somewhat different in the various devolved administrations. In addition, these two regulatory systems have different requirements in terms of the types of designated site that require assessment.

1.5.3 The air quality specialists undertaking assessments are required to make professional judgements. This is due to the diverse range of projects and the wide range of factors that influence the approach taken, which means it is not possible to be entirely prescriptive. IAQM advice is that the assessments of impacts should be undertaken by, or under the close supervision of, an experienced air quality practitioner. Where possible the name of the assessors and/or supervisors should be included in the assessment with a brief summary of their relevant qualifications, experience and role in the assessment.

Box 1.1 Key Issues for the Air Quality Assessment

1. Impacts vs. effects
   - The air quality practitioner calculates the air quality impacts.
   - The ecologist identifies the ecological effects.
2. IAQM document scope
   - This document is concerned with determining whether there will be a 'likely significant effect' on a habitat, and where this cannot be screened out, providing the ecologist with detailed information on the air quality impacts.
   - It is the job of the ecologist to determine whether in reality there will be a 'significant effect', or, for European sites, an 'adverse effect on the integrity of the site'.
3. Consultation
   - Always consult the regulator on the:
     - assessment approach;
     - pollutants to be included;
     - designated sites to be considered; and
     - the list of projects and plans to be included in the in-combination or cumulative impact assessment.
4. Collaboration
   - Work closely with the project ecologist throughout the air quality assessment.
   - If there is no project ecologist, as is the case for many environmental permit applications, it may be necessary in some circumstances to recommend to the client that one is appointed.
5. Professional judgement
   - There are too many different types of plans and projects and regulatory regimes in the UK to be prescriptive on how to undertake air quality assessments.
   - Professional judgement of an experienced air quality practitioner is essential.
   - Demonstration of experience of undertaking similar assessments should be provided in the assessment report or other appropriate document.
6. Proportionate
   - The scope and detail of assessment should be proportionate to the risk.
7. Precautionary principle
   - The assessment should be precautionary, but not so precautionary as to produce results that are unrealistic.
8. Guidance changes with time
   - Always check for the most recent guidance from the relevant regulator.

The key issues to be considered when undertaking an air quality assessment of ecological impacts with reference to this practical guidance are set out in **Box 1.1**.

1.5.4 In this document, current guidance from other bodies is referred to, recognising that it may be revised in the future as the regulatory position and case law evolves. If that happens, this document will be amended accordingly.

1.5.5 Throughout this document the term 'regulator' is used to describe the decision maker in both the planning and environmental permitting regimes; where the assessment includes a European Site, the 'regulator' is also the competent authority.

1.6 Other IAQM guidance

1.6.1 Impacts associated with dust soiling, e.g. from construction projects and minerals sites are not within the scope of this guide. Such impacts may be included in a subsequent edition. A methodology for the qualitative risk assessment of construction dust on ecological sites is available from IAQM[13]. IAQM also produces guidance for the assessment of impacts of mineral sites for planning applications[14].

1.6.2 Existing guidance on planning and air quality has been produced by Environmental Protection UK (EPUK) and IAQM[15]. This guidance applies in the context of human receptors only and specifically excludes consideration of ecological sites and so does not conflict in any way with this document. These guidance documents are complementary to this document and it is not anticipated that there will be any substantial overlap of application, as they serve different purposes.

1.7 Ecological impact assessment guidance

1.7.1 For many developments that could give rise to air quality impacts on designated sites, there will be a need to incorporate the assessment of these impacts into a wider ecological impact assessment (EcIA). CIEEM defines EcIA as '*...a process of identifying, quantifying and evaluating the potential effects of development-related or other proposed actions on habitats, species and ecosystems*'[16].

1.7.2 CIEEM has produced guidelines for implementing the EcIA process in the UK. The various stages of the EcIA process as recommended by CIEEM will interact with the various stages of an air quality assessment on designated sites. For detailed information on the EcIA process, the reader is referred to the CIEEM Guidelines. A summary of how the two processes may interact is provided in **Appendix C**.


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[1] Also known as a Habitats Directive Article 6 Assessment.
[2] These sites are designated under The Convention on Wetlands, known as the Ramsar Convention.
[3] Throughout this guidance, areas of ancient woodland are included within the definition of 'designated sites'. Environment Agency guidance states that ancient woodland within 2 km of a new emitting installation should be included in detailed assessments for environmental permit applications. (www.gov.uk/guidance/air-emissions-risk-assessment-for-your-environmental-permit). 'Ancient woodland' is in many cases included on countrywide Ancient Woodland Inventories; these are coordinated by the relevant country Statutory Nature Conservation Organisation (SNCO) and / or government forestry department. Ancient woodland can also occur in areas not included in the inventory.
[4] The Conservation of Habitats and Species Regulations 2010 (as amended); in Scotland, by The Conservation (Natural Habitats, &c.) Regulations 1994 (as amended) and in Northern Ireland, by The Conservation (Natural Habitats, etc.) Regulations (Northern Ireland) 1995.
[5] The Council Directive 92/43/EEC on the conservation of natural habitats and of wild fauna and flora.
[6] Readers need to check the status of elements of the Regulations in devolved administrations.
[7] The definition of site integrity covers the distribution, structure, function and abundance; 'typical species'; whether a species is a 'viable component of its natural habitat'; and a sufficiently large habitat to maintain populations on a long-term basis. www.clientearth.org/reports/natura-2000-site-integrity-briefing.pdf.
[8] MHVLG, 2019, National Planning Policy Framework 2019, www.gov.uk/government/publications/national-planning-policy-framework--2.
[9] European Commission, 2018, Commission Notice "Managing Natura 2000 Sites -The provisions of Article 6 of the 'Habitats' Directive 92/43/EEC", Brussels 21.11.18, C(2018) 7621 final. ec.europa.eu/environment/nature/natura2000/management/guidance_en.htm.
[10] '*Competent authorities*' are defined by the Habitats Regulations, and include local planning authorities, government departments and statutory undertakers.
[11] For example, NRW had its own guidance on 'Assessing the impact of ammonia and nitrogen on designated sites from new and expanding intensive livestock' (Guidance Note 020) cdn.naturalresources.wales/media/684017/guidance-note-20-assessing-the-impact-of-ammonia-and-nitrogen-on-designated-sites-from-new-and-expanding-intensive-livestock-units.pdf.
[12] See for example, Defra, 2016, Local Air Quality Management Technical Guidance TG.16 laqm.defra.gov.uk/technical-guidance/
[13] iaqm.co.uk/text/guidance/construction-dust-2014.pdf.
[14] www.iaqm.co.uk/text/guidance/mineralsguidance_2016.pdf.
[15] www.iaqm.co.uk/text/guidance/air-quality-planning-guidance.pdf.
[16] CIEEM, 2018, *Guidelines for Ecological Impact Assessment in the UK and Ireland: Terrestrial, Freshwater and Coastal*. Chartered Institute of Ecology and Environmental Management, Winchester.

2. Background

2.1 The broad effects of air pollution on habitats are now reasonably well understood, after several decades of research. Although the threat from acid deposition in the UK has diminished considerably in recent years, with the dramatic reduction in emissions of sulphur dioxide (SO_2), there is still a legacy effect in some habitats from the accumulated deposition since the Industrial Revolution. The effects of other pollutants are also apparent at many of the designated sites, especially from the deposition of nitrogen (N), and this problem is likely to persist for some time at the national and international level.

2.2 To provide clarity, IAQM uses the term 'impact' where discussing changes in concentration or deposition and the term 'effect' when discussing the ecological changes due to the air pollution impact.

2.3 To quantify and describe the effects on a designated site that might result from introducing a new source of airborne pollution, there needs to be an understanding of the atmospheric processes that define the scale of the impact following the release of a pollutant and the consequences of this impact for the habitat.

2.4 A useful summary of knowledge on this subject is provided by a consortium of environmental and conservation agencies on the Air Pollution and Information System (APIS) website[17], as hosted by the Centre for Ecology and Hydrology. This site provides a key database of information pertaining to air pollution effects at designated sites.

2.5 This IAQM document assumes that many users will be well acquainted with the subject of air pollution. For those who require an introduction to the concepts and terminology, a very brief summary is provided below, supported by further information found in **Appendix D**.

2.6 There are two categories of pollutants that are typically the subject of AQAs for designated sites. These are pollutants that have an effect on vegetation/habitats in a gaseous form and those which have an impact through deposition.

2.7 For some important gaseous pollutants, critical levels below which significant harmful effects are not thought to occur[18] have been adopted by, amongst others, the European Union and the United Nations Economic Commission for Europe (UNECE) and are used as regulatory standards. These are summarised in **Table 2.1.** Their origin and use are explained in further detail within **Appendix D**.

2.8 Some other pollutants, for example, heavy metals and hydrogen chloride, are emitted by industrial processes and these pollutants may also need to be assessed. It is recommended that, prior to the assessment of industrial emissions, the scope of any assessment is discussed with the regulator.

2.9 Another gaseous pollutant that has important effects on vegetation is ozone. This is a secondary pollutant, formed in the atmosphere from emissions of nitrogen oxides (NO_x) and volatile/semi-volatile organic compounds. Its production

Table 2.1 Critical levels

Pollutant	Averaging Period	Critical Level
Oxides of nitrogen (NO_x)	24 hours	75/200 µg/m^3 *
	Annual	30 µg/m^3
Sulphur dioxide (SO_2)	Annual	10 µg/m^3 (for lichens and bryophytes)
	Annual	20 µg/m^3
Ammonia (NH_3)	Annual	1 µg/m^3 (for lichens and bryophytes)
	Annual	3 µg/m^3
Hydrogen fluoride (HF)	24 Hours	5 µg/m^3
	Weekly	0.5 µg/m^3

* The critical level is generally considered to be 75 µg/m^3; but this only applies where there are high concentrations of SO_2 and ozone, which is not generally the current situation in the UK. See **paragraph D.4.11** in **Appendix D**.[19]

through photochemical reactions occurs at a considerable distance from the release point and is not amenable to the assessment methods set out in this document. Consequently, no guidance on its assessment is provided.

2.10 For the deposition of air pollutants critical loads, given as a range, for different habitats have been provided by UNECE (see the APIS[20]). APIS provides critical loads for nitrogen deposition (leading to eutrophication) and acid deposition (leading to acidification).

2.11 Critical loads for nitrogen deposition are in units of kilogrammes of nitrogen per hectare per year (kg N/ha/year) and vary with habitat sensitivity.

2.12 Nitrogen and sulphur deposition both contribute to acid deposition, as do some other compounds such as hydrogen chloride. APIS provides a Critical Load Function that defines the contributions from sulphur and nitrogen deposition that will not cause harmful effects. Critical loads for acidification are in units of kilograms of H^+ ion equivalents per hectare per year (keq/ha/year).

[17] www.apis.ac.uk.
[18] There is some more recent evidence that damage can occur at lower levels.
[19] Values taken from www.apis.ac.uk/critical-loads-and-critical-levels-guide-data-provided-apis#_Toc279788051.
[20] Available at www.apis.ac.uk.

3. Case Law

3.1 Several recent judgements in the European and national courts affect the way assessments of the impacts of developments and local plans on designated sites are undertaken. Four are discussed below. A more comprehensive review of case law is provided by the European Commission.[21]

3.2 Natural England's guidance[22] summarises Habitats Directive case law on the meaning of 'likely significant effect' as follows:

- An effect is likely if it *'cannot be excluded on the basis of objective information'*
- An effect is significant if it *'is likely to undermine the conservation objectives'*
- In undertaking a screening assessment for likely significant effects *'it is not that significant effects are probable, a risk is sufficient'....* but there must be credible evidence that there is *'a real, rather than a hypothetical, risk'*.

3.3 The implication of the Wealden Judgement, summarised in **Box 3.1**, means that it is no longer appropriate to scope out the need for a detailed assessment of an individual project or plan using, for example, the 1000 annual average daily traffic (AADT) increase in the Design Manual For Roads and Bridges (DMRB)[23] or the 1% of the critical level or load used by Defra/Environment Agency[24] without first considering the in-combination impact with other projects and plans. This position has been adopted by Natural England in its internal guidance for competent authorities assessing road traffic emissions under the Habitats Directive[25].

3.4 In 2016, the European Commission challenged a 2008 decision by the Federal Republic of Germany to authorise the construction of a coal-fired power station at Moorburg, near Hamburg.[26] The Court of Justice of the European Union (CJEU) ruled that even though the power station was a considerable distance from the Natural 2000 site, there was still a requirement to assess whether there would be was a likely significant effect on the site. The assessment undertaken showed that the power plant drawing cooling water from the river Elbe would result in a high risk for migratory species of fish. As the Court has previously held, competent authorities may authorise an activity only if they have made certain that it will not adversely affect the integrity of the protected site. There should be no reasonable scientific doubt as to the absence of such adverse effects. In this case, the Court ruled that the impact assessment did not contain sufficient definitive data regarding the effectiveness of the proposed mitigation measure. Although this case was not air quality-related, the ruling suggests there must be definite evidence of the efficacy of any mitigation measures proposed.

3.5 This case also concerned the failure of the impact assessment to take account of the cumulative impacts of a pumped-storage power plant and a potential hydroelectric plant on the fish stocks. When assessing cumulative effects, the Habitats Directive requires the assessment to take into account all other projects and plans which, in-combination with the project or plan for which an authorisation is sought, are likely to have a significant effect on a protected site even where those projects/plans precede the date of transposition of the directive.

3.6 In 'People Over Wind', the Irish High Court referred the following question to the CJEU for a preliminary ruling: *"Whether, or in what circumstances, mitigation measures can be considered when carrying out screening for appropriate assessment under Article 6(3) of the Habitats Directive?"*[27]

3.7 In reaching its decision, the Court noted the importance of the precautionary principle to the interpretation of Article 6(3) of the Habitats Directive. The Court judgment, made in 2017, was that it is more appropriate to consider mitigation at the assessment stage than the screening stage. What is unclear is where the boundary lies between what is an integral part of a proposed development and what is a mitigation measure.

3.8 The fourth recent case relevant to air quality assessments was on nitrogen emissions from farms in the Netherlands. In 2018 the CJEU ruled[28] that a reduction in emissions can only be taken into account in an appropriate assessment if the expected benefits are certain at the time of the assessment.

3.9 Previous case law on the interpretation of the Habitats Directive has clarified that 'certain' does not mean absolute certainty but *'where no reasonable scientific doubt remains'*[29] (emphasis added). In the Netherlands case, the CJEU recognised that the measures with which they were concerned had *"not yet been taken or have not yielded any results, so that their effects are still uncertain"*. It is in that context that the CJEU stated *"The appropriate assessment of the implications of a plan or project for the sites concerned is not to take into account the future benefits of such 'measures' if those benefits are uncertain, inter alia because the procedures needed to accomplish them have not yet been carried out or because the level of scientific knowledge does not allow them to be identified or quantified with certainty"*.

3.10 A summary of the ruling in the context of air quality assessments is provided in **Box 3.2**.

3.11 Also, of note is that the Court ruled that the grazing of cattle and the application of fertiliser may be classified as a 'project' under the Habitats Directive, and therefore require an appropriate assessment if it is likely to cause a significant effect on the designated site. The grazing of cattle and application of fertiliser is often a long-established activity, predating the Habitats Directive. The judgement suggests that a change in location, the rate of application, or spreading technique may be sufficient to trigger an assessment. This may mean that more assessments will be required for agricultural developments.

Box 3.1 The Wealden Judgement

Judgment in Wealden District Council v Secretary of State for Communities and Local Government, Lewes District Council and South Downs National Park Authority) [2017] EWHC 351 (Admin)

DATE: 21 March 2017

Wealden District Council challenged a part of the Lewes Joint Core Strategy (JCS) prepared jointly by Lewes District Council (LDC) and the South Downs National Park Authority (SDNPA). The case concerned the approach to in-combination assessments pursuant to the Habitats Regulations.

The principal issue was whether LDC and the SDNPA had acted unlawfully in concluding, on advice from Natural England, that the JCS would not be likely to have a significant effect on the Ashdown Forest Special Area of Conservation (SAC), in-combination with the Wealden Core Strategy. An in-combination assessment of the impact of vehicle emissions on nitrogen deposition on the heathland within the SAC had not been undertaken using advice from the then Highways Agency, in the Design Manual for Roads and Bridges (DMRB)[30]. This states that where annual average daily traffic movements (AADT) resulting from development did not exceed 1000 on affected roads, environmental effects could be regarded as neutral and "scoped" out of any further assessment.

Wealden District Council argued that, whereas its Core Strategy had been prepared on the basis that it would generate 950 AADT on part of the A26 road next to the SAC, the effect of the JCS would be to increase the AADT beyond the 1000 threshold and, on a proper interpretation of the DMRB guidance, this required an in-combination assessment of the effects of both the Wealden Core Strategy and the JCS which had not been carried out in the Habitats Regulations Assessment (HRA) associated with the preparation of the JCS. LDC and the SDNPA argued that no in-combination assessment was required, because the JCS on its own involved the generation of traffic below the threshold and, in applying the guidance, no further in-combination assessment was required.

The Secretary of State also referred to separate guidance relied upon by Natural England and prepared by the Air Quality Technical Advisory Group (AQTAG), to the effect that the 1000 AADT threshold equated to a 1% change in critical loads/levels which, if not exceeded, allowed the decision-maker to conclude that there was no likely significant effect. The advice also stated that experience of permitting allowed the Group to be *"confident that it was unlikely that a substantial number of plans or projects will occur in the same area at the same time, such that their in-combination impact would give rise to concern at the appropriate assessment stage. If such a situation were to arise then the assessment could be determined on a case-specific basis"*. Wealden District Council argued that this confirmed the unlawfulness of the approach taken in the HRA.

The judge found that, on a proper interpretation of the DMRB, at least in principle, in-combination effects are potentially relevant at the initial "scoping" stage as well as the subsequent stage requiring further assessment. It was also concluded that advice from Natural England to LDC and SDNPA on the approach to be taken to the HRA, which relied on the AQTAG guidance, was *"plainly erroneous"*:

It was therefore held that the HRA was *'contaminated'* by Natural England's advice, because LDC and the SDNPA should have undertaken further inquiry of Natural England in circumstances where no explanation had been given for not aggregating the two amounts; and because Natural England's error directly affected the decision-making process. The judge also directed Natural England to reconsider its advice in the light of this judgment and that the DMRB should be re-examined, and clarified, to reflect the concerns indicated.

Box 3.2 The Netherlands Air Quality Judgement

Judgment of the Court (Second Chamber)

DATE: 7 November 2018

Coöperatie Mobilisation for the Environment UA and Vereniging Leefmilieu v College van gedeputeerde staten van Limburg and College van gedeputeerde staten van Gelderland. Requests for a preliminary ruling from the Raad van State Joined Cases C-293/17 and C-294/17

These two cases challenged the Netherlands approach to the assessment and permitting of ammonia emissions from beef, dairy, pig and poultry farms. The Netherlands Government has adopted a strategic approach to regulating nitrogen deposition, known as the Programma Aanpak Stikstof or PAS. This aims to conserve or where necessary to restore the Natura 2000 sites to favourable conservation status whilst also allowing economic growth. The premise of PAS is that nitrogen deposition will reduce, and that half of that reduction can be offset by the emissions from new economic activity.

The national court referred a number of questions to the CJEU. These include whether an appropriate assessment may take into account the existence of conservation measures, preventative measures, measures specifically adopted for a programme or autonomous measures (i.e. those that are not part of the programme; in this case the PAS).

Of most relevance to the way air quality assessments are undertaken in the UK is the following question *"May the positive effects of the autonomous decrease in the nitrogen deposition … be taken into account in the appropriate assessment…, is it important that the autonomous decrease in the nitrogen deposition be monitored and, if it transpires that the decrease is less favourable than had been assumed in the appropriate assessment, that adjustments, if required, be made?"* The judgement states that according to previous case law *"…it is only when it is sufficiently certain that a measure will make an effective contribution to avoiding harm to the integrity of the site concerned, by guaranteeing beyond all reasonable doubt that the plan or project at issue will not adversely affect the integrity of that site, that such a measure may be taken into consideration in the 'appropriate assessment' within the meaning of Article 6(3) of the Habitats Directive"*.

The court concluded that an appropriate assessment may not take into account the existence of conservation measures, preventive measures, measures specifically adopted for a programme such as that at issue in the main proceedings (the PAS) or autonomous' measures (i.e. measures not part of that programme), if the expected benefits of those measures are not certain at the time of that assessment.

The CJEU also considered whether a threshold can be used to exclude projects from authorisation if the court is satisfied that the appropriate assessment carried out in advance meets the criterion that there is no reasonable scientific doubt as to the lack of adverse effect on the integrity of the sites concerned. The CJEU concludes that, under these circumstances, thresholds can be used. It should be noted that the PAS threshold (1 mol N/ha/yr which is equivalent to 0.014 kg N/ha/yr) is lower than 1% of the critical load (1 mol N/ha/yr is 0.28% where the critical load is 5 kg N/ha/yr). It must be ascertained, however, that, even below the threshold values, there is no risk of significant effects being produced which may adversely affect the integrity of the sites concerned.

An appropriate assessment must contain complete, precise and definitive findings and conclusions capable of removing all reasonable scientific doubt as to the effects of the plans or the projects proposed on the protected site concerned.

[21] European Commission, 2018, Commission Notice, Managing Natura 2000 sites - The provisions of article 6 of the Habitats Directive' 92/43/EEC Brussels 21.11.2018, C(2018) 7621 final.
[22] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.
[23] Highways Agency, 2019, Design Manual for Roads and Bridges, Sustainability & Environment Appraisal, LA 105 Air Quality.
[24] www.gov.uk/guidance/air-emissions-risk-assessment-for-your-environmental-permit.
[25] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.

[26] C-142/16 Judgement of the Court 26 April 2017, Action under Article 258 TFEU. For failure to fulfil obligations, brought on 9 March 2016, European Commission v Federal Republic of Germany curia.europa.eu/juris/document/document.jsf?text=&docid=190143&pageIndex=0&doclang=en&mode=lst&dir=&occ=first&part=1&cid=11885397.
[27] C-323/17 Judgement of the Court 12 April 2018, Request for a preliminary ruling under Article 247 TFEU from the High Court (Ireland), made by decision on 10 May 2017, received at the Court on 30 May 2017, in the proceedings of People Over Wind and Peter Sweetman v Coillte Teoranta, curia.europa.eu/juris/document/document.jsf?text=&docid=200970&pageIndex=0&doclang=en&mode=req&dir=&occ=first&part=1&cid=619449.
[28] C-293/17 andC-294/17 Judgment of the Court (Second Chamber) of 7 November 2018 in Coöperatie Mobilisation for the Environment UA and Vereniging Leefmilieu v College van gedeputeerde staten van Limburg and College van gedeputeerde staten van Gelderland. Requests for a preliminary ruling from the Raad van State curia.europa.eu/juris/liste.jsf?language=en&num=C-293/17.
[29] Case C-239/04 Commission v Portugal, 2006, ECR I-10183, para. 24; Holohan *et al* vs. An Bord Pleanála (C-461/17), para. 33.
[30] The relevant section of the DMRB has been replaced by LA 105 Air Quality (see reference 23).

4. Assessment outline

4.1 Introduction

4.1.1 The principal purpose of this document is to set out a procedure for air quality specialists to follow when evaluating the impacts of airborne pollution at designated sites. Whilst an air quality specialist may be able to conclude that there are no likely significant effects using established thresholds, they will not generally be able to assess the effects of the air pollution on the integrity of the designated site. This is the job of an ecologist. This chapter provides an overview of the complete assessment process and, where applicable, the basis for reaching a conclusion that there is no likely significant effect because the air quality impact is too small.

4.1.2 The procedure assumes that the assessment will be collaborative between air quality and ecology specialists since this represents the ideal combination of expertise. Collaboration between the two can be valuable at various stages of the assessment and it is important that the most appropriate specialist undertakes certain tasks. Collaboration can also help to minimise duplication of effort to ensure assessments are undertaken efficiently. The outline stages of an ideal assessment are set out in **Table 4.1**.

4.1.3 It should be noted, however, that ecologists are not engaged on all projects for which an air quality assessment is undertaken. This is a decision for the promoter of the proposed project or plan.

Table 4.1 Outline of assessment stages

Stage	Who	What	Planning/permitting output (all sites)	HRA output (European sites only)	Guidance in this document/ elsewhere
Scoping	Air quality specialist & ecologist	Initial evaluation of potential receptors, consultation with competent authority/ stakeholders	Study area, relevant receptors, pollutants	n/a	Chapter 4 and 5
Quantification & Screening					
Simple assessment	Air quality specialist	Calculate/estimate PC and compare with screening thresholds (1%, 1000 AADT)	Identify if insignificant OR if further assessment required (e.g. detailed modelling)	Screening Identification of likely significant effects (project alone & in-combination)	Chapter 5
	Ecologist		Assessment of significance of effects (inter & intra project)		
Detailed assessment	Air quality specialist	Calculate PC & PEC and compare against critical levels/loads at relevant receptors	Identification of impacts (project alone & cumulative impacts)	Identification of adverse effects on integrity (project alone & in-combination)	
	Ecologist		Assessment of significance of the project alone and cumulative effect (i.e. inter and intra project effects)		
Mitigation & monitoring	Air quality specialist & ecologist	The application of measures to address air quality impacts and associated ecological effects following a mitigation hierarchy, and the use of monitoring		Apply mitigation hierarchy. Identify imperative reasons of overriding public interest (IROPI)	IAQM position statement sets out the basic hierarchical principles for identifying mitigation measures[31]

4.1.4 A more complete description of the air quality assessment procedure follows in **Chapter 5**.

4.2 Scoping

4.2.1 Both the ecology and the air quality specialists should consult with each other and the relevant decision-makers and/or stakeholders prior to commencing their assessments. The results of those consultations should be shared between the specialists to allow the scope of the assessment to be defined. It may be appropriate at this stage to scope out the requirement for an air quality assessment of effects on habitats, because of the absence of relevant pollutants, and/or the lack of proximity of sensitive sites or species.

4.2.2 A summary of the key elements to be considered during the project initiation and evaluation (i.e. scoping) is given in **Box 4.1**.

4.3 Quantification of air quality impacts

4.3.1 **Box 4.2** sets out the key elements of the air quality assessment.

4.4 The ecological assessment

4.4.1 In those cases where effects (alone and in-combination) cannot be definitively described as insignificant on the basis of the air quality assessment alone (see **Section 5**), the ecologist will review the information provided by the air quality specialist and consider the likely significance of the effects.

4.4.2 For European sites the next formal stage is the completion of an HRA. This is largely undertaken by an ecologist.

4.4.3 It is the ecologist's responsibility (where included in the project team) to report the ecological assessment and the conclusions of the assessment. The air quality specialist would normally separately describe their assessment methodology, assumptions, and the impacts on air quality and deposition.

4.4.4 It is important that the ecologist provides the draft ecological assessment report to the air quality specialist to ensure that there has been no misinterpretation of the information.

4.4.5 If the ecologist identifies a significant effect or, for European Sites, adverse effect on the integrity of the site, mitigation and emission control measures need to be explored. These measures may include the need for changes to the project to avoid or reduce the air quality impact and this should be discussed with the air quality specialist, who may need to liaise with other members of the project team, such as the transport consultant or the process engineer designing the installation.

Box 4.1 Initial evaluation (Scoping)

Considerations	Actions
Pollutants: Are there any that may cause adverse effects on vegetation or habitats? Study area: Has the relevant regulator specified any screening distances from air pollution sources? Has the ecologist identified any other designated sites that might be affected by the change in emissions?	Identify designated sites. • Scope out any sites with habitats/species not sensitive to air pollution. • Provide mapping showing the sites to be assessed.

Box 4.2 Key elements of the air quality assessment (Quantification and Screening)

Considerations

Agreement between the air quality and ecology specialists on appropriate critical loads.

Agreement on the locations where estimates of pollutant concentrations and deposition rates are required.

Agreement with the regulator on the 'in-combination' effects that need to be accounted for.

Are the estimated impacts sufficiently small that their effects could be described as insignificant?

Outputs

- A list of sites and/or pollutants that have been screened out and require no further assessment;
- A list of sites and/or pollutants that require further assessment to determine whether, or not, there may be a likely significant effect at the relevant site(s);
- The grid references or areas of the modelled impacts;
- The basis for emissions calculations, and whether it takes into account the operational characteristics e.g. batch processes do not operate continuously;
- A list of the emission sources considered in the in-combination assessment and why they were included or excluded;
- Existing concentrations and deposition rates (except in some cases for permitting);
- The change due to the project, or the 'future baseline with project', and 'future baseline without project' concentrations and deposition rates;
- The change in the case of European sites should be quantified for the project without taking into account mitigation;
- A description of the assumptions used in the assessment e.g. hours of operation, assessment year, location of ecological habitats of concern, and future year conditions. The degree of conservatism and whether there are known uncertainties in the input data. A summary of the habitat categories selected, the critical levels and loads applied and existing concentrations and rates of deposition at each site;
- Tabulated results for the project or plan alone and, in-combination with other projects and plans showing for all pollutants the totals and changes in concentrations and deposition rates at the key locations of interest, and contributions as a percentage of the air quality criteria (critical levels and loads);
- If they are likely to be useful in the interpretation (notably, when assessing impacts of point sources) concentration contours (isopleths) overlaid in a clear manner over an Ordnance Survey (or equivalent) base map.

* Mitigation is generally considered to be any additional measure to reduce or remove emissions, or diminish their impacts, above and beyond those that would be expected to be present as part of a proposal or project design. See also **paragraph 3.7**.

[31] iaqm.co.uk/text/position_statements/mitigation_of_development.pdf.

5. The assessment of air quality impacts

5.1 Introduction

5.1.1 There is a range of existing statutory and non-statutory guidance and supporting tools provided by *inter alia* the Environment Agency (EA), Natural Resources Wales (NRW), Scottish Environmental Protection Agency (SEPA), Highways England, Defra, Natural England and the Air Pollution Information Service (APIS) on how to estimate the impact of a project or plan on ambient concentrations and pollutant deposition. It is not the intention of this chapter to reproduce this guidance, nor to describe how to model concentrations or deposition of air pollutants. This chapter aims to supplement existing guidance with further explanation of the air quality assessment process.

5.1.2 Most of the existing published guidance predates the Wealden Judgement and CJEU rulings described in **Chapter 3**. The exception is guidance for road traffic produced by Natural England in 2018 to address specifically the issues raised in the Wealden judgement[32].

5.1.3 The approaches to air quality assessment differ according to whether the project or plan comprises transport sources, industrial sources[33] or agricultural sources. A single project may include a mixture of these source types and therefore more than one guidance document may be applicable.

5.1.4 There are three stages of the air quality assessment which can be summarised as:

i. Scoping;
ii. Quantification; and
iii. Screening.

5.1.5 This chapter describes the air quality assessment process. Where an assessment concludes that there is a significant effect, or for European sites, a significant effect on the integrity of the site, there may also be an need for air quality mitigation measures to be investigated.

5.2 In-combination impacts

5.2.1 The Habitats Regulations place a duty on competent authorities to assess the effect of new projects and plans both alone and in-combination with other projects and plans, i.e. the effects of the plan or project being assessed must also be considered together with the effects of other relevant projects and plans. This is because a series of individually modest impacts may, in-combination, produce a significant effect on a habitat/species.

5.2.2 For development requiring Environmental Impact Assessment (EIA) under the EIA Regulations[34], there is a requirement to consider the cumulative effects of the development with other relevant developments. This is known as the inter cumulative impacts.

5.2.3 In EIA, cumulative effects can also apply to the combined effects of different impacts, e.g. an increase in air pollution and an increase in noise pollution. This is known as the intra cumulative impacts.

5.2.4 There is, therefore, an overlap between the meaning of 'in-combination effects' and 'cumulative effects' but they are not the same in all cases. In this IAQM document, the term 'in-combination' impact is used to refer to the cumulative air quality impacts of the project or plan being authorised with other relevant projects and plans that are in the public domain[35].

5.2.5 It should, however, be noted that where the impacts are due to road traffic emissions, the cumulative impact may not be explicitly identified (see **paragraph 5.4.1.19**).

5.2.6 Relevant projects and plans to be considered include those that may have been approved but are, as yet, incomplete (e.g. a committed development), the subject of an outstanding appeal, or ongoing review. The air quality specialist and ecologist should liaise with each other and the regulator to agree the list of relevant projects and plans. This information may also reside with other specialists in the wider assessment team, such as transport or planning. Ultimately, for European sites, a decision on the inclusion of other projects or plans is the responsibility of the competent authority.

5.2.7 It is important that the assessor considers the potential for in-combination impacts of plans and projects resulting from all relevant sources of emissions where there could be an overlap of air quality impacts.

5.2.8 Road transport emissions near to designated sites are often the result of many projects and plans located some distance from the site. It is normal in an air quality assessment to include traffic growth estimates using the Department of Transport's TEMPRO[36] growth factors or from a strategic transport model that explicitly includes traffic from other projects and/or plans.

5.2.9 It is, however, rare for a proposed new or enlarged industrial installation to be located close to other proposed new or enlarged industrial facilities and the risk of the plumes overlapping and giving rise to a significant effect on a designated site is generally low. Should these circumstances arise, the dispersion modelling can be extended to account for multiple sources, should the emission data be available. There is a higher likelihood that there will be a cluster of overlapping intensive agricultural emission sources close to designated sites and these need to be considered in assessments[37].

5.2.10 Regarding the permitting of industrial sources, the Air Quality Technical Advice Group (AQTAG) states that *"Experience of permitting allows us to be confident that it is unlikely that a substantial number of plans or projects will occur in the same area at the same time, such that their in-combination impact would give rise to concern at the appropriate assessment stage. If such a situation was to arise then the assessment could be determined on a case-specific basis"*[37].

5.2.11 The impacts from different pollutants also need to be considered, such as the impact on deposition of nitrogen derived from NO_x and NH_3. For example, the NH_3 contribution from agricultural activities may need to be considered together with NO_x and NH_3 emissions from road transport.

5.2.12 Where the impact of an isolated project meets the regulator's screening threshold (see later in this chapter) on its own and there will not be an in-combination effect with other projects or plans, the screening criterion can be used for the project alone. Defining an 'isolated source' precisely is not possible, and it is a matter for an experienced air quality specialist to use their professional judgement in consultation with the regulator. If there is any doubt, it should be assumed that there may be an in-combination effect.

5.2.13 Further advice on in-combination assessments is provided in the European Commission's 2018 guidance[38].

5.3 Stage 1. Scoping

5.3.1 The first stage in any assessment is to consult with the relevant regulator and stakeholders to ensure that the scope and approach to the assessment meet their requirements. Depending on the type of project, the stakeholders may include the relevant Statutory Nature Conservation Organisation (SNCO) (e.g. Natural England), environmental agencies and other potentially affected public and private bodies and special interest groups. The objective of this stage is to identify the *scope* of the assessment, in terms of the relevant habitats to be included, and to *screen* out any emission sources on the grounds that they are too small or too far away from a habitat to have a meaningful effect.

5.3.2 The locations and boundaries of international and national designated sites can be found online, e.g. on the MAGIC website or similar online resources from the relevant SNCO. If local sites are to be assessed, details can be obtained by consulting the Environment Agency or local biodiversity records office who may charge a nominal fee for this service. Sufficient time should be allowed to obtain this data.

5.3.3 For individual planning applications for conventional residential or mixed-use development where European sites are a consideration, the assessor should first investigate whether the air quality issues have already been fully explored for the Local Plan HRA. If this has been done, then it would be appropriate and in line with government guidance[39] to defer to that over-arching Local Plan assessment. This should be a suitable approach for windfall development[40] as well as actual allocations, as Local Plans all make an allowance for a specified quantum of windfall development in particular locations and this should be included in the strategic Local Plan air quality assessment and HRA.

5.3.4 Similarly, if a given local authority believes that Neighbourhood Plans will be coming forward in their authority boundary, they should consider including any sites allocated in those plans in their air quality modelling. This would also avoid problems for the planning application or Neighbourhood Plan that might otherwise result from the Wealden judgment (see **Box 3.1**). Deferring 'upwards' to the Local Plan also addresses the undesirable situation of having multiple traffic and air quality models for a single local authority area and the potential inconsistencies that can be introduced in such circumstances.

5.3.5 For projects requiring assessment, the air quality specialist, assisted by the ecologist, should identify the designated sites likely to be affected by the source of emissions to air that require assessment, taking into account distance criteria in the relevant guidance documents, where these exist, relevant to the scale and type of development being assessed. These criteria are described below.

5.3.6 The Design Manual for Roads and Bridges (DMRB)[41] describes the approach for the assessment of the impact of emissions from schemes on the strategic road network. A quantitative air quality assessment is required if European Sites are within 200 m of affected roads. Within this context, the distance of the affected road from the designated site is an important consideration. Air pollution levels fall sharply within the first few tens of metres from a road before reducing more slowly with distance. The air quality impact of a given change in traffic on a designated site where the relevant habitat/species is 100 m from a road will be very different to one that abuts the road.

5.3.7 For strategic planning, where substantial changes in traffic volumes are being considered, there is the potential for wider-scale impacts, which can potentially affect the future background concentrations, as well concentrations within 200m of individual roads within the affected network. In these circumstances, the modelling may need to encompass a large road network.

5.3.8 Natural England[42] advises that the next step is to identify the spatial distribution of qualifying features within a designated site. If there are no qualifying features sensitive to air pollution within 200 m of a road, then no further assessment is required. For example, a chalk river will not typically be sensitive to acid deposition because of its natural buffering capacity. In these circumstances, a screening conclusion of no likely significant effect on the site can be reached with regard to air quality without undertaking any modelling.

5.3.9 In some cases, a road surface and its adjacent verges may be included within a designated site's boundary. This does not necessarily mean that they will be of nature conservation interest and form part of a qualifying feature. This inclusion might simply be for convenience, e.g. for defining a boundary. These areas will, therefore, be of no special nature conservation interest. Conversely, at some sites, roadside verges may have been deliberately included within a site boundary and be an integral part of a designated site. It is important that the air quality specialist works with the project ecologist to make these decisions.

5.3.10 If a project/plan has not been screened out using the criteria outlined above, the next step is to consider the risk of the road traffic emissions using either the annual average daily traffic flow (AADT) or the predicted air quality impacts.

5.3.11 The DMRB provides a series of traffic screening criteria. These include the change in AADT flows on a given road of 1000 vehicles or 200 heavy duty vehicles (HDVs). These thresholds have been widely used to screen out the need for quantitative assessment of projects/plans in the absence of any other thresholds recognised as being applicable in this context.

5.3.12 The 2017 Wealden judgment[43] (see **Box 3.1**) has clarified that, if the DMRB screening criteria are used, they should be used to screen in-combination impacts as well as the project/plan alone.

5.3.13 The Defra/Environment Agency's *Air emissions risk assessment for your environmental permit* (which applies to industrial emission sources) currently identifies distances of 2 km for local and nationally important sites and areas of ancient woodland, and 5, 10 or 15 km depending on the emission source for European Sites. Smaller industrial facilities or waste sites may not require such a large study area. Different distances apply for agricultural emissions. The air quality specialist should check first with the relevant regulator/SNCO what distances apply as they can vary. Different regulators throughout the United Kingdom have different criteria in some cases, most notably for ammonia emissions from livestock.

5.3.14 It is important that the air quality specialist and the ecologist discuss the types of habitat located within the distance criteria. It may be that a site is screened-in, but the relevant habitat feature/species is not present in the study area (e.g. based on APIS or site survey data) nor needs to be present for the site to achieve its conservation objectives. In this circumstance, a conclusion of no likely significant effect on the site can be reached with regard to air quality without undertaking any modelling.

5.4 Stage 2. Quantification

5.4.1 Approach and methods

5.4.1.1 Once all the required information on the project/plan and the projects/plans for the in-combination assessment, has been gathered, concentrations and deposition rates will be calculated.

5.4.1.2 The change in pollutant concentrations due to an industrial or agricultural source is often determined simply by modelling the dispersion of the emissions. This is known as the process contribution (PC)[44]. The in-combination[45] impacts would then be assessed by adding the PCs from the other relevant projects and plans.

5.4.1.3 The PCs may be calculated by a variety of methods, depending on the circumstances and scale of the project. For a simple approach, for instance, at a screening stage, a spreadsheet tool such as the Environment Agency's risk assessment tool[46] or the Simple Calculation of Atmospheric Impact Limits (SCAIL)[47] may be used. However, these models have limitations[48]. In reality, detailed dispersion modelling is used in most cases.

5.4.1.4 To determine the concentration/deposition rates, the PC is added to the baseline concentration/deposition rates. These may be taken from measurement data or other appropriate sources such as Defra or APIS background maps. The concentration/deposition rate is known as the predicted environmental concentration (PEC).

5.4.1.5 Case law (see the Moorburg case in **Chapter 4**) suggests that it may no longer be sufficient to rely solely on the background data provided by Defra and APIS in all assessments, as these provide 'average' data and are typically based on emissions data for a time period which does not encompass newly operating facilities. Some assessments may need to include the impact of existing sources explicitly or those that

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have recently started operating. For this reason, the term 'baseline' is used instead of 'background' in this document.

5.4.1.6 It may also be worthwhile investigating whether operators of existing facilities or local authorities are required, or intend, to make improvements that will change air emissions and consequently future baseline concentrations.

5.4.1.7 Measurements of pollutant concentrations are made by local authorities and Defra and are available online or on request. The choice of data source will often depend on the location of the study area, i.e. whether urban or rural, which dictates the amount of monitoring data available. Site-specific monitoring (e.g. using diffusion tubes) is sometimes undertaken to determine baseline concentrations, to obtain the most up to date information or where appropriate baseline data are not available.

5.4.1.8 The APIS website holds a database of three-year average pollutant concentrations and deposition rates. These are available for five by five kilometre grid squares covering the whole of the UK. CEH also maintains another database of results from its Concentration Based Estimated (CBED) model[49]. This provides deposition rates for nitrogen and sulphur for three year averages at 5 km^2 resolution for two surface types (forest and moorland).

5.4.1.9 As noted earlier, care should be taken to ensure all relevant emission sources are included in the baseline concentration selected for each receptor location. For example, it is important that, where a sensitive receptor is close to a busy road, the contribution from traffic emissions on that road is explicitly included in the estimation of the PEC.

5.4.1.10 For projects/plans that generate road traffic, the dispersion modelling will estimate the PEC "without the project/plan" (i.e. the future baseline) and PEC concentrations "with the project/plan". The PC is derived by subtracting one from the other. This future baseline typically takes account of the traffic from other projects/plans. To calculate the in-combination PC another scenario will need to be modelled. This may use the baseline traffic data with future emission factors to provide an alternative future baseline PEC. Subtracting this from the project/plan PEC described in the last paragraph will provide the in-combination impact. This approach enables the future decline in road traffic NO_x emissions per vehicle km to be taken into account, although this may be offset to some extent if NH_3 emissions per vehicle km increase in the future.

5.4.1.11 The road traffic PC could be calculated using the publicly available version of the 2007 DMRB spreadsheet model; this, however, dates back over a decade and uses out of date emission factors and fleet composition[50]. The IAQM recommends that the latest version of the Emission Factor Toolkit (EFT) and dispersion modelling is used.

5.4.1.12 When modelling the dispersion of emissions, it is good practice to assess several points within each designated site, both along the site boundary and within the site itself (for point emission sources, this may be a grid of receptors, or for a road, a transect) to identify the maximum impact (i.e. the PC) at the site, as well as the range that may be experienced across the entire area[51].

5.4.1.13 Concentrations should not, however, be predicted too close to the roadway, since such predictions can be unreliable and may not represent areas of relevance to the assessment. It is recommended, for example, that predictions are not made closer than 2 m from the edge of a road.

5.4.1.14 The maximum PC within, or on, the boundary of the designated site should be used to provide a robust assessment (where that coincides with the presence of a habitat or species of concern).

5.4.1.15 Consideration should also be given to the distribution of habitat features of interest within the site. A single receptor point may be adequate if the site area is small and is situated a relatively large distance from the source, as there is less potential for variation in concentrations and deposition rates across the site. The air quality specialist should consult the appropriate guidance for determining the approach to selecting receptor points and grids[52].

5.4.1.16 The surface roughness in the wider area will affect the modelled ground level concentration of pollutant. A suitable value (or values) should be used, in line with model guidance.

5.4.1.17 Multiple years of representative meteorological data (typically three to five consecutive years, depending on the type of assessment) should be used in the dispersion modelling of point sources; for road schemes, one year is normally sufficient (according to LAQM TG16)[53].

5.4.1.18 For road transport sources, individual receptors along a transect, or along a series of transects at suitable intervals, perpendicular to the road up to 200 m are generally used[54]. As NO_x emissions from road traffic and other sources are forecast to decrease in the future, it is appropriate to estimate future air quality (see below). For a project, this usually is the year when it will be first operational. For large projects, several future years may be used, with and without the project, to provide information on the impacts during phases of development. For land use plans, the end year of the plan period is normally used as this is when the development set out in the plan may have been fully built out. This may, however, miss the potential for significant effects as there is a balance between traffic growth and declining emissions per kilometre from vehicles. Modelling one or more intermediate years should be considered.

5.4.1.19 Transport consultants often do not provide separate data to enable the impact of the other projects or plans to be explicitly estimated; however, a decision maker may require this to be assessed so they can review the impact of the project/plan alone and in-combination with other projects/plans. It is therefore important for the air quality specialist to consult with the decision maker and transport consultant at the earliest opportunity. (Also see **paragraph 5.4.1.10**).

5.4.1.20 The changes in deposition rates (i.e. PC) resulting from the project or plan for the pollutants of interest are typically derived from the product of the atmospheric concentration and the deposition velocity[55] (taking into account the units). The best available estimate of the deposition velocity available should be used for this calculation. The deposition velocity depends on the vegetation type (this can be general, for example, forest or heathland) at the location of interest. This information on vegetation type can be informed by the ecologist.

5.4.1.21 The most commonly used values are shown in **Table 5.1**, taken from AQTAG guidance[56]. An air quality specialist may choose to derive their own deposition velocities based on a

Table 5.1 Deposition velocities (after AQTAG)

Pollutant	Habitat	Deposition velocity (m/s)
NO_2	Grassland	0.0015
	Forest	0.003
SO_2	Grassland	0.012
	Forest	0.024
NH_3	Grassland	0.020
	Forest	0.030
HCl	Grassland	0.025
	Forest	0.060

review of published data. The source of the deposition velocity and justification for its use should be provided.

5.4.1.22 The Environment Agency's *"Guidance on modelling the concentration and deposition of ammonia emitted from intensive farming"*, where relevant, should be referred to when calculating deposition of ammonia from intensive farming[57].

5.4.2 Future concentrations and deposition rates

5.4.2.1 Natural England guidance[58] signposts the APIS website which provides information on deposition trends drawn from the results of national modelling over a number of years. APIS is updated annually, though background trends are a 3-year average to account for weather variation. The trend data for these 3-year averages are provided for maximum and minimum deposition (nutrient nitrogen and acid).

5.4.2.2 The APIS website also provides background concentrations data, but the higher spatial resolution background data available from Defra for certain pollutants should be used when possible. Note that it may be necessary to forecast future concentrations taking into account sources of emissions not directly relevant to the project/plan under consideration, such as road traffic for industrial projects.

5.4.2.3 The air quality specialist may choose to assume no change in future baseline concentrations or deposition rates, where there is no evidence to indicate that they may decrease in value. This may be appropriate if, for example, the project/plan under consideration is likely to be completed within a relatively short period of time (one or two years in the future). If there is a long lead-in period (due to construction and/or commissioning periods), it may be more appropriate to reduce future baseline concentrations/deposition rates to allow for anticipated improvements in national emissions. (There is an IAQM Position Statement on the uncertainties in the estimation of future road traffic emissions).

5.4.2.4 The judgement in the Netherlands cases concludes that 'autonomous measures' (see **Box 3.2**) can only be taken into account if it is sufficiently certain that the measure will deliver as anticipated. There is clear evidence that UK NO_x emissions, including those from road traffic, are declining and will continue to do so in the future. NO_2 concentrations are also declining.

5.4.2.5 What is not certain is the exact rate of reduction of NO_x emissions and therefore it is important that a conservative estimate is used for the modelling. There are reasons to believe that Defra's current Emission Factor Toolkit (version 9.0) may overestimate emissions over the longer term. This is because the assumptions in the fleet turnover model that is used in EFT do not reflect recent developments in either national policy nor in purchasing trends relating to diesel and non-conventional cars[59].

5.4.2.6 The Netherlands case also clarifies that a mechanism must be in place to ensure that the expected reductions take place. In the UK, the Government has published a Clean Air Strategy, which sets out the mechanisms by which the target of a 73% reduction in NO_x emissions will take place by 2030 (relative to a 2005 baseline). This will ensure compliance with the National Ceilings Emission Directive. The strategy also includes a target for the reduction of deposition of reactive forms of nitrogen in England's protected priority sensitive habitats.

5.4.2.7 There is more uncertainty regarding ammonia emissions, but the government is legally committed, under the 2016 National Emissions Ceiling Directive to reduce these emissions, along with SO_2 and NO_x emissions. The UK has a good track record on meeting its international emission reduction obligations. Any assumption should be clearly explained, with justification given.

5.4.2.8 It should be recognised that there is a non-linear relationship between emissions, concentrations and deposition and these relationships may change in the future because of changes in atmospheric chemistry.

5.4.2.9 Whichever approach is adopted, it is advisable to gain agreement in advance from the regulator and explain to the ecologist the basis of assessment, so they can use the information in their judgement of significance, particularly where precautionary assumptions have been applied.

5.5 Stage 3. Screening

5.5.1 Introduction

5.5.1.1 A database of site-specific critical loads for nitrogen and acid deposition rates is available on the APIS website[60]. Unless the lowest value for a high-level screening assessment is adopted, the selection of critical loads requires knowledge of the habitat type, site interest features, and specialist knowledge, such as whether the environment is nitrogen or phosphorous limited, or whether grassland is acidic or calcareous. It also requires knowledge as to the relative reliability of the critical load in question as some are supported by firmer evidence than others. This is identified on the APIS website.

5.5.1.2 In many circumstances, the air quality specialist will suggest the assessment criteria, although this may be modified by an ecologist in the light of knowledge of the habitat in question. It should be noted that ecologists are not appointed for the assessment of all developments and the air quality specialist may need to use professional judgment.

5.5.1.3 APIS does not cover all habitat types. In these cases, for an assessment to be undertaken advice from a suitably qualified ecologist is required.

5.5.1.4 For each site, and for each habitat within each site, the air quality specialist should calculate the PC as a percentage of the relevant critical level/load both alone and in-combination with other plans and projects.

5.5.1.5 The calculated maximum PC as a percentage of the relevant critical load/level is used to determine whether the impacts will have an insignificant effect or, conversely, may be large enough to warrant further evaluation by an ecologist.

5.5.1.6 In the case of Environment Agency permitting, an increment of 1% (or less)[61] of the relevant long term critical level or critical load alone is considered inconsequential. A change of such magnitude, i.e. two orders below the criterion for harm to occur, is challenging to measure (even by the most precise air quality instrument)[62] and difficult to distinguish from natural fluctuations in measured data (due to other variables such as variations in emissions and weather). For this reason, and others, it has been used as a precautionary screening criterion.

5.5.1.7 The 1% threshold has become widely used throughout the air quality assessment profession to define a reasonable quantum of long term pollution which is not likely to be discernible from fluctuations in background/measurements[63]. For example, for many habitats, 1% of the critical load for nitrogen deposition equates to a very small change of less than 0.1 kgN/ha/yr, well within the expected normal variation in deposition. Its use has not been challenged by the courts, but it should be used in the context of an in-combination assessment.

5.5.1.8 Crucially, the 1% screening criterion is not a threshold of harm and exceeding this threshold does not, of itself, imply damage to a habitat.

5.5.1.9 For all types of project/plan, if the air quality specialist identifies that the impact is sufficiently large (alone and/or in-combination) that it cannot be screened out and therefore it could have a potential significant effect, the information should be passed to the ecologist to use their expertise to determine whether or not there is, in fact, a likely significant effect of the project or plan on the habitat, and, if so, whether for European Sites it is possible to ascertain that there will be no adverse effect on the integrity of the site and for other types of designated sites, no likelihood of damage.

5.5.1.10 If the ecologist concludes, however, that an adverse effect on site integrity cannot be ruled out, the air quality specialist may be required to undertake an assessment of the impact of mitigation measures including providing advice on emission control measures that could be employed to prevent avoid, minimise or reduce impacts. The air quality specialist should provide evidence on the efficacy of any recommended mitigation measures.

5.5.2 Industrial point sources

5.5.2.1 The Environment Agency's risk assessment guidance[64] includes a series of criteria to define when impacts can, in their view, be screened out for an individual installation for the purposes of permitting. It should be noted these criteria are intended to be applied to simple and cautious calculation methods (e.g. the risk assessment tool). They are, however, commonly applied to all assessments, both those that have used a dispersion model to estimate the PC and point sources not regulated by the Environment Agency or equivalent organisation. For Ramsar, European and national designated sites, the guidance advises that to screen out the need for further assessment, a PC for any substance emitted from an industrial source[65] must meet both of the following criteria:

- the short-term PC is less than 10% of the short-term environmental standard[66]; and
- the long-term PC is less than 1% of the long-term environmental standard.

5.5.2.2 For local wildlife sites and ancient woodlands, the Environment Agency uses less stringent criteria in its permitting decisions. Environment Agency policy for its permitting process is that if either the short-term or long-term PC is less than 100% of the critical level or load, they do not require further assessment to support a permit application. In ecological impact assessments of projects and plans, it is, however, normal practice to treat such sites in the same manner as SSSIs and European Sites, although the determination of the significance of an effect may be different. It is difficult to understand how the Environment Agency's approach can provide adequate protection.

5.5.2.3 In March 2015, AQTAG clarified to the Planning Inspectorate that *'For installations other than intensive pig and poultry farms, AQTAG is confident that a process contribution (PC, as predicted by H1 or a detailed dispersion model) < 1% of the relevant critical level or load (CL) can be considered inconsequential and does not need to be included in an in-combination assessment'*[67].

5.5.2.4 AQTAG has also drawn a clear distinction between *'projects and plans considered to be inconsequential and never likely to have an in-combination effect (and so not included in any assessment of likely significant effect in-combination with a new plan or project)'* and those concluded to have *'no likely significant effect'* (insignificant alone but which may need to be considered in the assessment of any other new plans or projects)[67].

5.5.2.5 These recommendations made by AQTAG were made prior to the most recent court rulings. This advice may change in the future and alter the circumstances in which the screening criteria can be used with confidence. This is why it is important to consult with the relevant regulator.

5.5.2.6 In the IAQM's opinion, the 1% and 10% screening criteria should not be used rigidly and, not to a numerical precision greater than the expression of the criteria themselves. Whilst it is straightforward to generate model results for the PC to any level of precision required, the accuracy of the result is much less certain and it is unwise to place too much emphasis on whether the PC is 0.9% or 1.1%, for example. In practice, because the magnitude of impacts attributable to new sources is often around 1% of the criterion, a regulator may require the results to be presented at greater resolution, i.e. having one (or more) decimal places. The distinction here is between the presentation of the model results and the weight given to fine differences around the criterion itself in making a judgement.

5.5.2.7 It is important to remember that a change of more than 1% does not necessarily indicate that a significant effect (or adverse effect on integrity) will occur; it simply means that the change in concentration or deposition rate cannot in itself be described as numerically inconsequential or imperceptible and therefore requires further consideration.

5.5.3 Predicted Environmental Concentration (PEC)

5.5.3.1 The PEC (which applies to both annual mean concentrations and deposition rates) should be calculated with the project or plan alone and in-combination with other projects and plans to identify whether the critical levels or critical loads will be exceeded. This information should be passed on to the ecologist if the PC exceeds 1% of the critical level/load either alone or in-combination.

5.5.3.2 The Environment Agency risk assessment guidance states that if the PEC is less than 70% of the long-term criterion it can be deemed to be insignificant, regardless of the PC. For some pollutants (nitrogen deposition, in particular) background values

are high over much of the UK and it is unlikely there will be many occasions where the PEC is less than 70%. Also, this was intended to be a trigger for detailed dispersion modelling. It is not intended to be a damage threshold.

5.5.4 Traffic impacts

5.5.4.1 There is evidence that ammonia emissions from road vehicles may contribute more than half the local traffic related increment to nitrogen deposition[68].

5.5.4.2 The DMRB methodology[69] only requires the assessment of NO_x emissions and nitrogen deposition. It does not consider NH_3 or its contribution to nitrogen deposition. As road transport is a source of ammonia, albeit a small source compared to agriculture at a national level, consideration should be given to including it and its contribution to local nitrogen deposition.

[32] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.
[33] Including residential and commercial large boilers, combined heat and power plants, and data centres which may not typically be considered to be industrial sources of emissions.
[34] The Town and Country Planning (Environmental Impact Assessment) Regulations 2017.
[35] Sufficient data to quantify the impacts is only likely to be available for projects where planning permission (or other regulatory consent) has been applied for or granted but not yet implemented. For proposed plans data is only likely to be available in the public domain for those that are published for consultation.
[36] Trip End Model Presentation Program (TEMPRO) www.gov.uk/government/publications/tempro-downloads.
[37] Air Quality Technical Advisory Group, AQTAG21, 'Likely significant effect' – use of 1% and 4% long-term thresholds and 10% short -term threshold, Updated version approved 2 October 2015.
[38] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.
[39] In July 2012, Defra issued guidance on 'Competent Authority Co-Ordination under the Habitats Regulations', which recommends that '*Competent authorities should adopt the reasoning, conclusion or assessment of another competent authority, if they can*' and goes on to state that where another competent authority is a specialist in the issues under consideration, robustness of the previous HRA *'can be assumed ... if the reasoning, conclusion or assessment was undertaken or made by a competent authority with the necessary technical expertise'*. www.gov.uk/government/publications/guidance-on-competent-authority-coordination-under-the-habitats-regulations.
[40] Development that does not have a specific allocation, usually because local authorities do not allocate development sites below a certain size.
[41] Highways Agency, 2019, Design Manual for Roads and Bridges, Sustainability & Environment Appraisal, LA 105 Air Quality.
[42] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.
[43] Wealden District Council vs Secretary of State for Communities and Local Government. Lewes District Council and South Downs National Park Authority and Natural England. [2017] EWHC 351 (Admin) (See Box 3.1).
[44] HRA requires the in-combination effects to be assessed; Environmental Impact Assessment requires the cumulative impacts to be assessed. For assessing air quality impacts these terms may be different.
[45] Process contribution (PC) is a term used in the Environment Agency to define the contribution to ambient concentrations and deposition rates due to the emissions from the process being permitted. In this guidance the term is also applied to the contribution from any source such including road traffic, and commercial boilers.
[46] Formerly called the H1 screening tool, available at www.gov.uk/government/collections/risk-assessments-for-specific-activities-environmental-permits#H1-software-tool.
[47] A suite of screening tools for assessing the impact from agricultural and combustion sources on semi-natural areas. Produced by CEH, available at www.scail.ceh.ac.uk.
[48] iaqm.co.uk/text/position_statements/screening_tools_interim.pdf.
[49] www.pollutantdeposition.ceh.ac.uk/data.
[50] An updated version of the spreadsheet is available from the Overseeing Organisations (e.g. Highways England) for use on their road schemes.
[51] For some sources, notably tall point sources, it is not always the case that the highest modelled concentrations will occur on the site boundary closest to source.
[52] admlc.files.wordpress.com/2015/08/dispersion-model-guidelines-v1-5.pdf.
[53] laqm.defra.gov.uk/documents/LAQM-TG16-February-18-v1.pdf.

[54] A distance of 200 metres is generally used as concentrations from the road source decrease rapidly with distance from the source and beyond this distance the road source contribution is not typically discernible from fluctuations in the background concentration. See DMRB Volume 11.3.1, Appendix C, Figure C.1. The receptor locations along the transect should be chosen so as to capture the salient features of the changes in concentration or deposition rate.
[55] This quantity is correctly described as the 'deposition flux', a term used by the Environment Agency, for example. In this document, the term deposition rate is used instead, on the grounds that we are using deposition flux as a proxy for the quantity of pollutant deposited on the habitat over a defined period of time.
[56] Air Quality Advisory Group, 2014, AQTAG06 Technical guidance on detailed modelling approach for an appropriate assessment for emissions to air.
[57] Defra/Environment Agency, 2018, Intensive farming risk assessment for your environmental permit, www.gov.uk/guidance/intensive-farming-risk-assessment-for-your-environmental-permit.
[58] Natural England, 2018, Natural England's approach to advising competent authorities on the assessment of road traffic emissions under the Habitats Regulations.
[59] The proportion of new cars that are diesel has fallen from a peak of just over 51% in 2012 to 30% in the third quarter of 2018. Electric and plug in hybrids accounted for 2.5% in the same quarter. EFT v8.021 assumes that there will be zero miles driven on rural roads in 2030 by electric vehicles. www.gov.uk/government/statistical-data-sets/veh02-licensed-cars, Table 253.
[60] www.apis.ac.uk/indicative-critical-load-values.
[61] For intensive farming the Environment Agency environmental permitting guidance use 4%.
[62] The Ambient Air Quality Directive (2008/50/EC) sets data quality standards for monitoring; e.g. for NO_x (including NO_2) automatic monitors the uncertainty requirement is 15%; indicative methods (such as diffusion tubes) is 30%. It should be noted that deposition is not routinely monitored, but calculated from ambient concentrations.
[63] Some readers will be aware that the EPUK/IAQM planning guidance defines a method for describing the severity of impacts. Within this framework, an impact that is 0.5% of an assessment level is defined as negligible and can be regarded as not having a significant effect on air quality. There is no contradiction between this part of the impact descriptor framework in the planning guidance and the choice of 1% as a screening criterion for habitats. The two values serve a different purpose and have different origins.
[64] www.gov.uk/guidance/air-emissions-risk-assessment-for-your-environmental-permit.
[65] There are different screening criteria for agricultural sources.
[66] The short-term thresholds are only applied to the critical level rather than the critical load (since there are no short-term exposure critical loads) and is only relevant to point source emissions rather than vehicle exhaust emissions.
[67] AQTAG position regarding In-combination guidance and assessment. Correspondence between AQTAG and PINS. March 2015.
[68] Air Quality Consultants, 2020, Ammonia Emissions from Roads for Assessing Impacts on Nitrogen-sensitive Habitats.
[69] Highways Agency, 2013, Interim Advice note 174/13, Updated advice for evaluating significant local air quality effects for users of DMRB Volume 11, Section 3, Part 1 'Air Quality (HA207/07). Note that IAQM considers that there is a typo in this note and that NO_x concentrations and critical levels should be used (i.e. not NO_2 concentrations and critical levels).

6. Local plans

6.1 It has been increasingly common practice for air quality assessments to be undertaken to support the development of Local Plans for local planning authorities where sensitive internationally important wildlife sites may be affected. This is to enable an HRA to be completed for that Local Plan. Due to the nature of Local Plans, this means that all growth expected across a given district over a long period is assessed collectively.

6.2 The DMRB AADT thresholds were useful to decide whether the air quality assessment for a given Local Plan needed to consider a particular European site. Since the Wealden judgement, the 1000 AADT threshold cannot be applied, at least not rigidly, to growth arising from a single district.

6.3 One of the issues with an assessment of a local plan is how far the air quality assessment needs to 'cast its net'.

6.4 The scale of physical separation between a Local Plan area and European site will clearly be an important factor in making the decision, given that there is a limit to the accuracy of transport modelling at considerable distances (i.e. tens of kilometres) from source, as well as the fact that very small changes in air quality (such as are likely to occur at these distances) are unlikely to be detectable in air quality calculations, or in monitoring data. Distance alone, however, does not automatically mean that the contribution of growth in a given Local Plan area will be imperceptible. This will also be a function of the nature and scale of the development, the presence of strategic routes roads linking the development to the European site and consideration of journey-to-work and trip distribution data.

6.5 It is no longer appropriate for individual local authorities to rely purely on a change in flows of less than 1000 AADT as a reason to dismiss traffic-related air quality impacts in-combination with other local plans, unless there is reason to believe that flows on the road in question would be likely to be dominated by journeys arising from that district (for example, a minor road).

6.6 Since the judge in the Wealden case did not dismiss the use of the 1000 AADT threshold entirely, but only as a threshold to automatically rule out *individual* Local Plans there will be a greater need for local authorities to consider modelling their Local Plan air quality impacts collectively, as a group of authorities around a particular European site, rather than creating separate individual models. This already happens in some parts of the UK, such as within the Partnership for Urban South Hampshire and among Councils to the north and west of Epping Forest SAC.

6.7 The 'alone' assessment should be a comparison of a scenario which includes the background traffic growth (sometimes referred to as a 'do nothing scenario' but not the Local Plan, with a 'with Plan' scenario which adds on the Local Plan traffic.

6.8 Importantly, the air quality calculations should also make reasonable assumptions about expected changes in the baseline NO_2 concentrations over the plan period; given the 15 to 20 year or so timescale of most Local Plans. To assume no improvement over a 15 or 20 year period, would effectively ignore the more stringent legal requirements for vehicle NO_x emission standards to be achieved under real world driving conditions, trends in new vehicle registrations and ongoing government and international initiatives to improve air quality through reductions in emissions. Making a suitable allowance for improvements in baseline air quality can, given the long timescale of most Local Plans, mean that overall air quality at the end of the plan period is very likely to be better than air quality at the start, even allowing for the effects of Local Plan growth on traffic flows. It should be noted that there is no presumption that this improvement can be exploited for allowing unacceptable air quality impacts, with consequent effects on designated sites.

6.9 For ammonia emissions, it is more difficult to be certain regarding future trends, and it seems reasonable to either assume no change or to assume that emissions will change in line with the requirements of the 2016 National Emissions Ceiling Directive.

6.10 The application of national forecasts to local conditions may need to be justified to ensure the assessment is robust and not subject to challenge.

6.11 Assessing the results of both the 'alone' and 'in-combination' assessments, it is possible to identify the relative contribution of the Local Plan being assessed. This is necessary if the ecologist concludes that there is an adverse effect on the integrity of the designated site to enable the appropriate scale of mitigation measures that may be needed (such as transport management plans, rerouting of heavy duty vehicles,). If, for example, the Local Plan makes little or no difference to the nitrogen deposition when reported to the limits of reliability then little or no action would be specifically required to address the contribution of that Local Plan.

6.12 Additionally, if the ecologist concludes that there is no likely adverse effect on the integrity of the designated site no mitigation would be required. Given the likelihood that many Local Plan air quality assessments will identify an overall net improvement in air quality over the plan period, the contribution of the individual Local Plan(s) will often be in the form of potential retardation in improvement (i.e. a delay), rather than a deterioration. That is an important distinction in making judgments on adverse effects, although it may still be appropriate (depending on the scale of that delay) to introduce measures to address the plan contribution.

6.13 The preceding discussion is concerned with Local Plans, but the same principle would apply to the traffic impacts of Minerals & Waste Plans where those plans are expected to result in any net increase in vehicle movements within 200 m of sensitive designated sites. Minerals allocations may not result in a net change in vehicle movements due to the nature of minerals sites being worked sequentially (in other words, the 'growth' is in duration of operation rather than scale of activity); similarly, traffic associated with minerals and waste sites may be restricted to certain roads that would not lead them past designated sites. These factors will, therefore, be an important consideration in determining the need for traffic modelling or air quality calculations.


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7. General principles

7.1 There are a number of principles that should be applied when undertaking assessments of the air quality impacts on designated sites, which are set out below.

1. Suitably qualified, experienced and competent assessors should be responsible for the assessment.
2. A precautionary approach is required.
3. The assessment should be appropriate to the risk.
4. The assessment should be undertaken with an ecologist.
5. Always consult with the regulator.

7.2 Suitably qualified, experienced and competent assessors should be responsible for the assessment

7.2.1 All assessments require the use of professional judgement, as it is not possible to provide detailed guidance that covers the individual circumstances of all projects and plans that require assessment. Therefore, all assessments should be undertaken by suitably qualified and competent assessors or under the close supervision of such a person. It is considered useful for the air quality assessment report to include a short biography of each person involved in its production together with their role in the project.

7.3 A precautionary approach is required

7.3.1 Where there is uncertainty in an evaluation of the impact of a project or plan, a precautionary approach is required. This requirement is set out in Article 191 of the Treaty on the Functioning of the European Union. It aims to ensure a higher level of environmental protection than would be the case if this approach was not used. Similar provisions are to be set out in the Environment Bill, and therefore the intention is that the same approach will apply when the UK leaves the EU.

7.3.2 The European Commission guidance[70] on the precautionary principle states its application shall be informed by:

- *"the fullest possible scientific evaluation of the determination, as far as possible, of the degree of scientific uncertainty;*
- *a risk evaluation and an evaluation of the potential consequences of inaction;*
- *the participation of all interested parties in the study of precautionary measures, once the results of the scientific evaluation and/or the risk evaluation are available."*

7.3.3 In addition, the general principles of risk management remain applicable when the precautionary principle is invoked. These are the following five principles:

- *"proportionality between the measures taken and the chosen level of protection;*
- *non-discrimination in the application of the measures;*
- *consistency of the measures with similar measures already taken in similar situations or using similar approaches;*
- *examination of the benefits and costs of action or lack of action;*
- *review of the measures in the light of scientific developments."*

7.3.4 This would suggest that a degree of pragmatism should be used because absolute scientific certainty is rare. That is the nature of scientific endeavour. It often takes decades for scientific doubt to be satisfied. (Climate change is such an example).

7.4 The assessment should be appropriate to the risk

7.4.1 The European Commission guidance also suggests that the assessment should be proportional to the risk.

7.4.2 This means that the assessment must provide sufficient detail to enable a robust conclusion to be drawn regarding the air quality impacts. The level of detail required will depend on the specific circumstances of the project.

7.5 The assessment should be undertaken with an ecologist

7.5.1 The assessment of the impact of air pollution on designated wildlife sites is best undertaken in collaboration with a suitably qualified and experienced ecologist. An air quality specialist should not be making judgements on whether there is a likely significant effect or an adverse effect on the integrity of a site.

7.6 Always consult with the regulator

7.6.1 It is important that the assessment team consults with the regulator to agree the scope of the assessment. This includes agreeing the other projects that need to be considered in the in-combination impact assessment. The regulator is the decision maker and failure to consult can result in an assessment being rejected at a late stage in the process. This consultation should be undertaken early in the project to avoid unnecessary work.

7.7 Future Clarity

7.7.1 As alluded to above, there has been much litigation on the interpretation of the Habitats Directive and this is likely to continue. Whether or not the UK will voluntarily be bound by its rulings is a matter of conjecture at the current time. There are also increasing legal challenges on environmental decisions in the UK courts and therefore it is likely that litigation on this issue will continue and further clarity will be produced on the principles of assessment.

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[70] The Precautionary Principle eur-lex.europa.eu/legal-content/EN/TXT/?uri=LEGISSUM%3Al32042.

Appendix A: Glossary & Terminology

Term	Abbreviation	Description
Acid deposition		Atmospheric input to ecosystems of pollutants which may acidify soils and freshwaters.
Air Pollution Information System	APIS	An information system that provides a comprehensive source of information on air pollution and the effects on habitats and species (online at www.apis.ac.uk).
Air Quality Assessment	AQA	The process of assessing the impact of a project or plan on air quality.
Air Quality Modelling and Risk Assessment Team	AQMRAT	A Natural Resources Wales team that specialises in air quality related issues and assessments.
Air Quality Modelling and Assessment Unit	AQMAU	An Environment Agency team that specialises in air quality related issues and assessments.
Air Quality Standards Regulations 2010		UK regulations that transposed Directive 2008/50/EC into UK legislation. It contains mandatory limit values, targets and information levels for ambient air quality for the protection of human health and vegetation.
Air Quality Strategy	AQS	The 2007 AQS for England, Scotland, Wales and Northern Ireland provides details of national air quality objectives for air pollutants.
Air Quality Technical Advisory Group	AQTAG	AQTAG was established in 2000 by the Environment Agency's Habitats Directive Project to provide technical guidance on the assessment of air emissions from IPC/IPPC processes. Membership has since expanded to include all UK regulators and conservation agencies.
Ammonia	NH_3	A gas which may cause acidification of soils and physically damage vegetation.
Annual Average Daily Traffic	AADT	The number of vehicles using a road in a 24-hour period averaged over a year.
Ancient Woodland		Typically, a woodland that has existed continuously since 1600 or before (this can include areas where trees have been cut down and or replanted).
Annual Mean		The average of concentrations measured for one year (usually a calendar year).
Appropriate Assessment	AA	An assessment required by the Habitats Directive and Habitats Regulations, where a project (or plan) would be likely to have a significant effect on a European site, either alone or in-combination with other plans or projects. Undertaken by the competent authority (i.e. the decision maker).
Area of Outstanding Natural Beauty	AONB	A landscape designation protected under the Countryside and Rights of Way Act, 2000.
Area of Special Scientific Interest	ASSI	A Northern Ireland designation.
Avoidance		Prevention of adverse impacts occurring through, for example, decisions about project location or design.
Background		When used in the context of concentration or deposition rate this refers to the average over a 1km by 1km or 5km by 5km grid provided by Defra or CEH e.g. the LAQM background maps.
Baseline		The conditions that exist in the absence of the proposed project either at the time an assessment or survey is undertaken or in the future when the project would be constructed, operated or decommissioned.
Chartered Institute of Ecology and Environmental Management	CIEEM	Professional body governing ecology/ecologists.

Term	Abbreviation	Description
Centre for Ecology and Hydrology	CEH	Natural Environment Research Council research organisation focusing on land and freshwater ecosystems and their interaction with the atmosphere.
Compensation		Measures taken to make up for the loss of, or permanent damage to, ecological features despite mitigation Under the Habitats Directive and Habitats Regulations. Any replacement area should be similar in terms of biological features and ecological functions that have been lost or damaged, or with appropriate management, have the ability to reproduce the ecological functions and conditions of those biological features.
Conservation objective		The objective for the conservation of biodiversity (e.g. specific objective for a designated site or broad objectives of policy).
Conservation status		The state of a species or habitat including, for example, extent, abundance, distribution and their trends.
Critical level		The concentration of an air pollutant above which adverse effects on ecosystems may occur based to present knowledge.
Critical load		Deposition flux of an air pollutant below which significant harmful effects on sensitive ecosystems do not occur, according to present knowledge. Usually measured in units of kilograms per hectare per year (kg/ha/yr).
Cumulative effect		Changes caused by a proposed project in conjunction with other projects and plans.
Department for Environment, Food and Rural Affairs	Defra	The government department responsible for environmental protection, food production and standards, agriculture, fisheries and rural communities.
Deposition		The main pathway for removing pollutants from the atmosphere, by settling on the earth's surface.
Deposition flux		Deposition velocity x concentration.
Designated Site		Land designated for its wildlife interest. These include the following designations (note different names may be given to locally designated sites): • Ramsar site • Special Area of Conservation (SAC) • Special Protection Area (SPA) • Site of Special Scientific Interest (SSSI) • Areas of Special Scientific Interest (ASSI) • Local Wildlife Site (LWS) • Local Nature Conservation Site (LNCS) • Site of Importance for Nature Conservation (SINC) • Area in the Ancient Woodland Inventory (AWI) • National Nature Reserves (NNR) • Local Nature Reserves (LNR) • Ancient Woodland (AW).
Ecological feature		Habitat, species or ecosystem.
Ecosystem		An entire functional ecological system i.e. the plant and animal species that make up the constituent habitat (or habitats) plus the air, water, soil etc. that they require to persist and thrive.

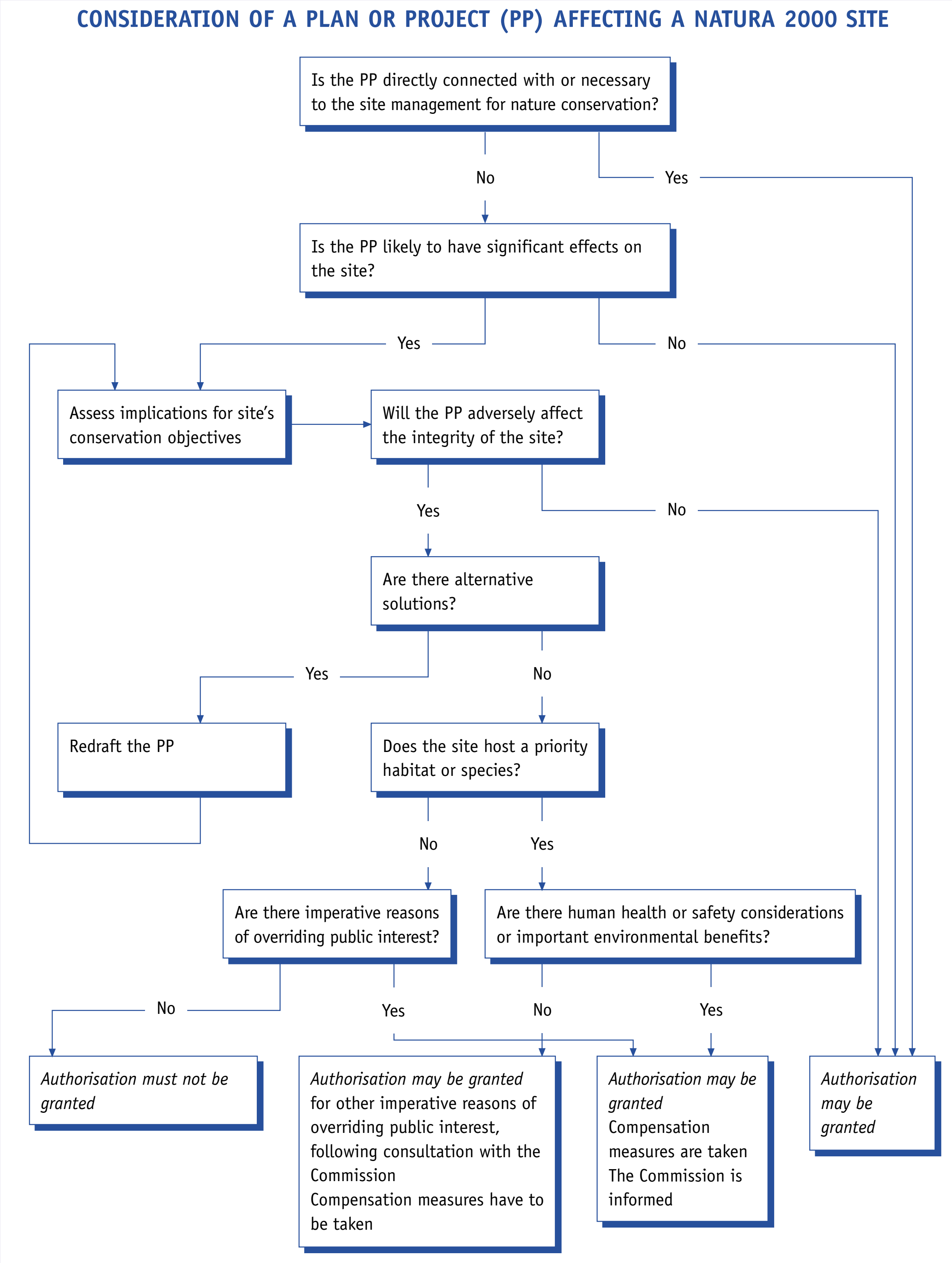
Term	Abbreviation	Description
Effect		The changes that occur to a habitat as a result of changes in concentrations or deposition of air pollution. Also, see 'Impact'.
Emission		The release of a substance into the air. May be discharged from a stack, vent, vehicle exhaust or from diffuse sources.
Emission Limit Value		The legal limit on the emission of a pollutant.
Enhancement		Improved management of ecological features or provision of new ecological features, resulting in a net benefit to biodiversity, which is unrelated to a negative impact or is 'over and above' that required to mitigate/compensate for an impact.
Environment Agency	EA	The Environment Agency is responsible for permitting certain industrial process in England.
Environmental Impact Assessment	EIA	The process of assessing the likely significant environmental effects of a proposed project as part of gaining consent carried out under the EIA Directive and Regulations.
Ecological Impact Assessment	EcIA	A process of identifying, quantifying and evaluating potential effects of development-related or other proposed actions on habitats, species and ecosystems.
Environmental Permit	EP	A permit required by industrial operators in accordance with the Environmental Permitting Regulations.
Environmental Permitting Regulations	EPR	The various sets of national regulations that regulate pollution through a permitting system.
Environmental Protection UK	EPUK	UK environmental Non-Governmental Organisation (NGO) working to improve the quality of the local Environment.
Environmental Statement	ES	The document which reports the process, findings and recommendations of an EIA.
Environmentally Sensitive Area	ESA	A designation for agricultural areas needing special protection by virtue of their landscape, wildlife or historical value.
European sites		A network of European designated sites including Special Protection Areas (designated under Directive 2009/147/EC) and Special Areas of Conservation as listed in Annex I and II of the EU Directive 92/43/EEC ("Habitats Directive"). Also referred to as Natura 2000 sites.
European Union	EU	A political and economic union of 28-member states that are located primarily in Europe.
Eutrophication		The process by which an ecosystem is subject to excessive growth of a few species of competitive plants and/or microorganisms as a result of excessive nutrient supply, thus forcing out less competitive plants and (in aquatic ecosystems) resulting in oxygen depletion and a reduction in animal life.
H1		The screening tool in the Environment Agency's former Horizonal guidance H1. This has been replaced by the Risk Assessment Tool in 'Air emissions risk assessment for your environmental permit'.
Habitat		An assemblage of physical and biological elements which form a recognisable unit. For example, heathland is a different habitat from chalk grassland or wet woodland, most obviously due to differences in specific plant and animal composition and physical structure.

Term	Abbreviation	Description
Habitat Regulations Assessment	HRA	An assessment of a plan or project potentially affecting European (Nature 2000) sites in the UK, required under the Habitats Directive (European Directive 92/43/EEC) and Regulations (Conservation of Habitats and Species Regulations, 2010, as amended).
Impact		The change in concentrations or deposition of an air pollutant. This may or may not rise to an effect on an ecological feature.
Isolated project		A project which, due to its geographical location, is not likely to give rise to in-combination effects on a designated site. This is determined using professional judgement and needs to take account of where there may be an overlap of the air quality impacts of projects and/or plans.
Institute of Air Quality Management	IAQM	The professional body representing air quality specialists.
Joint Nature Conservation Committee	JNCC	The public body that advises the UK government and devolved administrations on UK-wide and international nature conservation.
Kilogram per hectare per year	kg/ha/yr	Unit of measurement used to describe the rate of deposition.
Kilogram equivalent per hectare per year	keq/ha/yr	Unit of measurement used to describe the rate of acid deposition, in terms of hydrogen ion (H^+) equivalent.
Leaching		Leaching is the process whereby nutrients from agricultural fertilisers are washed out of the soil through the percolation of rainfall.
Local Nature Reserve	LNR	Statutory designation for places with wildlife or geological features that are of special interest locally.
Local wildlife sites		'Non-statutory' sites of nature conservation value that have been identified 'locally' (i.e. excluding SSSIs, SPAs, SACs, and Ramsar sites). LNRs are included as they are a designation made by the Local Authority rather than statutory country conservation agencies. These are often called Local Wildlife Sites, Local Nature Conservation Sites, Sites of Importance for Nature Conservation or other, similar names.
Microgram per cubic metre	$\mu g/m^3$	Unit of measurement of the concentration of an air pollutant. Often used for ambient concentrations.
Milligram per cubic metre	mg/m^3	Unit of measurement of the concentration of an air pollutant. Often used to describe emissions and their limit values for industrial processes.
Mitigation		Measures taken to avoid, reduce, or otherwise address the negative effects of air quality impacts. See also compensation (which is separate from mitigation).
Multi-Agency Geographic Information for the Countryside	MAGIC	A web-based mapping browser showing various geographical designations including designated nature conservation site boundaries.
National Nature Reserve	NNR	Statutory designations, supporting wildlife or geological features that are significant at a national level.
Natural Resources Wales	NRW	Welsh Government Sponsored Body, created in 2013, which took over the work of Countryside Council for Wales, Environment Agency Wales and Forestry Commission Wales.
Nitrate Vulnerable Zone	NVZ	A designated area where land drains into and contributes to nitrate found in nitrate-polluted waters.
Nitrogen	N	Nitrogen (N_2) is a relatively inert gas, but certain molecules containing nitrogen are more reactive with other chemicals.

Term	Abbreviation	Description
Nitric oxide	NO	Produced during combustion processes.
Nitrogen dioxide	NO_2	Produced during combustion and formed by the oxidation of NO in the atmosphere.
Oxides of nitrogen	NO_x	A term describing a mixture composed of nitrogen oxides (NO and (NO_2).
Pathway		The route by which a pollutant moves from a source to a receptor.
Predicted Environmental Concentration	PEC	The term used in AQAs of industrial processes to describe the concentration or deposition (i.e. process contribution (PC) plus baseline).
Process Contribution	PC	The term used in AQAs of industrial processes s to describe the incremental impact of the proposed development on the concentration or deposition flux).
Project (also known as plan or permission)		The term used for proposals to which this guidance might be applied (e.g. development proposal, road scheme, industrial facility or other land use change).
Ramsar		A wetland site designated of international importance under the international Convention on Wetlands, known as the Ramsar Convention. These sites are considered in the same way as European (Natura 2000) Sites as a matter of government policy.
Receptor		An identified location where an effect may occur.
Restoration		The re-establishment of a damaged or degraded system or habitat to a close approximation of its pre-degraded condition.
Scoping		A process early on in AQA, EIA or EcIA, to determine the matters to be addressed and ensure effective input to the assessment.
Screening		This term can be used either to determine whether or not an EIA or HRA is necessary or in the context of air quality assessment, to “screen out” emissions that are inconsequential using numerical criteria.
Scottish Environment Protection Agency	SEPA	Responsible for permitting certain industrial process in Scotland.
Significant effect		An effect that either supports or undermines biodiversity conservation objectives for ‘important ecological features.
Site of Special Scientific Interest	SSSI	A geological or biological conservation designation denoting a nationally protected area in the UK.
Scottish Natural Heritage	SNH	Funded by the Scottish Government with the purpose to promote, care for, and improve natural heritage.
Special Area of Conservation	SAC	Area of protected habitats and species as defined in the European Union’s Habitat Directive (92/43/EEC).
Special Protection Area	SPA	A designated area for birds under the European Union Directive on the Conservation of Wild Birds (2009/147/EC).
Statutory Nature Conservation Organisation	SNCO	E.g. Natural England, Natural Resources Wales, Scottish Natural Heritage.
Sulphur dioxide	SO_2	Combustion product formed from sulphur contained in fuels.
United Nations Economic Commission for Europe	UNECE	Regional commission of the United Nations helping countries to convene and cooperate on standards and conventions in support of the Sustainable Development Goals.

Term	Abbreviation	Description
Windfall development		Development that does not have a specific allocation in a local plan, often because local authorities do not allocate development sites below a certain size.
World Health Organization	WHO	Directs and coordinates international health within the United Nations' organisation.

Appendix B: Flowchart reproduced from Annex III of European Commission's guidance on managing Natura 2000 sites



Appendix C: Typical relationship between Ecological Impact Assessment (EcIA) and Air Quality Assessment (AQA)

EcIA Stage[71]	Interactions with this guidance
Scoping: Determining the matters to be addressed in the EcIA. Scoping is an ongoing process – the scope of the EcIA may be modified following further ecological survey/research and during impact assessment.	The initial stages of the air quality assessment process will determine whether air quality impacts on designated sites require consideration. See Chapters 3 and 4.
Establishing the baseline: Collecting information and describing the ecological conditions in the absence of the proposed project, to inform the assessment of impacts.	The ecologist and air quality specialist will often jointly identify designated sites that are relevant to the plan or project and describe these. See Chapters 3 and 4. It should be noted that ecologists are not instructed on all projects that require an AQA, for example, many applications for environmental permits.
Important ecological features: Identifying important ecological features that may be affected, with reference to a geographical context in which they are considered important.	
Impact assessment: An assessment of whether important ecological features will be subject to impacts and characterisation of these impacts and their effects. Assessment of residual ecological impacts of the project remaining after mitigation and the significance of their effects, including cumulative effects.	The assessment of air quality impacts on designated sites is an iterative process, with consideration of potential impacts starting early in the process set out in this guidance. Initial consideration of impacts starts during the air quality scoping (see Chapter 4) with the assessment becoming more detailed in the latter stages (see Chapter 4).
Avoidance, mitigation, compensation and enhancement: Incorporating measures to avoid, reduce and compensate ecological impacts, and the provision of ecological enhancements. Monitoring impacts of the development and evaluation of the success of proposed mitigation, compensation and enhancement measures.	An IAQM Position Statement[72] sets out the basic hierarchical principles for identifying mitigation measures.
Consequences for decision making: Consideration of the legal and policy framework throughout the EcIA process and assessment of how the proposed development has responded to this.	This will be assessed iteratively throughout the process of assessing air quality impacts on designated sites.

[71] Modified from page iv of: CIEEM (2016) Guidelines for Ecological Impact Assessment in the UK and Ireland: Terrestrial, Freshwater and Coastal, 2nd edition. Chartered Institute of Ecology and Environmental Management, Winchester.
[72] iaqm.co.uk/text/position_statements/mitigation_of_development.pdf.

Appendix D: Air pollutants and deposition processes

D.1 Introduction

D.1.1 Maintaining good air quality is important for the protection of ecosystems. Air pollution and its deposition onto vegetation, soil and water can damage vegetation directly or indirectly through the addition of nutrients or changes in acidity levels within a habitat. These can cause a shift in the competitive balance between species, changes in plant species composition or subtle changes in vegetation structure, which can affect the use of a habitat by an animal species.

D.2 Pollutant emission and deposition processes

D.2.1 The main air pollutants affecting vegetation and ecosystems are nitrogen oxides (NO_x), sulphur dioxide (SO_2) and ammonia (NH_3). Ozone (O_3) is also important but this pollutant is not addressed by this guide as it is a regional pollutant not assessed at scheme or project level.

D.2.2 These have both direct effects e.g. through exposure to the gas itself; and indirect effects, e.g. through deposition of the gas to soil and freshwater (dry deposition) or with precipitation (wet deposition).

D.2.3 Figure D1[73] illustrates in simple form the sources, pathways and receptors processes.

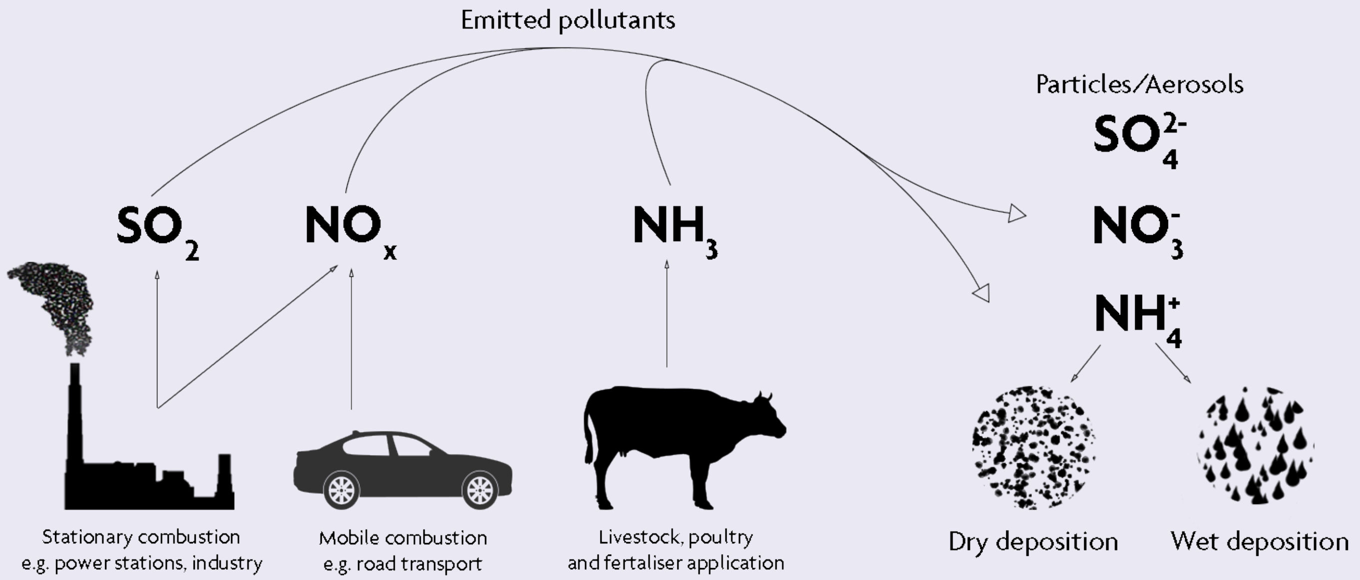
D.3 Critical levels and loads

D.3.1 The concepts of critical levels and critical loads were introduced by the United Nations Economic Commission for Europe (UNECE) Convention on Long-range Transboundary Air Pollution (CLRTAP).

D.3.2 **Critical levels** are defined by the UNECE[74] as: *"concentrations of pollutants in the atmosphere above which direct adverse effects on receptors, such as human beings, plants, ecosystems or materials, may occur according to present knowledge"*. In terms of ecosystem effects, they relate to effects on plant physiology, growth and vitality, and are expressed as atmospheric concentrations over a particular averaging time (hours to years). They are thus important as an indicator of direct adverse effects on ecological receptors and are thus useful tools for ecological assessment.

D.3.3 The critical levels for NO_x and SO_2 are set in the European Union (EU) Ambient Air Quality Directive[75] and transposed into

Figure D1. Schematic of the sources of air pollution



law by the Air Quality Standards Regulations 2010, as amended, and similar Regulations in the devolved administrations. The Directive defines a critical level as *"A level fixed on the basis of scientific knowledge, above which direct adverse effects may occur on some receptors, such as trees, other plants or natural ecosystems but not on humans"*. Under the Directive, assessment of compliance with the critical levels is strictly only required at locations more than 20 km from towns with more than 250,000 inhabitants or more than 5 km from other built-up areas, industrial installations or motorways[76]. In practice, however, assessment against critical levels for vegetation is frequently undertaken to inform planning and permitting processes across the country, regardless of this definition.

D.3.4 The Air Quality Strategy for England, Wales, Scotland and Northern Ireland[77] has adopted these critical levels, as national objectives for the maximum ambient air concentrations of NO_x and SO_2 (and ozone[78]) to be attained, for the aim of protection against the direct effects of air pollution.

D.3.5 The main critical levels used in air quality assessments of designated sites are set out in Table 2.1.

D.3.6 **Critical loads** relate to the potential effects of pollutant deposition [over periods of decades] and are defined by UNECE as *"a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge"*.

D.3.7 There are critical loads for nitrogen deposition (leading to eutrophication) and acid deposition (leading to acidification). Critical loads for nitrogen deposition are given as a range and quoted in units of kilograms of nitrogen per hectare per year (kg N/ha/year). A critical load for acidification is described in units of kilograms of H^+ ion equivalents per hectare per year (keq/ha/year). Most assessments consider nitrogen and sulphur deposition, but for some industrial processes, including energy from waste, other chemical species need to be considered, such as hydrogen fluoride (HF).

D.3.8 Critical loads are habitat dependent, further detail and supporting information is provided by the online resource, the Air Pollution Information Service (APIS)[79].

D.4 Oxides of nitrogen (NO_x)

D.4.1 Oxides of nitrogen (NO_x; also referred to as nitrogen oxides), are produced mainly as a result of combustion processes[80]. Almost half of the NO_x emissions in the UK are from road vehicles, mostly diesel engines; approximately one quarter is from power generation and the remainder from other industrial and domestic combustion processes. Emissions of NO_x are also produced naturally by lightning, forest fires and, to a small extent, microbial processes in soils. NO_x is conventionally considered to be a mixture of nitrogen dioxide (NO_2)[81] and nitric oxide (NO)[82]. The general long term UK trend in NO_x has been one of improvement, particularly since 1990, despite an increase in vehicles on the roads[83].

D.4.2 NO_x can affect plants directly or indirectly. It may directly enter a plant via the stomata (as NO or NO_2), where it has phytotoxic effects. Lower plants such as lichens and bryophytes (including mosses, landworts and hornwarts) are particularly vulnerable to direct exposure to the gases in this way[84]. NO_x can also deposit onto soil and, following transformation to nitrate, enrich the soil, leading to eutrophication, as discussed later.

D.4.3 There is no published evidence for any direct toxic effect of NO_x on animals and therefore effects on animals are not directly included in ecological impact assessments, which focus on the effects on vegetation. The effects on animals are sometimes indirectly included in an assessment where species are dependent on particular habitats for their survival and an assessment will focus on this supporting habitat.

D.4.4 The effects of elevated NO_x concentrations on vegetation can be broadly categorised as[85]:

- growth effects: particularly increased biomass, changes in root to shoot ratio and growth of more competitive species, but also including growth suppression of some species;
- physiological effects: e.g. CO_2 assimilation and stomatal conductivity; and
- (bio)chemical effects: e.g. changes in enzyme activity and chlorophyll content (probably through the effects of increased nitrogen, as demonstrated in lichens[86], but also documented in higher plants).

D.4.5 The long term (annual average) critical level for NO_x is 30 µg/m^3. At concentrations above this critical level, both beneficial and adverse responses have been recorded, and there is evidence suggesting an adverse synergistic effect when plants are exposed to both NO_x and SO_2[87].

D.4.6 The long term critical level for NO_x was set on the basis that growth effects are likely to affect vegetation diversity and survival and occur at lower annual average concentrations than other effects.

D.4.7 Data presented by the World Health Organization (WHO) 2000[85] indicates that, other than growth effects, biochemical or physiological effects have been demonstrated in vascular plants from exposure to annual average concentrations of more than 100 µg/m^3. With regard to lower plants, Das *et al* (2011)[88] recorded evidence of chlorophyll changes in lichens, also correlated with NO_x at higher concentrations (over 260 µg/m^3). These studies have also attributed the effects to the increase in available nitrogen, but at such high concentrations NO and NO_2 can also increase cellular acidity and inhibit lipid biosynthesis (Wellburn, 1990)[89].

D.4.8 The critical level does not differentiate between the role of nitrogen deposition and NO_x in the air. It is a precautionary general threshold, not specific to a particular habitat, plant species or impact pathway, below which there is currently a high degree of confidence that no adverse effects on vegetation will arise. Long term NO_x concentrations below the critical level are therefore desirable. Some species or habitats may not show adverse effects until higher concentrations are present.

D.4.9 The long term (annual mean) concentration of NO_x is most relevant for its impacts on vegetation, as the effects, particularly through the nitrogen deposition pathway, are additive over months and years. This is reflected in the adoption of the long term guideline in the EU Air Quality Directive as a limit value for vegetation. However, atmospheric exposure to very high concentrations of NO_x for short periods (hours/days) may also have an adverse effect under certain conditions even if the long term concentrations are below the limit value. The WHO guidelines[90] include a short term (24-hour average) NO_x critical level of 75 µg/m^3. Originally set at 200 µg/m^3 as a four-hour mean, the more detailed CD-ROM version of the 2000 WHO guidelines[91] comments: "*Experimental evidence exists that the CLE decreases from around 200 µg/m^3 to 75 µg/m^3 when in-combination with O_3 or SO_2 at or above their critical levels. In the knowledge that short-term episodes of elevated NO_x concentrations are generally combined with elevated concentrations of O_3 or SO_2, 75 µg/m^3 is proposed for the 24 h mean.*" Ozone and SO_2 concentrations are typically low in the UK compared to many other countries. If a regulator does require the use of the short term NO_x critical level, given the low UK SO_2 concentrations IAQM consider it is most appropriate to use 200 µg/m^3 as the short term critical load.

D.4.10 The relative importance of the long term mean compared to the short term mean is reflected in several studies which state that the '*UNECE Working Group on Effects strongly recommended the use of the annual mean value, as the long term effects of NO_x are thought to be more significant than the short term effects*'[92, 93]. This IAQM guidance, therefore, recommends that only the annual mean NO_x concentration is used in assessments unless specifically required by a regulator; for instance, as part of an industrial permit application where high, short term peaks in emissions, and consequent ambient concentrations, may occur.

D.5 Sulphur dioxide (SO_2)

D.5.1 The main anthropogenic source of sulphur dioxide (SO_2) is the combustion of sulphur containing fuel in electricity generation, other industry and domestic heating. Since the 1970s, UK emissions have fallen by 95% with the largest reductions occurring between 1990 and 2000, when emissions reduced by 70%[94].

D.5.2 SO_2 is directly toxic to both higher and lower plants. Lower, non-vascular, plants such as lichens and bryophytes are particularly vulnerable. In the UK, however, many lichen species have increased in abundance after the return to low ambient concentrations (<10 µg/m^3).

D.5.3 The critical level for protection of all vegetation types from the effects of SO_2 is 20 µg/m^3, as an annual mean, except for lichens and bryophytes (including mosses, landworts and hornwarts) for which the criterion is 10 µg/m^3, reflecting their greater sensitivity.

D.5.4 Another key effect of SO_2 is through the indirect effects arising from the acidification of soils. This is discussed in more detail below.

D.6 Ammonia (NH_3)

D.6.1 Agriculture is the main source of anthropogenic ammonia (NH_3) in the UK (82% in 2016[95]). A small amount of ammonia is emitted from petrol vehicles with early three way catalysts, although this source is declining as these older vehicles are retired from the fleet. Vehicles that use Adblue to control NO_x emissions from diesel engines potentially emit ammonia, but vehicles using this technology may have an effective system to remove ammonia from the exhaust gases. Anaerobic digesters used in the waste industry are also an important source of ammonia.

D.6.2 The direct uptake of NH_3 through the stomata increases the amount of nitrogen within the plant. In addition, its alkalinity adversely affects plant biochemistry; lichens and bryophytes are particularly sensitive to this effect[96]. Ammonia also reacts in the atmosphere to produce ammonium ions (NH_4^+) which contribute to nutrient nitrogen and acid deposition. Higher plants are considered to be less sensitive and, for this reason, the annual critical level for higher plants is 3 µg/m^3 but is reduced to 1 µg/m^3 where lower plants (lichens and bryophytes, including mosses, landworts and hornwarts) are a particular interest feature of a habitat. It is the ecologist's role identify the presence of these lower plants.

D.7 Hydrogen fluoride (HF)

D.7.1 Hydrogen fluoride (HF) is an acidic gas released from industrial processes (such as coal fired power stations, waste incinerators and aluminium production). In elevated concentrations, HF can have an adverse impact on the chlorophyll content of plants. The WHO recognises that HF concentrations in ambient air should be less than 1 µg/m^3, to prevent effects on livestock and plants; this guideline applies to long term (annual) exposure. The Environment Agency online risk assessment guidance[97] for permitting contains weekly and daily average standards for HF for the protection of vegetation.

D.8 Other pollutants

D.8.1 Other pollutants, for example, heavy metals and hydrogen chloride, are emitted by industrial processes and these pollutants may need to be assessed. It is recommended that prior to the assessment of industrial emissions that would be regulated by the Environment Agency or equivalent country regulator, the scope of any assessment is discussed with the regulator.

D.9 Pollutant deposition

D.9.1 There are two processes for atmospheric deposition of pollutants:

- Dry deposition is the deposition of gases and aerosols directly to the Earth's surface.
- Wet deposition is the process whereby pollutants are removed from the atmosphere by precipitation (e.g. rain, snow, fog) and then deposited to ground or vegetation.

D.9.2 Wet deposition is the dominant component of the background deposition rate[98] and often determines whether the critical load is exceeded. Wet deposition primarily depends on the rate of precipitation, and therefore generally follows rainfall patterns.

D.9.3 Wet deposition is not normally assessed by air quality practitioners because the impacts of a project or local development plan typically occur over short distances and over timescales that are too short for wet deposition to be significant. One exception to this is hydrogen chloride (HCl), which is readily 'washed out' of plumes at short range and can, therefore, be required for some industrial permit applications[99].

D.9.4 Deposition rates are dependent on the habitat as well as atmospheric pollutant concentrations. For example, the typical leaf area and height of a species will affect the available surface area for deposition. The deposition velocities used in assessments should reflect the type of vegetation cover.

D.10 Nitrogen deposition

D.10.1 Dry deposition of nitrogen is high within large conurbations and close to major roads, due to the higher NO_x concentrations in the atmosphere. High rates are also found close to agricultural activities such as intensive livestock farming, due to 'reduced nitrogen', which is derived from emissions of ammonia.

D.10.2 Although nitrogen is an essential growth nutrient, not all plants require the same relative quantities. Plants which are of higher conservation value tend to be those which have lower nitrogen requirements and are associated with lower nutrient status habitats.

D.10.3 The growth stimulation effects of nitrogen deposition are generally subtler than the effects of the application of agricultural fertiliser since the quantities of nitrogen deposited over a given period of time are much smaller. Negative effects have been demonstrated in epiphytic lichens. This is caused by a combination of growth inhibition[100] of more sensitive species and growth stimulation of nutrient tolerant species. This has been demonstrated in several studies in London[101].

D.10.4 The role of nitrogen deposition in growth stimulation depends on the availability of the three macronutrients (nitrogen, phosphorus and potassium). In most terrestrial systems nitrogen and phosphorus are naturally relatively scarce and this ordinarily restricts growth and keeps more competitive species in check. If nitrogen is available in sufficient quantities it ceases to be limiting.

D.10.5 Freshwater habitats are typically phosphorus limited[102]. Therefore, nitrogen deposition is usually less important than in terrestrial systems and control of eutrophication in freshwater environments is often directed towards controlling phosphorus inputs. Be aware some rivers and lakes may have nitrogen limits where nutrient nitrogen deposition is more ecologically important to assess.

D.10.6 Coastal systems are generally nitrogen limited. Therefore, nitrogen inputs are typically more important in coastal environments than in freshwater environments. The situation is more complex for terrestrial habitats because those that have a strong freshwater influence may be phosphate limited, others may be nitrogen limited.

D.10.7 Understanding how nutrients affect particular habitats is essential to understanding the role of nitrogen deposition and avoiding unnecessary time and effort being expended controlling non limiting nutrients.

D.10.8 'Site relevant critical loads' for internationally and nationally important wildlife sites and habitat specific critical loads are available through APIS[103]. The APIS website provides advice on the selection of an appropriate value within the critical load range that should be applied in assessments, based on the interest features of the site. It is at this point that the advice of an ecologist can be particularly important. This is discussed in **Chapter 3**.

D.11 Acid deposition

D.11.1 A range of air pollutants can cause the acidification of soil and freshwater. Salt water systems naturally buffer (acid neutralizing) any acid deposition in almost all cases. The key pollutants are sulphur, in the form of sulphate ions (SO_4^{2-}), and nitrogen, as nitrate (NO_3^-), nitric acid (HNO_3) and ammonium (NH_4^+) (from ammonia). As these pollutants are removed from the atmosphere the H^+ ion concentrations in the precipitation increases, making it more acidic.

D.11.2 Acid deposition is most likely to affect vegetation indirectly through changes to soil properties. Evidence from national monitoring programmes[104] shows that this occurs through increasing the soil acidity, which tends to increase the mobility of certain toxic metals (e.g. aluminium and manganese) and reduce the buffering capacity of the soil. Acid deposition can also cause nutrient deficiencies, by reducing base cation availability (e.g. phosphorus). In forests, leaching of base cations from the soil has been linked to leaf chlorosis (yellowing) (Huettl *et al*, 1990)[105]. Acid deposition can also lead to leaching of calcium from conifer needles, which subsequently may be less able to withstand winter freezing/desiccation (the removal of moisture) damage (Borer *et al* 2005)[106]. There may also be changes in microbial transformations. Root damage may result, especially from aluminium toxicity. Nutrient imbalance can lead to stunted growth. These effects can lead to changes in species composition.

D.11.3 Some ecological sites are more at risk from acid deposition than others, depending on the soil type, bedrock geology, weathering rate and buffering capacity. In general, habitats dependent on slightly acidic substrate (i.e. heathland or acid grassland) and bog habitats are at greater risk of being adversely affected by increased rates of acid deposition than those associated with more calcareous habitats (e.g. chalk grassland). However, it should be noted that calcareous substrates are not immune to acidification as the buffering ability of the soil can become exhausted over time.

D.11.4 Emissions of all acidifying pollutants, typically nitrogen and sulphur but also any other relevant compounds e.g. HCl, should be taken into account when assessing potential acidification of soils and impacts on vegetation. Nitrogen and sulphur containing compounds can be assessed using the APIS Critical Load Function tool[107].

73 Figure adapted from www.apis.ac.uk/starters guide air pollution and pollution sources.
74 www.unece.org/env/lrtap/WorkingGroups/wge/definitions.htm.
75 The EU Ambient Air Quality Directive refers to the Critical Levels as limit values for the protection of vegetation.
76 The Directive notes that the risk posed by air pollution to vegetation and natural ecosystems is most important in places away from urban areas and that compliance with critical levels for the protection of vegetation should focus on places away from built up areas.
77 Defra and the devolved adminstrations, 2007, The Air Quality Strategy for England, Scotland, Wales and Northern Ireland, Volume 1.
https://www.gov.uk/government/publications/the-air-quality-strategy-for-england-scotland-wales-and-northern-ireland-volume-1.
78 There are limit values and target values for ozone set out in the EU Ambient Air Quality Directive.
79 Available at www.apis.ac.uk.
80 National Atmospheric Emissions Inventory naei.defra.gov.uk.
81 NO_2 is the component of NO_x that cause human health effects.
82 Another oxide of nitrogen, N_2O (nitrous oxide) is not generally considered part of NO_x in terms of ambient air quality but it is an important greenhouse gas.
83 Emissions of nitrogen oxides fell by 72% between 1970 and 2017. Source: www.gov.uk/government/uploads/system/uploads/attachment_data/file/579200/Emissions_airpollutants_statisticalrelease_2016_final.pdf [accessed 14/06/2019].
84 www.apis.ac.uk/node/1071.
85 WHO Regional Office for Europe, Copenhagen, Denmark, 2000. Air Quality Guidelines – Second Edition. Chapter 11 Effects of nitrogen containing air pollutants: critical levels. http://www.euro.who.int/__data/assets/pdf_file/0005/74732/E71922.pdf#page=244.
86 Tiwari, 2008. Lichens as an Indicator for Air Pollution: A Review. Indian Journal of Pollution Control, 1, 8 17.
87 This rarely happens in the UK in practice as sulphur dioxide levels in the UK are now generally very low.
88 Das K, Dey U, Bhaumik R, Datta JK and Mondal NK. 2011. A comparative study of lichen biochemistry and air pollution status of urban, semi urban and industrial area of Hooghly and Burdwan district, West Bengal. Journal of Stress Physiology & Biochemistry Vol 7, No. 4 pp311 323.
89 Wellburn AR. 1990. Why are atmospheric oxides of nitrogen usually phytotoxic and not alternative fertilisers? New Phytologist 115 pp 395 429.
90 WHO, 2000, Air Quality Guidelines for Europe, Second Edition http://www.euro.who.int/__data/assets/pdf_file/0005/74732/E71922.pdf.

[91] WHO, 2000, Air quality Guidelines for europe, Second edition (CD ROM version) http://www.euro.who.int/en/health-topics/environment-and-health/air-quality/publications/pre2009/who-air-quality-guidelines-for-europe,-2nd-edition,-2000-cd-rom-version.
[92] Sutton MA, Howard CM, Erisman JW, Billen G, Bleeker A, Grennfelt P, van Grinsven H, Grizzetti B. 2013. The European Nitrogen Assessment: Sources, Effects and Policy Perspectives. Page 414. Cambridge University Press. 664pp. ISBN 10: 1107006120.
[93] June 2011. Manual on Methodologies and Criteria for Modelling and Mapping Critical Loads & Levels and Air Pollution Effects, Risks and Trends. Chapter 3: Mapping Critical Levels for Vegetation.
[94] naei.defra.gov.uk/overview/pollutants?pollutant_id=8.
[95] naei.defra.gov.uk/overview/pollutants?pollutant_id=21 [accessed 06/03/2019].
[96] www.apis.ac.uk.
[97] www.gov.uk/guidance/air-emissions-risk-assessment-for-your-environmental-permit.
[98] In this guidance, the term deposition rate is used to describe the amount of pollutant deposited over an area over a period of time, i.e. kg/ha/yr. This is also called the deposition flux.
[99] Where hydrogen chloride (HCl) or nitrate (HNO3) are emitted from industrial activities, advice should be sought from the regulator.
[100] Munzi S, Pisani T, Loppi S. 2009. The integrity of lichen cell membrane as a suitable parameter for monitoring biological effects of acute nitrogen pollution. Ecotoxicology and Environmental Safety 72 2009 2012.
[101] Imperial College & The Natural History Museum. March 2002. Effects of NO_x and NH_3 on lichen communities and urban ecosystems: A pilot study.
[102] However, some riverine designated sites, e.g. the River Clun SAC in Shropshire/Herefordshire, do have targets for nitrogen levels set in their conservation objectives.
[103] APIS recommends values within nutrient nitrogen critical load ranges for use in air pollution impact assessments. www.apis.ac.uk/sites/default/files/downloads/APIS%20critical_load_range_document_0.pdf.
[104] National Soil inventory (NSI), Countryside Survey, Environmental Change Network of sites (ECN), Inter National Co operative Programme (ICP)) and long term experiments.
[105] Huettl, R.F., Fink, S., Lutz, H.J., Poth, M., Wisniewski, J., 1990, Forest decline, nutrient supply and diagnostic fertilisation in southwestern Germany and in southern California. Forest Ecology and Management 30, 341 350.
[106] Borer, C.H., Schaberg, P.G., DeHayes, D.H., 2005, Acidic mist reduces foliar membrane associated calcium and impairs stomatal responsiveness in red spruce, Tree Physiology 25, 673 680.
[107] APIS focuses on the effects of nitrogen and sulphur compounds. These have been incorporated into the Critical Load Function tool, www.apis.ac.uk/critical load function tool.

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A.6 Scarborough Borough Council (2018). Cell 1 Coatham Dunes Report

Cell 1 Coatham Dunes Report 2018

A great place to live, work & play

March 2018

Scarborough Borough Council

Cell 1 Regional Coastal Monitoring Programme
Coatham Dunes Report 2018

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Data and reports collected as part of the Cell 1 Regional Coastal Monitoring Programme are available to download via the North East Coastal Observatory via the webpage: www.northeastcoastalobservatory.org.uk.

[1] Scarborough Borough Council is acting as client on behalf of all Local Authorities within 'Coastal Cell 1'.

Preamble

The Cell 1 Regional Coastal Monitoring Programme covers approximately 300km of the northeast coastline, from the Scottish Border (just south of St. Abb's Head) to Flamborough Head in East Yorkshire. This coastline is often referred to as 'Coastal Sediment Cell 1' in England and Wales (Figure 0-1). Within this frontage the coastal landforms vary considerably, comprising low-lying tidal flats with fringing salt marshes, hard rock cliffs that are mantled with glacial till to varying thicknesses, softer rock cliffs, and extensive landslide complexes.

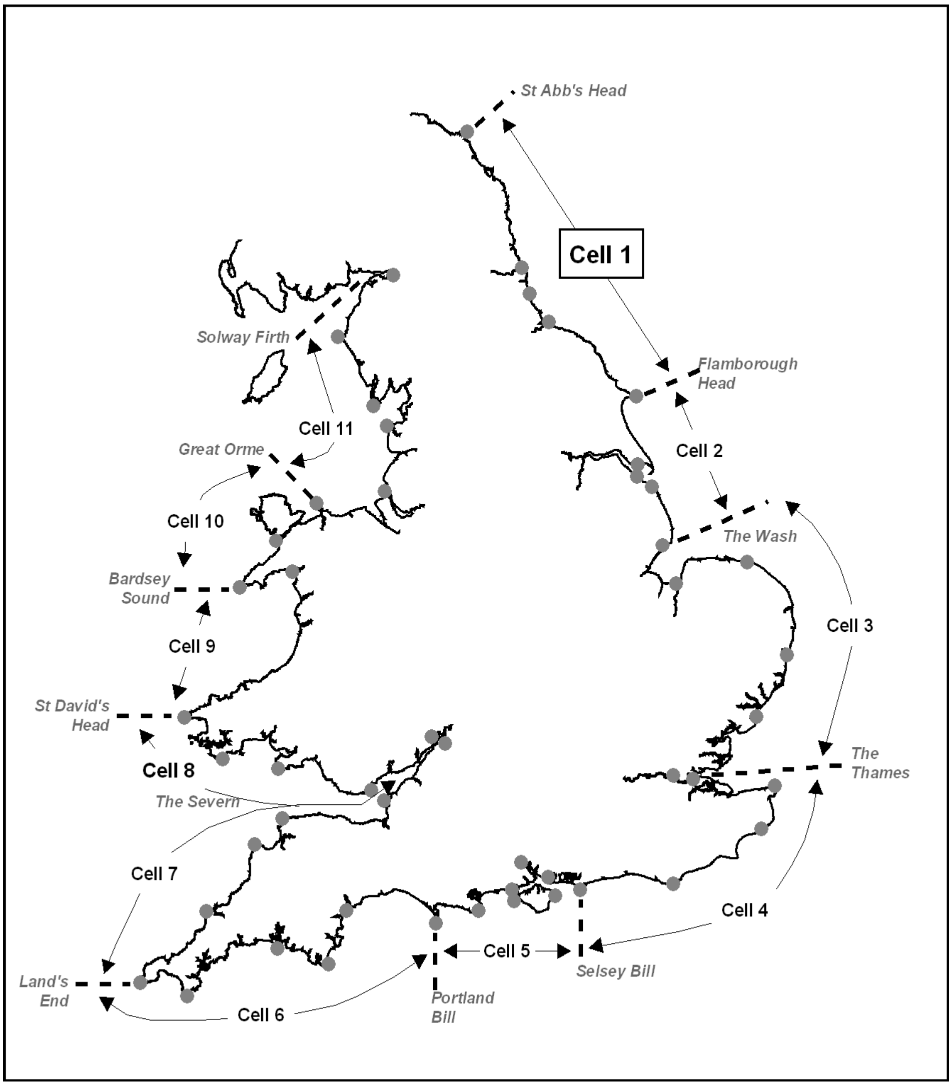

Figure 0-1 - Sediment Cells in England and Wales

The programme commenced in its present guise in September 2008 and is managed by Scarborough Borough Council on behalf of the North East Coastal Group. It is funded by the Environment Agency, working in partnership with the following organisations.

The main elements of the Cell 1 Regional Coastal Monitoring Programme involve:

- beach profile surveys
- topographic surveys
- cliff top recession surveys
- real-time wave data collection
- bathymetric and sea bed characterisation surveys
- aerial photography
- LiDAR survey
- walk-over inspection surveys

Royal HaskoningDHV has been appointed to provide Analytical Services in relation to the Cell 1 Regional Coastal Monitoring Programme 2016 - 2021.

Separate reports are produced for elements of the programme as and when specific components are undertaken, such as beach profile, topographic and cliff top surveys, wave data collection, bathymetric and sea bed characterisation surveys, and walk-over inspection surveys.

The present report is in addition to the above regular reports and covers a bespoke analysis of the coastal changes along the dunes at Coatham Sands in the borough of Redcar & Cleveland.

1. Introduction

The purpose of this report is to provide findings from a review of historic and contemporary maps and aerial photos to determine changes in land use and coastal erosion at the dunes along Coatham Sands, in the borough of Redcar & Cleveland.

All available aerial photos (historic and contemporary) from the Cell 1 Regional Coastal Monitoring Programme were downloaded from the North East Coastal Observatory website and viewed 'side by side' in ArcGIS to identify, describe and, where sufficient coastal change exists, quantify changes in the dunes along Coatham Sands, paying particular attention to the Majuba area towards the east of the frontage and any areas of identified change post 2013 and 2017 storm surges.

In addition, the selection of historic maps that is available from the National Library of Scotland website (which contains historic maps for the whole of the UK) was viewed on-screen for similar changes. [Note that the historic maps are not reproduced in this report due to copyright reasons].

2. Aerial Photography

1940

In the 1940 aerial imagery, the dunes adjacent to South Gare had not built out along the seaward edge of the spit in the manner that is observed in the present day, but instead occupied a bulbous shape, with a distinct ingress of sea water into a saline lagoon, with only a thin azimuth of land between the lagoon and the Bran Sands area of the River Tees estuary (Figure 1).

The Warrenby Slag Works are present in the 1940 imagery and slag deposits appear to push the shoreline seaward in locations immediately adjacent to the works, although the coastline here was still somewhat landward of its present day position in 1940 (Figure 2).

In the Majuba area, the present day caravan park had not been constructed in 1940 and whilst a seawall appeared to be present from the Redcar frontage towards the area of the present day Majuba car park, the car park itself was also not constructed at this time (Figures 3 & 4). It is notable that the dunes at this location were experiencing some vegetation loss and encroachment by the sea in the 1940s, even before the caravan park was built on this area.

Figure 1 – South Gare, 1940 (left) and 2017 (right)

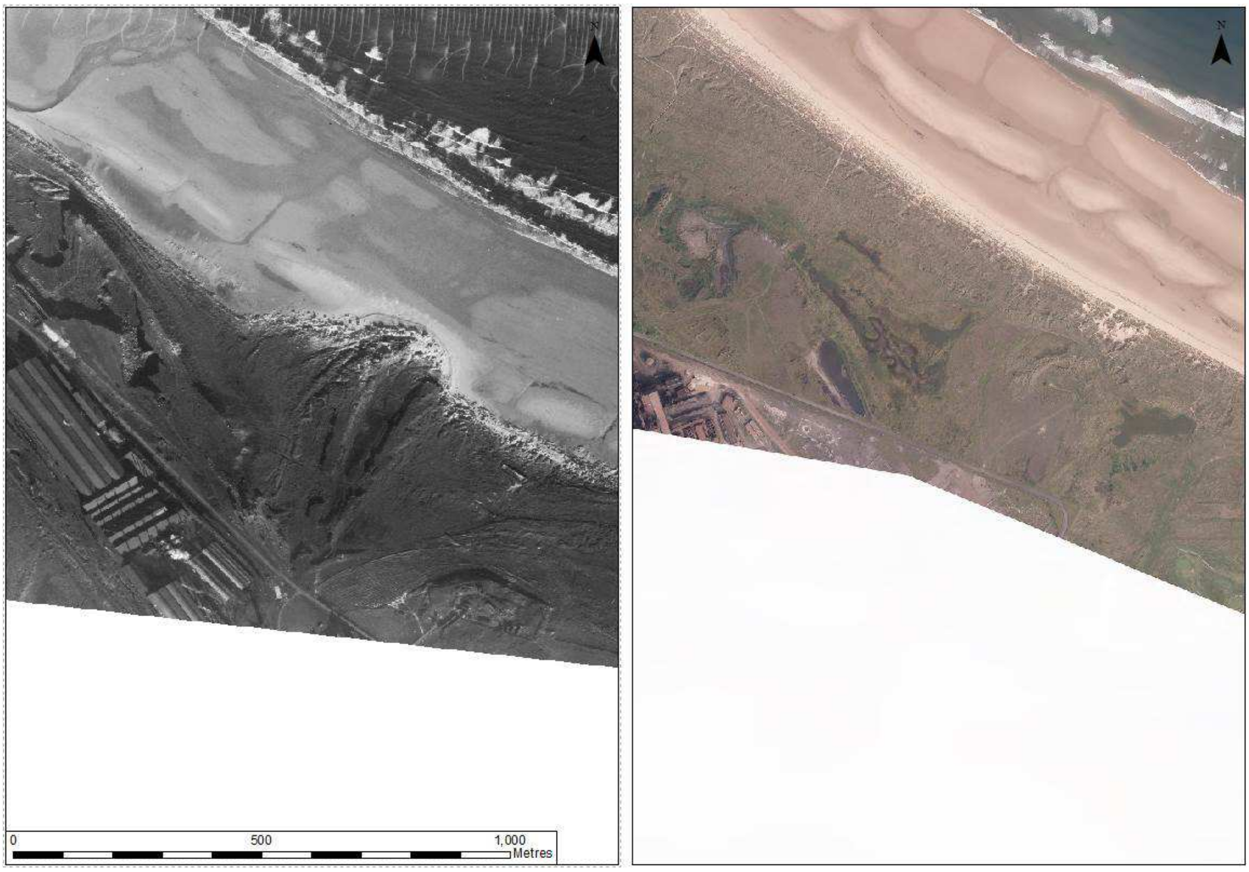

Figure 2 – Warrenby Slag Works, 1940 (left) and 2017 (right)

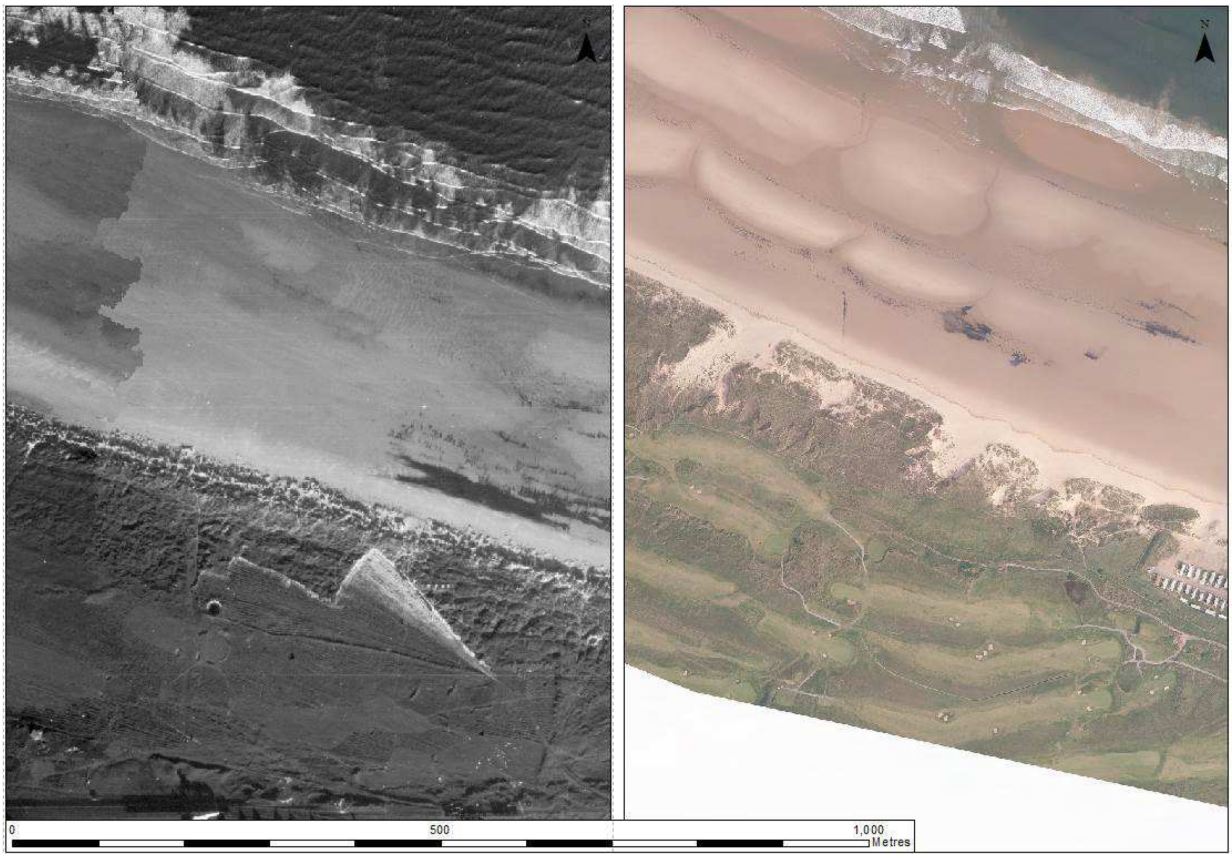

Figure 3 – Majuba Area (west), 1940 (left) and 2017 (right)

Figure 4 – Majuba Area (east), 1940 (left) and 2017 (right)

1999

By the time of the next available aerial photography from the Cell 1 Regional Coastal Monitoring Programme in 1999, the shore adjacent to the South Gare was undergoing change (Figure 5). It appears that sand or slag may have been artificially deposited to the east of the South Gare at this time, although the present day alignment had not yet been fully attained.

There had been continued progradation of the shore in the centre of the frontage, in the vicinity of the Warrenby Slag Works and the frontage by 1999 was appearing much more like a 'natural' dune system, with vegetated sand at the seaward limit, as opposed to a probable sand/slag mix present at the shore face in 1940 (Figure 6).

Both the caravan park and the car park had been constructed in the Majub area by 1999.

Figure 5 – South Gare, 1999 (left) and 2017 (right)

Figure 6 – Warrenby Slag Works, 1999 (left) and 1940 (right)

2009

By 2009, the shore adjacent to South Gare had continued to experience change, again likely in the form of sand/slag deposition and, due to the presence of the German Charlies in the nearshore, natural sand deposition in the now-sheltered areas. This resulted in quite a growth in the shore adjacent to the South Gare and stability in the dunes at the western end of the frontage (Figure 7).

Elsewhere along the frontage there was little change from 1999 to 2009, other than some exacerbation of areas of blow outs or bare dune vegetation to the immediate west of the caravan park in the Majuba area (Figure 8).

Figure 7 – South Gare, 2009 (left) and 1999 (right)

Figure 8 – Worsening of blow outs along eastern Coatham Sands, 2009 (left) and 1999 (right)

2010
There was little discernable change along the dunes in Coatham Sands between 2009 and 2010.

2012
The shore adjacent to South Gare appeared to contain greater quantities of material (sand/slag) and was more widely vegetated in 2012 than in 2010 (Figure 9).

At the Majuba area (Figures 10 & 11), part of the dunes adjacent to the caravan park were covered with hard-top and being used for car parking and portacabins in 2012. Presumably this was the Contractor’s compund for the duration of construction of the Redcar Sea Defence Scheme. It is also noticeable that the seaward row of caravans seen in the 2010 imagery had been removed by 2012, indicating a risk from erosion or sea flooding at that time.

Figure 9 – South Gare, 2012 (left) and 2010 (right)

Figure 10 – Majuba Area (west), 2012 (left) and 2010 (right)

Figure 11 – Majuba Area (east), 2012 (left) and 2010 (right)

2015
The shore adjacent to South Gare showed some further growth between 2012 and 2015 (Figure 12), but elsewhere along the Coataham Sands frontage there was no significant difference in the shore between the 2012 and 2015 imagery, indicating that if the December 2013 storm did cause localised damage, there had been natural recovery by 2015.

Following completion of the Redcar Sea Defence Scheme, the Contractor's portacabins at the caravan park in the Majuba area had been removed by 2015, but the hard-top remained intact. The most seaward row of caravans seen in the 2015 imagery had been restored since being (temprarily) removed before the 2012 imagery (Figure 13).

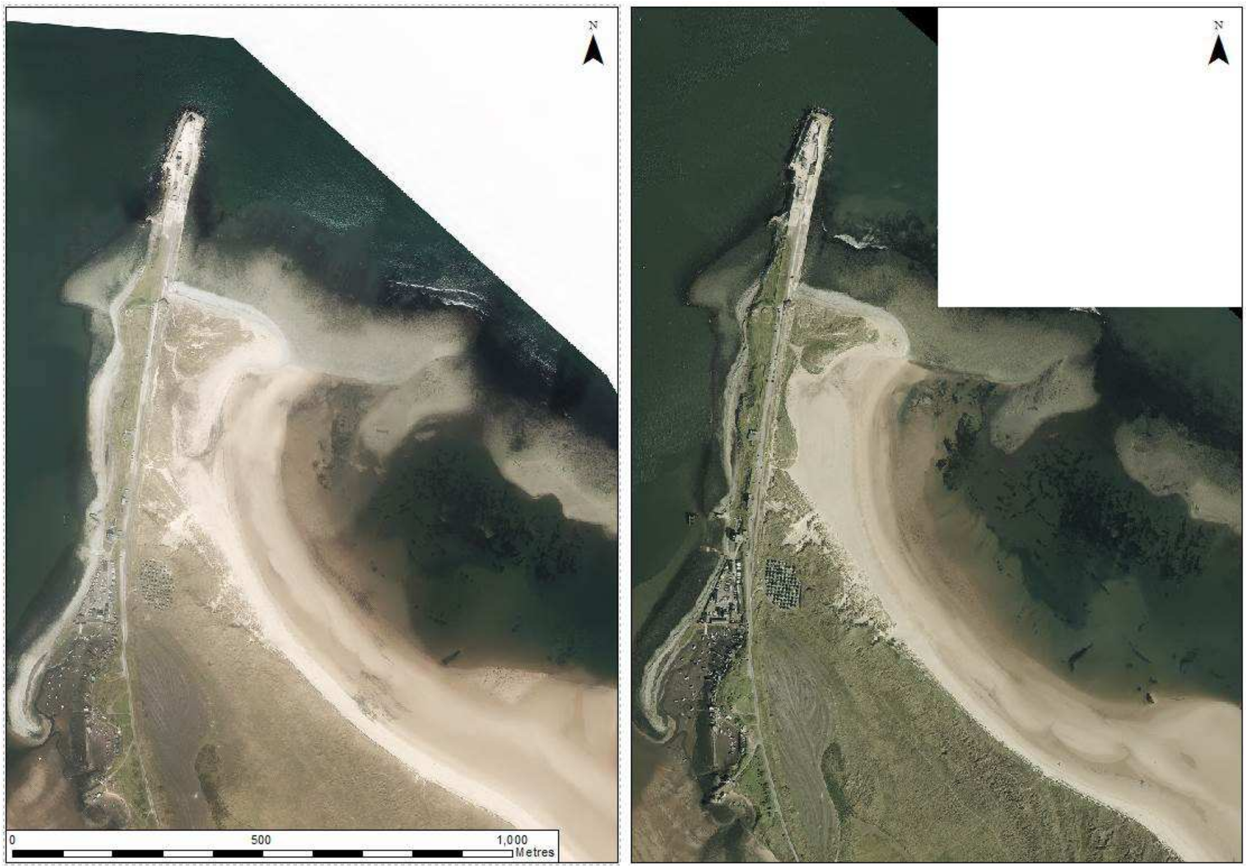

Figure 12 – South Gare, 2015 (left) and 2012 (right)

Figure 13 – Majuba Area, 2015 (left) and 2012 (right)

2017

In the immediate lee of the South Gare breakwater, the trend continued to be one of accretion in the shelter of the structure, with a notable increase in the extent of dune vegetation (Figure 14). Some areas of 'scalloped' dune evident in the 2017 aerial photography was also present in the photography that was collected in 2015 and appears not to have worsened. Arguably in some areas (e.g. Figure 15) it may have marginally recovered, although remaining heavily scalloped.

Some areas that were anecdotally described as 'breaching' or 'severely eroding' during the January 2017 storms, were clearly in such a state before the 2015 photography was collected and thus the damage to these dunes cannot be ascribed to the January 2017 storms alone (Figures 16 to 2-10).

Figure 14 – Vegetation growth on dunes in the lee of South Gare Breakwater between 2017 (left) and 2015 (right)

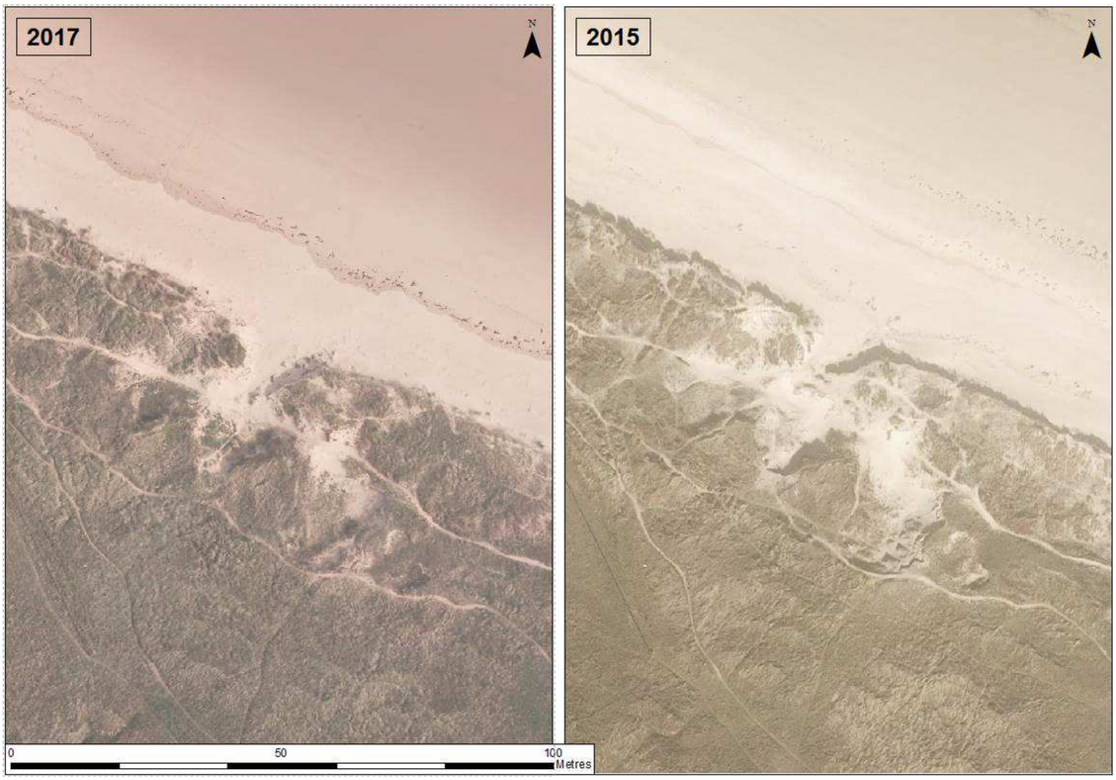

Figure 15 – 'Scalloping' of dunes along Coatham Sands in 2017 (left) and 2015 (right)

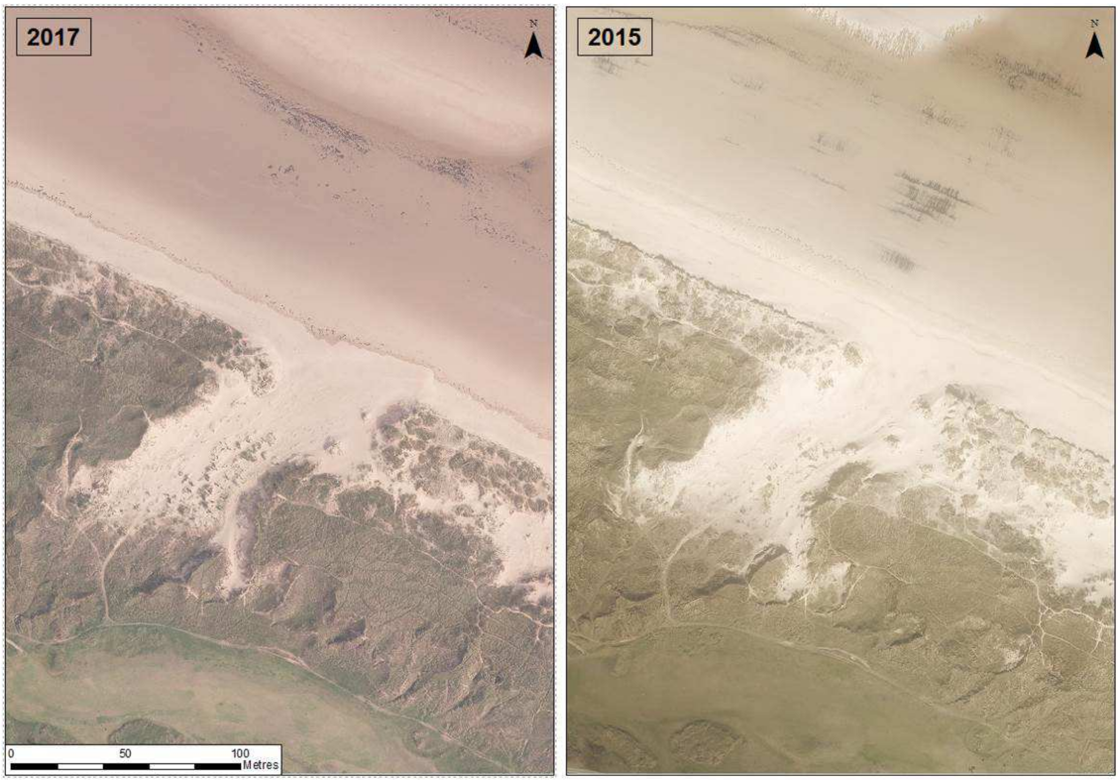

Figure 16 – Damage to dunes along Coatham Sands in 2017 (left) and 2015 (right)

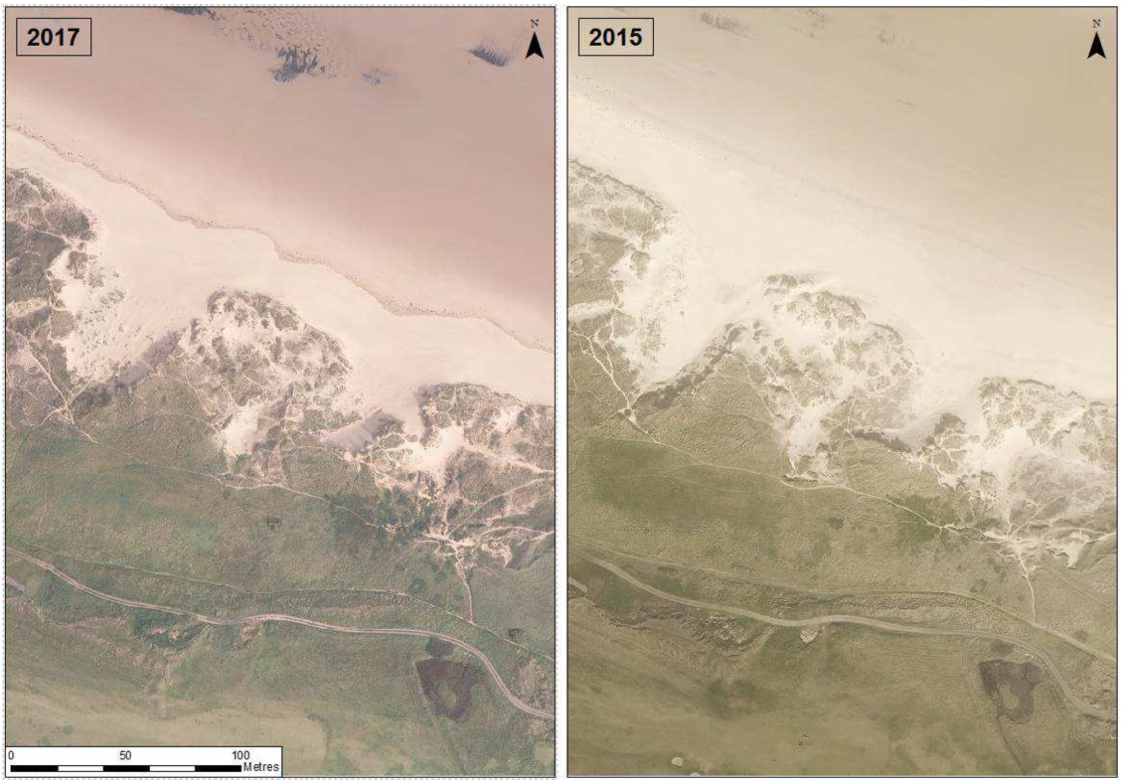

Figure 17 – Damage to dunes along Coatham Sands in 2017 (left) and 2015 (right)

Figure 18 – Dune condition fronting western section of caravan park along Majuba Area in 2017 (left) and 2015 (right)

Figure 19 – Dune condition fronting eastern section of caravan park along Majuba Area in 2017 (left) and 2015 (right)

3. Historic Maps

OS One Inch, 1885 – 1903

In the first available historic map, the morphology of the Tees estuary is very different from the present day, with extensive areas of inter-tidal mud flat and salt marsh exposed at low tide, especially across Seal Sands and Bran Sands.

South Gare appears to be a natural spit at the mouth of the Tees estuary, with rail tracks along its length. The German Charlies slag banks were not present in the nearshore zone at this time and so the sand accumulated in the lee of the spit was more parallel with the spit than in the present day (the beach immediately in the lee of the German Charlies has built out as a small embayment in the present day).

At the root of the spit the Mean High Water (MHW) mark was in a considerably more landward position than in the present day, indicating that sand dune accretion has occurred at this western end of the frontage over the long term. However, this accretion has not only occurred in the vicinity of the spit at Tees Mouth; along the whole length between the spit and what is now known as the Majuba Road car park the historic MHW line was more landward than in the present day (although the width of progradation decreases with progression to the east so that the historic MHW mark is very close to the present day in the vicinity of the caravan park).

The line of MHW is not a smooth ‘bay’ shape, but does have some jagged undulations towards the western section, perhaps indicating some differences in the topography or maybe even a former channel mouth.

Conversely, along Majuba Road towards Redcar (and beyond to the east), the historic MHW mark was more seaward than in the present day, indicating net recession of the shore in this area.

OS Six Inch, 1888 – 1913 & OS 25 Inch, 1892 - 1914

The improved scale of the OS six inch mapping from a similar period, shows the South Gare Breakwater clearly present and the dunes in the vicinity of the present day caravan park named as Coatham Bank. The MHW mark was considerably landward of its present day position from Tees Mouth to the present day caravan park, indicating accretion along this length, with recession evident further to the east. The point at which the dunes switch from accretion to erosion between the historic maps and the present day is exactly at the western end of the Majuba Road car park.

OS 1:25,000, 1937 – 1961 & OS One Inch 7th Series, 1955 - 1961

By the time of this map, the German Charlies had been placed and started to modify the morphology of the dunes in their lee. The jagged undulation in line of MHW was particularly pronounced just to the west of Warrenby Slag Works which by now were present (and presumably responsible for the German Charlies slag banks).

Even at this time, the MHW was landward of its present day position along most of the frontage, but the ‘switch-point’ between the accretion and erosion had migrated to the western end of the caravan park. This indicates that the caravan park frontage has been under some pressure since around the mid 1950s.

4. Conclusions

Analysis of historic maps from the National Library of Scotland website (which contains historic maps for the whole of the UK) and aerial photographs from the Cell 1 Regional Coastal Monitoring Programme reveals the following key findings:

- Coatham Sands has, in places along its length, been strongly influenced by historic deposition of slag from local ironworks. This means that large parts of the dunes must be a mix (in some manner) of slag deposits and natural marine-deposited and subsequently wind-blown sand.

- Accretion due to natural processes and/or progradation due to slag deposition has particularly been observed to the immediate east of South Gare, but is evident to some extent along the whole of Coatham Sands until reaching the Majuba car park. These processes were exacerbated when slag was deposited off the South Gare thereby creating the German Charlies which caused even calmer conditions conducive to natural accretion of sand at the western end.

- The most vulnerable section of Coatham Sands is undoubtedly the Majuba area. Historically, to the west of the car park the frontage experienced progradation and to the east (along the car park frontage) it experienced recession. However, the zone of transition between progradation and recession appears to have migrated westwards over time, meaning that more of the caravan park frontage and area to the immediate west has been exposed.

- However, the 'scalloped' nature of some sections of the dunes, especially towards the eastern end, has existed for some considerable time and cannot be ascribed to the effects of the December 2003 or January 2017 storms alone.

- The Majuba area, where it is understood there is an historic landfill at the core of the dunes with a covering of wind-blown sand, does appear to have lost vegetation (marginally) between 1999 and 2009 (Figure 20), perhaps due to local blow outs or storm erosion. However, the broad configuration of the dunes here has been roughly in its present condition consistently, with only minor changes, for some considerable time as shown by the similarities between the 2009 and 2017 photography (Figure 20). From previous field visits it is known that the historic waste material has become exposed on the seaward face where the covering of blown sand is absent, but this is not visible from the aerial photography.

Figure 20 - Majuba Area, 1999 (top), 2009 (middle) and 2017 (bottom)

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Open Access Earth System Science Data

Nitrogen deposition in the UK at 1 km resolution from 1990 to 2017

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Abstract. An atmospheric chemistry transport model (FRAME) is used here to calculate annual UK N deposition for the years 1990–2017, at a 1 km × 1 km resolution. Reactive nitrogen (N) deposition is a threat that can lead to adverse effects on the environment and human health. In Europe, substantial reductions in N deposition from nitrogen oxide emissions have been achieved in recent decades. This paper quantifies reductions in UK N deposition following the N emissions peak in 1990. In the UK, estimates of N deposition are typically available at a coarse spatial resolution (typically 5 km × 5 km grid resolution), and it is often difficult to compare estimates between years due to methodological changes in emission estimates. Through efforts to reduce emissions of N from industry, traffic, and agriculture, this study predicts that UK N deposition has reduced from 465 kt N in 1990 to 278 kt N in 2017. However, as part of this overall reduction, there are non-uniform changes for wet and dry deposition of reduced N (NH_x) and oxidised N (NO_y). In 2017, it is estimated 59 % of all N deposition is in the form of reduced N, a change from 35 % in 1990. This dataset uses 28 years of emissions data from 1990 to 2017 to produce the first long-term dataset of 28 years of N deposition at 1 km × 1 km resolution in the UK. Full data are available at https://doi.org/10.5285/9b203324-6b37-4e91-b028-e073b197fb9f (Tomlinson et al., 2020).

1 Introduction

The emissions and subsequent atmospheric deposition of nitrogen (N) have a well-documented list of effects on the global and local environment (e.g. Stevens et al., 2018). N deposition is associated with impacts on ecosystem biodiversity (Nowak et al., 2015; Payne et al., 2017), eutrophication (Greenwood et al., 2019), soil acidification (Aggenbach et al., 2017), changes in carbon stocks (Britton et al., 2019) and human health (Nowak et al., 2018).

These threats are driven by anthropogenic emissions of oxides of nitrogen (NO_x) from sources such as fuel combustion including from road transport, and emissions of ammonia (NH_3), to which agriculture contributes around 85 % annually in the UK (NAEI, 2019). Previous studies generally show total deposition of N in the UK peaking around 1990, following the peak in emissions. Fowler et al. (2004) estimate around 430 kt N was deposited to the UK in 1990, with a 54 % proportion of reduced N (predominantly ammonia). Using newer data, the Review of Transboundary Air Pollution report (RoTAP, 2012) re-estimated the total N deposition budget for 1990 in the UK to be ca. 380 kt N, and finally Levy et al. (2020) estimated 410 kt N deposited. Since the beginning of the 1990s, deposition has reduced as mitigation policies have sought to curb emissions of nitrogenous compounds, predominantly NH_3 and NO_x, but has stabilised at around 300 kt N yr^{-1} from ca. 2010.

In order to study the many effects of N deposition and its trends over time, there must be appropriately detailed and consistent deposition estimates to use, across time and space. N deposition data in the UK are typically available at a 5 km × 5 km resolution (e.g. Levy et al., 2020). It is very likely, however, that this relatively coarse spatial resolution smooths out significant variation at higher resolutions, which could be useful for studying effects. Smart et al. (2020) highlight this point by exploring the variance of a 5 km × 5 km and 1 km × 1 km N deposition output from the same model run, within a 10 km square. They found the variance within the 1 km × 1 km product to be up to 4 times higher than that of the 5 km × 5 km product (within the same 10 km square). The main driver for this increased variance of N deposition at higher spatial resolutions, compared to lower resolutions (within the same study area), is the more granular representation of dry N deposition from agricultural sources such as livestock houses and busy roads or local combustion sources. Dry deposition of N from reduced nitrogen ("NH_x") is very local to the emissions sources, which a 1 km × 1 km resolution can more easily reflect. Furthermore, the increased definition in a 1 km × 1 km rainfall map (for wet deposition) has more variation than a smoothed 5 km × 5 km rainfall map, while land cover is more readily represented in higher resolutions (which can determine deposition velocities and therefore N deposition).

Another facet of N deposition to consider is that of cumulative loading and whether the impacts develop over time, and whether they develop linearly (Payne et al., 2019, 2020). Payne et al. (2019) showed that N deposition effects on sensitive habitats should not only take account of the most recent best estimate, but that cumulative N deposition should be considered, e.g. over a period of 30 years. To enable such an approach, it is necessary to have a suitable consistent N deposition data series available. In the past, time series were often constructed by piecing together historical products that were using the best knowledge and datasets available at the time, rather than a single time series where all model output years are produced with consistent model input data from the latest back-cast inventory dataset, and with the same version of the model and calibration methodology.

This new dataset consists of 28 years of 1 km × 1 km resolution N deposition data on the UK terrestrial surface, from 1990 to 2017, using a consistent approach to inputs and model calibration. It is the first time annual N deposition data has been released at this resolution over this number of years in the UK, using a consistent methodology throughout. The consistent methodology means that the latest knowledge for emission distributions across the whole period can be used, with the latest emission factors used to back-cast the entire time series at a high spatial resolution. In addition, model parameters and calibrations for each time step use the same most up-to-date model version. It is envisaged that studying the effects of N deposition on the environment can be aided by such an increase in detail, as suggested by Hallsworth et al. (2010). This has been made available as part of The ASSIST programme (Achieving Sustainable Agricultural Systems; see https://assist.ceh.ac.uk, last access: 1 December 2020).

2 Data and methods

2.1 Atmospheric chemistry transport modelling

The Fine Resolution Multi-pollutant Exchange (FRAME) is an atmospheric chemistry transport model (ACTM) used to calculate annual deposition of reduced and oxidised nitrogen (N) over the United Kingdom. The model is fully described elsewhere (Aleksankina et al., 2018; Dore et al., 2012, 2016; Vieno et al., 2010; Singles et al., 1998), and only the relevant information for this work is reported here. The domain of the model covers Europe at 50 km × 50 km resolution to provide the boundary conditions for the UK model domain with a grid resolution of 1 km × 1 km. The UK model domain is represented by the British National Grid (EPSG:27700) projected coordinate system. A column of air with a depth of 2500 m is used to represent the relevant atmospheric processes. The column of air is advected across the model domain from all edge grid points and all wind directions with an angular resolution of 1°. Figure 1 shows the 1 km × 1 km UK model domain – which captures both the UK and the Republic of Ireland to allow for high-resolution modelling of the closest neighbouring territory – in the European context. Further figures in this work did not show lines of latitude or longitude.

Emission of gaseous pollutants, vertical diffusion, chemical transformation, and wet and dry removal processes take place within the air column. The model has 33 vertical layers with thickness varying from 1 m at the surface to 100 m in the upper layers. The model requires input data of both diffuse and point source emissions of ammonia (NH_3), oxides of nitrogen (NO_x) and sulfur dioxide (SO_2) (Vieno et al., 2010).

FRAME uses land-cover-specific deposition velocities to generate dry deposition for up to five land cover categories: woodland, low-growing semi-natural vegetation, improved grassland, arable and urban (Land Cover Map 2015; Rowland et al., 2017). The model uses different scavenging coefficients for soluble gases and particles and assumes constant drizzle for calculation of wet deposition. An annual precipitation map (Tanguy et al., 2019; Walsh, 2012) is used to drive the spatial variation in wet removal rate.

The FRAME model used for this work uses long-term radiosonde mean wind speed (Dore et al., 2006) for all the years included here (1990–2017). The wind frequency is derived from modelled data from the Weather and Research Forecast model (Skamarock et al., 2019). The wind frequency used here is kept constant to a 2001–2012 mean for the years 1990–2001, and the specific years afterwards (2001–2017).

The FRAME model, for both the European and British Isles domains, was run for each year from 1990 to 2017, using the corresponding emission and wind–rainfall data. The land cover was kept constant throughout. The FRAME model version used was 9.15.0.

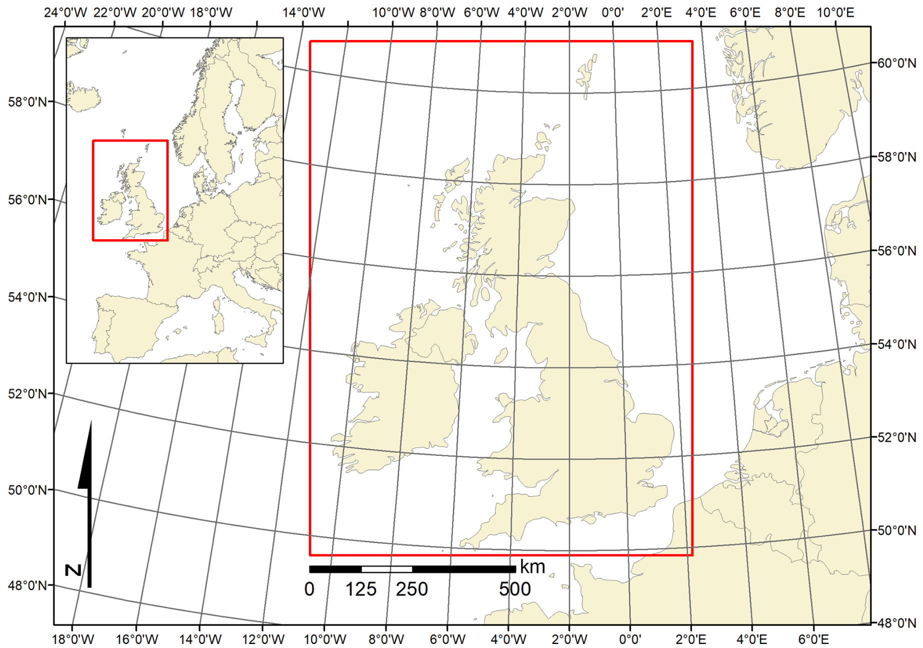


Figure 1. The UK FRAME modelling domain, shown by a red rectangle, within which 1 km × 1 km estimates of N deposition are made. The inset shows the context within Europe and lines of latitude and longitude are also shown, while the inputs and outputs of the model are in the British National Grid projected coordinate system.

2.2 Emissions data

2.2.1 Data sources

Input data were extracted and processed from the most recently available national emission inventory submissions from both the UK and the Republic of Ireland (EMEP, 2019; E-PRTR, 2019; NAEI, 2019). Emissions for the European domain were taken from Convention on Long-Range Transboundary Air Pollution (CLRTAP) submissions (EMEP, 2019). For agricultural NH_3 emissions, the latest set of annual emission maps from 1990–2017 was used, as derived for the UK's national atmospheric emission inventory. This inventory work utilises annual activity data at the holding level from the devolved authorities in the UK, i.e. Defra (England), the Scottish Government (Scotland), Welsh Assembly (Wales) and Daera (Northern Ireland) (see Carnell et al., 2019a, for details).

Emissions data are routinely made available via sectors (e.g. energy production) and to create a consistent structure for all data sources. NO_x and SO_2 emissions were restructured into the 11 Selected Nomenclature for sources of Air Pollution (SNAP) sectors (Table 1), developed by the European Topic Centre on Air Emissions (ETC/AE). Given the dominance of agriculture in NH_3 emissions, the FRAME model requires agricultural data to be split into livestock fertiliser emissions, with all non-agricultural sources as one sector (see Sect. 2.1.3).

The SNAP system is used in the UK for the annual updates to the National Atmospheric Emissions Inventory (NAEI, 2019). This corresponds to the main area of interest for the deposition outputs, and the Irish and wider European emissions were reformatted to match that reporting system. Whilst the UK, Ireland and the collated European data all use the Nomenclature for Reporting system (NFR, ca. 240 sectors – EEA, 2019), the UK collate the fine-resolution categories into SNAP sectors whereas the latter two report via the aggregated Generalised/Gridded Nomenclature for Reporting (GNFR). Table 1 also shows how these two aggregated reporting systems broadly relate to each other.

It is worth noting that emissions data for "international shipping" and "aviation cruise" do not count within a specific national inventory but are reported into a "pooled" total by all countries. Separate totals for national shipping, airports, and the take-off and landing of aircraft are reported on a country basis. Finally, emissions data should ideally be translated between the aggregated classification systems using the NFR codes upon which they are built (which still has some one-to-many relationships) but spatial data are not available at this level, and therefore the aggregated spatial data should not be broken down in an attempt to make the NFR level data.

2.2.2 Point and diffuse emissions of NO_x, SO_2 and NH_3

NH_3, NO_x and SO_2 emission inputs were produced for the years 1990 to 2017, for both diffuse and point source emissions. Diffuse sources are those deemed to be areal, non-exact locations such as agriculture, vehicles, population-related sources, etc. Point sources can be located by exact co-

Table 1. Selected Nomenclature for sources of Air Pollution (SNAP) sectors for emissions inventory reporting as outlined by CORINAIR, alongside the Generalised/Gridded Nomenclature for Reporting (GNFR) sectors (broadly matched).

SNAP sector	SNAP definition	GNFR sector
1	Combustion in Energy Production & Transformation	A_PublicPower
2	Combustion in Commercial, Institutional & Residential & Agriculture	C_OtherStationaryCombustion
3	Combustion in Industry	B_Industry
4	Production Processes	
5	Extraction & Distribution of Fossil Fuels	D_Fugitive
6	Solvent Use	E_Solvents
7	Road Transport	F_RoadTransport
8	Other Transport & Mobile Machinery	G_Shipping H_Aviation I_Offroad
9	Waste Treatment & Disposal	J_Waste
10	Agriculture Forestry & Land Use Change	K_AgriLivestock L_AgriOther
11	Nature	N_Natural
NA	Do not count towards national totals	O_AviationCruise P_IntlShipping

ordinates, for example the actual chimney/exhaust stacks of power stations and industry (Vieno et al., 2010). Point source information in the UK is nearly (but not totally) exclusive to energy generation and industry.

Figure 2 shows an overview of the processes to combine the various spatial and tabulated emissions data that are required for the 28 annual model runs. There are some important methodological details, for both diffuse and point emissions, worth noting. In the UK, diffuse data are produced and published for 11 SNAP sectors for the latest emissions inventory year, superseding any previous data. This is principally due to the fact that every year in the inventory compilation, minor to major changes are made to the way the data are compiled – this could be changes to emission factors with the latest research being incorporated or how underlying spatial methods and datasets are updated. While the non-spatial data are "back-cast" to 1990 (or earlier, depending on the pollutant), the maps are not currently updated as a time series. Consequently, it is unwise to compare previous years' gridded emissions surfaces to the latest available. For this reason, at the time of publication, only the latest 2017 emissions maps were used in the UK for the entire time series and were scaled back through the time series using the annual tabulated NFR totals, for SO_2, NO_x and non-agricultural NH_3. For agricultural NH_3, the latest mapped time series (using annual livestock and crop data) was used (Carnell et al., 2019a). For point sources – which in the more recent data number in the thousands – some earlier data were obtained back to 1990 but only for a subset of major polluters and not for all years (missing years were linearly interpolated). For the very largest emitters, information (when known) regarding the stack/chimney height, stack/chimney diameter and emission exit velocities is also used by the model to create plume characteristics. It is the non-coordinate parameters that are important in determining to what height into the atmosphere the emissions travel, and therefore what subsequent chemical interactions occur, which is important for the deposition modelling.

Emissions from the Republic of Ireland influence the deposition of N species in the UK. To allow for similarly high-resolution emissions inputs, the outputs from the National Mapping of Greenhouse Gas and Non-greenhouse Gas Emissions Sources project (MapEire, 2019; Pjeldrup et al., 2018) were used in a similar manner to the latest emissions surfaces produced for the UK in the NAEI. The MapEire project produced 1 km × 1 km resolution gridded emissions for all GNFR sectors for the year 2016, which were scaled to other years by the totals reported to the CLRTAP by the Republic of Ireland. These surfaces were then transformed to SNAP sectors (see Table 2) to be joined to the UK data. One important difference to note is that the MapEire gridded data include all sources of emissions, including point sources (the UK data do not). Therefore, the major emitting point sources, as reported to the European Pollutant Release and Transfer

Table 2. Deposition outputs as provided in this dataset from the Fine Resolution Multi-pollutant Exchange (FRAME) atmospheric chemistry transport model.

Name	Long name	Description	Units
NX_x dry	Dry deposition of reduced N	Grid average deposition of $NH_3 + NH_4$, plus forest- and moorland-specific deposition	$Kg\,N\,ha^{-1}\,yr^{-1}$
NH_x wet	Wet deposition of reduced N	Grid average deposition of $NH_3 + NH_4$, plus forest- and moorland-specific deposition	$Kg\,N\,ha^{-1}\,yr^{-1}$
NO_y dry	Dry deposition of oxidised N	Grid average deposition of $NO_2 + NO_3 + HNO_3 +$ PAN, plus forest- and moorland-specific deposition	$Kg\,N\,ha^{-1}\,yr^{-1}$
NO_y wet	Wet deposition of oxidised N	Grid average deposition of $NO_3 + HNO_3$, plus forest- and moorland-specific deposition	$Kg\,N\,ha^{-1}\,yr^{-1}$

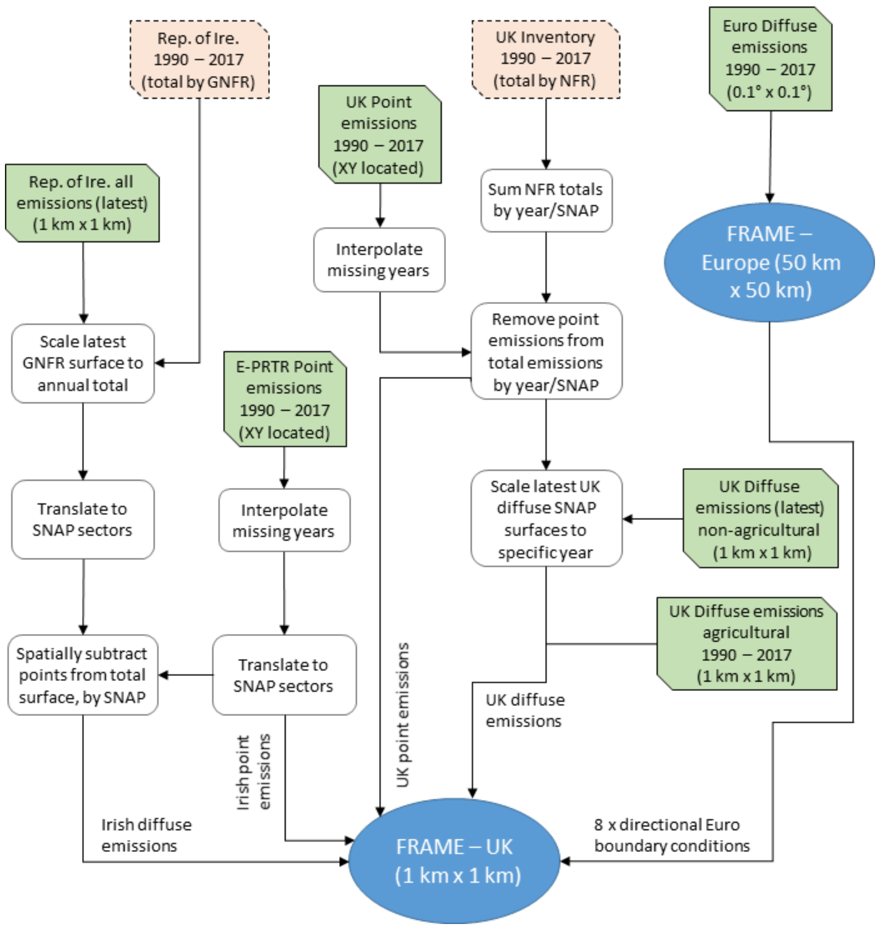


Figure 2. Visualised methodology of steps to create inputs for the Fine Resolution Multi-pollutant Exchange (FRAME) atmospheric chemistry transport model: rectangle with corners missing (solid border) denotes spatial data, rectangle with corners missing (dashed border) denotes tabulated data, rectangle with rounded corners denotes process and oval denotes model.

Register (E-PRTR, 2019), were extracted for NO_x and SO_2 for all available years back to 1990 (gaps were linearly interpolated). To conserve totals, Irish point values were removed from the Irish total gridded surface by subtracting the point value from the grid cell in which it was located, with any surplus emissions removed from the surrounding eight cells on an equal-share basis (if required). This created a diffuse surface and a point source input, consistent with the UK data.

A consistent time series of UK agricultural NH_3 emission estimates was created at a $1\,km \times 1\,km$ grid resolu-

Table 3. Four measurement networks used within the UK Acidifying and Eutrophying Atmospheric Pollutants (UKEAP) network, along with the 10 compounds used to evaluate the atmospheric modelling.

Network	Long name	Data provided	Measurement resolution	Units	Start year
NAMN	National Ammonia Monitoring Network	NH_3 – ammonia conc. in gas NH_4 – ammonium conc. in aerosol	Monthly	$\mu g\,m^{-3}$ $\mu eq\,L^{-1}$	1996
PrecipNet	Precipitation Network	NO_3 – nitrate conc. in NH_4 – ammonium conc. in precipitation	Fortnightly	$\mu eq\,L^{-1}$ $\mu eq\,L^{-1}$	1985
NO_2NET	Rural Background NO_2	NO_2 – nitrogen dioxide conc. in gas	Four-weekly	$\mu g\,m^{-3}$	1990
AGANET	Acid Gases & Aerosol Network	NO_3 – nitrate conc. in aerosol HNO_3 – nitric acid conc. in gas	Monthly	$\mu g\,m^{-3}$ $\mu g\,m^{-3}$	2000

tion for the years 1990–2017. These high-resolution agricultural NH_3 emission maps are produced annually for the NAEI, using an agricultural emission model jointly developed by the UK Centre for Ecology & Hydrology, Rothamsted Research, ADAS and Cranfield University. The emission model uses annual agricultural census data (e.g. livestock numbers and crop areas – see Carnell et al., 2019a) at the holding level, agricultural practice information (e.g. fertiliser application rates, stocking densities) and emission source strength data from the UK emissions inventories for agriculture (e.g. Brown et al., 2019; Richmond et al., 2019). Emission estimates are output for each individual emission source at a 10 km × 10 km grid resolution, which are spatially disaggregated to a 1 km × 1 km grid resolution using land cover data (Rowland et al., 2017) and methods outlined in Dragosits et al. (1998), Hellsten et al. (2008) and Carnell et al. (2019a). Emissions sources are numerous and include grazing, storage, spreading and housing for cattle, pigs, poultry, sheep and minor livestock (plus all sub-types) as well as differing fertiliser applications for varying crop and grass types.

2.3 Outputs

Outputs from the model as presented in this dataset are the annual values of wet and dry deposition of reduced nitrogen ("NH_x") and wet and dry deposition of oxidised nitrogen ("NO_y") as a weighted mean of all land cover types within a given cell, as well as vegetation-specific values for both forest and moorland – Table 2 provides more detail.

Deposition data are provided on a 1 km × 1 km resolution surface, using the British National Grid projection (same domain as the emission files) for UK terrestrial cells (no. of cells = 259 436). Other land cover types used in the calculations (but not output) are arable, urban and improved grassland.

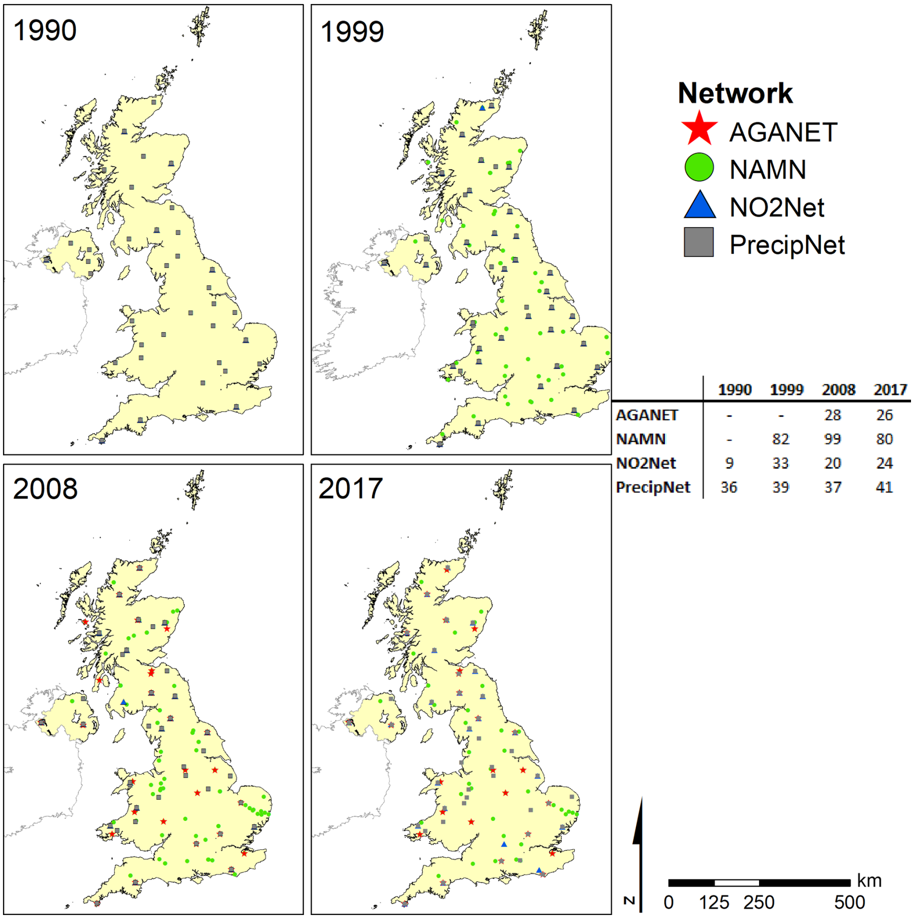
2.4 Evaluation

2.4.1 Observation data

ACTM results were evaluated using measured annual mean concentrations from rural background monitoring stations throughout the UK, via the UK Acidifying and Eutrophying Atmospheric Pollutants (UKEAP) network (UK AIR, 2020). Mean annual data were used (as the FRAME model output is an annual mean) if there was a data capture greater than 50 % across the measurements for a given site in a given year, which allows not only for direct comparison between modelled and measured data but also a certain amount of smoothing of potential variability in the measured data due to natural factors (Chang and Hanna, 2004). Table 3 outlines the available measurement networks and the data they provide, while Fig. 3 shows the spatial distribution of the observation sites with measurements in 1990, 1999, 2008 and 2017 (the first year of measurements for each observation network is noted in Table 3). It is believed that this is the first time model evaluation for gases, aerosols and concentration in precipitation has been done across a long time series at multiple points in time on the same dataset.

2.4.2 Evaluation metrics

It is unlikely for an ACTM to perfectly reproduce reality due to errors in, but not limited to, input data, model physics and chemistry schema, uncertainty in meteorological data, and the random effects of the real world. However, using methods outlined in Chang and Hanna (2004), several statistical metrics may be used to evaluate the agreement between the modelled predictions and the real-world observations: fraction of predictions within a factor of 2 of observations (FAC2), the fractional bias (FB), the normalised mean square error (NMSE) and the geometric mean bias (MG). These metrics are defined in the following way:



	1990	1999	2008	2017
AGANET	-	-	28	26
NAMN	-	82	99	80
NO2Net	9	33	20	24
PrecipNet	36	39	37	41

Figure 3. Locations of sites in four measurement networks, across four periods of the time series in this study: Acid Gases & Aerosol Network (AGANET), National Ammonia Monitoring Network (NAMN), Rural Background NO_2 (NO_2NET) and Precipitation Network (PrecipNet). Some sites from different networks are co-located, and therefore not all dots represented in the table are visible in the maps.

$$\mathrm{FAC2} = \text{fraction of data that satisfy } 0.5 \leq \frac{C_\mathrm{p}}{C_\mathrm{o}} \leq 2.0, \tag{1}$$

$$\mathrm{FB} = \frac{\left(\overline{C_\mathrm{o}} - \overline{C_\mathrm{p}}\right)}{0.5\left(\overline{C_\mathrm{o}} + \overline{C_\mathrm{p}}\right)}, \tag{2}$$

$$\mathrm{NMSE} = \frac{\overline{\left(C_\mathrm{o} - C_\mathrm{p}\right)^2}}{\overline{C_\mathrm{o}} \cdot \overline{C_\mathrm{p}}}, \tag{3}$$

$$\mathrm{MG} = \exp\left(\overline{\ln C_\mathrm{o}} - \overline{\ln C_\mathrm{p}}\right), \tag{4}$$

where C_o represents measured observations and C_p represents model predictions, the former being paired with the latter spatially. A perfect reproduction of measurement data would have $\mathrm{FAC2} = 1$, $\mathrm{FB} = 0$, $\mathrm{NMSE} = 0$ and $\mathrm{MG} = 1$.

FAC2 is a robust measure of performance, not overly influenced by outliers, indicating the proportion of modelled–measured pairs falling within a factor of 2 of each other. FB is a linear metric that measures the mean systematic bias of the model and may have predictions out of phase with measurements but still return a value of 0 due to cancelling errors. NMSE is a measure of mean relative scatter and reflects both systematic and random errors. Finally MG is also a measure of mean systematic bias, but is less influenced by extreme values as it is a logarithmic metric (see Chang and Hanna, 2004, for more detail). Hanna and Chang (2012) suggest that a model should satisfy at least 50 % of the criteria used (two of four in this study), while the acceptability criteria for each metric are as defined in Theobald et al. (2016): $\mathrm{FAC2} > 0.5$, $|\mathrm{FB}| < 0.3$, $\mathrm{NMSE} < 1.5$ and $0.7 < \mathrm{MG} < 1.3$.

3 Results and discussion

3.1 Emissions

In the UK, stricter air pollution policies, improving technology and changes in fuel use, have all contributed to the reduction of emissions. Initially, mitigation strategies concentrated on SO_2 emissions, but the focus was extended to nitrogen compounds such as NO_x (as well as VOCs) in an attempt to abate acidification and, latterly, to NH_3 (Grennfelt and Hov, 2005; Carnell et al., 2019b). Within the model do-

Table 4. Evaluation metrics of modelled concentrations of six nitrogen compounds in gas, aerosol and precipitation in the UK for 2017 (see Table 3 for definitions). Bold numbers represent where that metric has been satisfied (see Sect. 2.4.2 for metric definitions).

Metric	Acceptability	NH_3 (conc. in gas)	NH_4 (conc. in aerosol)	NH_4 (conc. in precip.)	NO_2 (conc. in gas)	NO_3 (conc. in aerosol)	NO_3 (conc. in precip.)
Points (n)	n/a	68	26	41	24	26	41
R^2		0.61	0.79	0.51	0.87	0.84	0.61
FAC2	> 0.5	**0.76**	**0.50**	**0.76**	**0.96**	**0.85**	**0.63**
|FB|	< 0.3	0.33	0.62	0.42	**0.26**	**0.20**	0.50
NMSE	< 1.5	**0.44**	**0.54**	**0.35**	**0.12**	**0.10**	**0.37**
MG	> 0.7 & < 1.3	**0.70**	2.29	0.64	1.33	1.31	0.56

n/a stands for not applicable.

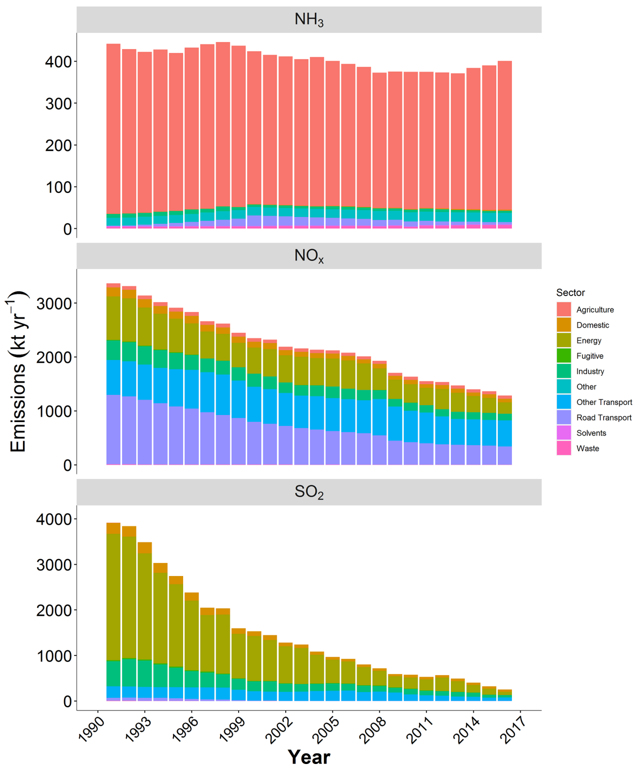


Figure 4. Emissions (in kt) of ammonia (NH_3), nitrogen oxides (NO_x) and sulfur dioxide (SO_2) in the model domain, covering the UK and Ireland, from 1990 to 2017, split into the main broad reporting sectors.

main, emissions of NH_3 and NO_x have decreased by $\sim 12\,\%$ and $\sim 64\,\%$ respectively from 1990 to 2017 (Fig. 4).

Much of the decrease in emissions of NO_x in the UK has been driven by the decline of coal use in power stations (95% decrease in emissions over the time series) and the improvement and modernisation of petrol combustion in road transport (98 % decrease in emissions over the time series). Decreases in NO_x have been offset by increases in emissions from DERV (diesel fuels) and aviation fuels. With regard to NH_3 emissions, which are dominated by agriculture, changes in farm practices have seen a patchwork of decreases and increases to various emissions sources, with a generally decreasing trend that has plateaued from ca. 2001. It is the non-agricultural sources, however, that have shown marked increases from 1990 to 2017, including those activities associated with the circular economy: anaerobic digestion, composting of organic materials, application of sewage sludge to land and the combustion of biomass for industry (total increase: ~ 5 to ~ 26 kt). Finally, SO_2 emissions have reduced by $\sim 94\,\%$ in the same time period (mean of $\sim 5\,\%\,yr^{-1}$), which is a direct result of the decline of coal use, especially in power stations, and restrictions being placed on the sulfur content of various fuels.

As all three pollutants are reactive in the atmosphere, differing rates of emissions reductions have varying effects on chemical reactions and subsequent deposition. Changes to emissions over time vary in space and so does, therefore, N deposition (Fowler et al., 2007).

3.2 Model evaluation

Scatter plots of the modelled predictions vs. measurements in 2017, for data collected in Table 3, are shown in Fig. 5. The associated performance metrics are given in Table 4.

For the latest year included in this study, all six N forms in Table 4 comply with the FAC2 metric and all six comply with the recommended NMSE limit of 1.5. FB and MG are met with less success, though all are close to the recommended thresholds, aside from NH_4 in aerosol (which contributes to dry deposition). FB and MG measure the systematic bias of the model, and for both NH_4 and NO_3, the model slightly under-predicts the aerosol phase and over-predicts the aqueous phase. Not shown in Fig. 5 and Table 4 is the evaluation of HNO_3 in gas, which similarly fulfils recommendations for FAC2 (0.54) and NMSE (0.48), but not for |FB| (0.48) or MG (0.56). Modelled predictions were also evaluated for 2016, with all seven compounds achieving 50 % compliance, with NH_3 in gas, NO_2 in gas and HNO_3 in gas satisfying all four. It is not fully known why 2016 achieves bet-

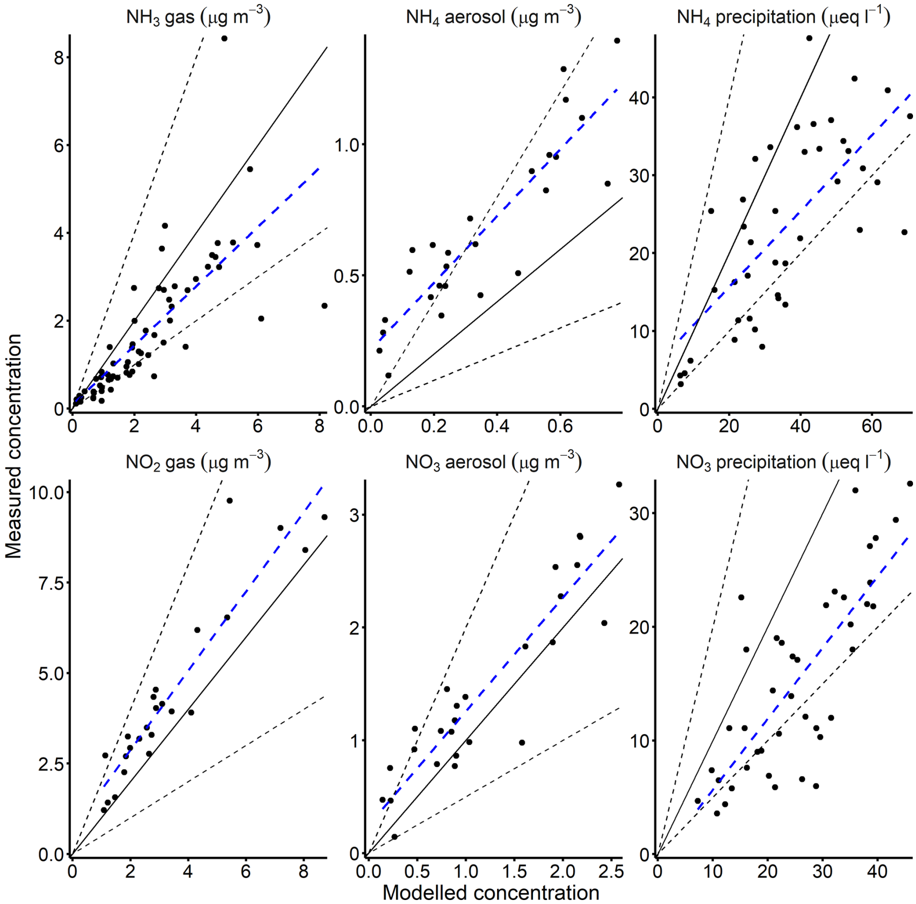


Figure 5. Evaluation of modelled (x axis) and measured (y axis) concentrations of six nitrogen compounds in the UK for 2017 (see Table 3 for definitions). The solid black line represents a 1 : 1 relationship, and the dotted lines represent a factor of 2 (FAC2) relationship; the blue dashed lines are linear regressions.

ter evaluation scores; it may be random variations in real-world conditions, but one reason may be that 2017 was a relatively warm year by annual mean temperature standards (and fourth warmest on record for England only). It is known that NH_3 emissions are affected by temperature (e.g. Hempel et al., 2016; Sutton et al., 2013; Riddick et al., 2018), and, as temperature fluctuations are not factored into the model or into the underlying emission inventories, this may have driven higher spring–summer emissions of NH_3 and therefore higher dry deposition episodes.

For context, Carslaw (2011) undertook a model intercomparison exercise for the UK Department for Environment, Food & Rural Affairs (Defra), with a specific focus on deposition from the CMAQ, EMEP4UK, FRAME, HARM and NAME models. Respectively, those models (at the time) were run at resolutions of 12, 5, 5, 10 and 12 km. In Carslaw (2011), the models performed with a similar correlation coefficient ("r") for all N compounds, aside from NH_4 and NO_3 in precipitation, for which the 2017 model run in this study had a weaker correlation (0.51–0.61 compared to 0.7–0.88).

The evaluation for 2017 would indicate that total wet deposition was over-predicted and total dry deposition was under-predicted. To provide further context and evaluation, measurement data were obtained for three previous years spanning the time series at equal intervals; 1990, 1999 and 2008. Data for historic years, especially prior to $\sim$ 1998, are limited, and so scatter plots in Fig. 6 show the relationship between modelled predictions and measured data for four N compounds while Table 5 shows the associated performance metrics.

All N forms for which data were available in 1990, 1999 and 2008 satisfy at least two of the four evaluation metrics, with four gas and aerosol N compounds fulfilling all metrics in 2008. An example of the benefit of multiple evaluation metrics is shown in Fig. 6 when looking at NO_2 and NH_3 in gas in 2008. Both have very low FB values (indicating very low mean bias) due to the cancelling effect around the 1 : 1 line, but the scatter of predictions to measurements of NH_3 is clearly much larger than for NO_2. Information of the NMSE and the FAC2, plus visual inspection of the plots, helps to illustrate that NH_3 has a larger error than NO_2.

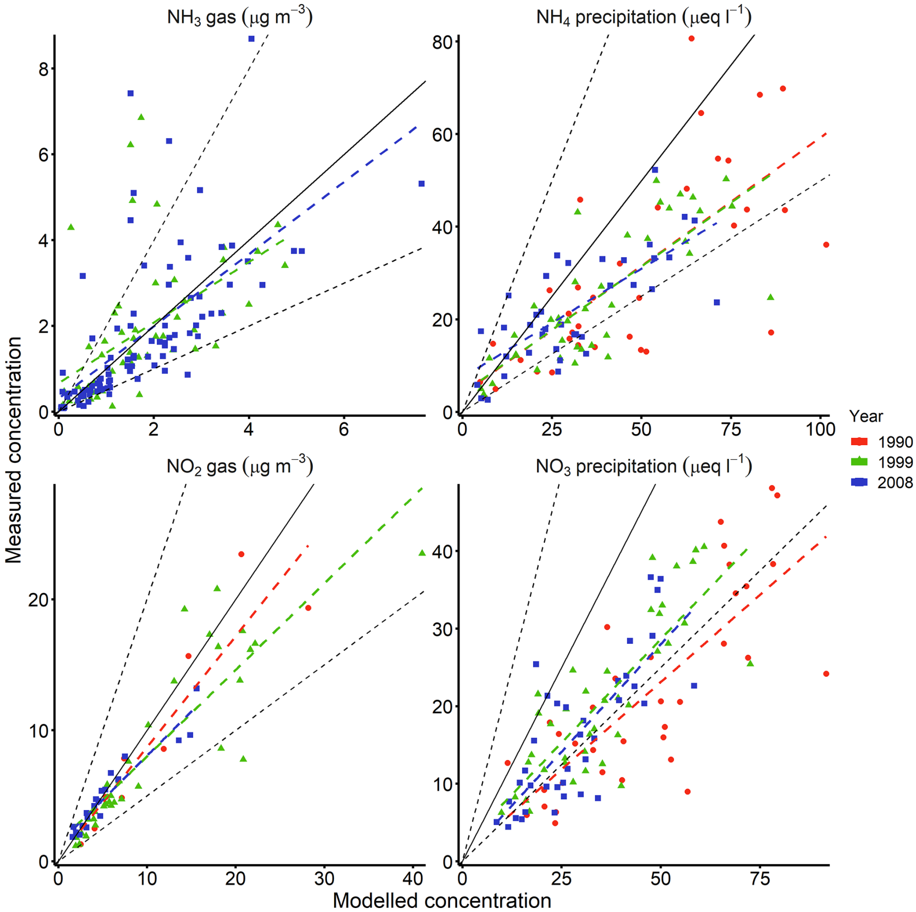


Figure 6. Evaluation of modelled (x axis) and measured (y axis) concentrations of four nitrogen compounds in the UK for 1990, 1999 and 2008 (see Table 3 for definitions; no NH_3 gas data exist for 1990). The solid black line represents a 1 : 1 relationship, and the dotted lines represent a factor of 2 (FAC2) relationship. The blue, green and red dashed lines are linear regressions.

From the perspective of model sensitivity and/or uncertainty, there were no further runs made with adjusted emissions inputs or adjusted deposition parameters within this study. However, Aleksankina et al. (2018) employed statistical techniques to obtain uncertainty estimates of the FRAME model, representing model runs with a ±40 % variation range for the UK emissions of SO_2, NO_x and NH_3. They found that the sensitivity of concentrations of primary precursors NO_x and NH_3, plus the deposition of N, was dominated by emissions. However, concentrations of secondary species such as particulate NO_3^- and NH_4^+ were more geographically dependent.

3.3 Nitrogen deposition

Grid average N deposition – NH_x wet and dry, NO_y wet and dry – is plotted in Fig. 7 at a 1 km × 1 km resolution over the UK terrestrial surface, for 2017. The total N deposition over the UK is 278.3 kt N ($\bar{x} = 10.7\ kg\ N\ ha^{-1}\ yr^{-1}$, $SD = 4.5\ kg\ N\ ha^{-1}\ yr^{-1}$), with a maximum of $74.3\ kg\ N\ ha^{-1}\ yr^{-1}$. Such high deposition values are reasonably rare (no. of cells $> 30\ kg\ N\ ha^{-1}\ yr^{-1} = 118$; no. of cells $> 50\ kg\ N\ ha^{-1}\ yr^{-1} = 8$) and are a direct result of the increased resolution of the model, when compared to the maximum deposition of 5 km × 5 km resolution N deposition.

The two wet deposition surfaces in Fig. 7 exhibit smoother spatial distributions and less heterogeneity (compared to dry deposition), a reflection of the precipitation surface across the UK, and they constitute ∼ 67 % of the total deposition. Wet deposition is nearly always of a longer-range than dry deposition, due to the transport in more elevated atmospheric layers, but some enhanced local washout around strong sources is also represented. This longer range transport acts as a smoothing effect on the deposition field due to the increased distance from the emission source. It should be noted that, as shown in Figs. 5 and 6, deposition in precipitation of both NH_4 and NO_3 is consistently overpredicted by the model throughout the time series. Upland areas are subject to the highest values of wet deposition, and most of the highest value cells between 25 and 50 kg total $N\ ha^{-1}\ yr^{-1}$ are dominated by wet deposition. Dry deposition of NO_y,

Table 5. Evaluation metrics of modelled concentrations of six nitrogen compounds in the UK for 1990, 1999 and 2008 (see Table 3 for definitions). Dashed lines represent no available data. Bold numbers represent where that metric has been satisfied (see Sect. 2.4.2 for metric definitions).

Metric	Acceptability	NH_3 (conc. in gas)	NH_4 (conc. in aerosol)	NH_4 (conc. in precip.)	NO_2 (conc. in gas)	NO_3 (conc. in aerosol)	NO_3 (conc. in precip.)
(a) 1990							
Points (n)				35	9	35	
R^2		–	–	0.51	0.85	0.60	–
FAC2	> 0.5	–	–	**0.69**	**1.00**	**0.40**	–
|FB|	< 0.3	–	–	0.44	**0.14**	0.73	–
NMSE	< 1.5	–	–	**0.45**	**0.11**	**0.81**	–
MG	> 0.7 & < 1.3	–	–	0.61	**0.80**	0.44	–
(b) 1999							
Points (n)		55	50	39	33		39
R^2		0.29	0.66	0.63	0.77	–	0.66
FAC2	> 0.5	**0.78**	**0.92**	**0.77**	**0.94**	–	**0.72**
|FB|	< 0.3	**0.11**	**0.23**	0.42	**0.23**	–	0.52
NMSE	< 1.5	**0.65**	**0.20**	**0.35**	**0.25**	–	**0.40**
MG	> 0.7 & < 1.3	**1.03**	**0.88**	0.66	**0.78**	–	0.58
(c) 2008							
Points (n)		90	42	37	20	28	37
R^2		0.44	0.88	0.55	0.91	0.91	0.61
FAC2	> 0.5	**0.82**	**0.88**	**0.81**	**1.00**	**0.93**	**0.57**
|FB|	< 0.3	**0.02**	**0.07**	0.34	**0.09**	**0.29**	0.56
NMSE	< 1.5	**0.54**	**0.08**	**0.33**	**0.10**	**0.20**	**0.47**
MG	> 0.7 & < 1.3	**0.94**	**1.11**	**0.74**	**0.98**	**0.82**	0.53

as modelled in this study, is the smallest contributor to total N deposition (~ 14 %) and is dominated by NO_2 and HNO_3, which both follow their respective concentration fields closely (RoTAP, 2012). Dry deposition of NO_2, therefore, is largest in urban areas and close to road networks such as motorways. Dry deposition of NH_x, ~ 20 % of total N deposition, is a highly heterogeneous surface with the highest values associated with areas of intensive livestock farming (including beef, dairy, pigs and poultry). Gaseous NH_3 has a short atmospheric lifetime and so is deposited close to the sources. The very highest values of total N deposition (> 50 $kg\,N\,ha^{-1}\,yr^{-1}$) are all dominated by dry deposition of NH_x and are located near high agricultural emissions. An important factor in the deposition of NH_x is the presence of oxidised SO_2, sulfuric acid (H_2SO_4), to form the aerosol $(NH_4)_2SO_4$. With decreasing SO_2 available to create H_2SO_4, more NH_3 is deposited within short distances as dry deposition. This effect is further enhanced by the increased rate of dry deposition of the available SO_2, a result of the increase in the concentration ratio of $NH_3 : SO_2$, which increases surface water pH, which further limits the available SO_2 to oxidise to H_2SO_4 (Baek and Aneja, 2004; Fowler et al., 2007; RoTAP, 2012; Tan et al., 2020).

Looking at the pattern of modelled N deposition from 1990 to 2017, Fig. 8 shows a steady decrease in wet and dry NO_y deposition, a slow decrease in wet NH_x deposition, and no apparent decrease in dry NH_x deposition. The latter is due to the change in atmospheric chemistry with declining sulfur emissions due to successful policy implementation. Total N deposition over the UK has decreased from 465 to 278 kt N, though no significant reductions in the total have occurred since around 2011.

Total oxidised N deposition has decreased by ~ 56 % from 1990 to 2017, while reduced N deposition has decreased by ~ 19 %. This reflects the larger emissions reductions achieved for NO_x than for NH_3 from 1990. Mean deposition values for all four N forms have changed in a similar fashion to their respective totals from 1990, but the standard deviation across all 5 km × 5 km cells for oxidised N (both wet and dry) has decreased over time, possibly due to the heavy reductions in emissions sources such as road traffic and power stations, which previously created very high localised dry deposition. Figure 9 shows every year of total N deposition from 1990 to 2017 and highlights the non-linear relationship between decreasing emissions and deposition.

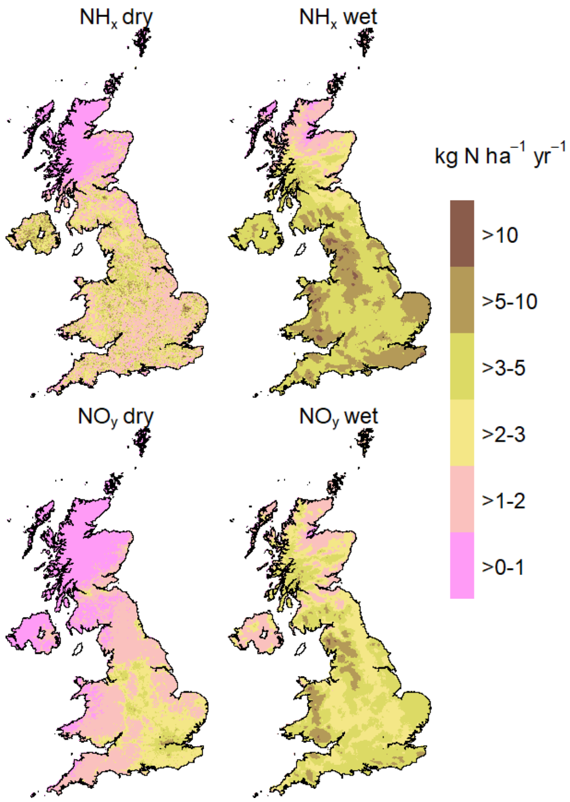


Figure 7. Four forms of nitrogen (N) deposition over the UK terrestrial surface in 2017 at 1 km × 1 km resolution, for grid average land cover: wet and dry deposition of reduced N (NH_x) and wet and dry deposition of oxidised N (NO_y) (kg N ha^{-1} yr^{-1}).

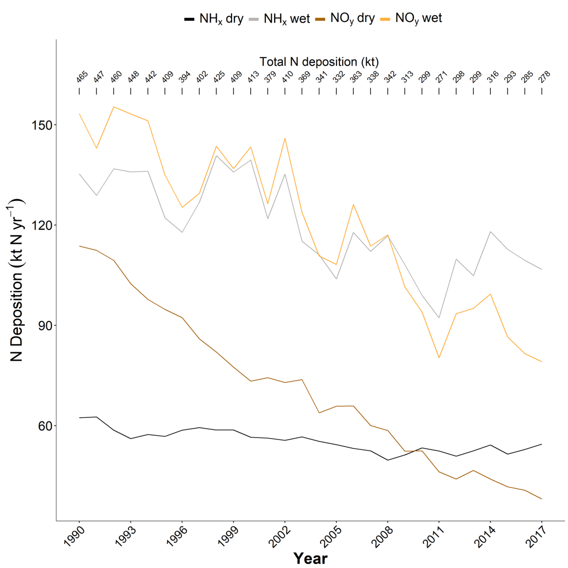


Figure 8. Four forms of total nitrogen (N) deposition over the UK terrestrial surface from 1990 to 2017, for grid average land cover: total wet and dry deposition of reduced N (NH_x) and wet and dry deposition of oxidised N (NO_y) (kt N yr^{-1}).

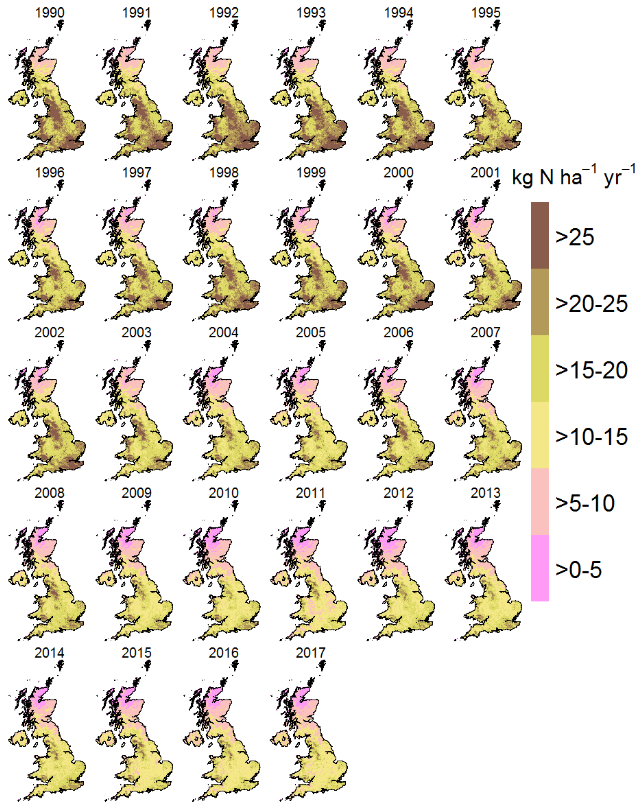


Figure 9. Spatial distribution of total nitrogen (N) deposition over the UK terrestrial surface, 1 km × 1 km resolution, from 1990 to 2017, for grid average land cover (kt N yr^{-1}).

Some of the areas with the highest N deposition in later years are remote upland areas, which are principally affected by longer-range wet deposition (and transboundary deposition) and have seen much lower relative decreases in N deposition than lowland areas such as southeast England. NO_x emissions have decreased by ∼ 64 % across the time series and resulted in wet and dry NO_y deposition decreases of ∼ 48 % and ∼ 66 %, respectively. This illustrates the non-linear processes involved with the chemical processing of NO_x emissions, in particular the resulting concentrations of NO_3 in precipitation which are not decreasing at the same rate as gas and/or aerosol forms of oxidised N (see Fowler et al., 2007; Sickles and Shadwick, 2015; Feng et al., 2020). It must be recognised again, however, that the model overestimates wet deposition of N to a degree.

As a result of emissions changes and non-linear chemistry, estimates of modelled dry deposition have decreased as a percentage of the total N deposition (1990 =∼ 38 %, 2017 =∼ 33 %) (see Fig. 10). This dataset models wet deposition as the dominant source of total N deposition.

As a result of the large decreases in NO_x emissions, and fewer regulations on most NH_3 emission sources in the UK compared to NO_x, reduced N is now the major component of N deposition. In this dataset, the proportion of dry deposition

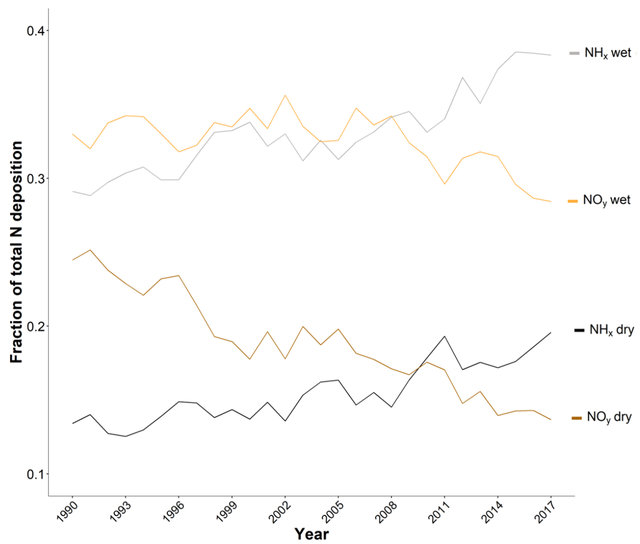


Figure 10. Fraction of the total nitrogen (N) deposition over the UK terrestrial surface for four forms of nitrogen (N) deposition, for grid average land cover, from 1990 to 2017: total wet and dry deposition of reduced N (NH_x) and wet and dry deposition of oxidised N (NO_y).

has moved from being dominated by oxidised N in 1990 (∼ 65 %) to reduced N in 2017 (∼ 59 %). This has resulted in a highly heterogeneous spatial distribution of N deposition that is more reflective of both agricultural practice and rainfall patterns.

4 Data availability

The deposition data described in this paper are made available via the NERC Environmental Information Data Centre at https://doi.org/10.5285/9b203324-6b37-4e91-b028-e073b197fb9f (Tomlinson et al., 2020).

5 Conclusions

This new dataset provides a consistent time series of modelled wet and dry deposition of both reduced and oxidised N (plus total N) for the whole UK terrestrial surface on a 1 km × 1 km resolution (no. of cells = 259 436), from 1990 to 2017. Atmospheric modelling was undertaken for all 28 years, and there is good agreement between modelled predictions and measured observations of various compounds of N not only for 2016 and 2017 but also for selected prior years when tests were carried out (1990, 1999 and 2008). It is estimated within this dataset that N deposition has undergone large decreases across the time period, from 465 to 278 kt N, but that a cessation in the decrease in NH_3 emissions (plus vast reductions in SO_2 emissions) has seen reduced N become the dominant fraction of all N deposition. Higher-resolution data enable more detailed effect studies across a wide range of disciplines, as well as cumulative effects from the annual time series. Further work should be aimed at improving the long-term spatial distribution of emissions.

Author contributions. SJT designed and coded the methodology to combine all data sources into compatible input data, quality assurance and quality control work, compiled a time series of rainfall data, reformatted FRAME Europe outputs to FRAME Europe inputs and analysed all model outputs, including historic model performance. EJC performed all of the agricultural emissions mapping for the time series, updated the FRAME UK land use files and undertook QAQC work. AJD undertook all atmospheric modelling requirements, principally the model runs and most recent evaluations. MV provided expert knowledge and advice with regard to atmospheric chemistry and modelling. UD managed the atmospheric modelling task and offered expert advice on the spatial distribution of emissions and N deposition. SJT prepared the manuscript with contributions from all co-authors. All co-authors commented on the draft manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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References

Aggenbach, C. J. S., Kooijman, A. M., Fujita, Y., van der Hagen, H., van Til, M., Cooper, D., and Jones, L.: Does atmospheric nitrogen deposition lead to greater nitrogen and carbon accumulation in coastal sand dunes?, Biol. Conserv., 212, 416–422, https://doi.org/10.1016/j.biocon.2016.12.007, 2017.

Aleksankina, K., Heal, M. R., Dore, A. J., Van Oijen, M., and Reis, S.: Global sensitivity and uncertainty analysis of an atmospheric chemistry transport model: the FRAME model (version 9.15.0) as a case study, Geosci. Model Dev., 11, 1653–1664, https://doi.org/10.5194/gmd-11-1653-2018, 2018.

Baek, B. H. and Aneja, V. P.: Measurement and analysis of the relationship between ammonia, acid gases, and fine particles in eastern North Carolina, Air Waste Manage. Assoc., 54, 623–633, https://doi.org/10.1080/10473289.2004.10470933, 2004.

Britton, A. J., Gibbs, S., Fisher, J. M., and Helliwell, R. C.: Impacts of nitrogen deposition on carbon and nitrogen cycling in alpine Racomitrium heath in the UK and prospects for recovery, Environ. Pollut., 254, 112986, https://doi.org/10.1016/j.envpol.2019.112986, 2019.

Brown, P., Broomfield, M., Cardenas, L., Choudrie, S., Jones, L., Karagianni, E., Passant, N., Thistlethwaite, G., Thomson, A., Turtle, L., and Wakeling, D.: UK Greenhouse Gas Inventory, 1990 to 2017: Annual Report for submission under the Framework Convention on Climate Change, available at: https://unfccc.int/sites/default/files/resource/gbr-2019-nir-15apr19.zip (last access: 1 November 2020), 2019.

Carnell, E. J., Thomas, I. N., Tomlinson, S. J., Leaver, D., and Dragosits, U.: The spatial distribution of ammonia, methane and nitrous oxide emissions from agriculture in the UK 2017, Contribution to the UK National Atmospheric Emission Inventory and Greenhouse Gas Inventory, Annual Report on Defra Project SCF0107, CEH Report, 13 pp., available at: http://nora.nerc.ac.uk/id/eprint/529856/ (last access: 15 October 2020), 2019a.

Carnell, E. J., Vieno, M., Vardoulakis, S., Beck, R., Heaviside, C., Tomlinson, S., Dragosits, U., Heal, M. R., and Reis, S.: Modelling public health improvements as a result of air pollution control policies in the UK over four decades – 1970 to 2010, Environ. Res. Lett., 14, 074001, https://doi.org/10.1088/1748-9326/ab1542, 2019b.

Carslaw, D.: Defra deposition model evaluation analysis – Phase 1, available at: https://uk-air.defra.gov.uk/assets/documents/reports/cat20/1105091512_DepsotionFinal.pdf (last access: 15 January 2020), 2011.

Chang, J. C. and Hanna, S. R.: Air quality model performance evaluation, Meteorol. Atmos. Phys., 87, 167–196, https://doi.org/10.1007/s00703-003-0070-7, 2004.

Dore, A. J., Kryza, M., Hall, J. R., Hallsworth, S., Keller, V. J. D., Vieno, M., and Sutton, M. A.: The influence of model grid resolution on estimation of national scale nitrogen deposition and exceedance of critical loads, Biogeosciences, 9, 1597–1609, https://doi.org/10.5194/bg-9-1597-2012, 2012.

Dore, A., Reis, S., Oxley, T., ApSimon, H., Hall, J., Vieno, M., Kryza, M., Green, C., Tsagatakis, I., Tang, S., Braban, C., and Sutton, M.: Calculation of Source-Receptor Matrices for Use in an Integrated Assessment Model and Assessment of Impacts on Natural Ecosystems, in: Air Pollution Modeling and Its Application Xxiv, edited by: Steyn, D. G. and Chaumerliac, N., Springer, Cham, 107–112, 2016.

Dore, A. J., Vieno, M., Fournier, N., Weston, K. J., and Sutton, M. A.: Development of a new wind-rose for the British Isles using radiosonde data, and application to an atmospheric transport model, Q. J. Roy. Meteorol. Soc., 132, 2769–2784, 2006.

Dragosits, U., Sutton, M. A., Place, C. J., and Bayley, A. A.: Modelling the spatial distribution of agricultural ammonia emissions in the UK, Environ, Pollut,, 102, 195–203, https://doi.org/10.1016/S0269-7491(98)80033-X, 1998.

EEA – European Environment Agency: EMEP/EEA air pollutant emission inventory guidebook 2019 – Technical guidance to prepare national emission inventories, EEA Report No. 13/2019, Luxembourg, 2019.

EMEP – European Monitoring and Evaluation Programme: The Emissions Database, available at: https://www.ceip.at/webdab-emission-database (last access: 20 August 2019), 2019.

E-PRTR – European Pollutant Release and Transfer Register: Welcome to the Industrial emissions portal, available at: https://industry.eea.europa.eu/, last access: 20 August 2019.

Feng, J., Chan, E., and Vet, R.: Air quality in the eastern United States and Eastern Canada for 1990–2015: 25 years of change in response to emission reductions of SO_2 and NO_x in the region, Atmos. Chem. Phys., 20, 3107–3134, https://doi.org/10.5194/acp-20-3107-2020, 2020.

Fowler, D., O'Donoghue, M., Muller, J. B. A., Smith, R. I., Dragosits, U., Skiba, U., Sutton, M. A., and Brimblecombe, P.: A Chronology of Nitrogen Deposition in the UK Between 1900 and 2000, Water Air Soil Pollut., 4, 9–23, https://doi.org/10.1007/s11267-004-3009-1, 2004.

Fowler, D., Smith, R. I., Muller, J. B. A., Cape, J. N., Sutton, M. A., Erisman, J. W., and Fagerli, H.: Long Term Trends in Sulphur and Nitrogen Deposition in Europe and the Cause of Non-linearities, Water Air Soil Pollut., 7, 41–47, https://doi.org/10.1007/s11267-006-9102-x, 2007.

Greenwood, N., Devlin, M. J., Best, M., Fronkova, L., Graves, C. A., Milligan, A., Barry, J., and van Leeuwen, S. M.: Utilizing Eutrophication Assessment Directives From Transitional to Marine Systems in the Thames Estuary and Liverpool Bay, UK, Front. Mar. Sci., 6, 116, https://doi.org/10.3389/fmars.2019.00116, 2019.

Grennfelt, P. and Hov, Ø.: Regional air pollution at a turning point, Ambio, 34, 2–10, https://doi.org/10.1579/0044-7447-34.1.2, 2005.

Hallsworth, S., Dore, A. J., Bealey, W. J., Dragosits, U., Vieno, M., Hellsten, S., Tang, Y. S., and Sutton, M. A.: The role of indicator choice in quantifying the threat of atmospheric ammonia to the 'Natura 2000' network, Environ. Sci. Policy, 13, 671–687, https://doi.org/10.1016/j.envsci.2010.09.010, 2010.

Hanna, S. R. and Chang, J.: Setting Acceptance Criteria for Air Quality Models, Air Pollut. Model. Appl., 5, 479–484, 2012.

Hellsten, S., Dragosits, U., Place, C. J., Vieno, M., and Sutton, M. A.: Modelling and assessing the spatial distribution of ammonia emissions in the UK. Environ. Pollut., 154, 370–379, https://doi.org/10.1016/j.envpol.2008.02.017, 2008.

Hempel, S., Saha, C. K., Fiedler, M., Berg, W., Hansen, C., Amon, B., and Amon, T.: Non-linear temperature dependency of ammonia and methane emissions from a naturally ventilated dairy barn, Biosyst. Eng., 145, 10–21, https://doi.org/10.1016/j.biosystemseng.2016.02.006, 2016.

Levy, P. E., Martin Hernandez, C., Smith, R. I., Dore, A. J., Tang, Y. S., and Stedman, J. R.: Sulphur and nitrogen atmospheric Concentration Based Estimated Deposition (CBED) data for the UK 2016–2018, NERC Environmental Information Data Centre, https://doi.org/10.5285/5999d471-fe1d-45fa-889d-3156edb785a7, 2020.

MapEire: National mapping of GHG and non-GHG emissions sources, available at: https://projects.au.dk/mapeire/ (last access: 1 November 2020), 2019.

NAEI – National Atmospheric Emissions Inventory: Data, available at: https://naei.beis.gov.uk/data/ (last access: 1 November 2020), 2019.

Nowak, D., Jovan, S., Branquinho, C., Augusto, S., Ribeiro, M. C., and Kretsch, C. E.: Chapter 4: Biodiversity, air quality and human health, in: Connecting Global Priorities – Biodiversity and Human Health: A State of Knowledge Review, edited by: Romanelli, C., Cooper, D., Campbell-Lendrum, D., Maiero, M., Karesh, W. B., Hunter, D., and Golden, C. D., World Health Organization and Secretariat of the Convention on Biological Diversity, WHO Press, Geneva, 63–74, 2015.

Nowak, D. J., Hirabayashi, S., Doyle, M., McGovern, M., and Pasher, J.: Air pollution removal by urban forests in Canada and its effect on air quality and human health, Urban Forest. Urban Green., 29, 40–48, https://doi.org/10.1016/j.ufug.2017.10.019, 2018.

Payne, R. J., Dise, N. B., Field, C., Dore, A., Caporn, S. J. M., and Stevens, C. J.: Nitrogen deposition and plant biodiversity: past, present, and future, Front. Ecol. Environ., 15, 431–436, https://doi.org/10.1002/fee.1528, 2017.

Payne, R. J., Campbell, C., Britton, A. J., Mitchell, R. J., Pakeman, R. J., Jones, L., Ross, L. C., Stevens, C. J., Field, C., Caporn, S. J. M., Carroll, J., Edmondson, J. L., Carnell, E. J., Tomlinson, S. J., Dore, A. J., Dise, N. B., and Dragosits, U.: What is the most ecologically-meaningful metric of nitrogen deposition?, Environ. Pollut., 247, 319–331, https://doi.org/10.1016/j.envpol.2019.01.059, 2019.

Payne, R. J., Campbell, C., Stevens, C. J., Pakeman, R. J., Ross, L. C., Britton, A. J., Mitchell, R. J., Jones, L., Field, C., Caporn, S. J. M., Carroll, J., Edmondson, J. L., Carnell, E. J., Tomlinson, S. J., Dore, A., Dragosits, U., and Dise, N. B.: Disparities between plant community responses to nitrogen deposition and critical loads in UK semi-natural habitats, Atmos. Environ., 239, 117478, https://doi.org/10.1016/j.atmosenv.2020.117478, 2020.

Plejdrup, M. S., Nielsen, O.-K., and Bruun, H. G.: Spatial high-resolution mapping of national emissions, WIT Trans. Ecol. Environ., 230, 399–408, https://doi.org/10.2495/AIR180371, 2018.

Richmond, B., Misra, A., Broomfield, M., Brown, P., Karagianni, E., Murrells, T. P., Pang, Y., Passant, N. R., Pearson, B., Stewart, R., Thistlethwaite, G., Wakeling, D., Walker, C., Wiltshire, J., Hobson, M., Gibbs, M., Misselbrook, T., Dragosits, U., and Tomlinson, S.: UK Informative Inventory Report (1990 to 2017), available at: https://uk-air.defra.gov.uk/library/reports?report_id=978 (last access: 15 December 2020), 2019.

Riddick, S. N., Dragosits, U., Blackall, T. D., Tomlinson, S. J., Daunt, F., Wanless, S., Hallsworth, S., Braban, C. F., Tang, Y. S., and Sutton, M. A.: Global assessment of the effect of climate change on ammonia emissions from seabirds, Atmos. Environ., 184, 212–223, https://doi.org/10.1016/j.atmosenv.2018.04.038, 2018.

RoTAP – Review of Transboundary Air Pollution: Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK, Defra Contract Number AQ0703, UK Centre for Ecology & Hydrology, available at: https://uk-air.defra.gov.uk/library/reports?report_id=701 (last access: 10 April 2020), 2012.

Rowland, C. S., Morton, R. D., Carrasco, L., McShane, G., O'Neil, A. W., and Wood, C. M.: Land Cover Map 2015 (1 km dominant aggregate class, GB), NERC Environmental Information Data Centre, https://doi.org/10.5285/711c8dc1-0f4e-42ad-a703-8b5d19c92247, 2017.

Sickles II, J. E. and Shadwick, D. S.: Air quality and atmospheric deposition in the eastern US: 20 years of change, Atmos. Chem. Phys., 15, 173–197, https://doi.org/10.5194/acp-15-173-2015, 2015.

Singles, R., Sutton, M. A., and Weston, K. J.: A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain, Atmos. Environ., 32, 393–399, https://doi.org/10.1016/S1352-2310(97)83467-X, 1998.

Skamarock, W. C., Klemp, J. B., Dudhia, J., Gill, D. O., Liu, Z., Berner, J., Wang, W., Powers, J. G., Duda, M. G., Barker, D. M., and Huang, X.-Y.: A Description of the Advanced Research WRF Version 4, NCAR Tech. Note NCAR/TN-556+STR, National Center forAtmospheric Research, Boulder, Colorado, 145 pp., https://doi.org/10.5065/1dfh-6p97, 2019.

Smart, S. M., Stevens, C. J., Tomlinson, S. J., Maskell, L. C., and Henrys, P. A.: Comment on Pescott & Jitlal 2020: Failure to account for measurement error undermines their conclusion of a weak impact of nitrogen deposition on plant species richness, Peer J., 9, e10632, https://doi.org/10.7717/peerj.10632, 2020.

Stevens, C. J., David, T. I., and Storkey, J.: Atmospheric nitrogen deposition in terrestrial ecosystems: Its impact on plant communities and consequences across trophic levels, Funct. Ecol., 32, 1757–1769, https://doi.org/10.1111/1365-2435.13063, 2018.

Sutton, M. A., Reis, S., Riddick, S. N., Dragosits, U., Nemitz, E., Theobald, M. R., Tang, Y. S., Braban, C. F., Vieno, M., Dore, A. J., Mitchell, R. F., Wanless, S., Daunt, F., Fowler, D., Blackall, T. D., Milford, C., Flechard, C. R., Loubet, B., Massad, R., Cellier, P., Coheur, P. F., Clarisse, L., van Damme, M., Ngadi, Y., Clerbaux, C., Skjøth, C. A., Geels, C., Hertel, O., Wickink Kruit, R. J., Pinder, R. W., Bash, J. O., Walker, J. D., Simpson, D., Horvath, L., Misselbrook, T., Bleeker, A., Dentener, F., and de Vries, W.: Toward a climate-dependent paradigm of ammonia emission & deposition, P. Roy. Soc. B, 368, 1621, https://doi.org/10.1098/rstb.2013.0166, 2013.

Tan, J., Fu, J. S., and Seinfeld, J. H.: Ammonia emission abatement does not fully control reduced forms of nitrogen deposition, P. Natl. Acad. Sci. USA, 117, 9771–9775, https://doi.org/10.1073/pnas.1920068117, 2020.

Tanguy, M., Dixon, H., Prosdocimi, I., Morris, D. G., and Keller, V. D. J.: Gridded estimates of daily and monthly areal rainfall for the United Kingdom (1890–2017) [CEH-GEAR], NERC Environmental Information Data Centre, https://doi.org/10.5285/ee9ab43d-a4fe-4e73-afd5-cd4fc4c82556, 2019.

Theobald, M. R., Simpson, D., and Vieno, M.: A sub-grid model for improving the spatial resolution of air quality modelling at a European scale, Geosci. Model Dev., 9, 4475–4489, doi10.5194/gmd-9-4475-2016, 2016.

Tomlinson, S. J., Carnell, E. J., Dore, A. J., and Dragosits, U.: Nitrogen deposition in the UK at 1 km resolution, 1990–2017, NERC Environmental Information Data Centre, https://doi.org/10.5285/9b203324-6b37-4e91-b028-e073b197fb9f, 2020.

UK AIR – Air Information Resource: United Kingdom Eutrophying & Acidifying Network (UKEAP), available at: https://uk-air.defra.gov.uk/networks/network-info?view=ukeap (last access: 2 February 2021), 2020.

Vieno, M., Dore, A. J., Bealey, W. J., Stevenson, D. S., and Sutton, M. A.: The importance of source configuration in quantifying footprints of regional atmospheric sulphur deposition, Sci. Total Environ., 408, 985–995, https://doi.org/10.1016/j.scitotenv.2009.10.048, 2010.

Walsh, S.: A Summary of Climate Averages 1981–2010 for Ireland, Climatological Note No. 14, Met Éireann, Dublin, 2012.