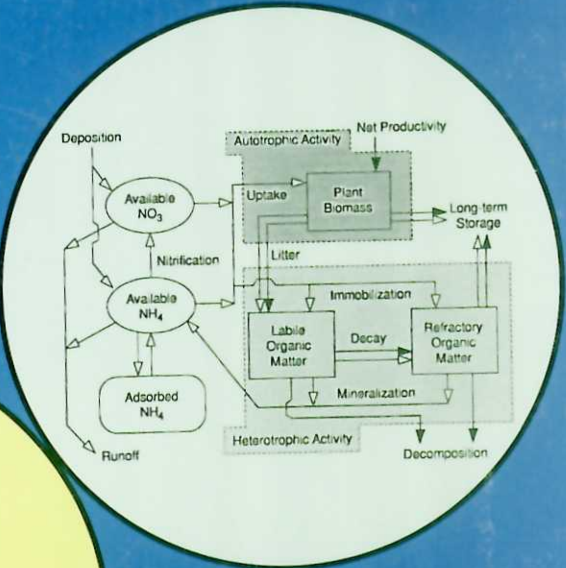
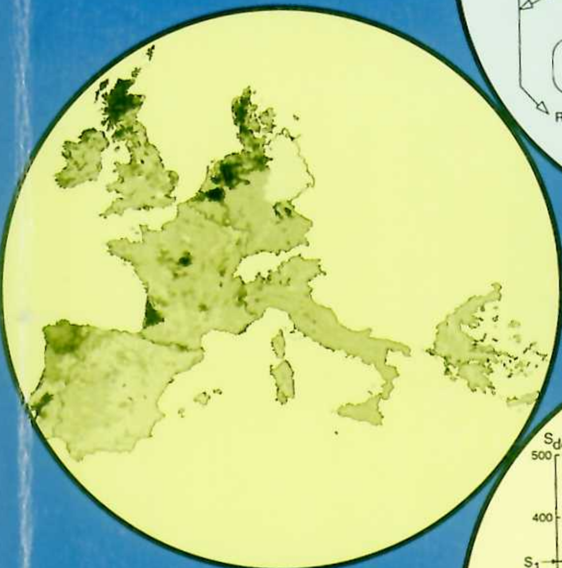
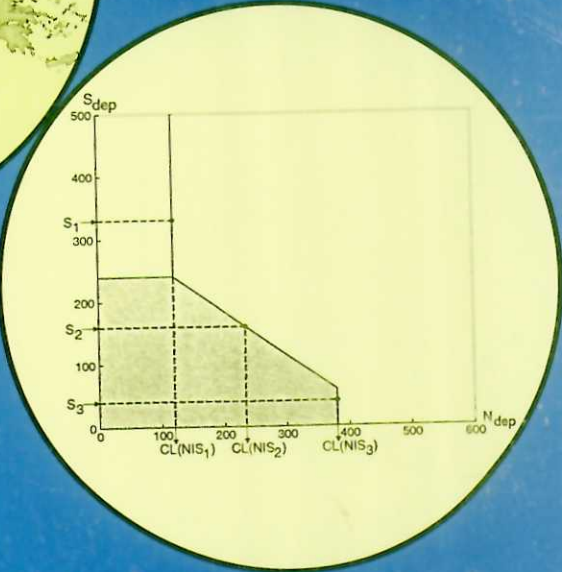
Mapping and Modelling of Critical Loads for Nitrogen : a Workshop Report







Proceedings of the
Grange-Over-Sands Workshop
24-26 October 1994

Mapping and modelling of critical loads for nitrogen - a workshop report.

Report of a workshop held at Grange-over-Sands, Cumbria, UK under the auspices of the UN-ECE Convention on Long Range Transboundary Air Pollution, Working Group for Effects, 24-26 October 1994.

Edited by: M. Hornung, M.A. Sutton and R.B. Wilson

Organized by the UK Department of Environment and hosted by the Institute of Terrestrial Ecology (Merlewood).

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FOREWORD

At its Eleventh Session, the Executive Body for the United Nations Economic Commission for Europe Convention on Long Range Transboundary Air Pollution agreed that after the conclusion in 1994 of the new Protocol on Further Reduction of Sulphur Emissions, highest priority should be given to the development of a strategy for the second stage of the Protocol concerning the Control of Nitrogen Oxides. It was agreed that a similar critical loads approach to that adopted for the Sulphur Protocol should be used. However, the quantification of critical loads and levels depends on the current scientific understanding of the impact of air pollutants on the environment and of necessity needs continuous review and updating.

The last review of nitrogen critical loads was held at Lökeberg, Sweden, 6-10 April 1992 and published later that year. Since then, however, significant advances in our scientific understanding of the impacts of nitrogen oxides and especially ammonia compounds have been made and these have allowed the development of better methods of mapping and modelling nitrogen critical loads and levels.

It was felt by the UNECE Working Group on Effects under the Convention that these advances should be reviewed and that the Lökeberg Report should be amended and updated where appropriate.The United Kingdom Department of Environment agreed to sponsor a workshop to undertake this task and this was held at Grange over Sands, Cumbria on 24-26 October, 1994. The main objectives were to

(1) update the Lökeberg Report

(2) produce a revised chapter on Nitrogen Critical Loads and Levels for theTask Force on Map ping (TFM) Manual on Methodologies and Criteria for Mapping Critical Loads/Levels

(3) provide an updated scientific basis for the mapping and modelling of nitrogen critical loads and levels for use in discussions on the Second Protocol on Nitrogen Oxides.

Three main working groups were set up to review the situation for terrestrial and freshwater ecosystems using empirical and mass balance approaches. Five discussion groups also reported on sections in the Mapping Manual relating to deposition, sea salt, dynamic modelling, marine ecosystems and mapping issues. The outputs from all these groups have been used to prepare amendments or update the Mapping Manual.

These working group reports, the background papers and information from 19 posters presented at the Workshop are all included in this report as a record of the scientific basis used for discussion and the decisions made. A number of recommendations made at the Workshop are also recorded.

The organisers are grateful to all participants for their valuable contributions to this Workshop, especially the presenters of the key note papers reviewing current scientific understanding and those participants who prepared and displayed posters. Particular thanks are due to the organizing secretariat, Kay Prior, Julie Delve, Marjorie Ferguson and Carole Pitcairn.

Professor M. Hornung, Dr M. Sutton, Mr R. Wilson,
March 1995

CHAPTER 1
INTRODUCTION

The concept of critical loads and levels is now widely accepted as a basis for developing optimised air pollutant abatement strategies within the UNECE Convention on Long Range Transboundary Air Pollution. It was first used in the negotiation of the Protocol on Further Reduction of Sulphur Emissions adopted in June 1994 and many workshops were held from 1988 onwards to produce information for this Protocol in such areas as:

(a) Inventories of current emissions and projections of future emission rates

(b) Estimates of the potential for, and costs of emission reductions,including structural changes and conservation of energy and natural resources

(c) Long range transport and deposition models

(d) Critical loads maps

(e) Integrated assessment models

One consequence of the requirements arising under (d) was the setting up of a Task Force on Mapping (TFM) to produce a Manual on Metholodologies and Criteria for Mapping Critical Loads/Levels. The requirements of the TFM have given rise to the setting up of a number of workshops and technical meetings to periodically review and update the manual in the light of current scientific understanding of air pollutants and their effects. Two important workshops were held in Skokloster, Sweden in 1988 (Nilsson and Grennfelt 1988) and Lökeberg, Sweden in 1992 (Grennfelt and Thörnelöf 1992) and the proceedings from both these were published, incorporated into the Mapping Manual (TFM 1993) and used to provide the scientific basis for the production of critical loads and critical loads exceedence maps for the Further Sulphur Protocol.

It was recognised in the Skokloster report that the nitrogen sections were weaker than those for sulphur and acidification. For this reason and in anticipation of the requirements of the Second Nitrogen Oxides Protocol, the Lökeberg workshop concentrated solely on nitrogen critical loads. In 1992, however, it was apparent that there were still important gaps in knowledge in such areas as the deposition and impacts of ammonium, the development of the mass balance approach, total nitrogen deposition and the mapping and modelling of nitrogen critical loads. These gaps were identified as needing further attention by the UNECE Working Group On Effects at its Eleventh Session in 1992 and it recommended that a further workshop be held to update the Mapping Manual on nitrogen critical loads and levels prior to the start of discussions on the Second Nitrogen Protocol. The United Kingdom offered to sponsor this workshop which was held at Grange Over Sands, Cumbria on the 24-26 October, 1994.

The main objective of the workshop was to update and revise the Lökeberg Report where appropriate, using the existing definitions of critical loads and levels as defined in the previous reports. These revisions were then submitted to the Task Force On Mapping for inclusion in the Mapping Manual.

This report is a record of the proceedings of the Grange-over-Sands meeting and includes the key scientific papers on which discussion was based and working group reports produced at the meeting.

In view of the large amount of new information made available at the Workshop, a thematic approach has been adopted in this report so that the reader can more easily refer to the basic science underpinning the conclusions reached. Six main Nitrogen related themes are presented. These are in the areas of the empirical terrestrial approach, freshwaters, the mass balance approach, dynamic modelling, atmospheric deposition and eutrophication. A seventh theme relating to critical loads of total acidity, including sea salt derived ions, is also included to reflect the conclusions of a discussion group convened at the meeting. Chapters which do not fall naturally into these five classifications are presented separately but cross-referenced where appropriate.

References

Grennfelt, P. and Thörnelöf, E. (Eds.) (1992) *Critical loads for nitrogen.* (Eds.) Report of the Lökeberg workshop. Nord (Miljörapport) 41. Nordic Council of Ministers, Copenhagen.

Nilsson, J. & Grennfelt, P. (1988) *Critical loads for sulphur and nitrogen.* (Eds.) Report of the Skokloster workshop. Miljørapport 15. Nordic Council of Ministers, Copenhagen.

TFM (1993) Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded. Texte 25/93. TFM / CCE / UNECE. Task Force on Mapping, Uweltbundesampt (UBA), Bismarckplatz 1, W-1000 Berlin 33. Germany.

For a better understanding of the UNECE Convention on Long-Range Transboundary Air Pollution and the work programme of the subsidiary working groups, the reader is advised to consult the following references:

Ashmore M.R. and Wilson R.B. (1994) *Critical levels of air pollutants for Europe* (Eds.) Report of the Egham workshop. Air Quality Division, Department of the Environment, London.

Downing R.J., Hettelingh J.-P. and de Smet P.A.M. (1993) *Calculation and mapping of critical loads in Europe.* (Eds.) Status report 1993, Coordination Centre for Effects of the Working Group on Effects. RIVM, Bilthoven, The Netherlands. (ISBN 90-6960-047-1).

Fuhrer J. and Achermann B. (1994) *Critical levels for ozone: a UN-ECE workshop report.* Report of the Bern workshop. FAC report 16. Federal Research Station for Agricultural Chemistry and Environmental Hygiene, CH - 3097, Liebefeld-Bern, Switzerland.

UNECE (1994) *Convention on Long-Range Transboundary Air Pollution*, GE.94-31533/ECE94/14. Environment and Human Settlements Division, United Nations Economic Commission for Europe, CH-1211 Geneva.

UNECE (1993) *The State of Transboundary Air Pollution, 1992 update. Report prepared within the framework of the Convention on Long-Range Transboundary Air Pollution.* Air Pollution Studies 9, ECE/EB.AIR/34, United Nations, New York (ISBN 92-1-116574-1).

WGE (1994) *The Working Group on Effects of Major Air Pollutants on Human Health and the Environment. UNECE Convention on Long-Range Transboundary Air Pollution.* Report T-1034, SFT, P.O. Box 8100 Dep. 0032, Oslo (ISBN 82-7234-994-8)

CHAPTER 2
EMPIRICAL NITROGEN CRITICAL LOADS FOR TERRESTRIAL ECOSYSTEMS

Working Group Report

1. Introduction

The critical loads set at the L^keberg meeting in 1992 have been reviewed in the light of new data including that presented by Bobbink and Roelofs (this volume). Critical loads are set on the basis of observed changes in ecosystem structure or function using experimental data, field observations or dynamic ecosystem models (the ëempirical approachí). There is insufficient information to quantify the differential effects of reduced and oxidised N on (semi-) natural ecosystems (except forests, see 2.5), and therefore critical loads have been set for total N deposition. The working definition adopted at L^keberg has been used again:

ì*A quantitative estimate of an exposure to deposition of N as NH_x and/or NO_y below which empirical detectable changes in ecosystem structure and function do not occur according to present knowledge.*î

In most cases the ëdetectable changeí is interpreted as a change in species composition or species dominance, but in some cases a change in ecosystem function such as N accumulation/ mineralisation has been used (e.g. ombrotrophic bogs). Such process changes are viewed as precursors of longer term changes in ecosystem structure.

The working group addressed some general issues and problems associated with the empirical approach (see section 2.) and then reviewed the critical loads for the different ecosystems.

2. Approach

2.1 Timescales

Critical loads should be set to protect ecosystems over a minimum of several decades. Many of the data used to derive empirical critical loads, however, are from short-term experiments - typically 1-5 years. There is a concern that the N load which produces an effect in a short term experiment is an overestimate of the critical load needed to protect the ecosystem in the longer term. Where data from long term field observations are available, they have been used in preference to short term experimental data to set the critical load. These studies rarely span a period greater than 20-30 years, so the critical loads given here cannot be assumed to protect ecosystems on a longer time scale than this. For some ecosystems, only experimental data were available to derive the critical load, and these values may be overestimates for long-term protection. The type of source data used for setting each critical load is shown in Table 1.

2.2 Vegetation types

The vegetation types identified at Lökeberg have been used here, with the addition of 'Upland *Calluna* moorland'. A number of other ecosystems which are potentially sensitive to N deposition have been identified although there is currently insufficient data to set a critical load (see recommendations).

2.3 Ranges and suggested mapping values

Critical loads have been expressed as ranges (Table 1). This can reflect:

a) The interval between the experimental treatments at which an effect was observed and not observed.

b) Differences in critical loads where a number of comparable studies are available, often from different countries.

c) Genuine intra-ecosystem variation.

d) Uncertainty in deposition values where critical loads are based on field observations.

While current evidence is insufficient to give a single value, it was recognised that a critical load range creates a problem for mapping and exceedence calculations. A default value for mapping has been suggested if the range is small, or the evidence merits it. This can be used by countries which have insufficient national data. Where a mapping value is not given, a central value within the range should be used.

2.4 Management and limiting factors

Recent data suggest that management practices may be important in modifying the response to N deposition in some semi-natural ecosystems. Critical loads for calcareous grasslands have been expressed as a range from low values for unmanaged systems to high values for intensively managed systems. The critical N load for heathlands may also vary according to the absence/presence and intensity of management practices such as burning, sod cutting, grazing and mowing. No guidance for management modification of the critical load is given here, however, due to lack of data. The effects of nitrogen are also likely to depend on whether N assimilation is limited by other factors e.g. phosphorus availability. Critical loads for P-limited and N-limited calcareous grasslands have been calculated, but this approach has not been followed for other ecosystems. It was aknowledged that lack of data on the distribution of limiting factors will create problems for mapping.

2.5 Forests

A critical load for NH_4-N has been set for tree nutrition/health based on N:nutrient cation ratios. The critical load for total N is based on empirical data of N-saturation, indicated by nitrate leaching from below the rooting zone. The mass balance approach should be used in conjunction with these values to set the critical load. Critical loads have been set separately for changes in ground flora which may not be protected by the SMB (steady state mass balance) method.

3 Empirical Critical Loads for Terrestrial Ecosystems (Table 1)

	Data Source	Critical Load (kg N ha^{-1} $year^{-1}$)	Mapping Value	Effect
Soft-water lakes	F,E	5-15##	10	Decline in Isoetes
Mesotrophic fens	F,E	20-35#	(-)	Increase in tall graminoids, decline in diversity
Ombrotrophic bogs	E	15-20#	15	Decrease in *Drosera & Sphagnum*
Calcareous species-rich grassland				
a) N limited	M(F,E)	14-40#	20	Increase in tall grass, decline in diversity. Low end of the range for poorly managed (mown), high end for optimally managed (grazed) systems
b) P limited	F,E	50-54#	50	High nitrate concentration in ground water
Neutral-acid species-rich grassland	F,E	20-30#	(-)	Increase in tall grass, decline in diversity
Montane-subalpine grassland	K	10-15(#)	(-)	Increase in tall grass, decline in diversity
Lowland dry-heathland	E,M	15-22#	17	Transition of heather to grass
Lowland wet-heathland	E,M	17-22#	19	Transition of heather to grass
Species-rich heaths/acid grassland	K	7-20#	(-)	Decline of sensitive species
Upland *Calluna* moorland		10-20#	(-)	Decrease in heather dominance
Arctic and alpine heaths	K	5-15#		Decline in lichens, mosses and evergreen dwarf-shrubs, increase in grasses and herbs
Acidic Coniferous Forest (managed)	F,E	10->50##	(-)	Nutrient imbalance. Use low end of the range for sites with low nitrification rate and/or low base saturation, high end of the range for sites with high nitrification rate and/or high base saturation.
	F,E	10-25##	(-)	N-saturation, N leaching and base cation depletion of soil.
	F,E	10-20##	(-)	Changes in ground flora.
Acidic Deciduous Forest (managed)	F,E	15->50#	(-)	Nutrient imbalance, shoot:root ratio. Use low of the range for sites with low nitrification rate and/or low base saturation, high end of the range for sites with high nitrification rate and/ or high base saturation.
	F	15-20#	(-)	Changes in ground flora.
Deciduous calcareous forest	F	15-20#	(-)	Changes in ground flora

Footnote to Table 1.
The critical loads shown are based upon published data combined with expert knowledge on expected responses. The type of data source used to derive the critical load is either field observations (F), experiments (E), models (M), ecological knowledge (K), or a combination. The reliability of the estimates is shown, based on the following criteria:

##	-	reliable: when a number of published papers of various studies show comparable results;
#	-	quite reliable: when the results of some studies are comparable;
(#)	-	best guess: when only limited or no data are available for this type of ecosystem.
(-)	-	insufficient data to provide single default value for mapping. A central value of the range should be used, taking site characteristics into account (e.g. soil type, manage ment) where appropriate.

4 Recommendations

- Further research/data collation is required to calculate a critical load for the following ecosystems:

  Steppe grasslands
  Acidic unmanaged forest
  Mediterranean vegetation types
  High altitude forests
  Coastal heathlands

- Where critical loads are based on "best guess" estimates or very few studies, further research and/or a detailed literature review of existing data are required. These are:

  Ombrotrophic bogs
  Montane sub-alpine grasslands
  Upland *Calluna* moorland
  Species-rich heath/acid grassland
  Arctic and Alpine heaths

- More information is required on:
  - the interaction between climatic and biotic factors and N deposition.
  - the effects of N deposition on fauna.
  - the effect of management on sensitivity to N deposition.
  - the interaction between forest fires and N deposition in mediterranean systems.
  - the relative effects of oxidized and reduced nitrogen.

- In order to refine current critical loads, nitrogen addition experiments with a high resolution of treatments between 5 and 40 kg N ha^{-1} yr^{-1} are required. This would overcome the problem of using experimental data for deriving critical loads when the first treatment level greatly exceeds the critical load.

- Dynamic vegetation models need to be developed so that data from short-term experiments can be used to set long-term critical loads. These models should be validated against long-term data sets if possible.

- The problem of large scale deposition data versus higher resolution critical load data needs to be addressed.

- Information on the distribution of some of the small-area ecosystems will not be availablein all countries. In the first instance, effort should be directed towards producing small scale maps for sensitive receptors of high conservation value. Information sources on their distribution include national data, the UN List of National Parks and Protected Areas and the CORINE database.

- Output from the UNECE Integrated Monitoring and EU/UNECE ICP Forest Programme networks should be fed into the critical load process to improve estimates.

Keynote Paper - Empirical Nitrogen Critical Loads: update since Lökeberg (1992).

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Introduction

The activities of man pose a number of threats to the structure and the functioning of (semi-) natural ecosystems, and thus to the natural variety of plant and animal species. One of the major ones has been the increase in air-borne nitrogen pollution (NH_x and NO_y) in recent decades. The most important impacts of increased nitrogen deposition upon biological systems are:

(i) short-term effects of nitrogen gases and aerosols to individual species;
(ii) soil-mediated effects of acidification and eutrophication;
(iii) increased susceptibility to secondary stress factors;
(iv) changes in (competitive) relationships between species, resulting in loss of diversity.

Empirical critical loads for excess nitrogen deposition were presented in a background document for the 1992 UNECE Workshop at Lökeberg (Sweden). The proposed values were discussed in detail prior to and agreed at the meeting (Bobbink *et al.* 1992; Grennfelt & Thörnelöf 1992). Validated changes in plant development and in vegetational and faunal composition of (semi-) natural ecosystems have been used as an indication of the impacts of excess nitrogen deposition. If possible, data from field surveys, experiments and dynamic ecosystem models have been used in conjunction. Nitrogen critical loads have been given within a range per ecosystem, because of

(i) genuine intra-ecosystem variation,
(ii) intervals between experimental treatments, and
(iii) uncertainties in deposition values.

Furthermore, the reliability of the presented figures has been indicated.

In this paper recent data on the ecological effects of nitrogen deposition have been reviewed to update the nitrogen critical loads, which were set at Lökeberg. Additional information from the period 1992-1994 is presented, after a short summary of the 1992-critical loads estimates. Finally, an overview of the updated critical loads and gaps in knowledge are given. Full information on ecological effects of excess nitrogen inputs and the list of original publications is given in Bobbink *et al.* (1992).

Softwater systems, ombrotrophic bogs and wetlands

The following critical loads (in kg N ha^{-1} yr^{-1}) were established at the L^keberg meeting, based upon changes in ecosystem processes and species composition:

	kg N ha^{-1} yr^{-1}		Indication
Shallow soft-water bodies	5-10	##	Decline isoetid species
Mesotriophic fens	20-35	#	Increase tall graminoids, decline diversity
Ombrotrophic bogs	5-10	#	Decrease typical mosses, increase tall graminoids

The effects of excess nitrogen deposition on softwater lakes have been intensively studied in the 1980ís and early 1990ís, especially in the Netherlands. The results of these studies have been used to set a reliable critical load (5-10 kg N ha^{-1} yr^{-1}) for these sensitive ecosystems. Since then, no new data have become available to necessitate a change of these values. The same holds for the critical loads of mesotrophic fens, although further study is necessary to increase the reliabilty of these figures.

Ombrotrophic bogs, which receive all their nutrients from the atmosphere, are particularly sensitive to increased air-borne nitrogen inputs and many studies had indicated the detrimental effects on the growth of bog-forming mosses. Furthermore, much evidence has been published which indicated increased N accumulation in these ecosystems. This has led to the development of reliable critical loads of 5-10 kg N ha^{-1} yr^{-1}. Besides the detrimental effects of atmospheric nitrogen on the development of the bog-forming *Sphagnum* species, N may influence the competitive relationships in nutrient-deficient vegetation such as bogs. The effects of the supply of extra nitrogen on the population ecology of *Drosera rotundifolia* has been recently studied in a 4-year fertilization experiment in Swedish ombrotrophic bogs (Redbo-Torstensson 1994). It was shown that experimental applications above 10 kg N ha^{-1} yr^{-1} (NH_4NO_3 at ambient deposition of 5 kg N ha^{-1} yr^{-1}) clearly affected this insectivorous species. The establisment of new individuals and the survival of the plants significantly decreased in the vegetation treated with extra nitrogen. This decrease in the total density of the population of the characteristic bog species *Drosera* was not caused by toxic effects of nitrogen, but by enhanced competition for light with tall species such as *Eriophorum* and *Andromeda,* which responded positively to the increased N inputs.

It has been suggested that increased nitrogen contents of the bog-mosses affect the decay rate of the peat, as nitrogen content strongly influences decomposition rates (e.g. Swift *et al.* 1979). The decay rate of *Sphagnum* peat in Swedish ombrotrophic bogs has been studied along a gradient of nitrogen deposition (Hogg *et al.* 1994). The results of this short-term decay experiment indicate that the decomposition rate was more influenced by the phosphorus content of the material, than by nitrogen, although some relation with nitrogen supply has been observed. Further evidence is necessary to evaluate the long-term effects of enhanced nitrogen supply on the decay of peat.

Based upon the British and Scandinavian studies it has become clear that increased nitrogen loads strongly affect ombrotrophic bog ecosystems, especially because of the high nitrogen retention capacity and closed nitrogen cycling. The growth of the bog-mosses is negatively affected by nitrogen and changed competitive relationship between the prostate dominants (e.g. *Eriophorum*) and the subordinate plant species, and thus, reduce biodiversity. A quite reliable critical load for nitrogen in these ombrotrophic bogs is in the range 5-10 kg N ha^{-1} yr^{-1}, although additional long-term studies with enhanced nitrogen (both nitrogen oxides and ammonia/ammonium) are necessary to validate this figure. A joint European study has just started to investigate the changes in essential ecosystem processes after long-term nitrogen enrichment (Silcock *et al.* 1994).

Species-rich grasslands

At Lökeberg the following empirical critical loads were established for grasslands with high conservational importance:

	kg N ha^{-1} yr^{-1}		Indication
Calcareous species-rich grassland	14-25	##	Increase tall grass, decline diversity
Neutral-acid species-rich grassland	20-30	#	Increase tall grass, decline diversity
Montane-subalpine grassland	10-15	(#)	Increase tall graminoids, decl. diversity

The effects of nitrogen in montane-subalpine grasslands were identified as a major gap in knowledge (Bobbink *et al.*, 1992; Grennfelt & Thörnelöf 1992). Unfortunately, this gap still exists and the critical load is based solely upon expert jugdement. More progress has been made upon the impacts of N inputs in other grassland ecosystems.

The effects of enhanced nitrogen input have been thoroughly investigated in Dutch calcareous grasslands. The results of this integrated programme have led to the 1992-critical load. An expansion of the grass *Brachypodium pinnatum* and a drastic reduction in species diversity have recently been found in a long-term permanent plot study using a factorial design of nutrient application (Willems *et al.* 1993). Following a survey of data from a number of conservation sites in southern England, Pitcairn *et al.* (1991) concluded that *Brachypodium* had expanded in the UK during the last century. They considered that much of the early spread could be attributed to a decline in grazing pressure but that more recent increases in the grass had, in some cases, taken place despite grazing or mowing, and could be related to nitrogen inputs. A study of chalk grassland at Parsonage Downs (UK) has, however, shown no substantial change in species composition over the twenty years between 1970 and 1990, a period when nitrogen deposition is thought to have increased to 15-20 kg N ha^{-1} yr^{-1} (Wells *et al.* 1993). *Brachypodium* was present in the sward but had not expanded as in the Dutch grasslands. In a linked experimental study, applications of nitrogen to eight forbs and one grass (*Brachypodium*) at levels of 20 to 80 kg N ha^{-1} yr^{-1} for two years did not result in *Brachypodium* becoming dominant. This indicates that the effects of nitrogen are sometimes counteracted by adequate management, or prohibited by P limitation, but the afore-mentioned data from British calcareous grasslands still fits within the range of the set critical loads. Additional information is, however, needed to quantify the long-term (ca. 10-20 years) effects of management and P limitation upon the impacts of excess N inputs.

A recent study of the response of mesothrophic grasslands in Great Britain have shown that additions as small as 25 kg N ha^{-1} yr^{-1} (without ambient deposition) lead to changes in species diversity after several years of fertilizer additions and that changes took place more rapidly at higher rates of addition

(Mountford *et al.*, 1994). This study indicates that these semi-natural grasslands are affected by nitrogen eutrophication and that the observed effects correspond with the critical load (20-30 kg N ha^{-1} yr^{-1}) set in 1992.

Heathlands

The following empirical critical loads have been established for the effects of excess nitrogen upon different heathland ecosystems:

	kg N ha^{-1} yr-1		indication
Lowland dry-heathland	15-20	##	Transition heather to grass
Lowland wet-heathland	17-22	##	Transition heather to grass
Species-rich lowland heaths/acid grassland	7-20	#	Decline sensitive species
Arctic and alpine heaths	5-15	(#)	Increase grasses

The critical loads for lowlands heath are based mostly upon data from the Netherlands. It has been shown that nitrogen eutrophication is a significant factor in the transition of dwarf shrub heaths to grasslands, especially after opening of the canopy. Apart from the changes in competitive interactions, heather beetle plagues and soil nitrogen accumulation are other important processes in the effects of excess nitrogen in lowland heaths. Furthermore, evidence is growing that the frost sensitivity of the dominant dwarf-shrubs may also be affected by increasing nitrogen inputs. At L^keberg, critical loads for dry and wet lowland heaths were set using dynamic ecosystem models, which integrated all these processes. These models (Berendse 1988; Heil & Bobbink 1993 a&b) have been calibrated with data from field and laboratory experiments in the Netherlands, but need to be tested in future with other European data.

Field surveys of lowland dry heathland in the UK do not show consistent patterns over the past 10 to 40 years. Pitcairn *et al.* (1991) assessed changes in abundance of *Calluna* at three heaths in East Anglia over the past decades. All three heaths showed a decline in *Calluna* and an increase in grasses; the authors concluded that increases in nitrogen deposition was at least partly responsible for the changes, but also noted that the management had changed, too. A wider assessment of heathlands in SE England showed that in some cases *Calluna* had declined and subsequently been invaded by grasses while other areas were still dominated by dwarf shrubs (Marrs 1993). This stresses the importance of (adjusted) management for the maintainance of dwarf-shrubs in heathlands, even at moderate nitrogen loads.

Application of ammonium sulphate for 4 years at rates equivalent to the critical loads or only slightly higher (7.7 & 15.4 kg N ha^{-1} yr^{-1}, ambient load ca. 18 kg N ha^{-1} yr^{-1}) has not resulted after 4 years in any negative effects upon *Calluna* in dry heathland in S England. However, a significant stimulation of *Calluna* growth, flower production, shoot nitrogen content and litter production has been found in the experimental period. The increased litter production returns nutrients to the soil which could have long-term effects on the nitrogen status of the system (Uren *et al.* in press). Because of the low application rates (only slightly above critical loads) it may be that deleterious effects upon the vegetation only become detectable after many years, especially because of the very low abundance of grasses at the experimental site (S.A. Power personal comm.). Preliminary simulations with the CALLUNA Model, using the UK data, suggest that the effects of N will become more evident after only 15-20 years.

The effects of nitrogen inputs are trigged by opening of the dwarf-shrub canopy by heather beetle attacks, frost damage or drought. *Calluna* plants of UK heathlands were fumigated with relatively high ammonia concentrations. The growth of heather beetle colonies increased significantly, probably caused by the enhanced nitrogen concentrations in the shoots (Uren 1992). This is in good agreement with the observations in the Netherlands. Because of the stochastic behaviour of heather beetle plagues and the many long-term processes, it has proved difficult to clarify experimentally the relationship between nitrogen and the beetle plagues. The heather beetle has, however, recently also been found in SW Norway and it is expanding its territory. The beetle may be an important cause of *Calluna* death even in this region, although further evidence is needed (Hansen 1991; Dommarsnes in prep.).

The *Calluna* canopy may also be influenced by effects of excess nitrogen inputs upon frost sensitivity. At this moment it is however not possible to draw final conclusions in this respect, because in some studies (Van der Eerden *et al.* 1991; Uren 1992) it has been shown that ammonia fumigation or ammonium application did increase frost sensitivity, whereas in another study ammonium application resulted in a decrease of frost sensitivity of *Calluna* (Caporn *et al.* 1994). It is concluded that at this moment it is not necessary to modify the established critical loads.

Species-rich heaths and acidic grasslands

In recent decades, besides the transition from dwarf-shrub dominated grass dominated heathlands, a reduced species diversity in these ecosystems has been observed. Herbaceous species of the acidic *NARDETALIA* grasslands and the related dry- and wet-heathlands (*CALLUNO-GENISTION* and *ERICION TETRALICES*) seem to be especially sensitive. The distribution of these species is related to small-scale, spatial variability of the heathland soils. It is suggested that atmospheric deposition may have caused such drastic abiotic changes of these species that they can not survive (Van Dam *et al.* 1986). Dwarf-shrubs as well as grass species are nowadays dominant in former habitats of these endangered species.

Enhanced nitrogen fluxes onto the nutrient-poor heathland soils lead to an increased nitrogen availability in the soil. However, most of the deposited nitrogen in W Europe originates from ammonia/ammonium deposition and may also cause acidification as a result of nitrification or ammonium uptake by plants. Whether eutrophication or acidification or a combination of both processes is important, depends on pH, buffer capacity and nitrification rates of the soil. The pH decrease may indirectly result in an increased leaching of base cations, increased aluminium mobilization and thus enhanced Al/Ca ratios of the soil (Van Breemen *et al.* 1982). Furthermore, the reduction of the soil pH may inhibit nitrification and result in ammonium accumulation and consequently increased NH_4/NO_3 ratios. In a recent field study the characteristics of the soil of several of these threatened heathland species have been compared with the soil characteristics of the dominant species (*Calluna vulgaris*, *Erica tetralix* and *Molinia caerulea*) (Houdijk *et al.* 1993). Generally the endangered species grow on soils with higher pH, lower nitrogen content, and lower Al/Ca ratios than the dominant species. The NH_4/NO_3 ratios were higher in the dwarf-shrub dominated soils compared with the ratios in the soil of the endangered species. Fennema (1992) has demonstrated that soils from locations where *Arnica montana* is still present, had higher pH and lower Al/Ca ratios than soils of former *Arnica* stands. However, he found no differences in total soil nitrogen and NH_4/NO_3 ratios. Both of these studies indicate that high Al/Ca ratios or even increased NH_4/NO_3 ratios are associated with the decline of these species. Results of a hydroculture experiment with *Arnica* showed that this species is very sensitive to enhanced Al/Ca ratios at intermediate or low nutrient levels (De Graaf 1994). Pot experiments have indicated that increased NH_4/NO_3 ratios have caused a decreased vitality of *Thymus*. Hydroculture experiments with this plant species confirmed that increased NH_4/NO_3 ratios affect the cation uptake (Houdijk 1993).

At present, however, there is too little information available on these rare heathland and acidic grassland species to formulate a reliable critical load. The observation that these heathland species mostly disappear before dwarf-shrubs are replaced by grasses, leads to the assumption that their critical load is lower than the critical load for the transition to grasses (<15-20 kg N ha^{-1}) and is probably between 7-15 kg N ha^{-1} yr^{-1}.

Forest ecosystems

Nutritional Imbalance of Trees

At the UNECE meeting at Skokloster in 1988 the following critical loads for tree health were set, based upon nutritional imbalance after excess ammonia/ammonium inputs (Boxman *et al.* 1988):

	kg N ha^{-1} yr^{-1}	Indication
Coniferous tree health (acidic)	10-15	Nutrient imbalance (low nitrification rate)
Coniferous tree health (acidic)	20->50	Nutrient imbalance (mod.-high nitrification rate)

These values are well established and it is thus not necessary to discuss them again. Almost all of these effects upon nutritional imbalances have been quantified for coniferous tree species, but recently it has been shown that the magnesium and phosphorus concentrations in leaves of *Fagus sylvatica*, a common deciduous tree in Europe, decreased significantly in permanent plots in NW Switzerland from 1984 to 1992 (Fl¸ckiger & Braun 1994). Further, the magnesium concentrations in the leaves of young *Fagus sylvatica* decreased significantly within a 4-year period at fertilization rates of 25 kg N ha^{-1} yr^{-1} (Fl¸ckiger & Braun 1994). In Sweden, the addition of nitrogen enhanced a nutritional imbalance in a 120-year old *Fagus* stand (Balsberg PÂhlsson 1992). It is thus obvious that deciduous tree species are also sensitive to nutritional imbalances induced by enhanced nitrogen supply. Nitrogen-rich free amino acids, especially arginine, have also significantly increased in *Fagus* leaves (Balsberg PÂhlsson 1992) as already disccussed in the L^keberg-paper for coniferous species. There is now clear evidence that high ammonia/ammonium loads produce adverse changes in the nutritional status and the growth of the investigated broad-leaved trees, and that a quite reliable critical load for nitrogen deposition on decidious tree vitality is in the range 15-20 kg N ha^{-1} yr^{-1}, as demonstrated in the experiments of Fl¸ckiger & Braun (1994).

Excess nitrogen deposition can seriously affect tree vitality via a complex web of interactions (e.g. susceptibility to frost and drought). Pathogens may play an important role in tree decline, but it is yet not possible to combine the observed processes and effects to an overall value for a critical load of nitrogen for tree health.

Nitrogen effects on the forest undergrowth

At L^keberg the following empirical critical loads have been set for the effects upon forest undergrowth (including mycorrhiza and fauna):

	kg N ha^{-1} yr^{-1}	Indication
Acidic (managed) coniferous forest	15-20 #	Changes in ground flora & mycorrhizas
Acidic (managed) decidious forest	15-20 #	Changes in ground flora & mycorrhizas
Calcareous forests	unknown	unknown
Acidic unmanaged forest	unknown	unknown

It has been shown that ectomycorrhizal fruit-body production is much more sensitive to (atmospheric) nitrogen enrichment than the mycorrhizal infection of tree roots. Currently, a decrease in fruit-body production of mycorrhizal species was demonstrated already after 1.5 year at a nitrogen application of 35 kg N ha^{-1} yr^{-1} (as NH_4NO_3) in a Swedish *Picea abies* stand (Nitrex-site; Brandrud 1995). This supports the critical load set for acidic coniferous forests, taking into account the short experimental period.

The relation between acidity and soil fauna has been studied in northern coniferous forests, but only very few studies have incorporated the effects of nitrogenous air-borne compounds. A reduction in the nitrogen deposition in a *Pinus sylvestris* stand (Nitrex site; Ysselstein) to pre-industrial levels increased the species diversity of micro-arthropods due to a decreased dominance of some species (Boxman *et al.* 1995). However, at present it is not possible to use these few data to formulate a critical load for changes in forest soil fauna due to increased nitrogen deposition. The effect of enhanced nitrogen input on (soil) fauna is a serious gap in knowledge and certainly needs more attention in future research.

Some recent evidence of the impact of nitrogen depositions on the understorey in calcareous forests has become available. In a large semi-natural *Fagus-Quercus* forest in NE France, ca. 50 permanent vegetation plots were described in 1972 and in 1991. Besides a number of moderately acidic habitats, the changes in species composition on calcareous soils have been followed. During the study period a significant increase in nitrophilous ground flora was observed in these high-pH (ca. 6.9) stands. This indicates that at this location (with ambient deposition of 15-20 kg N

$ha^{-1}\ yr^{-1}$) a distinct effect of increasing nitrogen availability could be detected (Thimonier *et al.* 1992). The setting of a critical load of 15-20 kg N $ha^{-1}\ yr^{-1}$ for effects upon the undergrowth of calcareous forests seems realistic (Table 1). A summary of the updated critical loads for effects upon the undergrowth of forests are given in Table 1.

Conclusions and gaps in knowledge

In this paper, the effects of nitrogen deposition on (semi-)natural freshwater and terrestrial ecosystems have been updated using the evidence published since 1992. Based upon observed changes in vegetation and reductions in biodiversity, critical loads for nitrogen have been (re)formulated (Table 1).

Table 1. Summary of guidelines for nitrogen deposition (kg N $ha^{-1}\ yr^{-1}$) to (semi-) natural freshwater and terrestrial ecosystems. ## reliable; # quite reliable and (#) best guess.

	Critical load	Indication
Shallow soft-water lakes	5-10 ##	Decline isoetid species
Mesotrophic fens	20-35 #	Increase tall graminoids, decl. diversity
Ombrotrophic (raised) bogs	5-10 #	Decrease *Sphagnum* and subordinate species, increase tall graminoids
Calcareous species-rich grassland	14-25 ##	Increase tall grass, decline diversity
Neutral-acid species-rich grassland	20-30 #	Increase tall grass, decline diversity
Montane-subalpine grassland	10-15 (#)	Increase tall graminoids, decl. diversity
Lowland dry-heathland	15-20 ##	Transition heather to grass
Lowland wet-heathland	17-22 ##	Transition heather to grass
Species-rich heaths/acid grassl.	7-20 (#)	Decline sensitive species
Arctic and alpine heaths	5-15 (#)	Decline lichens, mosses and ever green dwarf-shrubs, increase in grasses and herbs
Coniferous tree health (acidic)	10-15 #	Nutrient imbalance (low nitrification rate)
Coniferous tree health (acidic)	20->50 #	Nutrient imbalance (moderate-high nitrification rate)
Deciduous tree health	15-20 #	Nutrient imbalance; increased shoot-root ratio
Acidic (managed) coniferous forest	15-20 ##	Changes ground flora
Acidic (managed) deciduous forest	15-20 #	Changes ground flora
Calcareous forests	15-20 (#)	Changes ground flora
Acidic unmanaged forest	unknown	unknown

Most of the earths biodiversity is found in (semi-)natural ecosystems. It is crucial, therefore, to control the nitrogen load, in order to prevent negative effects on these natural ecosystems. In this document, critical loads for nitrogen have been updated as reliably as possible. As most of the research efforts have focused on acidification in forestry, serious gaps in knowledge exist on the effects of enhanced nitrogen deposition (NO_x, NH_x) on (semi-) natural terrestrial and aquatic ecosystems. The following gaps in knowledge are most important:

- *quantified effects of enhanced nitrogen deposition on fauna in all the reviewed ecosystems are extremely scarce.*

- *the critical load for nitrogen deposition to arctic and alpine heathlands is largely speculative.*

- *more research is needed on the nitrogen effects on forest ground vegetation and (ground) fauna, because most research has until now focussed on the trees only.*

- *a serious gap in knowledge exists on the effects of nitrogen in neutral/calcareous forests, which are not sensitive to acidification.*

- *more long-term research is needed into the response of montane/subalpine meadows, species-rich grasslands and ombrotrophic bogs to enhanced nitrogen deposition.*

- *the long-term effects of enhanced atmospheric nitrogen deposition on grasslands and heathlands of high nature conservation importance under different management regimes are insufficiently known and may affect the critical load value.*

- *the possible differential effects of the deposited nitrogen species (NH_x or NO_y) are insufficiently known to make a differentation between these N species for the establishment of critical loads of individual N species.*

- *the long-term effects of nitrogen eutrophication on (sensitive) freshwater ecosystems needs further research.*

To establish reliable critical loads, it is crucial to understand the long-term effects of increased nitrogen deposition on ecosystem processes in a representative range of ecosystems. It is thus very important to quantify the effects of nitrogen loads on (semi-)natural ecosystems by manipulation of nitrogen inputs in long-term ecosystem studies in both unaffected and affected areas. These data are essential to validate the currently set critical loads values and for the development of robust dynamic ecosystem models, which are sufficiently reliable to calculate the critical loads for nitrogen deposition in (semi)natural ecosystems.

References

Balsberg Påhlsson, A.-M. (1992) Influence of nitrogen fertilization on minerals, carbohydrates, aminoacids and phenolic compounds in beech (*Fagus sylvatica* L.) leaves. Tree Physiol. 10: 93-100.

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).

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. & Thörnelöf, E. (Eds.), Critical loads for nitrogen. Nord (Miljörapport) 41, 111-159. Nordic Council of Ministers, Copenhagen.

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. Miljørapport 15: 295-323.

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 stands in The Netherlands. Forest. Ecol. Managem. (in press).

Branderud, T.E. (1994) The effects of experimental nitrogen addition on the mycorrhizal fungus flora in an oligotrophic spruce forest in Gårdsjön, Sweden. Forest Ecol. Managem. (in press).

Caporn, S.J.M., Risager, M. & Lee, J.A. (1994) Effects of nitrogen supply on frost hardiness in Calluna vulgaris (L.) Hull. New Phytol. 128: 461-468.

Danmarsnes, S. in prep. Vegetation changes in west Norwegian coastal heath. Thesis Univ. Bergen, Norway.

De Graaf, M.C.C. (1994) Ammonium and nitrate in heathland and heathland related vegetations: preferences for nitrogen source and ammonium toxicity. Acta Botanica Neerlandica 43: 393.

Fennema, F. (1992) SO_2 and NH_3 deposition as possible causes for the extinction of *Arnica montana* L. Water Air Soil Poll. 62: 325-336.

Flückiger, W. and Braun, S. (1994) Waldschaden-Bericht. Untersuchungen in Buchenbeobachtungsflächen 1984-1993. Institut für Angewandte Pflanzenbiologie, 4124 Schönenbuch, Switzerland.

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

Hansen, K.F. (1991) Sociologisk-ekologisk differensiering, strüktur og dynamikk i dagens vestnaske lynghei. Thesis Univ. Bergen, Norway.

Heil, G.W. and Bobbink, R. (1993a) "CALLUNA" a simulation model for evaluation of impacts of atmospheric nitrogen deposition on dry heathlands. Ecol. Modelling 68: 161-182.

Heil, G.W. and Bobbink, R. (1993b) Impact of atmospheric nitrogen deposition on dry heathlands: a stochastic model simulating competition between Calluna vulgaris and two grass species. In: Aerts, R. & Heil, G.W., Heathland: Patterns and Processes in a changing environment. Geobotany 20, Kluwer, Dordrecht, pp. 181-200.

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

Houdijk, A.L.F.M., Verbeek, P.J.M., Van Dijk, H.F.G. and Roelofs, J.G.M. (1993) Distribution and decline of endangered herbaceous heathland species in relation to the chemical composition of the soil. Plant Soil 148: 137-143.

Marrs, R.H. (1993) An assessment of change in *Calluna* heathland. Biol. Conserv. 65: 133-139.

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 fertilizer application; (b) after cessation of fertilizer input for three years. Report to the Institute for grassland and environmental Research. Institute of Terrestrial Ecology(Monks Wood) Abbots Ripton, UK.

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. Report to the Nature Conservancy Council, Institute of Terrestrial Ecology (Edinburgh Research Station), Penicuik, UK.

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

Silcock, D.J., Buttler, A., Francez, A-J., Gilbert, D. , Grosvernier, Ph., Juanhaianen, J., Matthey, Y., Vasander, H. and Williams, B.L. (1994) Nitrogen inputs on ombrotrophic raised bogs. In: Abstracts Symposium on Ecosystem Manipulation, Bowness-on-Windermere,UK. Organized by A. Jenkins, Institutue of Hydrology, Wallingford, Oxon, UK.

Swift, M.J., Heal, O.W. and Anderson, J.M. (1979) Decomposition in terrestrial ecosystems. Blackwell, Oxford.

Thimonier, A., Dupouey, J.L., Bost, F. and Becker, M. (1994) Simultaneous eutrophication and acidification of a forest in North-East France. New Phytol. 126: 533-539.

Uren, S.C. (1992) The effects of wet and dry deposited ammonia on *Calluna vulgaris*. PhD-thesis, Imperial College, University of London.

Uren, S.C., Ainsworth, N, Power, S.A., Cousins, D.A. and Huxedurp, L.M. (1995) Long-term effects of ammonium sulphate on *Calluna vulgaris*. J. Appl. Ecol. (in press).

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

Van Dam, D. Van Dobben, H.F. Ter Braak, C.F.J. and De Wit, T. (1986) Air pollution as a possible cause for the decline of some phanerogamic species in The Netherlands. Vegetatio. 65: 47-52.

Van Der Eerden, L.J., Dueck, Th.A., Elderson, J., Van Dobben, H.F., Berdowski, J.J.M. and Latihinin, M. (1990) Effects of NH_3 and $(NH_4)_2SO_4$ deposition on terrestrial semi-natural vegetation on nutrient-poor soils. Report IPO/RIN Wageningen, The Netherlands.

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, Abbots Ripton, UK.

Willems, J.H., Peet, R.K. and Bik, L. (1993) Changes in chalk-grassland structure and species richness resulting from selective nutrient additions. J. Veg. Science 4: 203-212.

Keynote Paper - **Methods to derive Critical Loads for Nitrogen for Terrestrial Ecosystems**

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Abstract

The effects of an excess input of N on terrestrial ecosystems can include vegetation changes, forest damage and nitrate pollution of ground water. Critical loads (deposition levels) for N related to these effects can be derived by empirical data, steady-state soil models and/or dynamic soil vegetation models. Examples of the various approaches are given with a discussion of the advantages and disadvantages for the mapping of critical loads.

1 Introduction

The effects of N deposition on terrestrial ecosystems include:

(i) vegetation changes due to increased N availability;

(ii) decreased forest vitality caused by inhibition of base cation uptake and increased sensitivity to frost, drought and diseases.

Furthermore, it may cause NO_3 pollution of groundwater. A thorough review of the impacts of N inputs on terrestrial ecosystems, i.e. ombotrophic bogs and wetlands, heathlands, species-rich grasslands and forests, including empirical critical N loads related to vegetation changes, is given in Bobbink *et al.* (1992). Apart from vegetation changes, a continuous high input of N may cause damage to forests due to:

(i) water shortage, since a high N input favours growth of canopy biomass, whereas root growth is relatively unaffected (De Visser, 1994);

(ii) nutrient imbalances, since the increase in canopy biomass also causes an increased demand for base cation nutrients (Ca, Mg, K) whereas the uptake of these cations is reduced by increased dissolved levels of NH_4 (Boxman and Van Dijk., 1988);

(iii) an increased sensitivity to natural stress factors such as frost (Aronsson, 1980) and attacks by fungi (Roelofs *et al.* 1985).

An excess input of N can finally cause pollution of ground water due to NO_3 leaching. In the Netherlands, high N inputs have caused high NO_3 concentrations in ground water in shallow ground, often exceeding the current EC drinking water standard of 50 mg l^{-1}. This is harmful in areas where ground water is used for water supply.

In this article, an overview is given of various methods to derive critical N loads, and their application. Regarding the methods used, a distinction is made between an empirical approach and a model approach. The empirical approach is further divided in a deterministic and a probabilistic approach and the model approach is divided in steady-state soil models and dynamic soil vegetation models. Special emphasis is given to steady-state models, which are commonly used to derive critical load maps. The advantages and disadvantages of the various methods are also extensively discussed.

2 Methods and Criteria to derive Critical Nitrogen Loads

Critical N loads for terrestrial ecosystems loads can be derived directly from the relationship between atmospheric deposition and effects on "specified sensitive elements" within an ecosystem (ecosystem status) by correlative or experimental research. For example, increased N loads may lead to changes from heathlands to grasslands. Critical N loads can be estimated by comparing the N deposition on grass dominated and heather dominated heathlands, or by experimental investigation of the biomass development of grasses as a function of N input. Critical loads can also be derived indirectly from critical values for ion concentrations or ion ratios in the ecosystem, based on dose response relationships between these chemical criteria and the ecosystem status. In this respect, steady-state nitrogen models have been developed, calculating critical loads from critical chemical ion concentrations and/or ratios. (c.f. De Vries, 1992). An overview of the criteria that can be used to assess critical N loads for various ecosystems is given below in Table 1.

Table 1: Critical chemical values used for various N parameters in terrestrial ecosystems

Effects	Criteria			
	Compartment	Parameter	Unit	Value
Vegetation changes	Soil solution	N	($mol_c\ m^{-3}$)	0.02 - 0.04[1)]
Nutrient imbalances	Soil solution	NH_4/K	(mol^{-1})	5[2)]
	Foliage	N/K	(mol^{-1})	5.5 - 11.0[3)]
Increased susceptibility	Foliage	N	(%)	1.8[4)]
Nitrate pollution	Ground water	NO_3	($mol_c\ m^{-3}$)	0.8[5)]

1) A critical N concentration related to vegetation changes is difficult to assess. A nitrogen mass balance for a dry calcareous grassland in the Netherlands indicates that vegetation changes may take place in a situation where N leaching hardly increases above natural background values (Van Dam 1990). Similarly, N leaching is nearly negligible in Dutch heathlands, changing into grasslands. It is the increase in N availability through enhanced N cycling that triggers the vegetation changes (e.g. Berendse *et al.* 1987). In calculating critical N loads, however, the loss of nitrogen from the ecosystem should be accounted for, even though one should use a natural background value rather than a critical value. NO_3 concentrations in stream water of nearly unpolluted forested areas in Sweden are

as low as 0.02 $mol_c\ m^{-3}$ (Rosén, 1990). However, the total N concentration, including NH_4 and organic N is likely to be slightly larger and a natural N concentration range of 0.02 - 0.04 $mol_c\ m^{-3}$ seems plausible. Using a precipitation excess of 200 - 400 mm yr^{-1}, this gives a N leaching rate of 40 - 160 $mol_c\ ha^{-1}\ yr^{-1}$ (0.5 - 2 kg $ha^{-1}\ yr^{-1}$), which is a common range of natural N losses from an ecosystem

[2] Boxman and Van Dijk (1988) found a strong decrease in the uptake of Ca and Mg at an increasing molar NH_4/K ratio in a laboratory experiment with two-year-old Corsican pines. Using these data Boxman *et al.* (1988) proposed a critical NH_4/K ratio of 5 (mol mol^{-1}).

[3] Nutrient imbalances are also reflected by the ratio of N to base cations in the foliage. Based on an extensive literature review, Van den Burg *et al.* (1988) proposed an optimal N/K ratio 2 g g^{-1} (5.5 mol mol^{-1}) and a critical ratio of 4 g g^{-1} (11.0 mol mol^{-1})

[4] For most coniferous tree species, a N content in the needles of 1.6 to 2.0% is considered optimal for growth. At these levels the sensitivity to frost and fungal diseases, however, increases too. In a fertilization experiment in Sweden, it was found that frost damage to the needles of Scots pine strongly increased above an N content of 1.8% (Aronsson, 1980). At this N level, the occurrence of fungal diseases such as *Sphaeropsis sapinea* and *Brunchorstia pinea* also appears to increase.

[5] Derived from the EC drinking water standards of 50 mg l^{-1}, (approximately 0.8 $mol_c\ m^{-3}$). However, countries may wish to use different values based on a precautionary approach. For example in the Netherlands a target value of 25 mg l^{-1} (0.4 $mol_c\ m^{-3}$) is used for NO_3.

3 Empirical Data

3.1 Deterministic approach

At present, critical N loads for terrestrial (and aquatic) ecosystems related to changes in vegetation and fauna are generally derived by a so-called deterministic empirical approach. These empirical loads are based on an extensive but inhomogeneous summary of correlative field studies and large-scale laboratory (greenhouse) experiments (c.f. Bobbink *et al.* 1992). To illustrate this aspect, an overview of empirical data for critical N loads on terrestrial and aquatic ecosystems in the Netherlands is given in Table 2, together with their derivation.

Empirical critical N loads related to vegetation changes were derived either from simulated precipitation experiments with NH_4 (e.g. on small-scale heathland or soft water ecosystems in a greenhouse) or from correlative field studies between N deposition and species diversity, using present geographic differences or historical data on N deposition and species decline (c.f. De Vries, 1992). Empirical critical N loads related to increased susceptibility to frost damage and fungal diseases and to nutrient imbalances were also derived from correlative studies (Table 2). In this case, however, there was no relation between N deposition and observed damage but with an observed increase in critical N parameters, such as the N content in foliage (Van der Burg *et al.* 1988) and the NH_4/K ratio in soil solution (Boxman *et al.* 1988). In this situation, models are likely to produce more accurate estimates because of their ability to predict the long-term perspective of critical loads. For example, field data by Van den Burg *et al.* (1988) show that an N content of 1.8% in needles of Scots pine is associated with an N deposition of approximately 3000 $mol_c\ ha^{-1}\ yr^{-1}$. However, the same concentration can be reached by exposure to lower inputs over a longer period. By mathematical modelling, Van Grinsven *et al.* (1991) simulated an N content in needles of Douglas fir close to 1.8% at an N deposition level near 1500 $mol_c\ ha^{-1}\ yr^{-1}$ in a 40 year period (nearly the average rotation period of a coniferous tree in the Netherlands). In comparison, it is likely that empirical data by Boxman *et al.* (1988), who measured a molar NH_4/K ratio near 5 at an artificially induced NH_4 input of 800 $mol_c\ ha^{-1}\ yr^{-1}$ in soils with a very low nitrification rate, lead to an under-estimation of critical NH_3 loads because the present N/K ratio in needles is enhanced compared to the situation at critical loads.

Table 2: Empirical data of critical N loads ($mol_c\ ha^{-1}\ yr^{-1}$) for various effects in forests, heathlands and surface waters (After De Vries, 1992)

Critical N-load	Ecosystem	Effects	Derivation
700-1400	Forest	Vegetation changes	Correlation with N deposition and species diversity
1500	Forest	Increased susceptibility	Correlation between N deposition and N content in needles
800	Forest	Nutrient imbalances (most sensitive systems)	Correlation between N inputs (experiment) and NH_4/K ratio in soil solution
850-1400	Heathland	Change to grassland	Precipitation experiment
500-700	Heathland	Decreased species diversity	Correlation with N deposition
1400	Surface water	Change to nitrophilous species	Precipitation experiment
500[1)]	Surface water	Decreased species diversity	Correlation with N deposition

[1)] Refers to oligothropic surface waters in the Netherlands with low denitrification and N immobilization

3.2 Probabilistic Empirical Approach

The drawback of deterministic empirical critical nitrogen loads is that they do not give equal protection of the various ecosystems. The concept of risk assessment may be used in this situation as an alternative, because it provides a framework to achieve more standardization in the assessment of protection levels for different environmental problems (c.f. Latour *et al.* 1994). Most progress in assessing and quantifying ecological risks has been made in the field of toxicological stress. There the maximum tolerable concentration (MTC) is chosen as the environmental concentration of a compound at which (theoretically) 95% of the species are fully protected. MTCs are calculated by extrapolation of No Observed Effects Concentrations (NOEC levels) for single-species to an ecosystem. Various extrapolation techniques have been proposed, assuming a log-logistic, lognormal or triangular distribution of species sensitivities, but the concept of risk assessment remains the same (Slooff, 1992).

Until now, the probabilistic approach has been rarely used in setting critical levels or critical loads. Notable exceptions are Van der Eerden *et al.* (1994), who calculated maximum tolerable air concentrations for NH_3 and SO_2 based on NOECs determined in laboratory tests, and Latour *et al.* (1994), who derived critical N loads for fertilized grasslands based on NOECs for 275 species with the model MOVE (multiple-stress model for the vegetation; c.f. Latour and Reiling, 1993). MOVE predicts the occurrence probability of ca. 700 species as a function of three abiotic soil factors, including nitrogen availability, using regression relationships (c.f. Section 5). Since the combined sampling of vegetation and environmental variables is rare, the indication values of plant species by Ellenberg (1979) are used to assess the abiotic soil conditions. The deduction of values for the abiotic soil factors from the types of vegetation guarantees ecological relevance. Combinations of samples of vegetation and environmental variables are used exclusively to calibrate Ellenberg indication values with the quantitative values of the abiotic soil factors.

At the species level, the risks are assessed on the basis of the species-response function, which describes the occurrence probability of a species as a function of an environmental variable (Figure 1). The species-response function can be characterized by its optimum (O) and tolerance (T). Assuming a normal distribution, the tolerance is quantified as the standard deviation of the distribution. Latour *et al.* (1994) used the 5 and 95 percentiles of the species-response curves as NOEC-like measures for the risk at the species level (Figure 1). The 5 percentile corresponds with a reduced occurrence probability due to "limitation", the 95 percentile due to "intoxication". Assuming normal distributions, the 5 percentile can be calculated with the formula $P_5 = O - 1{,}645\ T$ and the 95 percentile with $P_{95} = O + 1{,}645\ T$. Species were considered protected when the 5 percentile of the species was lower and the 95 percentile higher than the value of the environmental variable.

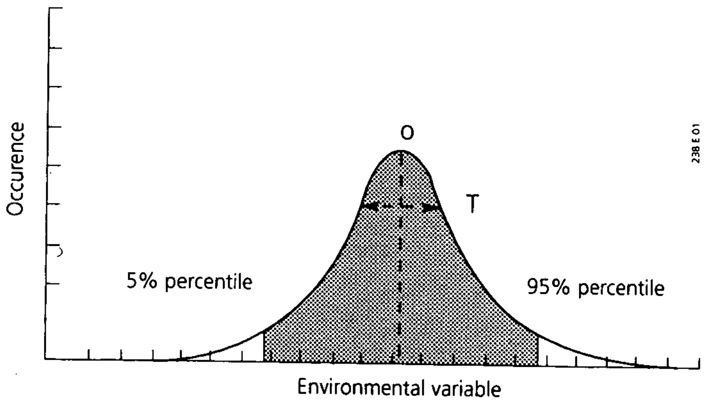

Figure 1. **Occurrence probability of a species as a function of an environmental variable (species-response function). Within the shaded area, species are considered to be protected.**

In setting critical N loads, MOVE was applied to fertilised grasslands in the province of South Holland. Species-response functions for 275 species were described for nitrogen availability, assuming a Gaussian logistic distribution of the species occurrence. Data on 12,000 vegetation recordings were used for the analysis. Optima and tolerance were expressed for each plant species in terms of Ellenberg-like indication values. These values were translated into nitrogen loads (in kg N ha^{-1} yr^{-1}) based on a linear relationship between the Ellenberg values for the optima of 42 plant species and the nitrogen load (Figure 2A). On the basis of this regression the nitrogen load, (N in kg N ha^{-1} yr^{-1}), was calculated from Ellenberg indication values (E) by: N=(E-2.6)/0.016. Figure 2B gives the percentage of protected grassland species in South Holland as a function of nitrogen load based on the 5 and 95 percentiles of each species. According to Figure 2B the maximum tolerable nitrogen load, corresponding to a protection of 95% of the species, is 60 kg N ha^{-1} yr^{-1}. This is one order of magnitude lower than current loads of 450 kg N ha^{-1} yr^{-1} resulting in a protection level of about 30% of the plant species. At loads higher than 100 kg N $ha^{-1}yr^{-1}$ the number of species is reduced through intoxication (95 percentile). At low nitrogen loads (up to 50 kg N $ha^{-1}yr^{-1}$) the number of species is reduced mainly through N limitation (5 percentile). Maximum reduction through limitation is 14% at zero nitrogen load.

4 Steady-State Soil Models

4.1 Model derivation

Total nitrogen

Critical loads of total N for terrestrial and aquatic ecosystems can be derived with a simple model of the N balance. The complete N balance including all N fluxes (in mol_c ha^{-1} yr^{-1})

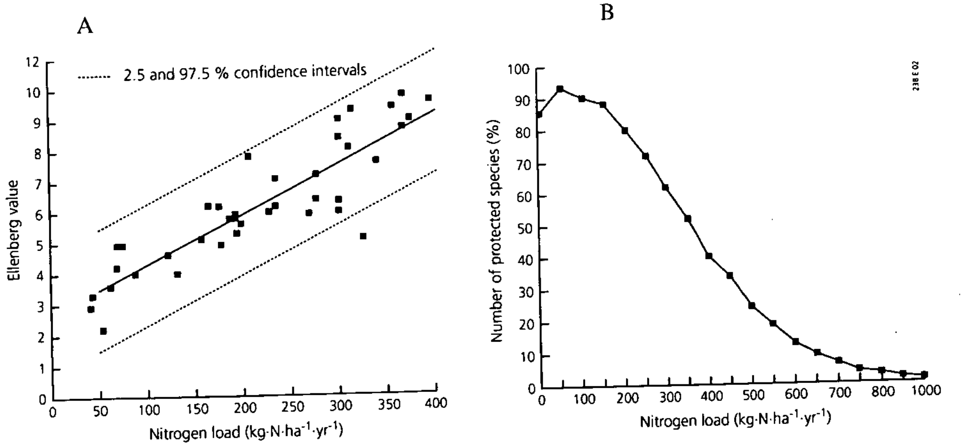

Figure 2. The relationship between nitrogen load and Ellenberg indication values for 42 plant species (A) and the number of protected plant species in grasslands in the province South Holland (B)

in an ecosystem reads:

$$N_{td} = N_{gu} + N_{im} + N_{de} + N_{vo} + N_{er} + N_{ad} + N_{le} - N_{fi} \quad (1)$$

where the subscript *td* refers to total deposition, *fi* to fixation, *gu* to growth uptake, *im* to net immobilization, *de* to denitrification, *vo* to volatilization, *er* to erosion, *ad* to adsorption and *le* to leaching. Adsorption of N can, however, be neglected as this process only plays a temporary role. Furthermore, even in the short term, adsorption of N can generally neglected since (i) N mainly occurs as NO_3 except for the topsoil and (ii) the preference of the adsorption complex for NH_4 is mostly low, especially in (acid) sandy soils. Volatilisation of N can play a role in grazed woodlands and in (Mediterranean) areas with frequent forest fires, whereas N removal by erosion may play a role at extremely steep slopes. However, in most cases these N fluxes are negligible. N fixation is also small in most forest and heathland ecosystems except for N-fixing species, such as red alder (c.f. De Vries, 1992). By assuming N loss by volatilization and erosion and N adsorption to be negligible, Eq. (1) can be written as:

$$N_{td} = N_{gu} + N_{im} + N_{de} + N_{le} - N_{fi} \quad (2)$$

Denitrification can be described as a fraction of the net N input:

$$N_{de} = fr_{de} \cdot (N_{td} + N_{fi} - N_{gu} - N_{im}) \quad (3)$$

This equation is based on the assumption that the excess N input leaches as NO_3. In most (forest) soils, N below the root zone (at 1m depth) is indeed dominated by NO_3, even in the Netherlands with an extremely high NH_4 input (c.f. De Vries, 1994). It is therefore reasonable to assume that NH_4 leaching is negligible. From Eqs. (2) and (3) a critical N load, $N_{td}(crit)$, can be derived according to:

$$N_{td}(crit) = N_{gu}(crit) + N_{im}(crit) + \frac{fr_{de}}{(1 - fr_{de})} \cdot N_{le}(crit) + N_{le}(crit) - N_{fi} \quad (4)$$

where $N_{im}(crit)$ stands for a critical (long-term acceptable) level of N immobilization and $NO_{3,le}(crit)$ for a critical level of NO_3 leaching. The third term in the right hand side of Eq. (4) stands for the denitrification rate at critical N loads. In wet forest and heathland soils, deep ground water and surface waters denitrification is generally not negligible and should be accounted for. However, in well-drained forest and heathland soils, denitrification is assumed negligible (c.f. De Vries, 1994). The same holds for N fixation. For these systems (including phreatic ground water) Eq. (4) simplifies to:

$$N_{td}(crit) = N_{gu}(crit) + N_{im}(crit) + N_{le}(crit) \quad (5)$$

The use of Eq. (5) is based on the idea that any N input exceeding the net N uptake by forest growth, the critical long-term immobilization rate and natural leaching of N, finally leads to vegetation changes or NO_3 pollution of groundwater.

Ammonia

Critical loads of NH_3-N for terrestrial ecosystems can be derived with a simple model including the major NH_4 and K inputs to the system. The ratio of NH_4 to K in the soil solution, which is the criterium for deriving a critical NH_3-N load, is determined by system inputs (in $mol_c\ ha^{-1}\ yr^{-1}$) according to:

$$RNH_4K = (NH_{4,tf} + NH_{4,mi} - NH_{4,ni} - NH_{4,ru}) \,/\, (K_{tf} + K_{mi} + K_{we} - K_{ru}) \tag{6}$$

where RNH_4K is the NH_4/K ratio, the subscript *tf* stands for throughfall, *mi* for mineralization, *ni* for nitrification, *ru* for root uptake and *we* for weathering. In calculating a critical load, the model was simplified by assuming that (i) throughfall of NH_4 equals total deposition of NH_3 minus foliar uptake, (ii) mineralization of NH_4 and K is equal to the N and K input by litterfall (steady-state situation) and (iii) root uptake does not affect the NH_4/K mol ratio. Eq. (6) thus simplifies to:

$$RNH_4K = NH_{3,td} - NH_{4,fu} + N_{lf} - NH_{4,ni} \,/\, K_{tf} + K_{lf} + K_{we} \tag{7}$$

where *fu* stands for foliar uptake and *lf* for litterfall. Foliar uptake can be described as a fraction of the total NH_3 deposition according to:

$$NH_{4,fu} = fr_{fu} \,.\, NH_{3,td} \tag{8}$$

where fr_{fu} is a foliar uptake fraction. Nitrification of NH_4 can be described as a fraction of the NH_4 input according to:

$$NH_{4,ni} = fr_{ni} \cdot (NH_{3,td} - NH_{4,fu} + N_{lf}) \tag{9}$$

where fr_{ni} is a nitrification fraction. Combination of Eqs. (7) and (8) gives:

$$RNH_4K = (1 - fr_{ni}) \cdot (NH_{3,td}\,(1 - fr_{fu}) + N_{lf}) \,/\, (K_{tf} + K_{lf} + K_{we}) \tag{10}$$

By defining a critical NH_4/K ratio (RNH_4K(crit)), a critical NH_3-N load ($NH_{3,dt}$(crit)) can be derived according to:

$$NH_{3,td}(\mathrm{crit}) = \frac{RNH_4K(\mathrm{crit}) \cdot (K_{tf} + K_{lf} + K_{we}) - (1 - fr_{ni}) \cdot N_{lf}}{(1 - fr_{ni}) \,.\, (1 - fr_{fu})} \tag{11}$$

Assuming negligible foliar uptake and nitrification, Eq.(11) simplifies to:

$$NH_{3,td}\ (crit) = RNH_4\ K\ (crit) \,.\, (K_{tf} + K_{lf} + K_{we}) - N_{lf} \tag{12}$$

4.2 Application examples and discussion

Total nitrogen

Critical N loads related to vegetation changes and to groundwater pollution can be derived using Eq

(4). The impact of the different criteria that are used for these effects is illustrated in Table 3. In deriving critical loads, one should be aware that the values for the various N fluxes should be long-term acceptable values. For example, the critical N uptake rate should be derived as the minimum of (i) N uptake that does not cause relative deficiencies of base cations (c.f. Posch *et al.* 1993) and (ii) the product of average stem growth during a rotation period and the N content in stemwood occurring at critical N load (cf De Vries, 1992). The first estimate will be limiting in areas with low Base Cation inputs produced by weathering and deposition, such as in most of Scandinavia, whereas the second estimate will be limiting in areas with high Base Cation inputs, such as the Mediterranean countries. The second estimate should, however, not be based on the present average annual N uptake in areas with a high N input. For example, in the Netherlands, the average annual growth data were derived from yield tables published before the large increase in NH_3 emissions in the Netherlands, whereas the N contents were taken from sites with a relatively low N deposition. At present, the net N uptake in stemwood in the Netherlands can be much higher due to increased N contents in stems and possibly also by increased growth rates (De Vries, 1992). The use of increased uptake rates in response to N deposition is, however, important in the application of dynamic models to assess acceptable interim loads.

In addition, N immobilization should not refer to the present situation but to the long-term critical net N-immobilization in stable organic N-compounds in the soil (stable forms of humus). This can be derived from the accumulation of N in soils since the period of soil formation. One may, however, accept a larger increase for a rotation period, e.g. a 5 to 10% change in N pool. In the short term (the coming decades), N accumulation in the humus layer of forests and heathlands may be higher. As with increased N uptake, short term N accumulation should, however, not be included in deriving a long-term critical load, but it is important for assessing short-term interim loads.

Table 3 Critical N loads related to vegetation changes and groundwater pollution using data that are typical for forest on well-drained and poorly-drained sandy soils in the Netherlands.

Effects	NO_3 (crit) PE		N_{gu}(crit)	N_{im}(crit)	N_{fi}	Critical N load ($mol_c\,ha^{-1}yr^{-1}$)[1)]	
	($mol_c m^{-3}$)	($m^3ha^{-1}yr^{-1}$)	---------	($mol_c ha^{-1}yr^{-1}$)	-----	fr_{de}=0.1	fr_{de}=0.5
Vegetation changes	0.03	2000	400	140	100	500	560
Nitrate pollution	0.40	2000	400	140	100	1330	2040
	0.80	2000	400	140	100	2210	3640

1) The value of 0.1 for fr_{de} is typical for a well-drained soil, whereas 0.5 is a typical value for a poorly drained sandy soil.

Ammonia

Critical loads of NH_3 related to nutrient imbalances in forests can be derived with Eq. (11) using a critical molar NH_4/K ratio of 5. An application example for the Netherlands is given in Table 4. As with the steady-state nitrogen model (Eq. 4), the problem in using Eq. (11) is that the data which are necessary to derive a critical NH_3 load, e.g. throughfall and litterfall data for N and K, strongly depend on the deposition levels of NH_3. These affect the K exudation rate and the N/K ratios in needles. Obviously, one should use the data at the critical load level. Failure to do this leads to wrong estimates of the critical load as illustrated in Table 4.

The first and second combination of data on N/K ratio in needles and K input by litterfall and throughfall in this Table are assumed to be indicative for the present situation and the situation at critical loads. The use of current input data are likely to give an underestimate of the critical load of NH_3-N, especially at low nitrification fractions. Table 4 shows that the critical NH_3-N load increases significantly with an increase in nitrification fraction and that the data that are likely to occur at critical loads leads to a minimum value of 1050 mol ha^{-1} yr^{-1} for soils with a negligible nitrification rate. However, this value is likely to be an underestimate since nitrification will never be inhibited completely.

Table 4 Model estimates of critical loads for NH_3-N for forests on acid sandy soils as a function of the N/K ratio in needles, the K input in throughfall and the nitrification fraction.

Situation	N/K ratio in needles[1] (mol mol^{-1})	K input by litterfall[2] (mol_c ha^{-1} yr^{-1})	K input by throughfall[3] (mol_c ha^{-1} yr^{-1})	Critical NH_3-N load (mol_c $ha^{-1}yr^{-1}$)[4]		
				fr_{ni}=0.0	fr_{ni}=0.2	fr_{ni}=0.5
Present	11	400	500	100	1225	4600
Past	5.5	400	250	1050	1860	4300

1) Based on data given by Van den Burg and Kiewiet (1989) for Scots pine, Corsican pine and Douglas fir. The past refers to the beginning of 1960 when the N load was much lower.
2) Based on De Vries (1992). The K input by litterfall is hardly affected by N deposition(Van den Burg and Kiewiet, 1989)
3) The average K input by throughfall varies between approximately 250 mol_c ha^{-1} yr^{-1} in relatively unpolluted areas and 500 mol_c ha^{-1} yr^{-1} in polluted areas, such as the Netherlands (cf De Vries, 1992).
4) The K weathering rate and foliar uptake fraction is assumed to be negligible. Nitrification fractions of 0.2 and 0.5 are based on data by Tietema (1992). A nitrification fraction of 0.0 is a worst case situation that hardly ever occurs.

5 Integrated Dynamic Soil Vegetation Models

The major drawback of steady-state soil models is their neglect of biotic interactions. For example, vegetation changes are mainly triggered by a change in N cycling (N mineralization; cc Berendse *et al.* 1987). Furthermore the enhancement of diseases, such as the heather beetle outbreaks, by elevated N inputs may stimulate vegetation changes (cc Heil and Bobbink, 1993). Consequently dynamic soil vegetation models, including such processes, have a better scientific basis for the assessment of critical N loads. Examples of such models are CALLUNA (Heil and Bobbink, 1993) and ERICA (Berendse, 1988).

The dry-heathland model CALLUNA integrates N processes by atmospheric deposition, accumulation and sod removal, with heather beetle outbreaks and competition between species, to establish the critical N load in lowland dry-heathlands (Heil and Bobbink 1993). The model has been calibrated with data from field and laboratory experiments in the Netherlands. Atmospheric nitrogen deposition has been varied between 5 and 75 kg N ha^{-1} yr^{-1} in steps of 5-10 kg N during different simulations. From these simulations it became obvious that the critical N load changes from dwarf-shrubs to grasses is ca 1000 - 1400 mol_c ha^{-1} yr^{-1}.

The wet-heathland model ERICA incorporates the competitive relationships between *Erica* and *Molinia*, the litter production from both species, and nitrogen fluxes by accumulation, mineralization, leaching, atmospheric deposition and sheep grazing. Berendse (1988) simulated the development of lowland wet-heathland after sod removal, because almost all of the Dutch communities are already strongly dominated by *Molinia* and it is impossible to expect changes in this situation without drastic management. Using the biomass of *Molinia* with respect to *Erica* as an indicator, his results suggested a critical N load of 1200 - 1600 mol_c ha^{-1} yr^{-1} for the transition of lowland wet-heathland into a grass-dominated sward (Berendse 1988).

Another example of an integrated dynamic soil vegetation model is SMART-MOVE. This model can predict the occurrence probability of plant species as a result of national environmental scenarios for acidification, eutrophication and desiccation. The model consists of a soil module (SMART; De Vries *et al.* 1989) and a vegetation module (MOVE; Latour and Reiling., 1993), predicting the abiotic and corresponding biotic effects, respectively (Figure 3).

In the soil module changes in abiotic soil factors indicating acidification (pH), eutrophication (N availability) and desiccation (moisture content) are predicted in response to scenarios for acid deposition and groundwater abstractions. A single-layer dynamic soil model SMART (Simulation Model for Acidification's for Regional Trends; De Vries *et al.* 1989) is used for these predictions. SMART has been used on European and national scales to gain insight into the long-term impact of deposition scenarios for N and S. The following processes have been included in this model: net uptake of nitrogen and base cations, net N transformations (nitrification, denitrification and net N immobilization), weathering of carbonates, silicates and Al hydroxides, cation exchange and CO_2 equilibria. Currently, nutrient cycling (litterfall, mineralization and uptake) has also been incorporated into the model. (Kros *et al.* 1994). In the vegetation module (MOVE) the occurrence of species is predicted as a function of the three abiotic soil factors: soil acidity, nutrient availability and soil moisture. With regression statistics, the occurrence probability of a species can be calculated for each combination of the soil factors or for each factor separately (species-response function). Since these response functions are based on Ellenberg indication values, a calibration of these indication values to quantitative values of the abiotic soil factors is necessary to link the soil module to the vegetation module (see section 3.2).

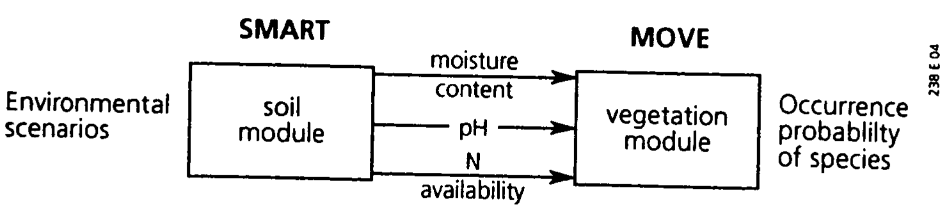

Figure 3 **Schematic presentation of the model SMART-MOVE**

The advantage of SMART-MOVE is that the two approaches to assess critical loads, i.e.: both exceedance of critical values for ion concentrations and ion ratios in soil water and analysis of changes in species composition, are combined in a consistent framework. The SMART model can be used to predict both exceedances of critical values for ion concentrations and ion ratios in soil water and the input data for the vegetation module, i.e. nutrient availability and soil acidity. In the vegetation module, critical loads can be assessed based on changes in species composition.

Neglect of biotic interactions also limits the derivation of critical N loads related to forest damage. Even the derivation of critical loads based on a critical foliar N content (c.f. Table 1) is impossible with a steady-state soil model. At present, there are several integrated forest-soil models that are potentially useful for a more scientifically based derivation of critical N loads. Examples are the models NAP (Van Oene, 1992) and FORSVA (Oja *et al.* 1995) that have been used to derive critical loads for N and S for the Solling spruce site (Germany). The criteria that were used to derive a critical load were: (i) optimal growth during a relation period (100 years) while avoiding Mg deficiency (NAP model) and (ii) a long-term sustainable biomass production avoiding toxic Al effects (FORSVA model). The critical loads for N and S thus derived were close to those derived by a steady-state soil model (De Vries *et al.* 1995). This is an important conclusion, since much work remains to be done, both in modelling efforts and data collection, before truly integrated forest soil models can be used for assessing critical loads on a large regional scale.

6 Discussion and Conclusions

An overview of the advantages and disadvantages of the various methods to derive critical N loads is given in Table 5. Empirical data and steady-state soil models are easily applicable for mapping. Uncertainties in deterministic empirical data on critical N loads are mainly related to the occurrence of time lags. Harmful effects on ecosystems may already occur before they are visible. The uncertainty in the probabilistic empirical approach described in this paper is mainly caused by the uncertainty in the relationship between the Ellenberg indication values and N availability.

Uncertainties in critical N loads derived by steady-state soil models are determined by the uncertainty in critical chemical values, model structure and data. The choice of the critical NO_3 concentration and NH_4/K ratio strongly affects the critical N and NH_3 loads. In this context, the uncertainty in critical N loads on forests will be larger than the critical N load related to NO_3 leaching to ground water, since 50 mg l^{-1} of NO_3 is an accepted critical value for drinking water, whereas the criteria for vegetation changes and forest damage are very uncertain. Uncertainties induced by the model structure include the neglect of biotic interactions (c.f. Section 5) and various other modelling assumptions. A systematic overview of the effects of uncertainties in modelling assumptions and data, both in steady-state and dynamic soil models, is given in De Vries (1994).

Until now, critical load maps for N have been exclusively based on effects on the vegetation, using either empirical data or steady-state soil models. Considering the possible trade off between species diversity and forest growth in relation to N load, it is necessary to derive critical N loads for various effects (vegetation changes, forest growth, forest vitality etc). In this context, it is important to derive optimal rather than critical N loads. Furthermore, it is necessary to apply integrated dynamic soil-vegetation models to check the results of conventional methods. The critical N loads derived by steady-state models are especially uncertain because of the uncertainty in the criteria related to effects (c.f. Table 1). However, integrated dynamic models are generally complex, thus limiting their use in mapping. However, SMART-MOVE, which is still relatively simple, has been applied for the whole of the Netherlands to evaluate effects of scenarios for acid deposition and groundwater abstraction (Kros *et* *al.* 1994). Furthermore, it is planned to apply the model on a European scale. When countries are able to apply integrated soil-vegetation models on a national scale, it is advised to do so. Otherwise use of conventional methods for critical load mapping is advised.

Table 5 *Evaluation of various methods for the assessment of critical loads*

Method	Advantage	Disadvantage
Empirical data	Clear relationship between species diversity and atmospheric N deposition	No clear relationship between forest vitality and atmospheric N (and S) deposition
	Easily applicable for mapping	Other effects than acidification and eutrophication may be involved
Steady-state soil models	Simple	No biotic interactions involved
	Easily applicable for mapping	Critical chemical values of soil parameters (e.g. NO3, A1) are uncertain
Dynamic soil-vegetation models	Comprehensive description of the ecosystem	Mode l complexity
	Important tool to assess target loads and eevaluate scenarios	Not easily applicable for mapping unless simple models are used

References

Aronsson, A. (1980) Frost hardiness in Scots pine. II Hardiness during winter and spring in young trees of different mineral status. Studia Forest Suecica 155: 1-27.

Berendse, F., Beltman, B., Bobbink, R., Kwant and Schimtz, M.B. (1987) Primary production and nutrient availability in wet heathland ecosystems. Acta Oec./Oecol. Plant. 8: 265-276

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).

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 (Miljörapport) 41, 111-159. Nordic Council of Ministers, Copenhagen.

Boxman, A.W. and Van Dijk, H.F.G. (1988) Het effect van landbouw ammonium deposities op bos - en heidevegetaties. Catholic University of Nijmegen, The Netherlands, 96 pp.

Boxman, A.W., Van Dijk, H.F.G., Houdijk, A.L.F.M. and Roelofs, J.G.M. (1988) Critical loads for nitrogen with special emphasis on ammonium. In: J. Nilsson and P. Grennfelt (eds.), Critical loads for sulphur and nitrogen; report from a workshop held at Skokloster, Sweden, 19-24 March, 1988. Milj rapport 1988: 15. Nordic Council of Ministers, K benhavn, 295.

De Visser, P.H.B. (1994) Growth and nutrition of Douglas fir, Scots pine and pedunculate oak in relation to soil acidification. Wageningen, Agricultural University, Ph.D Thesis, 185 pp.

De Vries, W. (1992) Empirical data and model results for critical nitrogen loads in the Netherlands. In: P. Grennfelt and E. Thörnelöf (Eds.), Critical loads for Nitrogen. Report from a workshop held at Lökeberg, Sweden 6-10 April, 1992. Nordic Council of Ministers, Report 1992, 41: 383-402.

De Vries, W. (1994) Soil response to acid deposition at different regional scales. Field and laboratory data, critical loads and model predictions. Wageningen, the Netherlands, Agricultural University, Ph.D Thesis, 487 pp.

De Vries, W., Posch, M. and Kämäri, J. (1989) Simulation of the long-term soil response to acid deposition in various buffer ranges. Water Air and Soil Poll. 48: 349-390.

De Vries, W., Posch, M., Oja, T., Van Oene, H., Kros, J., Warfvinge, P. and Arp, P.A. (1995) Modelling critical loads for the Solling spruce site. Ecological Modelling. (Submitted).

Ellenberg, H. (1979) Scripta Geobotanica 9, 1 - 122 Heil, G.W. and Bobbink, 1993 ìCALLUNAî a simulation model for evaluation of impacts of atmospheric nitrogen deposition on dry heathlands. Ecol. Modelling 68: 161-182.

Kros, J., Reinds, G.J., De Vries, W., Latour, J.B. and Bollen, M. (1994) Modelling of soil acidity and nitrogen availability in natural ecosystems in response to changes in acid deposition and hydrology. SC report.

Latour, J.B. and Reiling, R. (1993) A multiple stress model for vegetation (MOVE): a tool for scenario studies and standard setting. Sci. Tot. Environ. 93: 1513-1526.

Latour, J.B., Reiling, R. and Slooff, W. (1994) Ecological standards for eutrophication and desiccation: perspectives for a risk assessment. Water Air Soil Poll. 78: 265-277

Oja, T., Yin, X. and Arp, P.A. (1995) The forest model series For M-S: Application to the Solling Spruce site. Ecological Modelling (Submitted).

Posch, M., Reinds, G.J. and De Vries, W. (1993) SMART, Simulation Model for Acidificationís Regional Trends. Model Description and User Manual. Mimeograph Series of the National Board of Waters and the Environment 477, Helsinki, Finland, 43 pp.

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

Rosén, K. (1990) The critical load of nitrogen to Swedish forest ecosystems. Uppsala, Department of forest soils. Swedish University of Agriculture Science, Internal Report, 15 pp.

Slooff, W. (1992) RIVM Guidance Document: Ecotoxicological effect assessment. Deriving maximum tolerable concentrations from single-species toxicity data. RIVM, Bilthoven, The Netherlands, report no. 719102018. 49 pp.

Tietema, A. (1992) Nitrogen cycling and soil acidification in forest ecosystems in the Netherlands. Ph.D Thesis, University of Amsterdam, the Netherlands, 139 pp.

Van Dam, (D. 1990) Atmospheric deposition and nutrient cycling in chalk grassland. Ph.D thesis, University of Utrecht, 119 pp.

Van Den Burg, J., Evers, P.W., Martakis, G.F.P., Relou, J.P.M. and Van Der Werf, D.C. (1988) De conditie en de mineralevoedingstoestand van opstanden van grove den (Pinus silvestris) en Corsicaanse den (Pinus nigra var. Maritima) in de Peel en op de zuidoostelijke Veluwe najaar 1986. Wageningen, The Netherlands, Instituut voor Bosbouw en Groenbeheer, ìDe Dorschkampî, Rapport nr. 519, 66 pp.

Van Den Burg, J. and Kiewiet, H.P. (1989) Veebezetting en de naalsamenstelling van grove den, Douglas en Corsicaanse den in het optreden van bosschade. Wageningen, The Netherlands, Instituut voor Bosbouw en Groenbeheer, ìDe Dorschkampî, Rapport nr. 559, 76†pp.

Van Der Eerden, L.J., Dueck, T.A., Posthumus, A.C. and Tonneijck, A.E.G. (1992) Assessment of critical levels for air pollutant effects on vegetation: some considerations and a case study on NH_3. UNECE Workshop on Critical levels, Egham, U.K., 23-26 March 1992.

Van Oene, H. (1992) Acid deposition and forest nutrient imbalances: a modelling approach. Water Air Soil Poll. 63: 33 - 50.

Van Grinsven, J.J.M., Van Minnen, J. and Van Heerden, C. (1991) Effects on growth of Douglas fir. In: T. Schneider and G.J. Hey (eds.), Acidification research in the Netherlands. Final report of the Dutch Priority Program me on Acidification. Studies in Environmental Science 46, Elsevier Science, 180-190.

CHAPTER 3
CRITICAL LOADS OF NITROGEN FOR SURFACE WATERS

Working Group Report

Introduction

The concepts behind critical loads for nitrogen in surface waters, and the alternative approaches applied, have been described elsewhere (e.g. Kämäri *et al.* 1992; Henriksen *et al.* 1992, 1993; CLAG Freshwaters 1995; Jenkins & Shaw 1993). Discussions within the workshop therefore focused on evaluating the relative strengths and weaknesses of current models (see Table 1), and identifying gaps in knowledge relating to these different approaches. The workshop also discussed more general research recommendations relating to critical loads modelling for nitrogen. This report briefly summarises these discussions, and outlines the recommendations made by the group.

Empirical Steady-State Water Chemistry Model

The empirical steady-state water chemistry (SSWC) model, developed by Henriksen *et al.* (1992), provides critical acidity loads and critical loads exceedances relating to a fixed value of Acid Neutralising Capacity (ANC). The model is relatively simple, and can often be applied using existing water chemistry data-sets to provide regional assessments of the current effects of sulphur and nitrogen deposition, and the future effects of sulphur. The use of a fixed ANC value allows the model to be related to the response of individual species to acidification. An advantage of the approach is that data on nitrogen deposition are not required. The model also assumes no catchment sources of nitrogen.

A disadvantage of the model is that the exceedance values relate to current nitrate leaching rather than potential leaching. One further disadvantage is that the model cannot be used for testing scenarios of future nitrogen deposition.

The model requires the use of a single nitrate leaching input value, whereas nitrate concentrations in surface waters can display significant seasonal variability. The sub-group recommended that when using the SSWC a flow-weighted mean annual nitrate value should be used wherever possible.

Empirical Diatom Model

The empirical diatom model (Battarbee *et al.* 1993) provides critical acidity loads and critical load exceedances relating to the response of diatom communities to acidification. The model has been calibrated using sediment core diatom records from 41 lakes in the United Kingdom. Diatoms are amongst the most sensitive indicators of acidification, therefore critical loads calculated using the diatom model predict, in theory, the point of first ecological response to acidification. The approach is complimentary to the SSWC model, which is based on a fixed ANC value. The model is simple and can be used to provide regional assessments of current acidification status. The model assumes no catchment sources of nitrogen.

Like the SSWC model, exceedance values calculated using the diatom model relate to current nitrate leaching. The model cannot therefore be used to test scenarios of future nitrogen deposition.

The model has been calibrated using data from the UK. The applicability of the model to other countries requires validation, which could be achieved by desk study.

First-order Acidity Balance Model

This first-order acidity balance (FAB) model (e.g. Downing *et al.* 1993; Henriksen *et al.* 1993) provides a prediction of the maximum potential nitrate leaching for given values of sulphur and nitrogen deposition. Critical deposition (sulphur and nitrogen) and deposition exceedances are calculated relative to a fixed value of ANC.

A major advantage is that the model can be used to evaluate scenarios of future sulphur and nitrogen deposition. Unlike the empirical approaches, the FAB model provides maximum potential nitrate leaching values, and is therefore not limited to contemporary observations of nitrate leaching, although the timing of the maximum leaching is not indicated. It is important to note that the potential leaching derived from the FAB model relates to leaching below the rooting zone, while the empirical approaches use nitrate values for water which has travelled through all soil and geological routings.

The principal disadvantage of the approach is the requirement for substantially more input data than the empirical approaches. Data availability may therefore preclude the application of the model to mapping on a European scale. If the model is used for such regional assessment there may be considerable uncertainties in some of the input values, particularly those associated with nitrogen cycling processes. Literature values are available for many of these inputs, but clearer guidelines will be required for mapping purposes.

Dynamic Models

Dynamic models (e.g. MAGIC) provide predictions of dynamic changes in water chemistry for any given deposition scenario over a range of timescales. A major advantage is that such models provide the only mechanism for assessing dynamic aspects of nitrogen deposition conventions. Dynamic models also allow the development and testing of scenarios of future change in both deposition and land-use.

Dynamic models require detailed data inputs and therefore applications of such models tend to be site specific. Regional critical loads assessments using dynamic models are possible, although at present the approach has limited use for mapping.

The major uncertainties of dynamic modelling relate to the dynamics of nitrogen cycling processes. Additionally the prediction of Aluminium by dynamic models is poor.

Critical Loads for Nutrient Nitrogen (Eutrophication)

Primary production in freshwaters is generally limited by phosphorus rather than nitrogen. As a consequence it has generally been assumed that for typical softwater lakes, the critical load for acidity will be less than the critical load for nutrients. However, recent data from Sweden (Wilander, unpublished) suggests that some types of low phosphorus, softwater lakes typically used in critical loads assessments can become nitrogen limited on a seasonal basis. Additionally there is a lack of data relating to biological responses to increasing nitrate levels in phosphorus limited systems. In such systems, shifts in species composition relating to nitrate may occur independently of shifts in productivity.

There is a clear requirement for data on nutrient limitation in softwater lakes, and on biological responses to increasing nitrate levels. Critical loads models for nutrient nitrogen are being developed and tested, but to date maps are only available on a local scale.

Gaps in Knowledge and Research Recommendations

Empirical Modelling

- Comparison of maximum and mean annual nitrate levels in surface waters
- Validation of the empirical diatom model for non-UK sites

First-order Acidity Balance Model

- Improved data on nitrogen cycling processes, particularly uptake by vegetation and immobili zation by soils
- Guidelines on appropriate literature values for input terms
- Clearer definitions of model uncertainties
- Clarification of the relationship between empirical nitrate leaching values and potential nitrate leaching functions

Critical Loads for Nutrient Nitrogen

- Regional evaluations of nitrogen:phosphorus ratios (limiting nutrients) in softwater lakes
- Assessment of the biological impact of high nitrate levels in softwater systems with respect to eutrophication and ecosystem structure using both spatial data-sets and field experimentation
- Continued development and application of critical loads models for nutrient nitrogen

General Recommendations

- Detailed evaluation of seasonal and temporal variability of inorganic and organic forms of nitrate in surface waters
- Identification of the combination of deposition levels, soil/land-use characteristics and other factors that lead to nitrogen saturation and breakthrough from catchment soils
- Assessment of problems introduced to exceedance calculations by scale mismatches between critical loads data and deposition data

- Assessments of the influence of land-use on model application
- Application of dynamic models to as many sites as possible
- Integrated comparison of model outputs for selected key sites

Table 1 *Comparison of current approaches to calculating critical loads for nitrogen in surface waters*

Model	Empirical Steady-State Water Chemistry Model	Empirical Diatom Model	First-order Acidity Balance Model	Dynamic Models (e.g. MAGIC)
Complexity	Simple	Simple	Simple/Moderate	Moderate/Complex
Ease of Application	Yes	Yes	Yes	Limited
Allows Nitrogen Scenario Testing	No	No	Yes	Yes
Allows Nitrogen Scenario Testing (with Timing)	No	No	No	Yes
Provides Current Nitrate Leaching	Yes	Yes	Yes	Yes
Provides Potential Nitrate Leaching	No	No	Yes	Yes
Data Requirements	Low	Low	Moderate	High

References

Battarbee, R.W., Allott, T.E.H., Kreiser, A.M. and Juggins, S. (1993) Setting critical loads for UK surface waters: the diatom model. In Hornung, M. & Skeffington, R.A. *Critical Loads: Concept and Applications.* London:HMSO, 99-102

CLAG Freshwaters (1995) *Critical Loads of Acid Deposition for UK Freshwaters.* Report of the Critical Loads Advisory Group, sub-group on Freshwaters, for the UK Department of the Environment, 138pp.

Downing, R.J., Hettelingh, J-P. and de Smet, P.A.M. (Eds) (1993) *Calculation and Mapping of Critical Loads in Europe: Status Report 1993.* RIVM Report No.259101003, 163pp.

Henriksen, A., Kämäri, J., Posch, M. and Wilander, A. (1992) Critical loads of acidity: Nordic surface waters. *Ambio* 21, 356-363.

Henriksen, A., Forsius, M., Kämäri, J., Posch, M. and Wilander, A. (1993) *Exceedance of Critical Loads for Lakes in Finland, Norway and Sweden: Reduction Requirements for Nitrogen and Sulfur Deposition.* Report 32/1993, Norwegian Institute for Water Research, 44pp.

Jenkins, A. and Shaw, C. (1993) Comparisons of empirical and dynamic model approaches for surface waters. In Hornung, M. & Skeffington, R.A. *Critical Loads: Concept and Applications.* London:HMSO, 109-114.

Kämäri , J., Jeffries, D.S., Hessen, D.O., Henriksen, A., Posch, M. and Forsius, M. (1992) Nitrogen critical loads and their exceedance for surface waters. In Grennfelt, P. & Thörnelöf. E. (Eds) *Critical Loads for Nitrogen - a Workshop Report.* Nord 1992:41, 161-200.

Keynote Paper - Critical Loads of Nitrogen and their Exceedance in UK Freshwaters

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Introduction

In June 1994 the United Kingdom government signed the European sulphur protocol which committed them to a 70% reduction in SO_2 emissions by the year 2005 and 80% by 2010, based on 1980 levels. This was the first instance of international legislation being formulated using the critical load concept. Notwithstanding the anticipated benefits to the environment of such reductions it was also recognised that such improvements could be diminished unless a similar approach was used to address the problem of increasing nitrogen emissions.

The Freshwater Sub-group of the UK Critical Loads Advisory Group (CLAG) has responsibility for evaluating available techniques for calculating critical loads for nitrogen and for establishing a scientific programme to address key issues and gaps in our knowledge of nitrogen dynamics. This paper summarises the preliminary findings of the Freshwater Group with respect to the nitrogen status and critical loads of UK freshwater.

General Considerations

At its peak in the 1970's, the deposition of sulphur compounds accounted for about two thirds of the total acidic deposition in the UK. During the past 20 yrs this deposition has declined by up to 30% while emissions of oxidised nitrogen (NO_x) have increased by nearly 20% as a result of emissions from motor vehicles. Ammonia emissions, especially from agricultural sources, have also increased significantly in recent years. When this situation and the projected future large (60-80%) reductions on S depositions are considered, it is clear that the relative importance of N deposition will increase dramatically.

Because the processes and pathways of nitrogen utilisation by vegetation, soils and water are far more complicated than for sulphur a more complex set of questions must be addressed, for example:

a) Will different forms of N deposition (NO_3^-, NH_4^+) contribute equally and via similar processes to the acidification and eutrophication status of freshwaters?

b) Can both acidification and eutrophication potential be quantified using similar models?

c) How can seasonal variability in nitrate leaching be incorporated into critical load models?

d) Is there a direct link between N inputs and surface water nitrate concentrations, and if so, how can the extent of N limitation be quantified?

Finally, bearing in mind the many complexities noted above, do we have acceptable methods to map critical loads and exceedance valves for nitrogen (or total acidity).

Current Nitrogen Status of UK Freshwaters

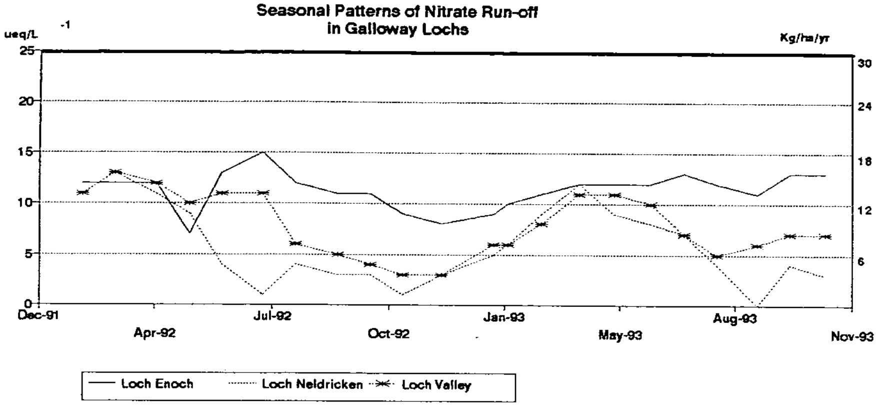
A preliminary evaluation of the current nitrogen status of UK Freshwaters has been made using the recently developed critical load database. This report will shortly be presented to the Department of Environment by the Freshwater Group. Although the relative contributions of NO_3^- and NH_4^+ in precipitation are approximately equal in the UK, ammonium ions are rarely detected in surface water run-off in non-agricultural areas. Consequently only nitrate (and organic N) have been considered at the present time although this situation could change in the future.

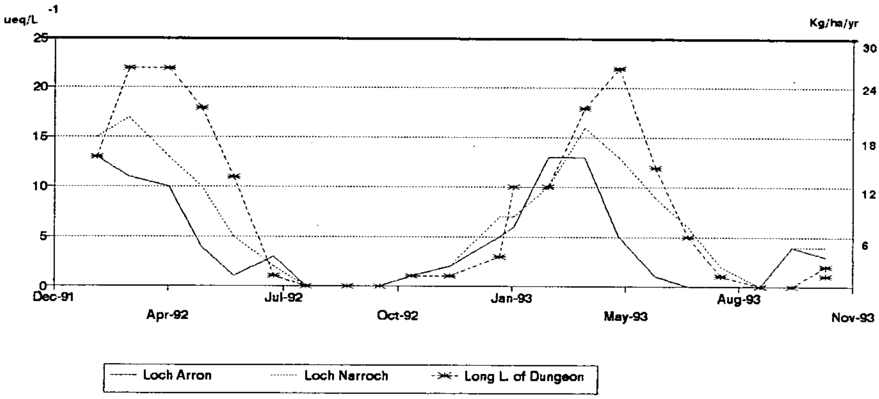
The pattern of nitrate concentrations in UK surface waters (INDITE 1994) clearly reveals agricultural and urban influences in southern and eastern regions. However, by screening sites to include only those of high sensitivity ($Ca^{++} < 300$ eql^{-1}), and excluding those with agricultural catchments, a clearer picture emerges which reveals two major features. Firstly, significant nitrate concentrations (>20 eql^{-1}) occur in areas (eg Pennines and Cumbria) where S critical loads are already exceeded (See Harriman and Christie, 1993) and secondly, some sites (especially in central and north west Scotland) exhibit negligible nitrate leaching even though N inputs are quite large. To quantify the relationship between leaching and deposition a matrix was derived for all 584 UK sites which fitted the above sensitivity/ land-use criteria (Table 1). While the general trend is for greater nitrate leaching at higher N inputs about 50% of these sites still retain most of the deposited N in the terrestrial ecosystem, even at some sites where N deposition exceeds 1.0 K eq ha^{-1} yr^{-1}.

A preliminary assessment of the factors which determine the seasonal pattern of nitrate levels was made using monthly data from sensitive, high-elevation lochs in Galloway, south west Scotland, where vegetation, soil type and N inputs were as similar as possible. Even under these circumstances significant differences were found in the pattern of nitrate leaching at these sites (Fig. 1), suggesting that each was at a different stage of nitrogen saturation. Stoddard (1994) suggested a four stage saturation classification (0-3) ranging from stage 0 when concentrations remained low all year, with little or no seasonality, to stage 3 when concentrations remained high all year, again with little or no seasonal change. The Galloway lochs appear to exhibit all the stages of saturation apart from stage 3 as shown by the distinct groups of lochs in Figure 1. Superficially the main differences between these systems appear to be soil depth, lake depth, flushing rate and lake to catchment area which suggests that hydrological and physical properties of systems may also be important.

Calculating Critical Load and Exceedance Values for Nitrogen

The following models are being used by the Freshwater Group to determine critical load and exceedance values for UK freshwater. Some preliminary maps are presented but at this early stage many caveats must be applied.





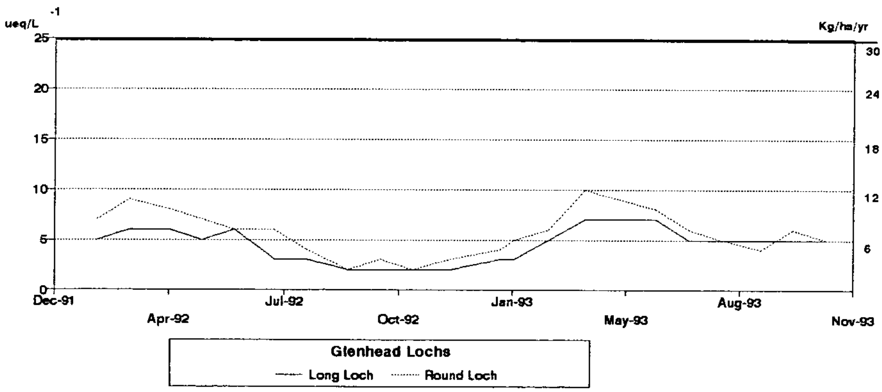


Figure 1 Seasonal patterns of nitrate run-off in Galloway lochs, south-west Scotland.

Steady-state Water Chemistry model (SSWC)

This method was developed by Kämäri *et al.* (1992) and uses the same methodology as that applied to sulphur

i.e. $$CL_{AC} = Q\ ([BC^*]_0 - [ANC]_{limit}) - BC^*_{dep} \quad (1)$$

This only differs from the sulphur value in that a lower BC_0 concentration is obtained to account for the extra base cation losses due to N leaching. The critical load values derived from equation (1) are effectively the true total acidity loading as they reflect both N and S leaching. One important advantage of this method is that N input data are not required to calculate exceedance values because nitrate leaching is assumed to equal N sources minus N sinks.

i.e. $$N_{leach} = N_{dep} - N_S \quad (2)$$

where N_S represents all sinks of N in the catchment.

The formulation in equation 2 represents a generalised situation where current N leaching is a consequence of long-term N deposition. Nevertheless it should be appreciated that current N leaching can change significantly in the short-term, independent of N deposition. This situation is usually caused by changes in the N dynamics of soils and vegetation, especially in managed catchments.

Consequently exceedance of the critical load of total acidity can be derived from

$$Ex_{AC} = S_{dep} - N_{leach} - BC^*_{dep} - CL_{AC} \quad (3)$$

No individual critical loads of N and S can be assigned by this method.

Provisional exceedance maps produced by this model for UK fresh waters (Fig. 2) are similar to those for sulphur indicating that N impacted sites are generally in the same areas of the UK as those affected by S inputs (i.e. Upland Wales, Pennines, Lake District and south west Scotland). Therefore, although the distribution of exceeded squares remains relatively unchanged the number of sites in the high exceedance band (> 1.0 $Keqha^{-1}yr^{-1}$) increases.

Diatom Critical Loads Model (DCL)

The DCL model is complementary to the SSWC model and is based on the general observation that diatom flora retained in sediment profiles show little or no change in species composition prior to the onset of anthropogenic acidification. Any shift to a more acidophilous diatom flora can be considered as a "point of change" which is analagous to exceedance of the critical load for that specific and usually very sensitive biological indicator. Development of the methodology for calculating critical loads is described by Battarbee *et al.* (1993) but essentially the model uses relationships between pre-acidification calcium concentrations in water (as a measure of site sensitivity) and present day sulphur and nitrogen loadings. This relationship was derived from 41 UK lakes which gave accurate diatom records and calibrated using logistic regression to calculate the probability of acidification for different calcium to loading ratios.At the optimum dicrimination, giving a probability of acidification of 50%, a ratio of 94:1 was found. The model is currently being adapted to provide critical loads, and critical load exceedances, for total acidity (sulphur and nitrogen). Exceedance values for total acidity require a measure of the fraction of deposited nitrogen leached into the surface waters. This is calculated from

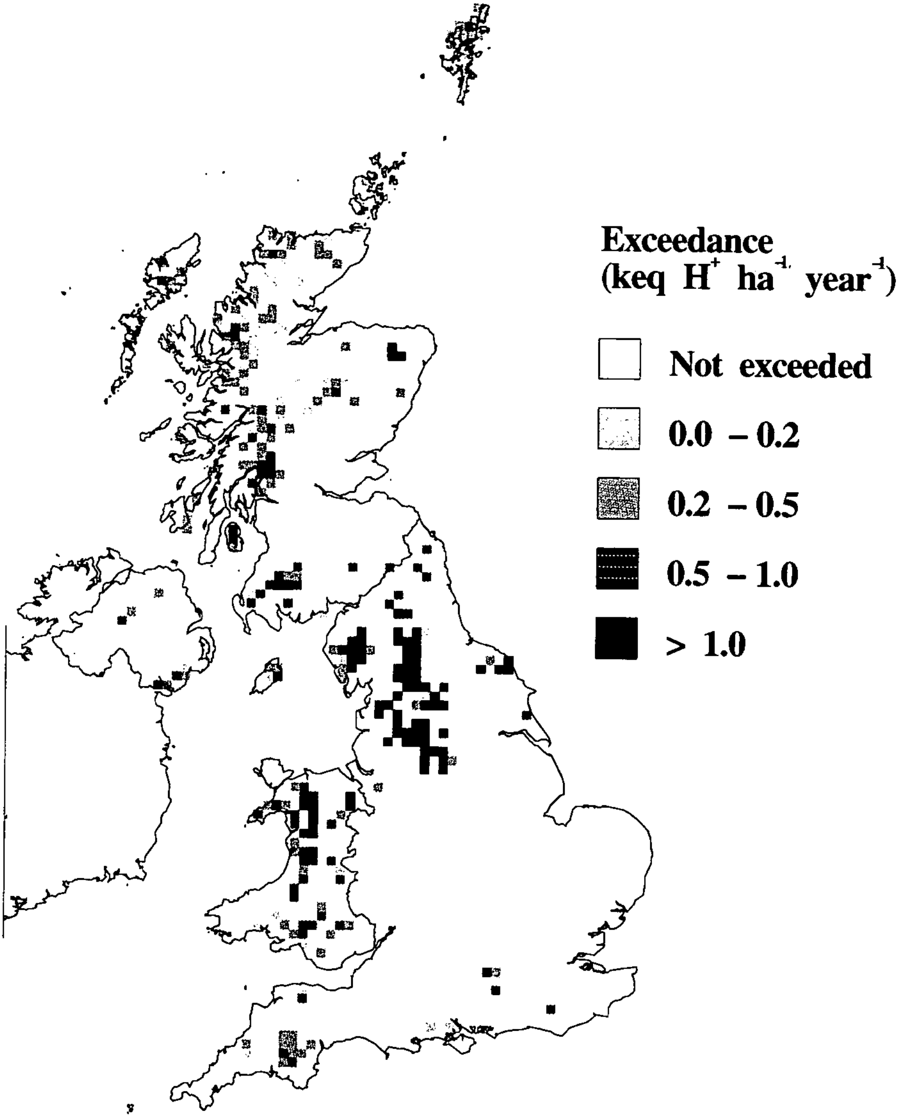


Figure 2: **Exceedance of critical loads for total acidity for UK freshwaters (revised SSWC model)**

the differences between the proportions of sulphate/nitrate in the water and sulphur/nitrogen deposition modelled for the site. In this way the fraction of the nitrogen deposition contributing to acidification can be added to the value of sulphur deposition to provide total "effective" acid deposition. The critical ratio used in the diatom model has also been recalibrated using total effective deposition rather than sulphur deposition, giving a ratio of 89:1. Critical loads for total acidity are calculated using this ratio.

A preliminary map of UK critical load exceedances for total acidity has been prepared using the diatom model (Fig. 3). Superficially this mirrors the exceedance map for sulphur deposition alone as the areas most severely impacted by nitrogen deposition are also those with high exceedances for sulphur. However, calculations of the increase in exceedance due to the inclusion of leached nitrogen deposition demonstrate that for large parts of the UK (e.g. Cumbria,the Pennines, Galloway) nitrogen deposition by itself could account for significant exceedances. This finding has implications not only for the regional extent of acidification, but also for the extent of recovery at these sites if only sulphur deposition is reduced.

First Order Acidity Balance (FAB) Model

The SSWC and DCL models only provide information on present day exceedances of critical loads. For estimates of potential future exceedances the FAB model should be used. This model is based on an acidity mass balance and includes rate-limited processes for denitrification and in-lake retention which are assumed to increase with increasing N inputs.

The full charge balance for a lake and its catchment can be described as follows:

$$N_{dep} + S_{dep} = fN_u + (1-r)(N_i+N_{de}) + rN_{ret} + rS_{ret} + BC_l - ANC_l \tag{3}$$

where

N_{dep}	= deposition of N
S_{dep}	= deposition of S
N_u	= net growth uptake of N by vegetation
N_i	= immobilisation of N in the catchment soils
N_{de}	= N denitrified in the catchment soils
N_{ret}	= in-lake retention of N
S_{ret}	= in-lake retention of S
BC_l	= base cations leaching from the catchment
ANC_l	= ANC leaching from the catchment
f	= fraction of forested area in the catchment
r	= lake: catchment area ratio

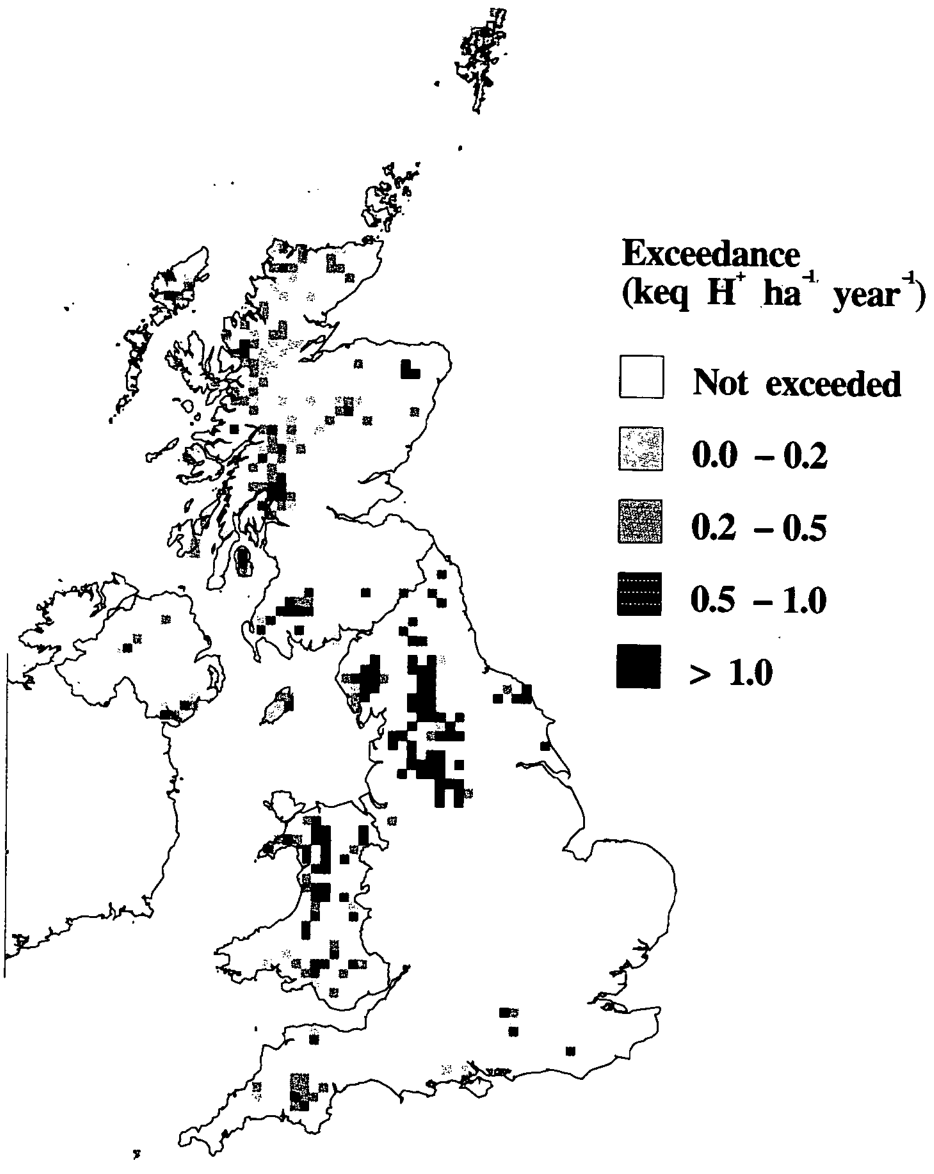


Figure 3: Exceedance of critical loads for total acidity for UK freshwaters (revised diatom model).

Full details of the development of this model are given by Kämäri *et al.* (1992) and Posch *et al.* (1993) and further modifications are described by Henriksen *et al.* (1993). The key derivations for mapping requirements are discussed below. By incorporating relationships for denitrification and in-lake retention equation (3) can be modified to give:

$$a_N N_{dep} + a_S S_{dep} = b_1 N_u + b_2 N_i + Q([BC^*]_0 - [ANC]) \tag{4}$$

where the dimensionless constants a_N, a_s, b_1 and b_2 are all smaller than one and depend on lake and catchment properties alone:

$a_N \quad = (1 - f_{de}(1-r))(1-\rho_N)$

$a_S \quad = 1-\rho_S$

$b_1 \quad = f(1-f_{de})(1-\rho_N)$

$b_2 \quad = (1-r)(1-f_{de})(1-\rho_N)$

By choosing an appropriate ANC limit, equation (4) then converts to the critical load expression:

$$a_N CL(N) + a_S CL(S) = b_1 N_u + b_2 N_i + Q([BC^*]_0 - [ANC]_{limit}) \tag{5}$$

Exceedance of critical load can now be calculated by subtracting the right side of equation (5) from the left side of equation (4)

$$Ex(N_{dep}, S_{dep}) = a_N N_{dep} + a_S S_{dep} - b_1 N_u - b_2 N_i - L_{crit} \tag{6}$$

where $\qquad L_{crit} = Q([BC^*]_0 - [ANC]_{limit})$

which is the same formulation as the SSWC model.

If the potential nitrate leaching is required (for example, to compare with current N leaching or to insert in the SSWC model to calculate future exceedances) this can be calculated by subtracting sources and sinks of N.

i.e. $$N_l = a_N N_{dep} - b_1 N_u - b_2 N_i \tag{7}$$

Alternatively the critical load for nutrient nitrogen can be calculated by fixing N_i at a value which for any system, would not cause eutrophication.

$$CL_{nut}(N) = (b_1 N_u + b_2 N_i + N_{l,crit})/a_N \tag{8}$$

Because of the substantial data requirements for different catchment types no provisional maps have yet been produced using this model. A full description of the updated version of the FAB model is given in Posch.

A few general points should be emphasised when comparing the output from these models.

1. The potential nitrate leaching derived from the FAB model may not be directly transferable to the SSWC model because the former model relates to leaching below the rooting zone while the latter (SSWC model) uses nitrate values for water which has travelled through all soil and geological pathways.

2. The minimum critical load of nitrogen $CL_{min}(N)$ represents the point where nitrate will begin to leach into freshwater (i.e. by setting $N_{l,crit}$ at zero in equation (8). Conversely the maximum critical load for nitrogen $CL_{max}(N)$ equates to $CL_{min}N + L_{crit}$.

3. The key input parameter in the SSWC model is nitrate leaching, therefore to account for seasonality some estimate of a flow weighted mean value should be used.

4. In the FAB model the key parameters are N_u (nitrogen growth uptake by vegetation) and even more importantly, N_i (nitrogen immobilisation in soils).

Accurate estimates of these parameters are urgently required for upland UK catchments of different soil and vegetation type.

Level II Analysis (Dynamic Models)

The requirements for a dynamic modeling approach to nitrogen is perhaps even more urgent than for sulphur because of the complexity of nitrogen dynamics in the terrestrial ecosystem. Predicting long-term changes requires a model which incorporates most of the key time-dependent variables. For sulphur the Freshwater Group used the MAGIC model to assess the impacts of different deposition scenarios while for nitrogen a derivative of MAGIC has been developed.

MAGIC-WAND (MAGIC-With Aggregated Nitrogen Dynamics) builds directly on MAGIC and allows the main fluxes and transformations of nitrogen to be independently specified at each time-step. MAGIC-WAND has been specifically developed for wide application and scenario assessment. It maintains the sulphur based chemistry dynamics of MAGIC and considers reduced and oxidised nitrogen species. The model requires specification of nitrification, mineralisation, fixation and denitrification rates and changes in these fluxes through time. Plant uptake is non-linear and dependent upon external nitrogen concentrations. A wide range of data describing these rates and fluxes have been reported and are reviewed to aid in model calibration.

The main sensitivity of the model lies in the selection of the parameters which describe the hyperbolic uptake function. Literature data can provide ranges for these values but specific catchment related values are not obtainable since the model is conceptual. Selection of uptake parameters must reflect catchment vegetation and vegetation change through time. Further observational and experimental work to determine nitrogen fluxes and dynamics at different ecosystems is required to facilitate site specific and regional model applications. It is already clear that the data requirements for validation of nitrogen models is extensive and will require detailed knowledge of the nitrogen status of UK catchments.

Details of recent developments in nitrogen models and their use in calculating critical loads for nitrogen are reported by Ferrier *et al.* (1995) in this volume.

Conclusions and Recommendations

If the mass balance approach is the prefered method for mapping critical loads and exceedances for the major ecosystem components, then the following studies are recommended, bearing in mind that many upland catchments are leaking nitrate and may have already reached a critical saturation stage.

Priorities are:

1. To establish a programme of intensive nitrogen monitoring at a range of UK sites (e.g. Galloway, Pennines, North Wales, SW Scotland and N Scotland) to assess spatial and temporal variability in relation to N deposition and catchment characteristics so that appropriate N leaching values can be obtained.

2. To determine the key catchment conditions that lead to N leaching by establishing a range of key reference sites where simultaneous measurements of water, soils, vegetation and N deposition can be made. This information is vital for the development of dynamic models that simulate N behaviour in catchments.

3. To assess the impact of elevated N concentrations on the biology of upland waters with regard to acidification, eutrophication and species composition changes. Information on the extent of N limitation (as opposed to P) in upland waters is essential and this may require N manipulation experiments to speed up the interpretation of biological impacts.

4. Because many upland sites appear to be extremely sensitive and exceeded by nitrogen alone it may be appropriate to make more extensive use of freshwater maps for N protocols.

Acknowledgements

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References

Battarbee, R.W., Allott, T.E.H., Kreiser, A.M. and Juggins, S. (1993) Setting critical loads for UK surface waters: the diatom model. In: Critical loads: concept and applications. Eds: M. Hornung and R.A. Skeffington, 99-102. ITE symposium 28 HMSO, London.

Ferrier, R.C., de Vries, W. and Warfvinge, P. (1995) The use of dynamic models for the determina tion of critical loads for nitrogen - developments since Lökeberg. (This volume).

Harriman, R. and Christie, A.E.G. (1993) Evaluation of the steady-state water chemistry method for surface waters. In: Critical loads: concept and applications. Eds. M.Hornung and R.A.Skeffington 103-108 ITE Symposium 28 HMSO, London.

Henriksen, A. Forsius, M. Kämäri, J., Posch, M. and Wilander, A. (1993) Exceedance of critical loads for lakes in Finland, Norway and Sweden: reduction requirements for nitrogen and sul phur deposition. Report 32/1993 NIVA report for Nordic Council of ministers. 46pp.

Kämäri, J., Jeffries, D.S., Hessen, D.O., Henriksen, M., Posch, M. and Forsius, M. (1992) Nitrogen critical loads and their exceedance for surface waters In: Grennfelt, P. and Thörnelöf, E. (Eds). Critical loads for nitrogen Nord 1992-41 Nordic Council of Ministers, Copenhagen, Denmark pp161-200.

Posch, M., Forsius, M. and Kämäri, J. (1993) Critical loads of sulphur and nitrogen for lakes. I: Model description and estimation of uncertainty. Water, Air and Soil Pollution 66. pp173-192.

Keynote Paper - **An Empirical Model of Critical Acidity Loads for Surface Waters based on Palaeolimnological Data**

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Introduction

The clear link between acid deposition and surface water acidification suggests that the best method for controlling and reversing acidification is the elimination or reduction of deposition. The first United Nations Economic Commission for Europe (UNECE) sulphur protocol encouraged member states to reduce national emissions by a minimum of 30% by 1993 based on a start date of 1980. However, this approach does not specifically target high sulphur sources or protect highly sensitive areas. Dissatisfaction with such "across the board" policies encouraged the UNECE to adopt a "critical loads" approach for the second sulphur protocol signed in Oslo in June,1994.

In the critical loads approach emission reductions are calculated according to the need to reduce acid deposition at sensitive acidified sites to below a threshold, or critical load, where ecological "damage" should not occur. The main modelling method used to derive critical loads for freshwaters is the steady-state chemistry model (Henriksen *et al.* 1992). Here we present an alternative empirical model based on palaeolimnological data.

The Diatom Critical Loads Model for Freshwaters

Diatom assemblages in cores from acidified lakes usually show that prior to acidification the diatom flora, and therefore water chemistry, changed little. The point of acidification is indicated by a shift towards a more acidiphilous diatom flora. A typical example of this pattern is shown in Figure 1. Diatoms are amongst the most sensitive indicators of acidification in freshwater ecosystems, hence it can be argued that the point of change in the diatom record indicates the time at which the critical load for the site was exceeded.

Although the acid loading at the time of the initial acidification is not known, the critical load of a particular site can be inferred from the comparison of the acidification status of 41 UK sites with site sensitivity and current deposition loadings (Battarbee *et al.* 1995). If lake-water calcium values are taken to be an indicator of sensitivity to acidification, acidified sites (as indicated by the sediment diatom record) are those where the calcium to sulphur deposition ratio is less than 94:1 (Battarbee *et al.* 1995). This ratio, determined by logistic regression, can be used to define critical sulphur loads for any site including streams. Critical loads values are calculated using pre-acidification calcium concentrations derived using the F-ratio (Henriksen & Brakke 1988). For example, the critical sulphur load for a site with a $[Ca^{2+}]_0$ value of 40 eq l^{-1} is approximately 0.43 keq S ha^{-1} yr^{-1}.

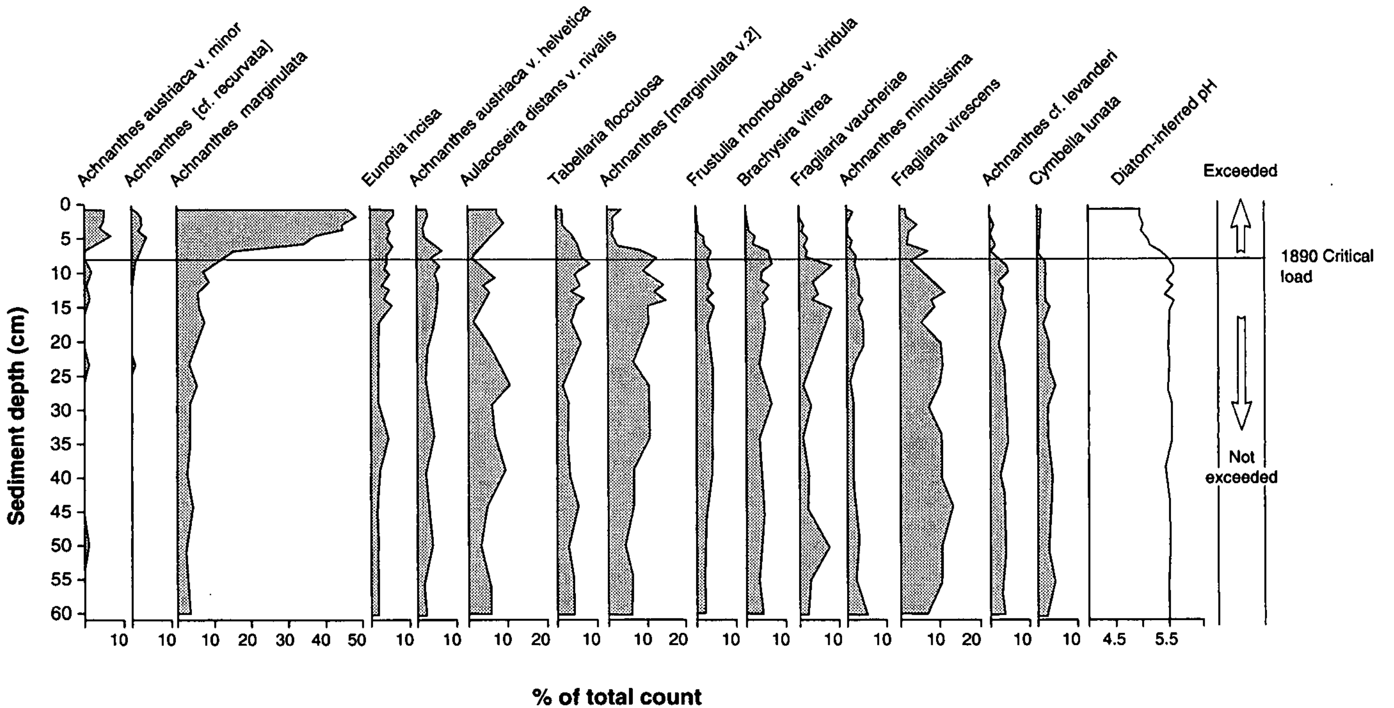

Figure 1 **Diatom diagram from Lochnagar, Scotland, showing point of critical load exceedance in the late 19th century (from Battarbee (*et al.* 1993)**

Mapping Critical Loads for Sulphur in the UK

As part of the UK Department of the Environment's critical loads programme, a data-set of water chemistry has been collected which contains a sample from the most sensitive water body (as predicted from soils and geological data) in each 10 x 10 km grid square in the UK (Kreiser *et al.* 1993). The diatom critical loads model has been applied to this data-set to provide a map of critical sulphur loads (Harriman *et al.* 1995). Critical loads are low in areas sensitive to freshwater acidification such as north-west Scotland, Galloway, north and central Wales and the Pennines.

If the sulphur critical load at a site is less than the current sulphur deposition, then the critical sulphur load is exceeded. Current UK sulphur deposition data have been used to produce a map of critical sulphur load exceedances (Allott *et al.* 1995). The main areas of exceedance in this map are those where problems of acidification have been previously documented (e.g. Galloway, Cumbria, North Wales). However, the map also predicts critical load exceedances in some areas where there is less documentation of freshwater acidification (e.g. the north-west of Scotland). Maps of exceedance have also been drawn up using different future emission and deposition scenarios to be used for policy making on both a national and international level.

Calculating Critical Loads for Total Acidity

The diatom model is currently being adapted to provide critical loads, and critical load exceedances, for total acidity (sulphur and nitrogen). Exceedance values for total acidity require a measure of the fraction of deposited nitrogen leached into the surface waters. This is calculated from the differences between the proportions of sulphate/nitrate in the water and sulphur/nitrogen deposition at the site (Eq. 1). In this way the fraction of the nitrogen deposition contributing to acidification (a_N) can be added to the value of sulphur deposition to provide total "effective" acid deposition. This method assumes an equilibrium between sulphur deposition and sulphate in the water, and only applies to sites with no catchment nitrogen inputs.

$$a_N = \frac{S^*_{dep}:N_{dep}}{[SO_4^{2-}]^*_t:[NO_3]_t} \tag{1}$$

The diatom model has been recalibrated for total acidity loads by substituting total effective deposition for sulphur deposition. The resulting critical ratio of 89:1 is slightly lower than when sulphur alone is considered. Critical loads for total acidity are calculated according to equation 2. Critical load exceedances can then be calculated according to equation 4.

$$CL = \frac{[Ca^{2+}]_0}{89} \tag{2}$$

$$[Ca^{2+}]_0 = [Ca^{2+}]^*_t - F_{Ca_0}([SO_4^{2-}]^*_t + [NO_3^-]_t - [SO_4^{2-}]^*_0 - [NO_3^-]_0) \tag{3}$$

$$CL_{ex} = CL - S^{*}_{dep} - a_N(N_{dep}) \quad (4)$$

A preliminary map of UK critical load exceedances for total acidity has been prepared using this model. Superficially this reflects the exceedance map for sulphur deposition alone, as the areas most severely impacted by nitrogen deposition are also those with high exceedances for sulphur. However, calculations of the increase in exceedance due to the inclusion of leached nitrogen deposition demonstrate that for large parts of the UK (e.g. Cumbria, the Pennines, Galloway) nitrogen deposition by itself could account for significant exceedances. This finding has implications not only for the regional extent of acidification, but also for the extent of recovery at these sites if only sulphur deposition is reduce.

Conclusions

Palaeolimnological studies using diatoms have been used to define critical loads and critical loads exceedances for freshwater lakes and streams. These critical loads represents the first ecological response to acidification, or the "baseline" critical load for the site, and are therefore complimentary to the steady-state water chemistry method of Henriksen which is based on a fixed ANC value. To date, the diatom model has been applied to more than 1500 sites in the UK. Current work is adapting the diatom model to calculate critical loads exceedances for total acidity. Preliminary maps of total acidity exceedance emphasise the importance of nitrogen deposition.

The diatom model has been calibrated using sites and data from the United Kingdom. However, a major advantage of the approach is that predictions for any lake site can be validated by analyzing diatoms in a sediment core. In this way the applicability of the model to sites outside the UK can be tested.

References

Allott, T.E.H., Battarbee, R.W., Curtis, C., Harriman, R., Hall, J., Bull, K. and Metcalfe, S.E. (1995) Sulphur deposition, present and projected critical load exceedances. In CLAG Freshwaters: *Critical Loads of Acid Deposition for UK Freshwaters.* Report of the Critical Loads Advisory Group, sub-group on Freshwaters, for the UK Department of the Environment, 60-76.

Battarbee, R.W., Allott, T.E.H., Juggins, S. and Kreiser, A.M. (1995) Estimating the base critical load: the diatom model. In CLAG Freshwaters: *Critical Loads of Acid Deposition for UK Freshwaters.* Report of the Critical Loads Advisory Group, sub-group on Freshwaters, for the UK Department of the Environment, 20-27.

Harriman, R., Allott, T.E.H., Battarbee, R.W., Curtis, C., Hall, J. and Bull, K. (1995) Critical load maps for UK freshwaters. In CLAG Freshwaters: *Critical Loads of Acid Deposition for UK Freshwaters.* Report of the Critical Loads Advisory Group, sub-group on Freshwaters, for the UK Department of the Environment, 47-59.

Henriksen, A. and Brakke, D.F. (1988) Sulfate deposition to surface waters. Estimates of critical loads for Norway and the Eastern United States. *Environ.Sci.Technol.* 22, 8-14.

Henriksen, A., Kämäri, J., Posch, M. and Wilander, A. (1992) Critical loads of acidity: Nordic surface waters. *Ambio* 21, 356-363.

Kreiser, A.M., Patrick, S.T. and Battarbee, R.W. (1993) Critical acidity loads for UK freshwaters - introduction, sampling strategy and use of maps. In Hornung, M. & Skeffington, R.A. (Eds) *Critical Loads: Concept and Applications.* ITE Symposium No.28, HMSO:London, 94-98.

Keynote Paper - Critical Loads for Nitrogen: Surface Waters

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Introduction

In the Lökeberg meeting document (Grennfelt and Thörnelöf 1992) two models, one empirical and one process oriented were presented, for calculating critical loads of acidifying deposition (both S and N) to surface waters. The first model, the Steady State Water Chemistry Method (SSWC) model, enables the calculation of critical loads of acidity and present exceedances of incoming total acidity (including nitrogen) over the critical loads. The process-oriented model, the First-order Acidity Balance (FAB) Model allows the simultaneous calculation of critical loads of acidifying N and S deposition and their exceedances. The FAB Model is based on the steady-state mass balance principle widely used in many models for computing critical loads for forest soils.

This paper presents some considerations on the importance of nitrogen for the critical load exceedance of acidity in Norway and applications of the modified SSWC Method and the FAB Model to Norwegian surface waters.

The Modified SSWC Method

At Lökeberg, the Steady State Water Chemistry Method (SSWC) was modified to include both S and N acidity, in such a way that present N-leaching (N_{leach}) was considered in the calculation of critical load exceedance (Present Ex_{AC}).

$$\text{Present } Ex_{AC} = S_{dep} + N_{leach} - BC^*_{dep} - CL_{AC} \tag{1}$$

where S_{dep} is present S-deposition, $N_{leach} = N_{dep} - N_{upt} - N_{imm} - N_{den} - N_{ret}$ (N_{upt}: harvested N, N_{imm}: immobilisation of N, N_{den}: denitrified N and N_{ret}: inlake N retention), BC^*_{dep} is the seasalt-corrected atmospheric Base Cation deposition and CL_{AC} is the critical load of acidity (Henriksen *et al.* 1992).

The predict future situations, we must know the magnitude of N_{imm}, N_{upt}, N_{den} and N_{ret}, of which the latter two are rate-dependent parameters. The SSWC Model assumes there is no rate limitation. The First-order Acidity Model (FAB), however, incorporates rate-limited descriptions of denitrification and in-lake N-retention (Henriksen *et al.* 1993).

The FAB Model

When considering the effects of both sulphur and nitrogen simultaneously, one cannot expect the obtain unique critical loads of S and N, since a reduction in the deposition of sulphur might allow a higher deposition of acidifying nitrogen compounds without causing 'harmful effects'. From an acidity balance one can derive the following equation, describing the trade-off between sulphur and nitrogen critical loads (Henriksen *et al.* 1992):

$$a_N\ CL(N) + a_s\ CL(S) = b_1\ N_{upt} + b_2\ N_{imm} + BC_{le,crit} \qquad (2)$$

where N_{upt} and N_{imm} are the net growth uptake (harvested N) and immobilization of N and a_N, a_s, b_1 and b_2 are dimensionless constants depending on lake and catchment properties alone:

$$a_N = (1-f_{de}(1-r))(1-\rho_N)$$
$$a_s = 1-\rho_s)$$
$$B_1 = f(1-f_{de})(1-\rho_N)$$
$$b_2 = (1-r)(1-f_{de})(1-\rho_N)$$

where f is the fraction of forest area within the catchment and r is the lake catchment area ratio. In deriving Eq. 2 not only the uptake and immobilization of N have been taken into account, but also denitrification and the in-lake retention of N and S, all three as linear functions of the net input of N (resp. S) with proportionality coefficients F_{de}, ρ_N and ρ_s, leading to the coefficients above. The in-lake retention coefficient ρ_N is modelled by a kinetic equation (Kelly *et al.* 1987):

$$\rho_N = s_N/(Q/r + s_N)$$

where s_N is the net mass transfer coefficient for N (m yr^{-1}). An analogous equation holds for ρ_s.

Finally, the critical Base Cation leaching from the catchment is estimated from water quality data by the steady-state model introduced by Henriksen *et al.* (1992):

$$BC_{le,crit} = Q\ ([BC]_0 - [ANC]_{limit})\ (3)$$

In addition to Eq. 2 the critical load of S is limited by the following constraint:

$$CL(S)\quad Clmax\ (S) = BC_{le,crit}/a_s$$

Eq. 2, together with this constraint, determines the so-called critical load function (Figure 1), separating the N- and S-deposition values which cause 'harmful effects' (exceedance) from those which do not (non-exceedance).

As mentioned above, unique critical loads for S and N cannot be specified: however, this may be an advantage, since it allows determination of (cost)-optimal deposition reductions. If the deposition of one of the compounds is fixed (prescribed), the critical load for the other can be computed from Eq. 2.

In addition to the parameters needed in the SSWC-model (Eq. 1), this model needs information on the following parameters: N_{up} f, N_{imm} f_{de} s_N, s_s and r. Data on N_{upt} and f have been obtained from the national forest research institutes; the value for N_{imm} (2 kg N ha^{-1} yr^{-1}) is at the lower end of a recommended range of values (Downing *et al.* 1993). f_{de} varies between 0.1 and 0.8 depending on the fraction of peatlands in the catchment; values for s_N (5 m yr^{-1}) and s_s (0.5 m/yr) have been taken from the literature (Dillon and Molot 1990, Baker and Brezonik 1988); finally, r has been taken from the lake surveys.

Nitrogen "Saturation"

Most terrestrial systems do retain N strongly. Aber *et al.* (1989) have defined **nitrogen saturation** as: *"the state at which the availability of ammonium and nitrate is in excess of the total combined plant and microbial nutrient demand, as manifest by leaching of significant amount of nitrate from the catchment".*

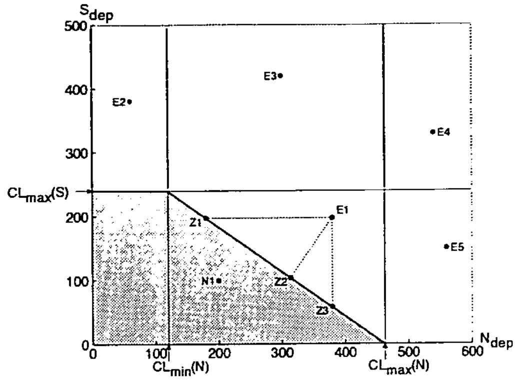

Figure 1: Example of a critical load function for an aquatic ecosystem. The shaded area indicates those N and S depositions not causing exceedance. Also shown are 3 possible ways to reach zero exceedance (Z1,Z2, Z3) from a given N and S deposi tion E1. Excess amounts of nitrogen may also cause eutrophication of aquatic ecosystems, as indicated in the diagram. Units: meq/ha/yr.

Catchments exhibiting significant NO_3^- leaching occur in areas receiving elevated S and N deposition (Kämäri *et al.* 1992). Stoddard (1994) has defined symptoms of N saturation through recognizable stages of long-term and seasonal patterns of lake- and streatwater NO_3^- concentrations that reflect the changes in rates and relative importance of N-transformations as these watersheds become more N-sufficient (see Stoddard and Traaen, this volume). The early stages of N saturation are marked by increases in the severity and frequency of NO_3^- episodes. The later stages are marked by elevated NO_3^- concentrations through the year.

In Norway NO_3^- concentrations in lakes in southernmost part of Norway doubled from 1975 to 1986 (Henriksen *et al.* 1988), all receiving high S and N deposition. In 1993 four of these lakes were sampled every second week (Table 1, Figure 2). All lakes show low pH, high labile aluminium as well as high NO_3^- concentrations and no significant changes in nitrate and total nitrogen during the year. Also in other areas in Europe we find similar patterns, as exemplified by data from two lakes in the Tatra Mountains (Figure 3).

According to Stoddard's classification these patterns should indicate N-saturation in the terrestrial soil in the catchments of these lakes. Typically, the catchment for all four lakes contain thin and patchy soil and no forest. The NO_3^- flux out of the lake is clearly related to the lake to catchment size ratio i.e. the flux is higher at lower retention time (Figure 4a). This indicates significant contributions of nitrate from direct NO_3^- deposition on lake surface. The nitrate flux is higher at lower Base Cation flux (Figure 4b), which in turn relates to the weathering rate (soil thickness). NH_4^+ concentrations, however, are very low during the year (less than 20 g N l^{-1}). Although vegetation is sparse in these areas, ammonia is completely held in the catchment and the lake. The high leaching rate of nitrate indicates that mechanisms other than vegetational uptake are involved. Possibly the nitrate input is too high to completely participate in the internal N-cycling in the soil and vegetation. This could indicate that the hydrological conditions in these areas are important for nitrate retention in the lake catchments. The nitrogen retention in the lakes themselves appears to be insignificant (Hindar and Henriksen, this volume). Thus, the N leaching appears to be more directly related to the amount of N deposition than the N-retention in these catchments.

Table 1: *Average chemistry of four lakes in southernmost Norway sampled every second week for 1993 (Henriksen, pers comm)*

Lake No.	pH	Ca mg l^{-1}	Mg mg l^{-1}	Na mg l^{-1}	K mg l^{-1}	Cl mg l^{-1}	SO_4 mg l^{-1}	NO_3 $\mu gN\ l^{-1}$	Tot-N $\mu gN\ l^{-1}$	Inorg-Al $\mu g\ l^{-1}$	TOC mgC l^{-1}	Runoff 1 $km^2\ s^{-1}$
1	4.63	0.85	0.59	5.36	0.21	10.1	3.5	153	312	214	2.09	48
2	4.37	0.51	0.69	5.90	0.24	10.9	3.6	493	643	266	1.07	38
3	4.65	0.59	0.72	6.39	0.21	11.5	3.7	399	496	360	0.52	45
4	4.60	0.69	0.98	8.40	0.24	15.2	4.5	460	546	442	0.63	45

Results and Discussion

SSWC Method

The modified SSWC-method applied to Norway increases the exceeded area from 35 to 39% (4%) only, but the amount of excess acid is 19% higher when present nitrate leaching is included (Table 2). The new sulphur protocol, which was signed in Oslo, Norway in June 1994 implies a 55-60% reduction in sulphur deposition in Norway in the year 2010 relative to 1990 deposition. Without any reductions in nitrogen deposition and no change in NO_3 leaching, the excess acid due to nitrogen deposition will then amount to 42% of the total excess acid.

The FAB Model

The FAB-model has been applied to the Norwegian critical load database for surface water. At present deposition of S and N, the critical load of acidity is exceeded in approximately 48% of Norway (Figure 5a). The sulphur deposition scenario of the new sulphur protocol in 2010 with present nitrogen deposition and leaching reduces the exceeded area in Norway to about 38% (Figure 5b).

At present deposition rates of S and N, equal amounts of reduction in the deposition of either sulphur or nitrogen will giver similar reductions in the areas of Norway (Figure 5a), exceeding critical loads for acidity. Since the Second Sulphur Protocol has now been signed, the effects of reductions in nitrogen deposition can be considered in a future (year 2010) scenario (Figure 5b). In this situation, further reductions in sulphur will not give as large effects as reductions in nitrogen deposition. Reductions in nitrogen deposition will thus become more important when the Second Sulphur Protocol is implemented, especially if the present load leads to increasing nitrate leaching. Whether increased nitrate leaching will occur at present, the load is not evident from present available data. It will, however, be very important to evaluate this further, and for this purpose, the maintenance of long term monitoring program is very important.

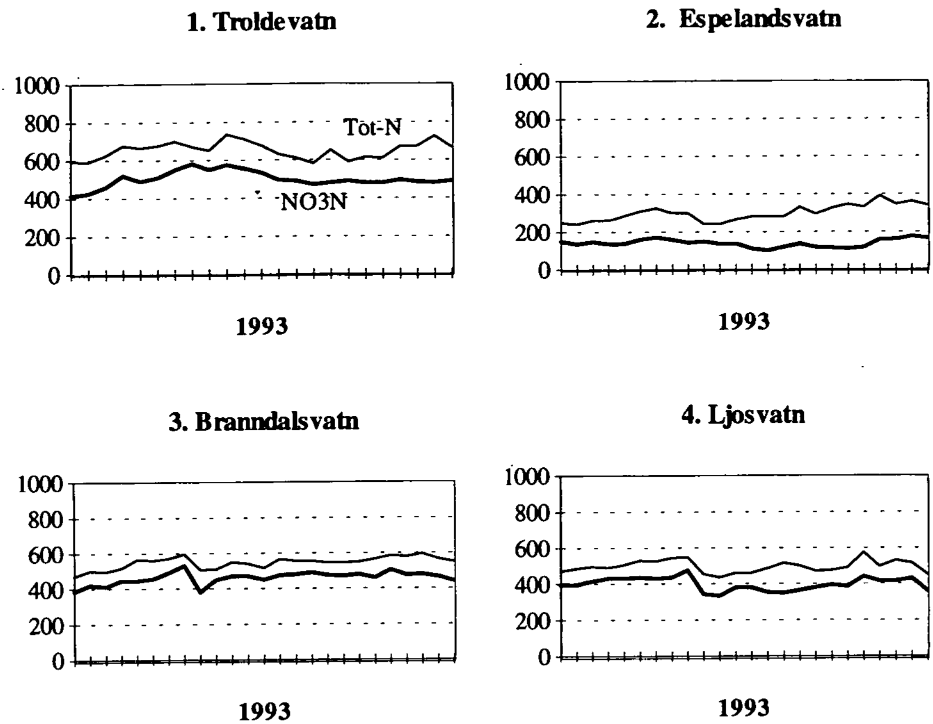

Figure 2: Seasonal variations in nitrate and total nitrogen in four coastal lakes in southernmost Norway (sampled every second week) (Henriksen, unpublished results).

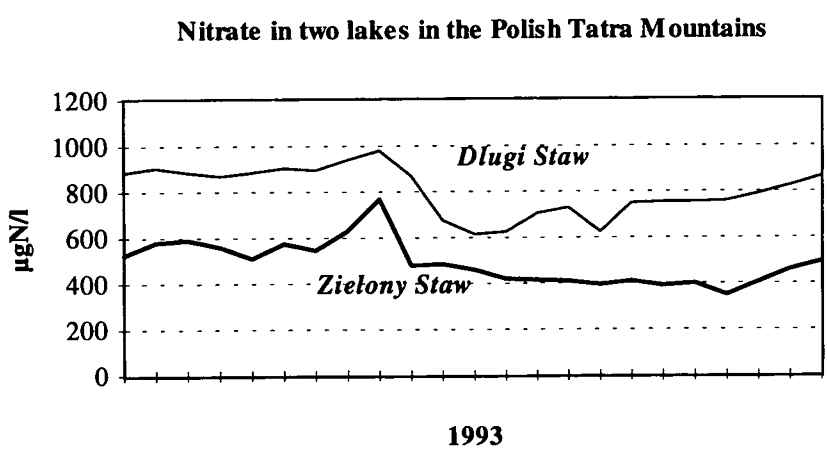

Figure 3: Seasonal variations in nitrate in two lakes in the Polish Tatra Mountains (sam pled every second week) (Henriksen, unpublished results

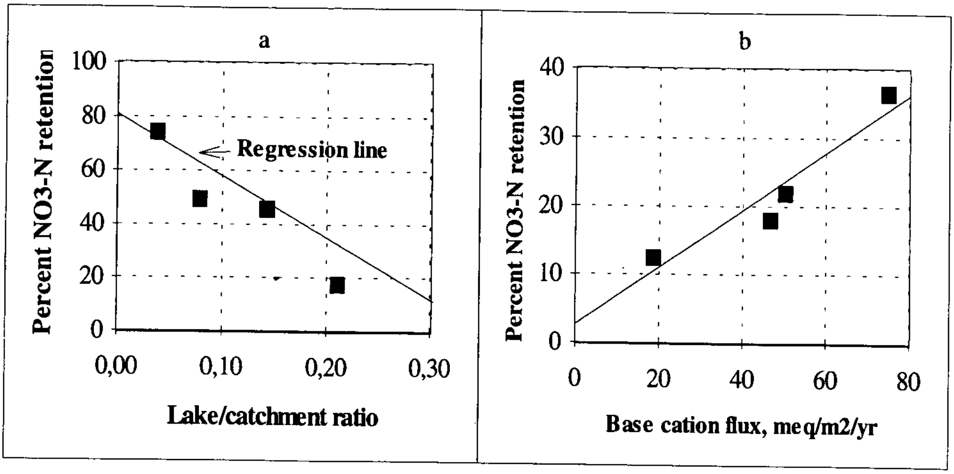

Figure 4: Ratio of lake to catchment area (a) and flux of base cations (b) vs. percent nitrate retention in the lake and catchments of the four lakes (Figure 2).

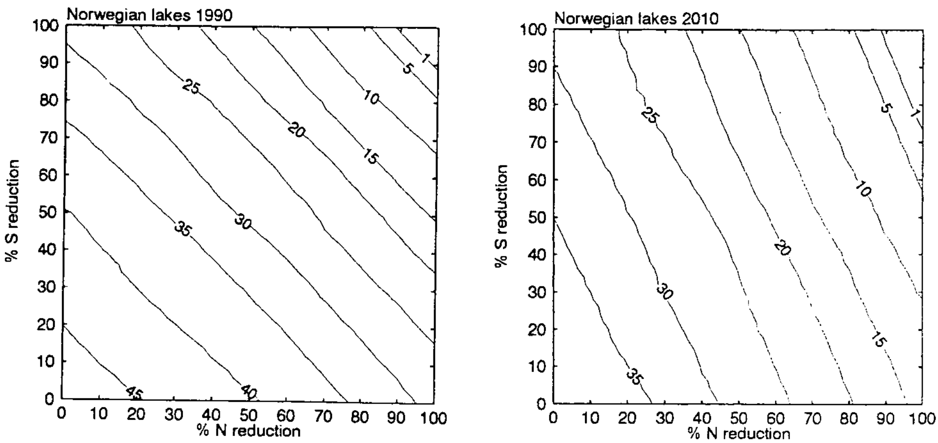

Figure 5. Isolines of the percentage of area of Norway where critical loads of acidity to surface waters are exceeded for every combination of percent reduction from (a) present depo sition of S and N, and (b) present N deposition and S deposition in year 2010 according to the 2nd Sulphur Protocol.

Table 2: Acid in excess of critical loads in Norway distributed on S- and present N leaching for two scenarios.

Scenario	Excess acid, percent	
	Sulphur	Nitrogen
1990	81	19
2010	58	42

Conclusions

In large parts of southern Norway, nitrogen deposition is apparently too high as indicated by high present leaching of nitrate. At present deposition of S and N, equal amounts of reduction in deposition of either sulphur or nitrogen will give similar reductions in the exceeded areas in Norway. Reductions in nitrogen deposition will, however, become more important when the Second Sulphur Protocol has been implemented and S deposition is reduced.

References

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M. (1989) Nitrogen saturation in northern forest ecosystems: hypothesis and implications. *Bioscience* **39**: 378-386.

Baker, L.A. and Brezonik, P.L. (1988) Dynamic model of in-lake alkalinity generation. *Wat. Resour. Res.* **24**: 65-74.

Dillon, P.J. and Molot, L.A. (1990) The role of ammonium and nitrate in the acidification of lakes and forested catchments. *Biochem* **11**: 23-43.

Downing, R.J., Hettelingh, J.P. and de Smet, P.A.M. (1993) Calculation and Mapping of Critical Loads for Europe: Status Report 1993. Co-ordination Centre for Effects, National Institute of Public Health and Environmental Pollution, Bilthoven, The Netherlands. RIVM Report No 259101003.

Grennfelt, P. and Thörnelöf, E. (eds.) (1992) *critical Loads for Nitrogen.* Nord. 1992: 41, Nordic Council of Ministers, Copenhagen, Denmark.

Henriksen, A., Lien, L., Traaen, T.S., Sevaldrud, I.S. and Brakke, DF. (1988) Lake acidification in Norway - present and predicted chemical status. *Ambio* **17**: 259-266.

Henriksen, A., Forsius, M., Kämäri, J., Posch, M. and Wilander, A. (1993) Exceedance of Critical Loads for Lakes in Finland, Norway and Sweden: Reduction requirements for Nitrogen and Sulphur Deposition. *Acid Rain Research Report 32/1993.* Norwegian Institute for Water Research (NIVA), Oslo, Norway.

Henriksen, A., Kämäri, J., Posch, M. and Wilander, A. (1992) Critical loads of acidity: Nordic surface waters. *Ambio* **21**: 356-363.

Kelly, C.A., Rudd, J.W.M., Hesslin, R.H., Schindler, D.W., Dillon, P.J., Driscoll, C.T., Gherini, S.A. and Heskey, R.H. (1987) Prediction of biological neutralization in acid sensitive lakes. *Biogeochem* **3**: 129-140.

Kämäri, J., Jeffries, D.S., Hessen, D.O., Henriksen, A., Posch, M. and Forsius, M. (1992) Nitrogen Critical Loads and their Exceedance for Surface Waters. In: Grennfelt, P. and Thörnelöf, E. (eds.) 1992. *Critical loads for nitrogen*, Nord 1992: 41, Nordic Council of Ministers, Copenhagen, Denmark.

Stoddard, J.L. (1994) Long-term changes in Watershed Retention of Nitrogen. Its causes and aquatic consequences. In: L.A. Naker (ed.). *Environmental Chemistry of Lakes and Reservoirs.* Advances in Chemistry Series No 237, American Chemical Society, Wash. D.C.

Keynote Paper - Critical Loads for Aquatic Ecosystems

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Introduction

When considering the effects of both sulphur and nitrogen simultaneously, one cannot expect to obtain unique critical loads of S and N, since a reduction in the deposition of sulphur might allow a higher deposition of acidifying nitrogen compounds (and vice versa), without causing 'harmful effects'. In this paper, a method is described which allows the estimation of the combined critical loads of S and N for aquatic ecosystems. The method has been called the First-order Acidity Balance (FAB) model and has been described in Kämäri *et al.* (1992) and modified in Henriksen *et al.* (1993) and Downing *et al.* (1993).

The First-order Acidity Balance Model

The charge balance for an aquatic ecosystem (lake+catchment) reads (see Henriksen *et al.* 1993 for details):

$$N_{dep}+S_{dep} = fN_u+(1-r)(N_i+N_{de})+rN_{ret}+rS_{ret}+BC_l-ANC_l \qquad (1)$$

where

N_{dep}	=	deposition of N
S_{dep}	=	deposition of S
N_u	=	net growth uptake of N by vegetation
N_i	=	immobilization of N in the catchment soils
N_{de}	=	N denitrified in the catchment soils
N_{ret}	=	in-lake retention of N
S_{ret}	=	in-lake retention of S
BC_l	=	base cations leaching from the catchment
ANC_l	=	ANC leaching from the catchment
f	=	fraction of forested area in the catchment
r	=	lake:catchment area ratio

Note, that all quantities (except f and r) have to be given in equivalents (moles of charge) per unit area and time.

Denitrification is assumed to be proportional to the net input of N (De Vries *et al.* 1993):

$$N_{de} = \begin{cases} f_{de}(N_{dep}-N_i-N_u) & \text{for forested land} \\ f_{de}(N_{dep}-N_i) & \text{for open land} \end{cases} \qquad (2)$$

where f_{de} 1 is a constant denitrification fraction. This equation is based on the assumption that immobilization and growth uptake are faster processes than denitrification. Similarly, in-lake retention of N is assumed proportional to the net input of N to the lake:

$$rN_{ret} = \rho_N\left(N_{dep} - fN_u - (1-r)(N_i + N_{de})\right) \tag{3}$$

where the factor ρ_N is modelled by a kinetic equation (Kelly *et al.* 1987):

$$\rho_N = \frac{s_N}{s_N + z/\tau} = \frac{s_N}{s_N + Q/r} \tag{4}$$

where z is the mean lake depth, τ is the lake's residence time, Q is the runoff and s_N is the net mass transfer coefficient for N.

Furthermore, the net base cation leaching is computed as in the SSWC-model by $Q[BC^*]_0$ (see above) and can be estimated using the so-called F-factor from present-day water chemistry data (Brakke *et al.* 1990, Sverdrup *et al.* 1990). Replacing ANC_l by Q[ANC] and inserting the expression for denitrification, in-lake N-retention and an analogous expression for the in-lake S-retention ($rS_{ret}=\rho_S S_{dep}$ and $\rho_S=s_S/(s_S+Q/r)$) into Eq.1 yields

$$a_N N_{dep} + a_S S_{dep} = b_1 N_u + b_2 N_i + Q\left([BC^*]_0 - [ANC]\right) \tag{5}$$

where the dimensionless constants a_N, a_S, b_1 and b_2 are all smaller than one and depend on lake and catchment properties alone:

$$\begin{aligned} a_N &= \left(1 - f_{de}(1-r)\right)(1-\rho_N) \\ a_S &= 1-\rho_S \\ b_1 &= f(1-f_{de})(1-\rho_N) \\ b_2 &= (1-r)(1-f_{de})(1-\rho_N) \end{aligned} \tag{6}$$

Inserting values for N and S deposition into Eq.5 yields the concentration of ANC in the lake. Conversely, this equation can be used to derive critical loads for N and S, if one can relate [ANC] to 'harmful effects' of the chosen indicator organism. For a variety of aquatic organisms (fishes) thresholds for [ANC] have been determined (Lien *et al.* 1992). Inserting one of these $[ANC]_{limit}$'s, we obtain the following relationship between *CL(S)* and *CL(N)*:

$$a_N CL(N) + a_S CL(S) = b_1 N_u + b_2 N_i + Q\left([BC^*]_0 - [ANC]_{limit}\right) \tag{7}$$

The critical loads are limited by the following constraints:

$$CL(N) \leq \left(b_1 N_u + b_2 N_i + L_{crit}\right)/a_N =: CL_{max}(N) \tag{8}$$

and

$$CL(S) \le L_{crit}/a_S =: CL_{max}(S). \tag{9}$$

where we have introduced the following abbreviation:

$$L_{crit} := Q\left([BC^*]_0 - [ANC]_{limit}\right) \tag{10}$$

Furthermore, if

$$N_{dep} \le \left(b_1 N_u + b_2 N_i\right)/a_N =: CL_{min}(N) \tag{11}$$

all N is consumed by uptake and immobilization, and sulphur can be considered alone. The relationship between depositions and critical loads, given by the above equations, is illustrated in Fig.1. The thick lines indicate all possible pairs of critical loads of N and S acidity, and it has been named the **critical load function**.

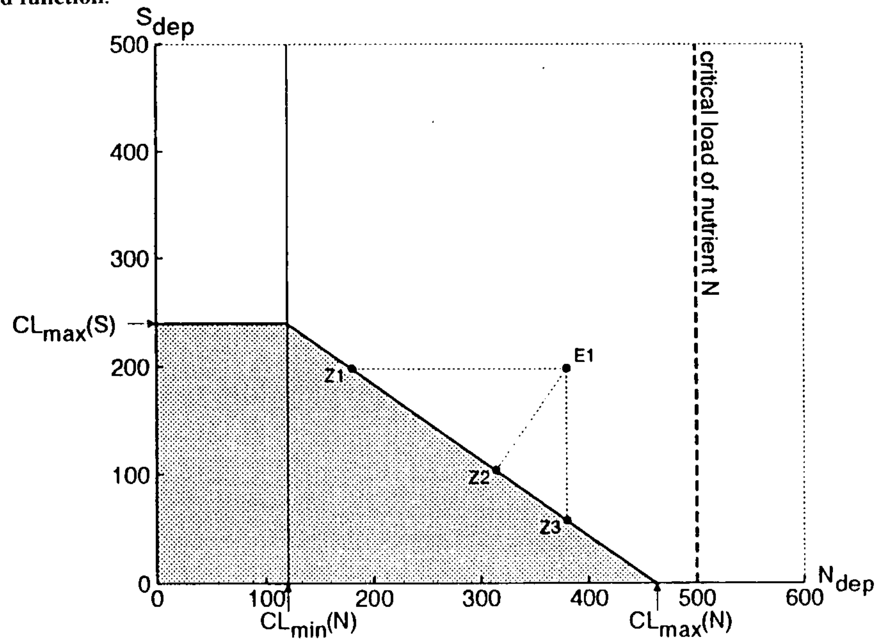

Figure 1 **Example of a critical load function for an aquatic ecosystem. The shaded area indicates those N and S deposition not causing exceedance. Also shown are 3 possibilities to reach zero exceedance (Z1, Z2, Z3) from a given N and S deposition E1.**

As mentioned above, unique critical loads for S and N cannot be specified. Each pair of deposition (N_{dep},S_{dep}) fulfilling Eq.7 (and the constraining equations 8-11), i.e. lying on the critical load function (the thick line in Fig.1), are called critical loads. We define the difference between left and right hand side of Eq.7 after inserting N_{dep} and S_{dep} as the **exceedance** of the critical loads.

$$Ex(N_{dep},S_{dep}) = a_N N_{dep} + a_S S_{dep} - b_1 N_u - b_2 N_i - L_{crit} \tag{12}$$

If $Ex \leq 0$ the point (N_{dep},S_{dep}) falls into the grey area in Fig.1 (or on to the critical load function) and we have non-exceedance. If $Ex>0$, it falls outside (like point E1 in Fig.1) and we have exceedance of critical loads. Note however, that a positive exceedance value is not necessarily the amount by which N or S has to be reduced. This can be easily seen from the example in Fig.1. By reducing N_{dep} one reaches the point Z1 and therefore non-exceedance without reducing S. On the other hand one can reach non-exceedance by solely reducing S_{dep} till reaching Z2. Finally, with a smaller reduction of both N_{dep} and S_{dep} one can reach non-exceedance as well (e.g. point Z3). In practice external factors, such as costs of emission reductions, will determine which path to follow to reach the critical load function and thus non-exceedance.

Nitrogen Leaching (Eutrophication)

In addition to acidification, excess amounts of nitrogen may cause eutrophication of aquatic ecosystems. The amount of N leached, N_l, is determined as the difference between N sources (inputs) and N sinks within a catchment. This mass balance for N reads:

$$N_l = N_{dep} - fN_u - (1-r)(N_i + N_{de}) - rN_{ret} \tag{13}$$

where the different terms are explained above. Inserting the expression for N_{de} and N_{ret} (Eqs.2 and 3) one can compute the amount of N leached from the catchment for any value of N_{dep}:

$$N_l = a_N N_{dep} - b_1 N_u - b_2 N_i \tag{14}$$

where b_1, b_2 and a_N are defined above. Conversly, by prescribing a maximum allowable ('critical') leaching of N, $N_{l,crit}$, one can compute a critical load of nutrient nitrogen:

$$CL_{nut}(N) = (b_1 N_u + b_2 N_i + N_{l,crit})/a_N \tag{15}$$

If $CL_{nut}(N)$ is smaller than $CL_{max}(N)$, this would impose another constraint on the N-deposition values not causing exceedance. However, for aquatic ecosystems it is not only the amount of N leached but also its form and the presence (or absence) of phosphorus that triggers eutrophication. For this reason and due to the fact that almost all lakes in Nordic countries are P-limited, critical loads for eutrophication are not considered thus far.

Input data

In addition to those needed for the SSWC-model, the FAB-model needs information on the following parameters: N_u, N_i, f_{de}, f, r, s_N and s_S. The following Table summarizes values and potential data sources.

Variable	Value	Source
N_u	depending on tree species and harvesting practices	forest inventories
N_i	2-5kgN $ha^{-1}yr^{-1}$(=142-357 eq ha^{-1} yr^{-1})	Downing *et al.* (1993)
f_{de}	0.1 - 0.8	De Vries *et al.* (1993)
f	0 - 1 land cover maps	
r	0 - 1 topographic maps	
s_N	2 - 8 m yr^{-1}	Dillon and Molot (1990)
s_S	0.2 - 0.8 m yr^{-1}	Baker and Brezomik (1988)

Note, that the first 3 variables are also needed for estimating critical loads for forest soils.

Example

The above method for computing critical loads has been used to compute critical loads for several thousand lakes in Finland, Norway and Sweden (Henriksen *et al.* 1993). In the following we illustrate this by computing the relevant parameters for a specific lake in south-western Finland (Lake Ilojärvi). This lake covers about 1% of the catchment area (r = 0.0107), and almost 70% of the catchment area is covered by forests (f = 0.691). The net growth uptake by this forests is estimated as N_u = 137.6 eq ha^{-1} yr^{-1}; for the denitrification fraction we estimate f_{de} = 0.3 (from a peatland coverage of 28.5% and the interpolating relationship f_{de} = 0.1+0.7f_{peat}), and for nitrogen immobilization we use the lower end of the range given in the Table above (N_i = 142.9 eq ha^{-1} yr^{-1}). For the runoff from this catchment we obtain from a digitized national runoff map Q = 247 mm. No data are available concerning the retention of S and N in lakes. Therefore we take intermediate values from the ranges given above: s_N = 5 m yr^{-1} and s_S = 0.5 m yr^{-1}. From this we obtain for the retention factors ρ_N and ρ_S:

$$\rho_N = 0.178 \quad \text{and} \quad \rho_S = 0.021$$

and for the factors from Eq.6 one obtains:

$$a_N = 0.578, \quad a_S = 0.979, \quad b_1 = 0.398, \quad b_2 = 0.570$$

The original base cation concentration is calculated as in the SSWC-model (see above). Here we use a modified version of it (Henriksen *et al.* 1993) and from water chemistry data of this lake we obtain $[BC^*]_0$ = 0.297 eq m^{-3}. For the ANC-limit we use 20 eq l^{-1}, a value derived from fish status surveys in Norway (Lien *et al.* 1992). Therefore we get for the leaching term, defined in Eq.10, L_{crit} = 684.2 eq ha^{-1} yr^{-1}. This results in the following limiting values for the critical loads (see Eqs.8-11):

$$CL_{min}(N) = 235.8, \quad CL_{max}(N) = 1419.5, \quad CL_{max}(S) = 698.9 \quad \text{eq ha}^{-1}\ \text{yr}^{-1}$$

These values determine the critical load function for this lake and allow to determine the deposition reduction requirements for any given N and S deposition.

References

Baker, L.A. and Brezonik, P.L. (1988) Dynamic model of in-lake alkalinity generation. *Wat. Resour. Res* **24**: 65-74.

Brakke, D.F., Henriksen, A. and Norton, S.A. (1990) A variable F-factor to explain changes in base cation concentrations as a function of strong acid deposition. *Verh. Internat. Verein. Limnol.* **24**: 146-149.

De Vries, W., Posch, M. Reinds, G.J. and Kämäri, J. (1993) *Critical loads and their exceedance on forest soils in Europe.* Report 58 (revised version), DLO The Winand Staring Centre for Integrated Land Soil and Water Research, Wageningen, The Netherlands.

Dillon, P.J. and Molot, L.A. (1990) The role of ammonium and nitrate in the acidification of lakes and forested catchments. *Biogeochem.* **11**: 23-43.

Downing, R.J., Hettelingh, J-P. and de Smet, P.A.M. (eds.) (1993) *Calculation and Mapping of Critical Loads in Europe*, Status Report 1993, Coordination Center for Effects, National Institute of Public Health and Environmental Protection (RIVM), Bilthoven, The Netherlands.

Henriksen, A., Forsius, M., Kämäri, J., Posch, M. and Wilander, A. (1993) *Exceedance of critical loads for lakes in Finland, Norway and Sweden: Reduction requirements for nitrogen and sulphur deposition.* Acid Rain Research Report 32/1993, Norwegian Institute for Water Research, Oslo, Norway.

Kämäri, J., Jeffries, D.S., Hessen, D.O., Henriksen, A., Posch, M. and Forsius, M. (1992) *Nitrogen critical loads and their exceedance for surface waters.* In P. Grennfelt and E. Thörnelöv (eds.) Critical Loads for Nitrogen, Nord 1992:41, Nordic Council of Ministers, Copenhagen, Denmark, pp. 161-200.

Kelly, C.A., Rudd, J.W.M., Hesslin, R.H., Schindler, D.W., Dillon, P.J., Driscoll, C.T.,Gherini, S.A. and Heskey, R.H. (1987) Prediction of biological acid neutralization in acid sensitive lakes. *Biogeochem.* **3**: 129-140.

Lien, L., Raddum, G.G. and Fjellheim, A. (1992) *Critical loads of acidity to freshwaters - fush and invertebrates*, NIVA-Report 0-89185, Norwegian Institute for Water Research, Oslo, Norway.

Sverdrup, H., de Vries, W. and Henriksen, A. (1990) *Mapping Critical Loads*, Nord 1990:98, Nordic Council of Ministers, Copenhagen, Denmark.

Keynote Paper - The Stages of Nitrogen Saturation: Classification of Catchments included in "ICP on Waters"

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Background

The leaching of N to surface waters can be seen to occur in stages (described in detail in Stoddard, 1994), which correspond to the stages of terrestrial N saturation described by Aber *et al.* (1989). The most obvious characteristics of these stages of N loss are changes in the seasonal and long-term patterns of surface water NO_3^- concentrations, which reflect the changes in N cycling that are occurring in the watershed. The N cycle at stage 0 is dominated by forest and microbial uptake, and the demand for N has a strong influence on the seasonal NO_3^- pattern of receiving waters (Figure 1a). The "normal" seasonal NO_3^- pattern in a stream draining a watershed at Stage 0 would be one of very low, or immeasurable, concentrations during most of the year, and of measurable concentrations only during snowmelt (in areas where snow packs accumulate over the winter months), or during spring rain storms (Figure 1c). The small loss of NO_3^- during the dormant season is a transient phenomenon, and results because snowmelt and spring rains commonly occur in these environments before substantial forest and microbial growth begin in the spring.

At Stage 1, the seasonal pattern typical of Stage 0 watersheds is amplified. It has been suggested that this amplification of the seasonal NO_3^- signal may be the first sign that watersheds are proceeding toward the more chronic stages of N saturation (Driscoll and Schaefer 1989; Murdoch and Stoddard 1992), and this suggestion is consistent with the changes in N cycling that are thought to occur at Stage 1 (Figure 1b; Aber *et al.* 1989). Overall limitation of forest growth (in the early stages of N saturation) is characterized by a seasonal cycle of limitation by physical factors (e.g. cold and diminished light), during late fall and winter, and nutrients (primarily N) during the growing season. The effect of increasing the N supply is to postpone the seasonal switch from physical limitation to nutrient limitation during the breaking of dormancy in the spring, and to prolong the seasonal N saturation that is characteristic of watersheds at this stage (Stoddard 1994). The key characteristics of Stage 1 watersheds are episodes of surface water NO_3^- that exceed concentrations typical of deposition (Figure 1d).

In Stage 2 of watershed N loss, the seasonal onset of N limitation is even further delayed, with the effect that biological demand exerts no control over winter and spring N concentrations, and the period of N limitation during the growing season is much reduced (Figure 2a). The annual N cycle, which was dominated by uptake at Stages 0 and 1, is instead dominated by N loss (through leaching and denitrification) at Stage 2. Sources of N (deposition and mineralization) outweigh the N sinks (uptake). The same mechanisms that produce episodes of high NO_3^- during extreme hydrologic events at Stage 1 also operate at Stage 2. But more importantly, the increased N sufficiency of the forest and soils creates a situation where NO_3^- can leach below the rooting zone, and elevated groundwater concentrations of NO_3^- result. This creates elevated NO_3^- concentrations during baseflow periods when deeper hydrologic flowpaths are contributing most to streamflow (Figure 2c).

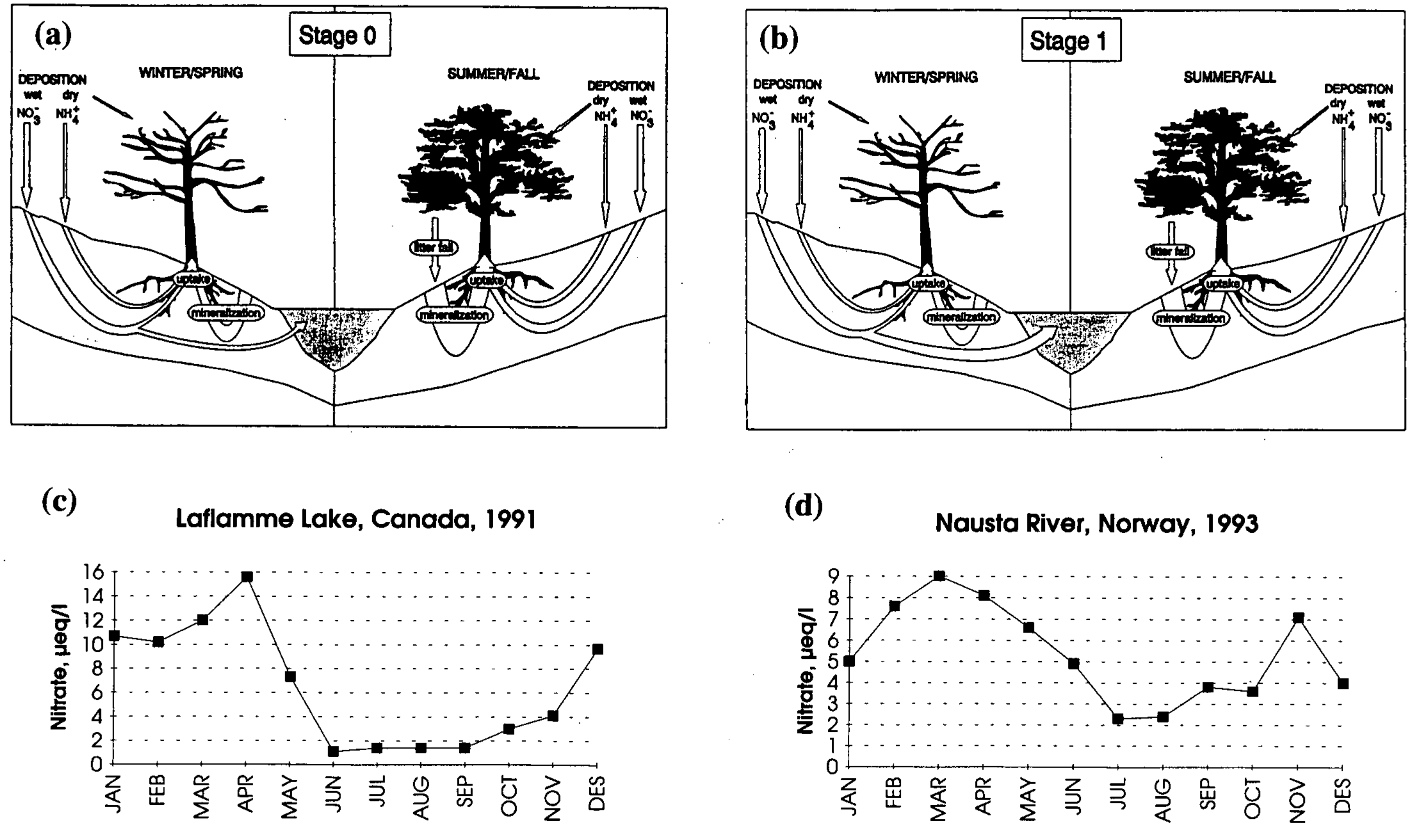

Figure 1 **Schematic representation of the watershed nitrogen cycle at Stage 0(a) and Stage 1(b) of nitrogen saturation.**
The size of arrows is roughly equivalent to the sizes of fluxes for each process, and cycles are divided into winter/spring (dormant) and summer/fall (growing) season. Examples of the seasonal pattern of NO_3^- concentrations at ICP sites classified as Stage 0(c) and Stage 1(d) are also shown.

In stage 2, the watershed becomes a net source of N rather than a sink (Figure 2b). Nitrogen retention mechanisms (e.g. uptake by vegetation and microbes) are much reduced, and mineralization of stored N may add substantially to N leaching the watershed through leaching or in gaseous forms. As in Stage 2, nitrification rates are substantial. Deposition, mineralization and nitrification all contribute NO_3^- to leaching waters, and surface water NO_3^- concentrations can exceed inputs from deposition alone. The key characteristics of Stage 3 watersheds are these extremely high NO_3^- concentrations, and the lack of any coherent seasonal pattern in NO_3^- concentrations (Figure 2d).

Conceptually, the stages of watershed N loss can be thought of as occurring sequentially, as a single watershed progresses from bring strongly N deficient, to strongly N sufficient. A number of factors may predispose a watershed to progress through the stages of N loss, including elevated N deposition, stand age, and high soil N pools (Stoddard 1994). High rates of N deposition play a clear role, as the ability of forest biomass to accumulate N must be finite. At very high, long-term rates of N deposition, the ability of forests and soils to accumulate N will be exceeded, and the only remaining pathway for loss of N (other than runoff) is denitrification. High rates of N deposition may favour increased rates of denitrification, but many watersheds lack the conditions necessary for substantial denitrification (e.g. low oxygen tension, high soil moisture, and temperature).

Another important factor in N loss from watersheds is the age of the forest stands. A loss in the ability to retain N is a natural outcome of forest maturation, as both older trees, and those that occur later in a successional sequence, grow more slowly and therefore exhibit lower N demand (Vitousek and Reindes 1975). Uptake rates of N into vegetation are generally maximal around the time of canopy closure for conifers, and somewhat later (and at higher rates) in deciduous forests due to the annual replacement of canopy foliage in these ecosystems (Turner *et al.* 1990). Finally, soil N status may also affect N loss rates; where large soil N pools exist, they imply that soil microbial processes that are ordinarily limited by the availability of N are instead limited by some other factor (e.g. availability of labile organic carbon, or another inorganic nutrient), and contribute to the likelihood that watersheds will leach NO_3^- (Johnson 1992; Joslin *et al.* 1992). To be N saturated, both the vegetational and soil microbial N demands of a watershed must be fulfilled; the existence of large soil N pools suggests that the second of these requirements may be easily met.

The NITREX project (Weight *et al.* 1995; Moldan *et al.* in press) and the Watershed Manipulation Project (Kahl *et al.* 1993) have both shown that it is possible to move forest ecosystems (and the runoff of nitrogen) through different stages of nitrogen saturation by experimentally increasing the deposition of nitrogen, thus confirming the conceptual basis for N saturation stages.

N saturation in CIP on Water Catchments

In this paper an attempt will be made to classify the ICP on Waters sites according to the N saturation scheme described above. The first step in such an assessment is establishing clear criteria to define each N saturation stage. The original criteria of Stoddard (1994) have been adapted here for the range of sampling frequencies that have been used to collect the ICP data. For sites with abundant data, the criteria to establish a site's N saturation stage were based on monthly average NO_3^- concentrations (Table 1).

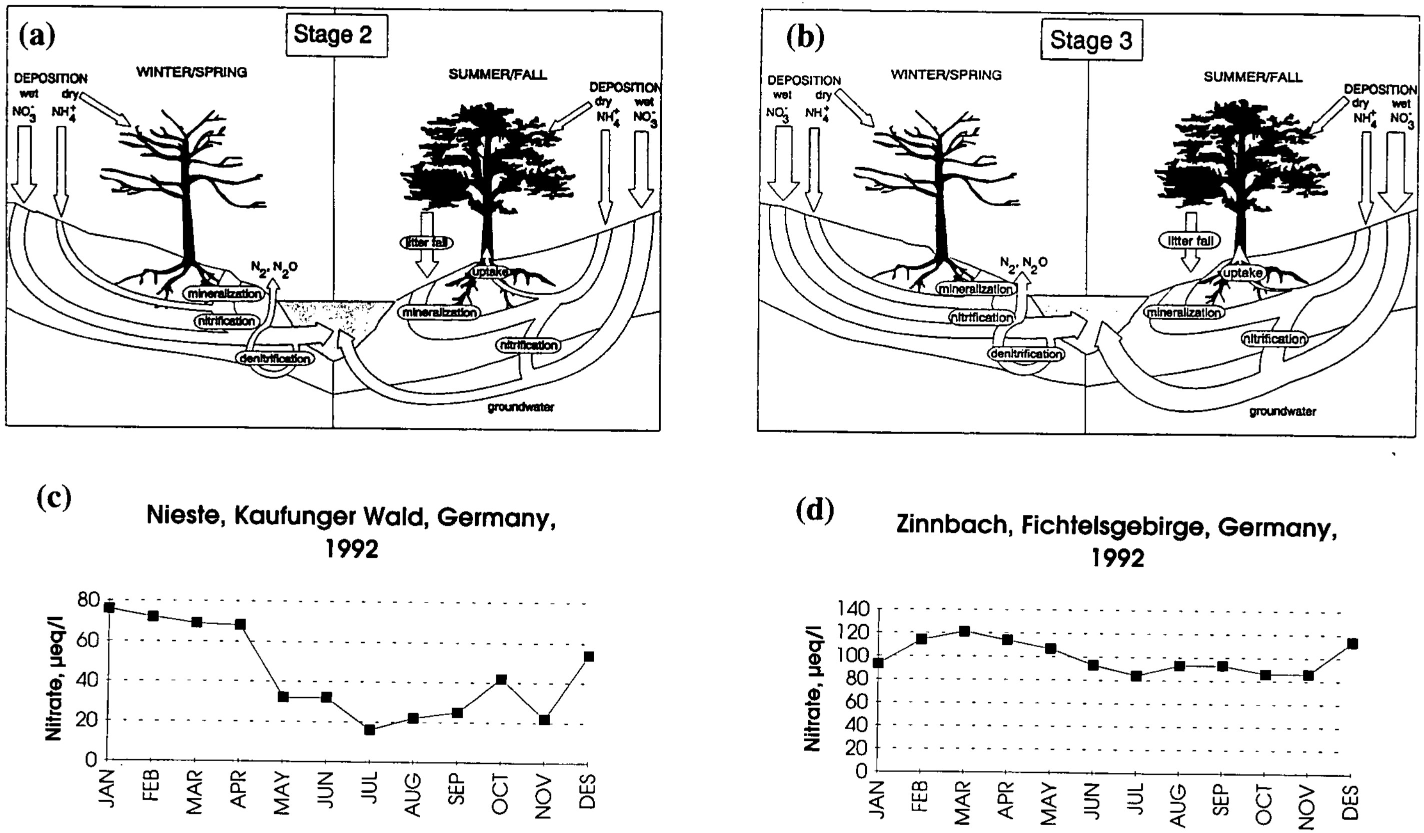

Figure 2 **Schematic representation of the watershed nitrogen cycle at Stage 2(a) and Stage 3(b) of nitrogren saturation.**
The size of arrows is roughly equivalent to the sizes of fluxes for each process, and cycles are divided into winter-spring (dormant) and summerfall (growing) seasons. Examples of the seasonal pattern of NO_3^- concentrations at ICP sites classified as Stage 2(c) and Stage 3(d) are also shown.

Table 1: N saturation stage criteria for sites with frequent samples.

Stage	Criteria
0	>3 months in the growing season with NO_3 <3 μeq/l and peak value <20 μeq/l.
1	<3, >0 months in the growing season with NO_3 <3 μeq/l, or >3 months in the growing season with NO_3 <3 μeq/l and peak value >20 μeq/l.
2	No month with NO_3 <3 μeq/l and >3 months in the growing season with NO_3 <50μeq/l.
3	<3 months with NO_3 <50 μeq/l.

Many of the ICP sites have only 3 or 4 measurements during the year, usually at different seasons. It is, however, still possible to make an approximate classification of these sites, even if it will not always be possible to distinguish between Stages 0 and 1, and between Stages 2 and 3. If the summer and/or fall value are below 3 μeq l^{-1}, the site belongs to either Stage 0 or Stage 1, and will be denoted Stage 0/1. If 3 measurements at different seasons all show values of nitrate lower than 3 μeq l^{-1}, it is reasonable to assume that the concentrations have been low also in months between measurements, and the site will be classified at Stage 0. If all measurements show nitrate concentrations about 3 μeq l^{-1}, but well below 50 μeq l^{-1}, the site will be classified at Stage 2. Sites with one or more measurements above 50 μeq l^{-1} will be classified at Stages 2/3.

In order to include as many sites as possible at this assessment of the ICP data, a second set of criteria were developed for sites with infrequent data. If insufficient data are available to distinguish between Stages 0 and 1, or between Stages 2 and 3, then the criteria designate the sites as either Stage 0/1 or Stage 2/3 (Table 2).

The criteria for sites with infrequent samples were tested on data from 25 sites with frequent samples, selecting 2 sets of 3 values from each site (March, June and September, or April, July and October). The 2 sets gave identical classifications to the frequent sample criteria in 18 and 20 cases, respectively. In all of the remaining cases the infrequent sample classification included the frequent sample classification (e.g. Stage 0 and Stage 1 sites were classified as Stage 0/1, etc).

A summary of the N saturation status of ICP on Waters catchments is shown in Table 3. Included in this analysis are 146 sites listed in the ICP on Waters Six Year Report (Skjelkvåle *et al.* 1994), plus 2 new Russian and 2 new Finnish sites. Due to insufficient data, it was not possible to determine stages for 6 of these sites, leaving 144 catchments in Table 3. In the case of the Dutch sites, the seasonal patterns in NH_4^+ are used, rather than patterns of NO_3^- because NH_4^+ is the dominant form of nitrogen in the runoff.

Table 2: *N saturation stage criteria for sites with infrequent samples (equally spaced in time, at least 2 samples in the growing season).*

Stage	Criteria
0	At least >2 values in the growing season <3 μeq/l NO_3 and peak value <10 μeq/l.
0/1	1 value in the growing season <3 μeq/l and peak value <10 μeq/l or >1 value in the growing season <3 μeq/l and peak value >10 μeq/l.
1	<1 value in the growing season <3 μeq/l and peak value >20 μeq/l.
1/2	>1 value in the growing season <5 μeq/l, but >3 μeq/l.
2	>2 values in the growing season >5 μeq/l, but <50 μeq/l.
2/3	At least 1 value in the growing season >50 μeq/l and at least 1 value <50 μeq/l.
3	All values in the growing season >50 μeq/l.

Table 3: *Number of ICP on Waters catchments within each stage of nitrogen saturation. (For explanation see text)*

Country	Stage 0	Stage 0/1	Stage 1	Stage 2	Stage 2/3	Stage 3	Total
Norway	4		4	2			10
Sweden	6		3	2			11
Finland	4	2					6
Denmark						2	2
Germany		1		8	1	20	30
Netherlands						5	5
Belgium				7		1	8
Austria				3		3	6
UK	1	2		3			6
Ireland	10		5	3			18
Canada	9	6		3			18
USA	8	4	3	7			22
Russia	2						2
TOTAL	**44**	**15**	**15**	**38**	**1**	**31**	**144**

Nearly half (70 of 144) of the ICP catchments exhibit a high degree of nitrogen saturation (Stage 2 or 3). Such sites are found in all participating countries except Finland and Russia. All ICP on Waters sites in Denmark, Belgium, The Netherlands and Austria, and 29 out of 30 German sites, exhibited Stage 2 or Stage 3 patterns.

The relationship between rates of nitrogen deposition (estimated from EMEP grid data) and nitrogen saturation stages in European ICP on Waters sites is shown in Figure 3. The mixed classifications (e.g. Stages 0/1 and 2/3) are not included. There is a clear connection between N deposition values and stage classification. At Stage 0 and 1 catchments only 7 out of 25 have N deposition values above 0.8 gN m^{-2} yr^{-1}, while at stage 2 only 2 out of 25 have deposition values below 1.0 gN m^{-2} yr^{-1}. All stage 3 sites have deposition values above 1.5 gN m^{-2} yr^{-1}. While these results suggest that a threshold value of N deposition, above which significant N saturation can exist, occurs at approximately 1 gN m^{-2} yr^{-1}, it should be reiterated that deposition is only one factor influencing NO_3^- leaching from catchments. Nitrogen demands on the part of vegetation (controlled largely by forest age and health) and on the part of soils (controlled largely by pool sizes for nitrogen and carbon) must be satisfied before N saturation occur. Some consideration should be given to these factors when identifying critical levels of N deposition. Because information on forest status and soil chemistry is unavailable for most ICP on Waters catchments, the data in Figure 3 indicate only a strong influence of appropriate threshold value of N deposition is.

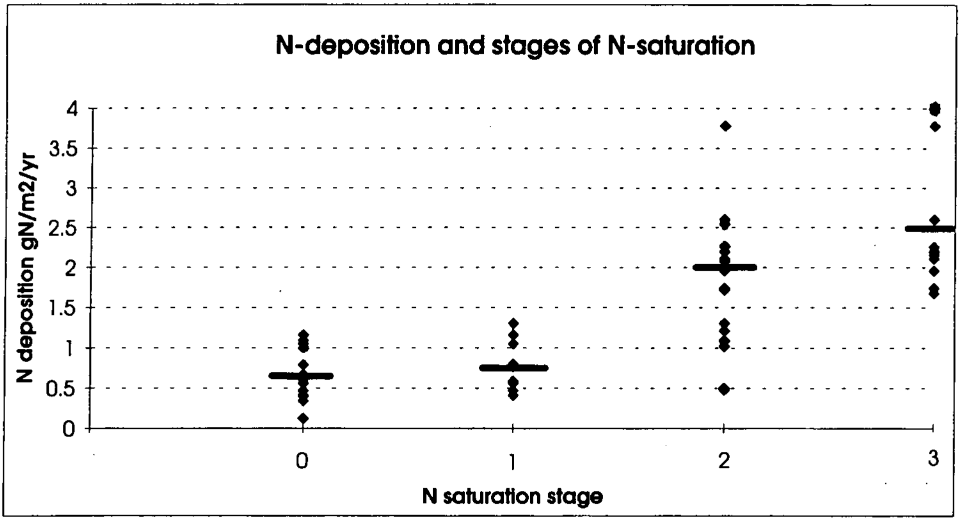

Figure 3: **Relationship between nitrogen deposition (estimated from EMEP grid data) and stages of nitrogen saturation for European ICP on Waters sites. Average values are indicated by horizontal bars.**

References

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M. (1989) Nitrogen saturation in northern forest ecosystems. *BioScience* **39**: 378-386.

Driscoll, C.T. and Schaefer, D.A. (1989) Background on nitrogen processes. Pages 4.1-4.12 in: J.L. Malanchuk and J. Nilsson (eds.) *The Role of Nitrogen in the Acidification of Soils and Surface Waters*. No. 1989: 10, Nordic Council of Ministers, Copenhagen, Denmark.

Johnson, D.W. (1992) Nitrogen retention in forest soils. *J. Environ. Qual.* **21**: 1-12.

Joslin, J.D., Kelly, J.M. and Van Miegroet, H. (1992) Soil chemistry and nutrition of North American spruce-fir stands: Evidence for recent change. *J. Environ. Qual.* **21**: 12-30.

Kahl, J.S., Norton, S.A., Fernandez, I.J., Nadelhoffer, K.J., Driscoll, C.T. and Aber, J.D. (1993) Experimental inducement of nitrogen saturation at the watershed scale. *Environ. Sci. Technol.* **27**: 565-568.

Moldan, F., Hultberg, H., Nystrom, U. and Wright, R.F. in press Nitrogen saturation at Gardsjon, SW Sweden, induced by experimental addition of ammonium nitrate. *Forest Ecology and Management.*

Murdoch, P.S. and Stoddard, J.L. (1992) The role of nitrate in the acidification of streams in the Catskill Mountain of New York. Water Resources Research **28**: 2707-2720.

Skelkvåle B.L., Newel, A.D., Raddum, G., Johannessen, M., Hovind, H., Tjomsland, T. and Wathne, B.M. (1994) *The six year report*: Acidification of surface waters in Europe and North America. Dose/response relationships and long-term trends. Serial No 3041, Norwegian Institute for Water Research, Oslo, Norway.

Stoddard, J.L. (1994) Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. Pages 223-284 in: L.A. Baker (ed.) *Environmental Chemistry of Lakes and Reservoirs*, Advances in Chemistry Series, No 237, American Chemical Society, Washington, DC.

Turner, R.S., Cook, R.B., Van Miegroet, H., Johnson, D.W., Elwood, J.W., Bricker, O.P., Lindberg, S.E. and Hornberger, G.M. (1990) *Watershed and Lake Processes Affecting Surface Water Acid-Base Chemistry.* State-of-Science/Technology Report No 10, National Acid Precipitation Assessment Program, Washington, DC.

Vitousek, P.M. and Reiners, W.A. (1975) Ecosystem succession and nutrient retention: A hypothesis. *BioScience* **25**: 376-381.

Wright, R.F., Roelofs, J.G.M., Bredemeier, M., Blanck, K., Boxman, A.W., Emmett, B.A., Gundersen, P., Hultberg, H/. Kjonaas, O.J., Moldan, F., Tietema, A., Van Breemen, N. and Van Dijk, H.F.G. (in press) NITREX: response of coniferous forest ecosystems to experimentally-changed deposition of nitrogen. *Forest Ecology and Management.*

CHAPTER 4
CRITICAL LOADS OF NITROGEN:
THE MASS BALANCE APPROACH

Working Group Report

CRITICAL LOAD FOR TOTAL NITROGEN

1. Background

This working group focused on the application of the mass balance approach in the context of eutrophication. The application of the approach in the context of acidification has been considered in detail at a number of other conferences and workshops.

The mass balance approach as applied in the context of N eutrophication, balances N inputs and outputs at equilibrium. It considers a long term perspective and the workshop endorsed the timescales suggested at the Lökeberg workshop i.e. at least one rotation for managed forest systems and ca. 100 years for other ecosystems. It is acknowledged that several of the variables in the mass balance equation vary over time and in response to natural and anthropogenic perturbations. In the mass balance approach, the impacts of these variations are averaged over the period under consideration and long-term averages are used for the fluxes. The impacts of management of N inputs and outputs can be incorporated into the mass balance approach. The effects of normal or traditional land management practices, e.g. burning of moorland, should be incorporated when calculating critical loads of N.

The mass balance approach can, in principle be used to calculate critical loads of nutrient nitrogen for any terrestrial ecosystem, or habitat type, to prevent vegetation change and/or to protect surface or groundwater quality. It cannot be used to calculate critical loads to prevent physiological changes, such as increased frost sensitivity, or changes in susceptibility to insect attack.

Vegetation soil systems respond differently to inputs of NH_y as opposed to NO_x. At inputs equal, or similar to the critical load, these differences are within the general uncertainty of the mass balance approach but may become important at inputs above the critical load. Those systems should be identified in which the differences in response to NH_y and NO_x are important.

2. Underlying concepts

The Lökeberg workshop identified three important underlying concepts to the setting of critical loads which were:

i) nitrogen saturation
ii) soil acidification and
iii) sustainability.

The importance of all three concepts was endorsed in the current workshop but, as noted above, it concentrated on consideration of eutrophication.

i) **Nitrogen saturation:** the mass balance approach aims to set the critical load of nutrient nitrogen to prevent the occurrence of nitrogen saturation as defined by Aber *et al.* (1989). Enhanced NO_3^- leaching is taken as an indication of N saturation.

The concept seems to be supported for forest systems by results from recent plot and catchment-based N addition experiments. Results from these experiments show that the response of forest systems to increased N inputs depends on the N status of the receptor and the form of the added N (NH_y versus NO_x). Thus, for example, additions of NO_3^- to systems close to saturation result in NO_3^- leaching; added NH_4^+ may still be retained by these systems. Receptor systems which are not close to saturation retain most of the added N.

It should be noted, however, that vegetation change probably takes place before N saturation and enhanced leaching. setting the critical load to avoid N saturation will not therefore necessarily prevent vegetation change.

ii) **Soil acidification:** the role of nitrogen compounds in acidification of soils should be considered in parallel with nutrient effects and the critical load set to avoid both acidification and eutrophication.

iii) **Sustainability:** the mass balance approach aims to set the critical load such that the removal of N at harvesting in managed ecosystems is balanced by a rate of concurrent removal of other nutrients which can be balanced by the rate of supply from weathering and atmospheric inputs. It is recognised that this may not give optimal growth. Where additions of fertilisers incorporating nutrients other than N is part of normal forest management, the critical load for these managed systems should be calculated incorporating the usual levels of fertilizer input. However, the addition of non-N fertilisers to exploit the fertiliser effect of pollutant N inputs is not regarded as sustainable management.

3. Formulation of the Mass Balance Equation

All significant inputs and outputs of N should be incorporated and made explicit in the formulation of the mass balance equation.

The following formulation of the equation is recommended:

$$CL_{(N)nut} = N_{l(crit)} + N_{i(crit)} + N_{u(crit)} + N_{de(crit)} - N_{fix(crit)} + N_{fire(crit)} + N_{erode(crit)} + N_{vol(crit)}$$

$CL_{(N)nut}$ critical load for nutrient nitrogen

$N_{l(crit)}$ total annual N leaching (NO_3^- + NH_4^+ - dissolved organic N) from the rooting zone under natural conditions in the absence of pollutant N inputs plus any enhanced leaching following forest harvesting or following natural fires or fires used as part of traditional management regimes.

$N_{i(crit)}$ an acceptable annual level of N immobilisation in soil organic matter (including forest floor), at N inputs equal to the critical load, at which adverse ecosystem change will not take place.

$N_{u(crit)}$ net annual removal of N in vegetation and harvested animals at N inputs equal to the critical load.

$N_{de(crit)}$ annual flux of N to the atmosphere as a result of denitrification at N inputs equal to the critical load.

$N_{fire(crit)}$ N losses in smoke from natural wildfires or from fires used as part of traditional management regimes.

$N_{erode(crit)}$ annual N losses through erosion under natural conditions plus enhanced erosion losses following forest harvest, natural fire or fires used as part of traditional management regimes.

$N_{vol(crit)}$ annual N losses to the atmosphere from NH_4^+ colatilisation.

4. Methods for quantification of the variables

$N_{l(crit)}$ this should be set whenever possible by using data from similar ecosystems in pristine environments; enhanced N leaching following harvesting or fire should be averaged over the rotation period or over the interval between fires.

$N_{i(crit)}$ there is no definitive method for calculating an acceptable level of immobilisation, model approaches are being developed but the following approaches are currently recommended:

i) the total N accumulated in the soil profile divided by the period of soil formation or since colonisation by vegetation.

ii) an acceptable percentage increase in the size of the N pool in litter and the biologically active soil horizon over a forest rotation - this method is only recommended for forests on sandy podzols.

iii) values assigned to strata defined on the C:N ratio, soil and vegetation type

Rates of N accumulation between disturbance of a system and the establishment of a new equilibrium, for example forest invasion of abandoned pastures, can be used to derive a ceiling value for N immobilisation rates; values derived in this way should not be used as input to the mass balance equation but as a guide to the upper limit.

$N_{de(crit)}$ derived from measured values for similar ecosystems in pristine conditions or calculated using one or the two methods detailed in the UNECE mapping manual (appendix 1).

$N_{u(crit)}$

i) N removal in harvested biomass (vegetation or animal) divided by the period between harvests; great care must be taken in determining removal in havested animal biomass to ensure that a net value is derived;

ii) the application of the nutrient limitation approach (Appendix 2).

$N_{fire(crit)}$ measured N losses in smoke for similar ecosystems divided by the interval between fires; N losses in smoke are only likely to be significant in a few natural ecosystems, e.g. Mediterranean forests, mediterranean sclerophyllous scrub (maquis) or boreal for est, or in systems where fire forms part of traditional management of extensive grazing or game habitats.

$N_{fix(crit)}$ measured annual N fixation in similar ecosystems under pristine conditions.

$N_{vol(crit)}$ measured N losses from volatilisation in similar ecosystems under pristine conditions, ammonia volatilisation is only likely to be significant from some moist calcareous soils or in ecosystems with large natural populations of animals (the application of the mass balance approach to intensive grazing systems is inappropriate).

$N_{erode(crit)}$ measured natural annual rates of erosion for similar ecosystems or following natural fires or traditional fire management, in the latter case the erosion should be divided by the period between fires.

National input data may not be available for one or more of the input variables and it is important, therefore that a library of default values is available for a range of important and widespread ecosystems. Some default values were suggested at the Lökeberg workshop and incorporated into the workshop report. An exploratory exercise was carried out at the current workshop to explore the development of a series of default values, or ranges of suggested values. The matrix of values is illustrated in Table 1.

Table 1: *Estimated values (kg ha^{-1} yr^{-1}) for the parameters in the mass balance equation for nutrient nitrogen critical load in a range of different terrestrial ecosystems.*

Ecosystem	N-leaching	N-immobil	N-removal	N-fix	N-denit[2]	N-fire	N-vol	CN(N)
Comment on the range	Water surplus Low-High	Climate Warm-Cold	Vegetation growth N[4]; Low-High	Soil Moisture Dry-Wet	Climate Dry-Wet	Frequency Low-High		Possible range
Tundra	0-3	1-4	0-2	0.5(<1)	0-1	0	0	4
Boreal forest	2-4	1-4[5]	1-7	0-2	0-1	0-1	0	3-10
Temp. conif.	1-4	1-3	2-10	0-2	0-4[6]	0-1	<.5	4-20
Temp. decid.	1-?	1-3	5-15	1 (1-3[7])	0-4	0	0	2-27
Medit. forest	0.5-1	<1	3-5	<1	<0.5	1-5	?	2+?-12+?
Acid grassland	1-3	0.5-2	0.5-1[8]	<1	0.5-2	0-1	0	2-8

1 Removal of plant (and animal) biomass
2 Not including symbiotic fixation e.g. Alder
3 Removal in animal biomass extra 0-0.5
4 Unmanaged systems (nature reserves) assumed to have no net removal of biomass
5 Some could be higher e.g. taiga, Russia
6 High value, peat soils with high pH
7 Epiphyte rich system
8 Sheep etc grazing

5. Recommendations

The mass balance approach provides a useful method for the calculation of critical loads of N in the context of both acidification and eutrophication.

Whenever possible, the critical load of N as a nutrient should be determined using both the mass balance and empirical approaches. The critical load of N in the context of acidification should be calculated in parallel. The critical load for N should be at the lower value of the critical load.

The N inputs used in the calculation of exceedence should include total N inputs, NH_y and organic N.

Data matrices should be developed, based on a literature search for all the input variables to the mass balance equation for a range of ecosystems.

The mass balance approach cannot be used in scenario analysis and in assessment of the temporal aspects of the response of systems to N inputs. The development of dynamic models should, therefore, remain a high priority.

6. Gaps in knowledge and research requirements

i) Controls on N immobilisation rates and the relationship between changes in immobilisation rates and changes in ecosystem stability, structure and function.

ii) The relationship between variations in leaching fluxes of N and changes in ecosystem stability, structure and function.

iii) The great majority of the N manipulation experiments carried out to date have involved forest systems; there is a need for similar studies on a range of non-forest systems.

CRITICAL LOAD FOR NH_3-N BASED ON NH_4:K RATIOS

1. Background

In areas with large inputs of NH_3-N and consequent large concentrations of NH_4-N in soil solution, ammonium uptake can result in nutrient imbalances and deficiencies, particularly of K. Critical NH_4:K ratios in soils have been proposed at which K deficiency is likely to occur. A steady-state model, which uses the concept of a critical NH_4:K ratio has been proposed for the calculation of critical loads of NH_3-N (de Vries 1994). The model includes the major NH_4 and K inputs to the system and the ratio of NH_4 to K is calculated from the system inputs.

2. Formulation of the Model

The following formulation has been proposed by de Vries (1994):

$$RNH_4K = (NH_{4th} + NH_{4min} - NH_{4nl} - NH_{4nl} - NH_{4ru})/(K_{th} - K_{min} - K_w - K_{ru}) \quad (1)$$

RNH_4K NH_4:K ratio

NH_{4th}	annual NH_4-N flux in throughfall
K_{th}	annual K flux in throughfall
NH_{4min}	annual production of NH_4-N by mineralisation
K_{min}	annual release of K from organic material by mineralisation
NH_{4ni}	annual nitrification of NH_4-N
NH_{4ru}	annual root uptake of NH_4-N
K_{ru}	annual root uptake of K
K_w	annual release of K by mineral weathering

In calculating the critical load, the model is simplified by assuming that

i) throughfall NH_4 and total deposition of NH_3 (NH_{3td}) are equal;

ii) mineralisation of NH_4 and K is equal to the N and K input by litterfall (steady state situation);

iii) root uptake does not effect the NH_4:K mol ratio and,

iv) release of K from weathering is negligible.

The above equation can then be simplified to:

$$RNH_4K = NH_{3td} + N_{lf} - NH_{4ni}/K_{lf} + K_{lf} \quad (2)$$

N_{lf} annual input of N in litterfall

K_{lf} annual input of K in litterfall

Nitrification is described as a fraction of the NH_4 input according to:

$$NH_{4ni} = Fr_{ni}(NH_{3td} + N_{lf}) \quad (3)$$

where Fr_{ni} is a nitrification fraction:

Combining equations (2) and (3):

$RNH_4K = (1 - Fr_{ni}) . (NH_{3td} + N)/(1 - Fr_{ni})$

Defining a critical NH_4/K ratio as $RNH_4K(crit)$ a critical NH_3-N load, $NH_{3td}(crit)$ can be derived as follows:

$NH_{3td}(crit) - RNH_4K(crit) . (K_{tf} + K_{lf}) - (1 - Fr_{ni}) . N_{lf}/(1 - F_{ni})$

3. Gaps in knowledge

Critical NH_4:K ratios for a range of vegetation-soil systems still need to be determined.

4. Recommendations

The model should be tested by application in a range of countries and the calculated critical loads compared with empirical critical loads and critical loads for nutrient N calculated using the mass balance approach.

References

Aber, J.D., Nadelhoffer, K.J., Steudler, P. and Melillo, J.M. (1989) Nitrogen saturation in northern forest ecosystems. *BioScience* **39**: 378-386.

de Vries, W. (1994) Soil responses to acid deposition at different regional scales. Wageningen: DLO Winard Staring Centre.

CHAPTER 5
CRITICAL LOADS FOR TOTAL ACIDITY

DISCUSSION GROUP REPORT

Introduction

Opportunity was taken at the meeting to convene a discussion group on the Critical Loads of Total Acidity, including sea salt derived ions. This was done to address the problem experienced by many countries regarding the contribution of sea salts to total deposition, especially in those areas where anthropogenic contributions of acidifying compounds are low.

1 Base Cation Inputs

Background

The current version of the critical loads mapping manual stipulates the use of "excess base cation" inputs when calculating critical loads of acidity for soils using the modified mass balance equation. Detailed evaluation of the equation in the UK had raised doubts about the scientific basis for the use of "excess" base cations as opposed to total inputs, including sea salt derived ions. The Soils Sub-Group of the UK Critical Loads Advisory Group therefore requested a small workshop to discuss the problem. As many of those involved in the development of the modified mass balance equation were attending the Grange worshop, an additional session was arranged during that workshop to discuss the Base Cation problem.

Formulation of the equations

The discussion was based around the version of the mass balance equation developed for use in high rainfall areas, following the workshop held in Vienna in March 1992, but with further modifications which

i limit uptake and leaching losses to ensure that the values of neither of these variables exceed the quantity of base cations available from weathering plus atmospheric inputs; and

ii satisfy plant uptake demands before calculating the leaching losses. The equation with these latter modifications is presented in the Keynote Paper (Sverdrup *et al.*).

The ANC_L term, which is incorporated in the formulation of the basic equation for the critical load of actual acidity, is derived from the maximum permitted leaching of Al and H. This maximum permitted leaching is calculated with reference to a critical chemical limit at which damage will not occur to a biological indicator, in this case the Base Cation:Aluminium ratio at which damage will not occur to fine roots. The calculation of the limiting Al leaching, with respect to this critical chemical limit, incorporates a Base Cation leaching term. The Base Cation leaching is calculated from a mass balance which includes Base Cation deposition.

The discussion session considered whether this Base Cation deposition term should comprise total or non-marine Base Cation deposition. It was concluded that fine root health would be influenced by the Total Cation: Aluminium ratio in soil solution and that the origin of the base cations was irrelevant. The use of total Base Cation deposition in the mass balance equation was therefore considered to be most appropriate.

2 Critical Loads for Peats

The mass balance equation as currently formulated in the UN-ECE mapping manual cannot be used to calculate the critical load of acidity for peats. These soils contain little aluminium and it is therefore inappropriate to calculate the ANC_L in terms of a critical chemical limit set with reference to a Base Cation:Aluminium ratio at which fine root damage will occur. In these soils, acidity rather than Base Cation:Aluminium ratio is likely to be the main control on fine root status. In these soils, therefore it is more appropriate to express the critical chemical limit in terms of the Base Cation:Hydrogen ion ratio. Once a Base Cation:Aluminium ratio has been derived, the same numerical value can be used for the Base Cation:Hydrogen ion ratio. The formulation of the modified mass balance equation incorporating the use of a Base Cation:Hydrogen ion ratio is detailed in the Keynote Paper (Sverdrup *et al.*).

3 Recommendations

- The Base Cation input to the mass balance equation for the calculation of critical loads of acidity for non-peat soils should be redefined to include marine-derived Base Cation inputs.

- The sulphur inputs used to calculate exceedances of critical loads for acidity should be re-evaluated to assess whether or not total sulphur deposiiton should be used.

- The proposed formulation of the mass balance equation for the calculation of critical loads for peats (c.f. Keynote Paper - Sverdrup *et al.*) should be evaluated by application in one or two countries with large areas of peats.

Keynote Paper - Modification of the Simple Mass Balance Equation for Calculation of Critical Loads of Acidity

H Sverdrup, W de Vries, M Hornung, M Cresser, S Langan, B Reynolds, R Skeffington, W Robertson

1 Introduction

Over the last few years, the simple mass balance equation for the calculation of critical loads of acidity has been gradually modified as the underlying critical load concepts have developed and as problems with particular forms of the equation have been identified, through application in particular countries. The first major update of the equation took place following a workshop held in Vienna, Austria (Hojesky *et al.* 1993). The workshop was held to discuss problems which had been identified when the then current form of the equation was applied in countries with high rainfall. The problems had largely arisen because of simplifications and assumptions incorporated into the early formulation of the equation. The equation was reformulated to overcome the problems identified at the workshop. However, further problems were identified when the reformulated equation was applied in the UK in situations with a combination of high rainfall, large marine inputs and widespread occurrence of organic soils. A small workshop was, therefore held in Grange-over-Sands, UK in late 1993 to dicuss the problems and to further re-evaluate the equation. The problems had arisen in the UK because of simplifications and assumptions made in the formulation concerning, in particular, cation leaching and uptake. As a result, a more rigorous treatment of these variables was incorporated into the equation. The reformulation of the equation, as derived at the September 1993 workshop is described below.

2 The Basic Equation

The reformulated equation is based on the following definition of the critical load for actual acidity:

$$CL = ANCw - ANC_L \quad (1)$$

where
$ANCw$ = alkalinity produced from weathering $eq\ ha^{-1}\ yr^{-1}$
CL = Critical load of acidity $eq\ ha^{-1}\ yr^{-1}$
ANC_L = ANC leaching $eq\ ha^{-1}\ yr^{-1}$

In this equation the limiting ANC leaching is determined by the maximum permitted leaching of H and Al at which damage will not occur to a sensitive biological indicator and as derived from the following simplified expression:

$$ANC_L = -H_L^{+} - Al_L^{3+} \quad (2)$$

where
Al_L^{3+} = Al^{3+} leaching $eq\ ha^{-1}\ yr^{-1}$
H_L^{+} = H^+ leaching $eq\ ha^{-1}\ yr^{-1}$

3 Plant Response Criteria

The maximum permitted leaching of ANC is set in terms of a critical chemical limit above, or below which, damage will not occur to a selected biological indicator. The most frequently used indicator is fine roots with the critical chemical limit most commonly set in terms of the Base Cation:Aluminium ratio in soil solution. However, the Base Cation:Hydrogen ratio can be used in soils with a large organic matter content and small Aluminium contents.

The following equation gives the limiting Al flux in equation (2) when the molar Base Cation:Al ratio is used as the critical chemical limit:

$$Al_L^{3+} = \frac{BC_L}{BC/Al_{crit}} \tag{3}$$

where

	BC_L	= Base Cation leaching	eq ha^{-1} yr^{-1}
	$(BC/Al)_{crit}$	= BC/Al ratio used as the critical chemical limit	

The Base Cation leaching is calculated from a mass balance:

$$BC_L = BC_{W(CaMgK)} + BC_D - BC_U \tag{4}$$

where

	BC_D	= Base Cation deposition	eq ha^{-1} yr^{-1}
	$BC_{W(CaMgK)}$	= Weathering rate of Ca+Mg+K	eq ha^{-1} yr^{-1}
	BC_U	= Base Cation uptake	eq ha^{-1} yr^{-1}

In the mass balance equation for base cations, approximately 30% of released base cations from weathering are Na, which provides no protection against Al for plants. The production of Ca, Mg and K from weathering is:

$$BC_{W(CaMgK)} = x_{BC} \cdot ANC_W \tag{5}$$

where

	X_{BC}	= Fraction of weathering as Ca+Mg+K = 0.7	
	ANC_W	= Neutralisation rate due to weathering	eq ha^{-1} yr^{-1}

However, two qualifications must be considered when calculating Base Cation uptake and leaching:

i. base cations at very low concentrations in soil solution, <2meq m^{-3}, will be unavailable for plant uptake because of physiological limitations, and

ii. the quantity of base cations leached cannot be larger than the supply from weathering plus Base Cation deposition. The minimum Base Cation leaching can be derived as follows:

$$BC_{min} = Q.[BC]_{min} \tag{6}$$

where Q is percolation, [BC] is the limiting concentration for uptake, provided sufficient base cations are available. However if:

$$BC_{min} > x_{BC} \,.\, ANC_W + BC_D \tag{7}$$

then:

$$BC_{min} = x_{BC} \,.\, ANC_W + BC_D \tag{8}$$

A further condition which must be satisfied is that plant uptake of base cations cannot exceed the quantity of bases cations available from weathering and deposition. Thus, if:

$$BC_U > x_{BC} \,.\, ANC_W + BC_D - BC_{min} \tag{9}$$

then:

$$BC_U = x_{BC} \,.\, ANC_W + BC_D - BC_{min} \tag{10}$$

The Al leaching term can then be written as:

$$AL_L^{3+} = 1.5 \,.\, \frac{(x_{BC} \,.\, ANC_W + BC_D - BC_U)}{(BC/Al)_{crit}} \tag{11}$$

Operationally the H^+ concentration can be calculated using the gibbsite equation:

$$[H^+] = \left(\frac{[Al]^{3+}}{K_{gibb}}\right)1/3 \tag{12}$$

where K_{gibb} = gibbsite coefficient 300 $m^6 eq^{-2}$ (-pK(gibb)=8.5)

Accordingly, the limiting H^+-concentration corresponding to a certain Al concentration in the soil is calculated from the Al^{3+}-flux calculated as above (equation 11), divided by the flow and the gibbsite coefficient:

$$[H^{+}]_{limit} = \left(\frac{Al_{L}^{3+}}{Q \cdot K_{gibb}}\right)1/3 \tag{13}$$

By inserting the expression for the Al-limiting flux in the expression and multiplying by flow Q to get from H^+-concentration to flow, we get:

$$H_{L}^{+} = \left(1.5 \cdot \frac{x_{BC} \cdot ANC_{W} + BC_{D} - BC_{U}}{(BC/Al)_{crit} \cdot Q \cdot K_{gibb}}\right)1/3 \cdot Q \tag{14}$$

The modified SMB equation for critical load of acidity in eq ha^{-1} yr^{-1} thus becomes:

$$CL = ANC_{W} + \left(1.5 \cdot \frac{(x_{BC} \cdot ANC_{W} + BC_{D} - BC_{U})}{(BC/Al)_{crit} \cdot K_{gibb}}\right)1/3 \cdot Q^{2/3}$$

$$+1.5 \cdot \left(\frac{x_{BC} \cdot ANC_{W} + BC_{D} - BC_{U}}{(BC/Al)^{crit}}\right) \tag{15}$$

4 Plant Response based on Base Cation:Hydrogen Ion Ratio

The use of the Base Cation: Aluminium ratio is inappropriate for highly organic, peat soils which have very small contents of aluminium. In these soils, the critical chemical limit may be more appropriately expressed in terms of the Base Cation: Hydrogen ion ratio at which root damage occurs. The limiting H^+ flux can then be determined as follows:

$$H_{L}^{+} = \frac{BC_{L}}{(BC/H)_{crit}} \tag{16}$$

The Base Cation leaching is calculated from a mass balance:

$$BC_{L} = BC_{W(CaMgK)} + BC_{D} - BC_{U} \tag{17}$$

Al is normally set to zero in pure peat:

$$[Al^{3+}] = 0 \tag{18}$$

We can now fill in the equation:

$$CL = ANC_W - H_L^{\cdot} \tag{19}$$

$$CL = ANC_W + \frac{0.5 \ . \ (x_{BC} \ . \ ANC_W + BC_D - BC_U}{(BC/H)_{crit}} \tag{20}$$

The factor 0.5 arises from the use of a molar BC/H^+ ratio in an expression based on equivalents. The critical load is then given by:

$$CL = ANC_W + \frac{0.5 \ . \ (x_{BC} \ . \ ANC_W + BC_D - BC_U}{(BC/H)_{crit}}$$

$$+1.5 \ . \ \frac{(x_{BC} \ . \ ANC_W + BC_D - BC_U)}{(BC/Al)_{crit}} \tag{21}$$

The equation should be applied to peat, mosses, shallow rooted grasslands, organic alpine soils, raised bogs, black earth soils.

5 Soil Stability Criteria

In high precipitation areas, net soil Aluminium depletion may cause structural changes in soils. For many soils, secondary Aluminium phases and complexes are important structure bearers in the soil. Stability of these soils depend on the stability of the reservoir of these substances. In high precipitation areas, acid deposition may potentially lead to Aluminium leaching in excess of Aluminium produced in weathering:

$$Al_l = Al_w \tag{22}$$

where Al_w = Production of Al from weathering in eq ha^{-1} yr^{-1}

The production of Al from weathering of minerals is related to the production of base cations from weathering through the stoichiometry of the minerals. An approximation, using typical mineralogy of North European soils would imply:

$$Al_W = RAL \ . \ BC_W \tag{23}$$

where RAL is the Aluminium to Base Cation release rate ratio in the weathering raction. RAL vary in the range from 1 in volcanic soils to 3 in soil mainly composed of aluminium-rich clays such as in the topical areas. A good average for RAL would be to use the value 2.

$$H_l = (\frac{RAL \ . \ BC_W}{K_{gibb}})^{1/3} \ . \ Q^{2/3} \tag{24}$$

The Aluminium criterium (leaching not greater than weathering of aluminium) leads to the equation for critical load of acidity:

$$CL^* = ANC_W + RAL \ . \ BC_W + (\frac{RAL \ . \ BC_W}{K_{Gibb}})^{1/3} \ . \ Q^{2/3} \tag{25}$$

where	CL*	= Critical load for soil stability	eq ha^{-1} yr^{-1}
	ANC_W	= BC_W neutralization from weathering	eq ha^{-1} yr^{-1}

$$CL^* = (1 + RAL) \ . \ ANC_W + (\frac{RAL \ . \ BC_W}{K_{Gibb}})^{1/3} \ . \ Q^{2/3} \tag{26}$$

The effective critical load will be the minimum of critical load of activity calculated from plant tolerance of Aluminium (BC:Al-ratio) and critical load of acidity calculated from soil stability criteria.

6 Basic Assumptions

The factor 1.5 derives from the conversion of critical loads and Base Cation concentrations in equivalents to molar ratio.

The equation is based on the following assumptions:

- The soil profile is assumed to be one stirred tank
- The same gibbsite coefficient is assumed to apply through the soil profile
- The weathering rate is evenly distributed over the soil profile
- Uptake is evenly distributed over the soil profile
- The weathering rate is independent of chemical conditions
- The BC/Al ratio is assumed to have a value such that the value of ANC_L always is negative.

The calculation does not distinguish between marine and non-marine ions of any sort. This implies that the full Base Cation deposition is used in the calculation. It also implies that the full sulphur deposition is used for calculation of sulphur exceedance and the full ion balance for the calculation of acidity exceedances. This implies that all Cl and Na also enter the calculation. Na is still excluded from being bioactive.

CHAPTER 6
CRITICAL LOADS OF NITROGEN: DYNAMIC MODELLING

Discussion Group Report

1 Merits of Dynamic Models

The steady state mass balance (SSMB) approach is an attractive tool for quantifying critical loads of N deposition to soils, because its data requirements are relatively modest. Although sometimes its application will involve applying simplifying assumptions and/or default data, preliminary N critical loads maps should be deliverable on a European scale on an acceptable time scale for policy makers. However the SSMB approach does not provide any indication of the quantitative effects of exceedance of critical load at any site. Nor does it provide any indication of timescales of change. This poses a serious problem when attempts are made to validate the use of the SSMB approach for critical load mapping, since it is always unclear how close the soil system is to steady state conditions. The problem is especially acute on sloping sites, where increased inputs from up slope of base cations in laterally flowing water may slow weathering rates, thus extending the time period after which physico-chemical damage symptoms will become detectable.

In contrast to steady state approaches, dynamic models allow *quantitative* assessment of the mid to long term impacts on soils of emission increase or reduction scenarios. They also allow soil changes to be predicted as a function of time. Long term effects of critical load exceedances can only be evaluated by the application of dynamic models, which also allow estimation of the time at which predicted changes will occur. Dynamic models therefore have an important role to play in the quantitative cost/benefit analysis of emission reductions and in the development of emission control strategies over realistic timescales. Thus dynamic models are a very helpful tool for policy makers.

International measures to reduce sulphur emissions may be offset by increasing nitrogen inputs to ecosystems. The effects of N and S deposition on soil ecosystems are interactive at the fundamental process level. Dynamic models may be used to disentangle the N and S deposition effects on soil acidification, for example. Ultimately, when incorporated into integrated catchment models, they can provide the essential link between critical loads for vegetation systems, soils, freshwater and groundwater.

2 Applications of Dynamic Models

Dynamic models may be applied over a diverse range of timescales, from days through months and years to decades or longer. They allow assessment of the importance of seasonality effects, for example on the contamination of groundwater with nitrate.

Dynamic models require detailed parameterization and are therefore generally applied on a site-specific basis. However, if sufficient data are available, they also may be applied on a regional scale. This allows assignment of critical loads for mapping on the basis of precisely specified "damage" criteria, and relationships between exceedance and different degrees of damage or differences in growth increment data from long-term forest records to be evaluated.

3 Current Dynamic Models for Assessing N Critical Loads

Dynamic models have been tested and improved since the Lökeberg Workshop in 1992, and several of the current models take nitrogen transformations into account. Models covering a range of complexity are currently under evaluation, the more sophisticated ones incorporating effects of carbon cycling and taking into account the dynamics of other key nutrient elements as well as nitrogen.

a) MAGIC

Model of Acidification of Groundwaters in Catchments and Derivatives
MAGIC has been used to produce critical loads exceedance maps for Norway for different scenarios for nitrogen and sulphur deposition. Results from the NITREX nitrogen saturation experiment and from the ENSF (evaluation of nitrogen and sulphur fluxes) database have been used to allow nitrate output predictions from known N inputs.

A model developed at the Institute of Hydrology in the UK, MAGIC WAND, a derivative of MAGIC (MAGIC with Associated Nitrogen Dynamics), couples nitrogen and sulphur deposition effects, using inputs such as mineralisation and nitrogen fixation rates as driving variables (Ferrier *et al.* 1995). Nitrification is quantified in terms of a turnover rate, and denitrification *via* a first order rate function dependent upon external nitrogen concentrations. Uptake is assumed to follow approximately Michaelis-Menton kinetics.

b) MERLIN

Model for Ecosystem Retention and Loss of Nitrogen
MERLIN is a new model based upon the assumption that major processes are controlled by variables that are dependent upon the carbon to nitrogen ratio of terrestrial pools (Ferrier *et al.* 1995).

c) SAFE

Soil Acidification in Forest Ecosystems
SAFE is the dynamic counterpart of the well known steady state PROFILE model developed by Sverdrup *et al.* at Lund, Sweden, and uses the same input data and weathering rate sub-model as PROFILE (Warfinge *et al.* 1993). However SAFE employs time series data for deposition, uptake, hydrology and climate, to give time series outputs showing changes in soil solution chemistry and soil base saturation. It has been generally applied using yearly average values for inputs and outputs.

d) SMART

Simulation Model for Acidification Regional Trends
SMART is the simplest of three models of increasing complexity being developed and evaluated at the Winand Staring Centre in Wageningen. It was developed as a relatively simple single layer soil model for application on a Europe-wide scale, which also evaluated combined effects of sulphur and nitrogen deposition (De Vries *et al.* 1989, 1995).

d) RESAM

Regional Soil Acidification Model
RESAM is more complex than SMART in that it is a multi-layer model, designed for application throughout the Netherlands (De Vries *et al.* 1994). It includes nutrient cycling on an annual basis and

key processes in the nitrogen cycle. It assumes first order reactions for nitrogen transformations. Weathering and exchange are treated as first order and equilibrium reactions as appropriate.

e) NUCSAM

Nutrient Cycling and Soil Acidification Model
Unlike RESAM, which processes data on an annual basis, NUCSAM is a multi-layer soil model based on one-day time increments (Groenenberg *et al.* 1995). It is targeted at evaluation of exceedance effects on a site-by-site basis.

Even more sophisticated models are under development in the Netherlands. For example, an integrated soil/vegetation model (SOILVEG) couples a soil model analogous to RESAM with a simulated forest tree growth and nutrient dynamics model. Such models are complex and have substantial data input requirements.

4 Policy Needs and Model Development

In spite of the increasing availability of more complex models, at present it seems that time limitations imposed by policy formulation constraints will dictate the use of simpler regional scale models in the first instance.

The above, necessarily brief, synopsis of existing models is not intended to be exhaustive, but rather to give some insight into the types of approach currently under consideration throughout Europe. It is not felt that any of the models, as they stand, are yet ready for widespread application. However, when attempts are made to apply and/or to validate models, and problems or apparant anomalies are identified, this often results in an improvement in our understanding of ecosystem functioning, and to subsequent model refinement. Thus although in some respects several of the models being tested might be regarded as the result of excessive optimism at the present time, there is a clear need for modelling and fundamental understanding to develop side by side.

References

De Vries, W., Kros, J. and van der Salm, C. (1994) The long-term impact of three emission-deposition scenarios on Dutch forest soils. *Water Air Soil Pollut.*, 75, 1-35.

De Vries, W., Posch, M. and Kämäri, J. (1989) Simulation of the long-term soil response to acid deposition in various buffer ranges. *Water Air Soil Pollut.*, 48, 349-390.

De Vries, W., Posch, M., Reinds, G.J. and Kämäri, J. (1995) Simulation of soil response to acidic deposition scenarios in Europe. *Water Air Soil Pollut.*, 79, 1-32.

Ferrier, R.C., de Vries, W. and Warfvinge, P. (1995) The use of dynamic models for the determination of critical loads for nitrogen - developments since Lökeberg. *Proceedings of the UNECE Workshop on Nitrogen Deposition and its Effects: Critical Loads Mapping and Modelling, Grange-over-Sands, 24-26 October 1994,* Department of the Environment, London.

Groenenberg, J.E., Kros, J., van der Salm, C. and De Vries, W. (1995) Application of the model NUCSAM to the Solling spruce site. *Ecological Modelling*, submitted for publication.

Warfvinge, P., Falkengren-Grerup, U., Sverdrup, H. and Andersen, B. (1993) Modelling long-term cation supply in acidified forest stands. *Environ. Pollut.*, 80, 209-221.

Keynote Paper - The use of Dynamic Models for the Determination of Critical Loads for Nitrogen - Developments since Lökeberg.

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1 Introduction

The acidifying effects of sulphur and nitrogen deposited from the atmosphere depend upon the mobility and retention of the sulphate and nitrate anions once in the ecosystem. Soils in glaciated regions typically have low-to-moderate sulphate adsorption capabilities and thus sulphate inputs and outputs are generally in steady state. Changes in sulphate deposition appear rather quickly as changes in the sulphate concentration of waters draining from the soils. If sulphate concentrations decline, acid stresses on the soils are reduced. Nitrogen on the other hand, is generally retained within the ecosystem, largely because nitrogen is growth limiting in many systems. Chronic elevated deposition of nitrogen, however, can produce quantities of inorganic nitrogen in the soils in excess of that needed by the biota for sustained growth. The un-assimilated nitrogen can be subject to leaching into surface waters. Generally this leached nitrogen is in the form of nitrate in runoff waters. As with sulphate, nitrate is a mobile strong anion and acts to acidify the soils. Nitrogen "saturation" therefore, reduces the critical loads of both nitrogen and sulphur.

The use of dynamic (Level 2) modelling approaches has been identified as essential to the assessment of critical loads. The load calculations elucidate the time taken for systems to reach equilibrium and the transition between present and potential exceedances. They form the only basis for investigating the coupled relationship between potential recovery from sulphur deposition and the interaction with nitrogen. Dynamic models include processes such as cation exchange, anion adsorption, and the role of land management (for example, forest growth dynamics) that result in time dependency of soil and water response to acidic inputs. It is also important to distinguish between dynamic soil-oriented models where vegetation response is calculated using litterfall and tree growth as forcing functions, and integrated models which include a prediction of biotic response. The latter (integrated) models are most suitable for a critical load calculation, whereas the former can only simulate dynamic changes in soil solution chemistry.

A range of different soil acidification models have been developed to address the dynamics of terrestrial nitrogen, such as, Agren (1983), VEGIE, (Aber *et al.*, 1991), RESAM (de Vries *et al.*, 1991), SOILVEG, (van Grinsven *et al.*, 1991) and NIICE (van Dam 1992). One of the major problems with respect to critical load calculations and mapping in particular, is that parameterisation is limited by data availability and this is manifest in problems of spatial integration. Other models, such as MAGIC (Cosby *et al.* 1985) have successfully been used for the calculation of critical and target loads for sulphur at a range of spatial scales.

This paper reviews the current status of the application of coupled nitrogen-sulphur dynamic models and the determination of critical loads, since the Lökeberg Workshop (Grennfelt and Thornelof (1992)). Proposed developments of the different modelling groups throughout Europe are also highlighted.

2 General approach

It is apparent that development of sound catchment management strategies (for both aquatic and terrestrial resources) requires appropriate tools for understanding the water-level processes and biogeochemical effects of nitrogen saturation phenomena. In particular, lumped mathematical models are required to integrate current understanding of biogeochemical processes affecting nitrogen at all scales in watersheds and within a regional framework. For this to be achieved , models require the following attributes:

i they should be applicable at the catchment scale (i.e. stated variables within the model should relate to whole ecosystem responses- such as water quality or plant productivity - which integrate effects of processes operating at various scales within the catchment).

ii the models should be applicable to several ecosystem types.

iii it should be possible to calibrate/validate the models using catchment scale variables (ie. aggregated soil characteristics; statistical descriptions of terrestrial biota; annual average, seasonal and/or episodic soil and surface water concentrations; survey data.)

iv the models should be amenable to implementation at multiple sites or on a regional basis using mapped, remotely sensed or surveyed data (i.e. model complexity should be maintained at a level consistent with process knowledge while simultaneously recognising that management needs often require model applications in situations where data are sparse or incomplete).

v the models should be designed with a view to incorporation into a decision making framework (i.e. the models can be used to assess alternative control scenarios which in turn require that potential control variables such as land use patterns, atmospheric deposition, can be represented in the model structure).

vi the models should provide a mechanism for coupling the dynamics of nitrogen to the hydrochemistry of soils and surface waters.

2.1 Overview of approaches.

2.1.1 Development of the MAGIC model framework, and the Model of Ecosystem Retention and Loss of Inorganic Nitrogen (MERLIN)

Current catchment and regional scale dynamic modelling of the effects of nitrogen deposition (oxidised and reduced forms) on nitrogen leaching within the MAGIC model framework is being approached at three levels. The choice of levels is mainly dependent upon the data available for model calibration but the intended use of model outputs is also an important consideration. The three levels are distinguished by the level of aggregation of the terrestrial nitrogen cycle within the model structures, ie. lumped soil (MAGIC), process soil (MAGIC-WAND), and intergrated soil-forest (MERLIN).

(a) MAGIC (Model of Acidification of Groundwaters In Catchments).

The current version of MAGIC comprises an extremely simplified representation of nitrogen involving a specified net catchment retention to calibrate simulated nitrogen against observed nitrogen in stream water. This retention is the net result of all fluxes and processes of the terrestrial nitrogen cycle. This has previously been used to assess the influence of nitrogen deposition on calculated critical loads for sulphur, by varying the net catchment retention of nitrogen (Figure 1) (Jenkins and Shaw, 1992). This approach has been furthered by representing the net nitrogen retention as a function of nitrogen deposition. Compiled input-output data from coniferous forest ecosystems in Europe (Dise and Wright, 1995) have been used to set empirical "best case" and "worst case" limits for nitrogen retention. The data suggested that (a) nitrate leaching from soils is minimal in catchment areas with nitrogen deposition levels less than 10kg N ha^{-1} yr^{-1}, (b) nitrate leaching varies from 0-100% of nitrogen deposition in catchments with nitrogen deposition in the range 10-25kgN $ha^{-1}yr^{-1}$, and, (c) nitrate leaching is 50-100% of nitrogen deposition in all catchments with nitrogen deposition more than 25kg N ha^{-1} yr^{-1} (Figure 2).

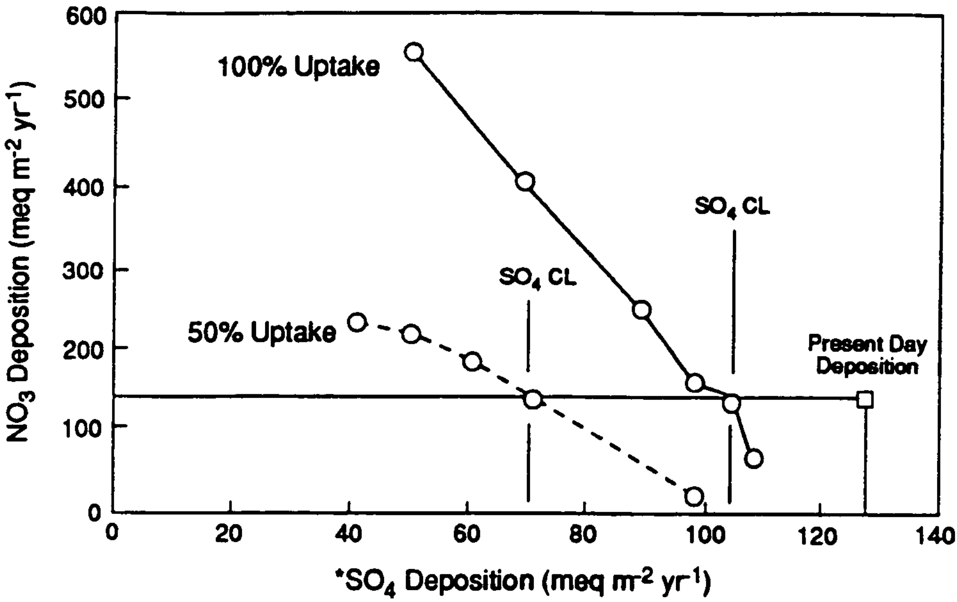


Figure 1 **The interaction between sulphur and nitrogen critical loads. The graph shows MAGIC calculations for Round Loch, SW Scotland, of critical loads for sulphate at a given N deposition, assuming 100% or only 50% N uptake. Sulphate critical loadings for these two assumptions at the present rate of N deposition are shown.**

Frogner *et al.* (1994) calculated the critical loads for forest soils in Norway based on these empirical relationships combined with appropriate future sulphur scenarios using MAGIC. The criterion value was that the Ca/Al ratio in the uppermost 50 cm soil solution should not be less than 1.0 at a point fifty years in the future. In the worst case calculated critical loads for nitrogen are low and exceeded in southernmost Norway. However, the variability in response to different scenario combinations highlights the need for more information regarding the magnitude of nitrogen retention and processes controlling N cycling. There is a requirement for a more explicit representation of the nitrogen cycle, particularly under changing patterns of land use. In particular, the model must be capable of reproducing the observed change in net nitrogen retention as a function of forest age.

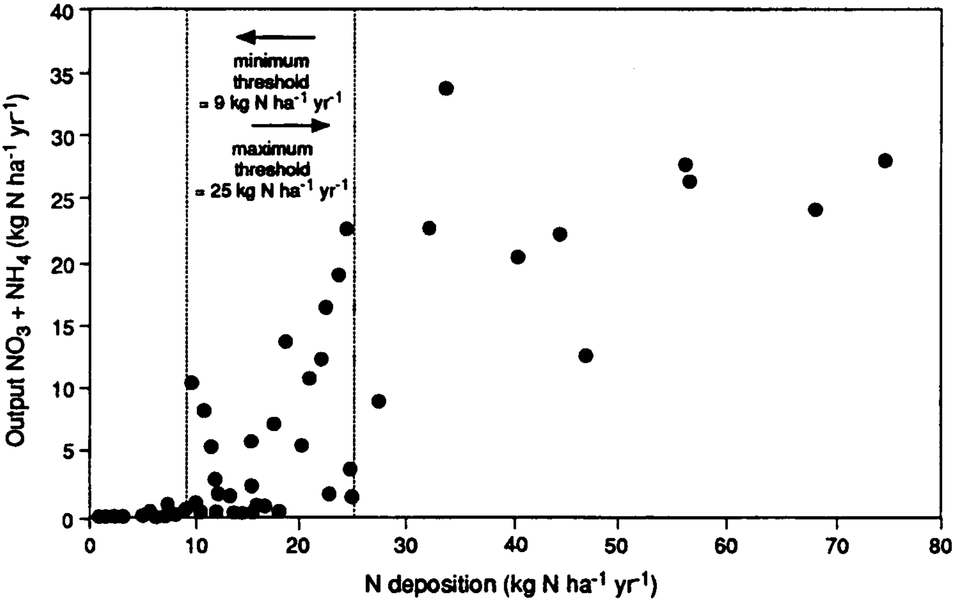


Figure 2 **Empirical relationship between input (Wet+dry) and output at coniferous ecosystems in Europe. Data are delimited by "worst" and "best case" lines of nitrogen retention: these were used as scenarios in determining nitrogen critical loads.**

(b) MAGIC-WAND (MAGIC - With Aggregated Nitrogen Dynamics).

This represents an extension to the MAGIC model to incorporate the major nitrogen fluxes and changes through time (Figure 3). The nitrogen dynamics are fully coupled to the existing suphur driven model. This model structure is designed to enable assessment of future surface water chemistry responses to a given nitrogen deposition scenario. Assumptions relating to the uptake capabilities of the vegetation and future land use change and the sensitivity of a catchment or region to these components can also be assessed.

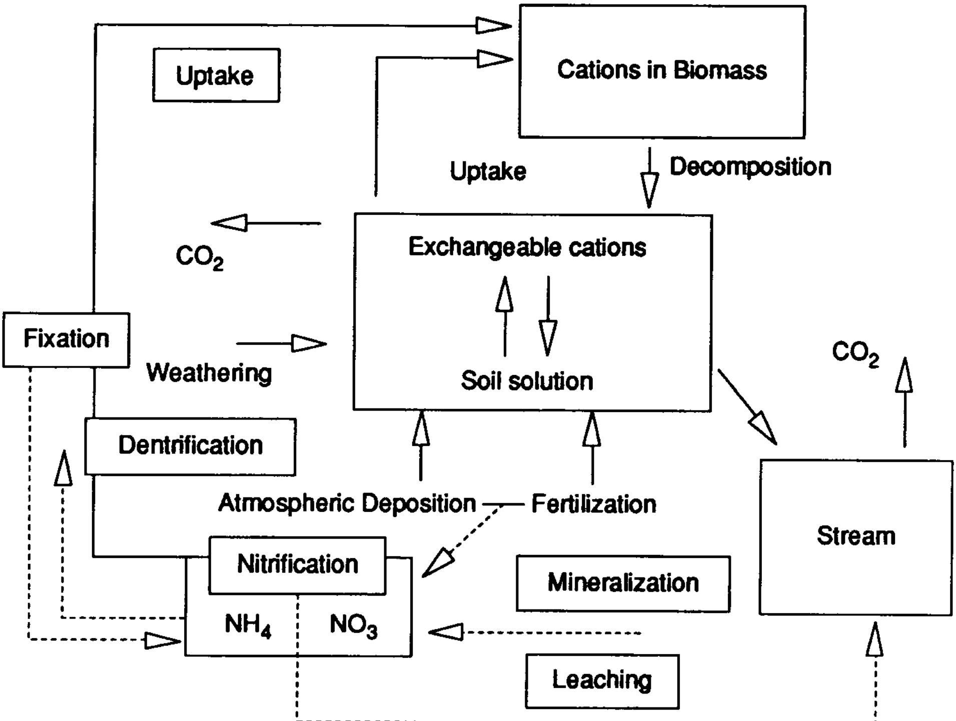


Figure 3 **Conceptual diagram of the fully coupled sulphur-nitrogen model MAGIC-WAND (MAGIC - With Aggregated Nitrogen Dynamics).**

MAGIC-WAND considers both species of inorganic nitrogen , nitrate and ammonium. Both species are assumed to be present only in solution in soil water. The model explicitly incorporates the major terrestrial fluxes of nitrogen such that if the net result of these fluxes is positive (surplus nitrate and/or ammonium), leaching to surface waters occurs:

Nitrate leaching = (deposition + nitrification +external addition) - uptake - denitrification.

(c) MERLIN (Model of Ecosystem Retention and Loss of Inorganic Nitrogen)

This model structure is the most scientifically rigorous of the three approaches. All of the processes described in MAGIC-WAND are calculated from internal state variables and not incorporated as input driving variables. Such internal state controls are based on the carbon and nitrogen pools of the terrestrial compartments (photosynthetic biomass, wood, litter, labile organic matter, refractory organic matter etc.)(Figure 4). This model requires detailed infor mation describing nitrogen and carbon fluxes and pool sizes for calibration, and the model is currently being calibrated to sites in the NITREX nitrogen manipulation research programme (Dise and Wright 1992). A generic approach, based on generalised forest productivity and flux determination is also being developed for regional spatial integration.

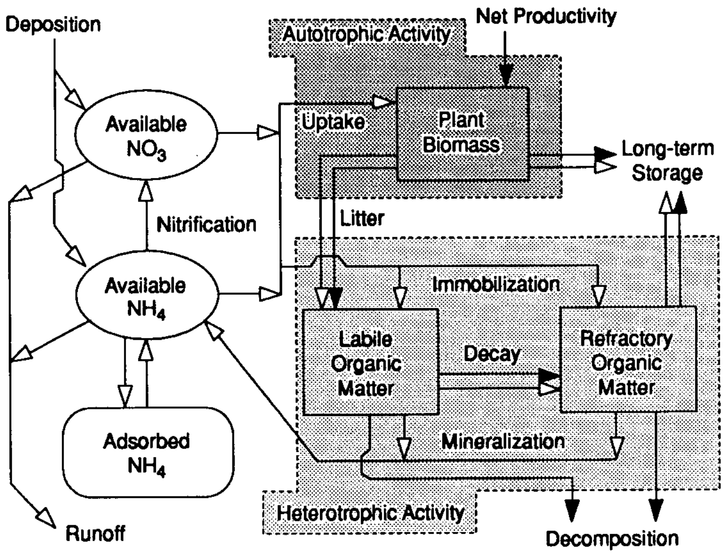


Figure 4 **Model of Ecosystem Retention and Loss of Inorganic Nitrogen (MERLIN) - a process based model for simulating catchment scale retention and loss of inorganic nitrogen.**

2.2 SMART, RESAM and NUCSAM

2.2.1 Approach

As with the MAGIC model framework, three levels of detail can be distinguished in soil acidification models that have been developed at DLO Winand Staring Centre, i.e. SMART (Simulation Model for Acidificationís Regional Trends; De Vries *et al.* 1989), RESAM (Regional Soil Acidification Model; De Vries *et al.* 1994), and NUCSAM (Nutrient Cycling and Soil Acidification Model; Groenenberg *et al.* 1995). NUCSAM has been coupled with FORGRO (Forest Growth Model; Mohren *et al.* 1991) to simulate the response of both the soil and the tree, in terms of growth and nutrient dynamics, to acid deposition. SMART, RESAM, and NUCSAM/FORGRO are comparable to MAGIC, MAGIC-WAND, and MERLIN respectively. The aim of the models and the scale of application were factors of crucial importance in selecting the level of detail in both the model formulation and associated input data. A less detailed objective, and a decreasing data availability at a larger application scale justified the development of a simpler modelling approach (Table 1).

Table 1 *Characteristics of the dynamic soil acidification models used at the Winand Staring Centre.*

Name	Complexity	Soil layering	Time step	Application scale
SMART	Simple	one-layer	one year	European
RESAM	Intermediate	multi-layer	one year	National
NUCSAM	Complex	multi-layer	one day	Site specific

To minimise input requirements, the models SMART and RESAM designed for regional predictions are simpler than the site scale model NUCSAM. The simplifications consist of:

i the reduction of temporal resolution i.e. using an annual time step, thus suppressing interannual variability of both model inputs and processes,
ii reduction in spatial resolution, by using a smaller number of soil compartments, and
iii the use of less detailed process formulations (Table 2)

Table 2 Processes (descriptions) in the soil acidification models developed.

Processes	SMART	RESAM	NUCSAM
Water flow	Annual Precipitation excess	Annual flow varying with depth	Hydrological sub-model (SWATRE)
Nutrient cycling	Not included	Included on an annual basis	Included on a daily basis[1)]
Nitrogen transformations	Simple linear relationships	First -order kinetic reactions .	First-order kinetic reactions[2)]
Weathering & exchange	Equilibrium reactions	First-order and equilibrium reactions	First order equilibrium reactions[2)]
Complexation reactions	Not included	Not included	Equilibrium Reactions[2)]
Heat flow	Not included	Not included	Heat transport sub-model

[1)] In the stand-alone version, tree growth is included as a forcing function. coupled with FORGRO, nutrient dynamics are simulated in the tree (stems, branches, leaves and roots) as well.

[2)] Temperature dependent.

An important aspect is the time-step. Since SMART and RESAM were specifically designed to evaluate long-term responses to deposition scenarios on a continental to national (regional) scale, these models do not include seasonal dynamics. The temporal resolution of the models is one year, and the description of hydrology in these models is very simple. Simulation of the seasonal variability is, however, included in NUCSAM, which is specifically developed for application (and validation) on a site specific scale.

2.2.2 **Validation**

Until now SMART, RESAM, and NUCSAM have not been used for critical load calculations, but to simulate N dynamics in the soil (solution). The models have been validated on an intensively monitored spruce stand at Solling, Germany, by comparing simulated concentrations

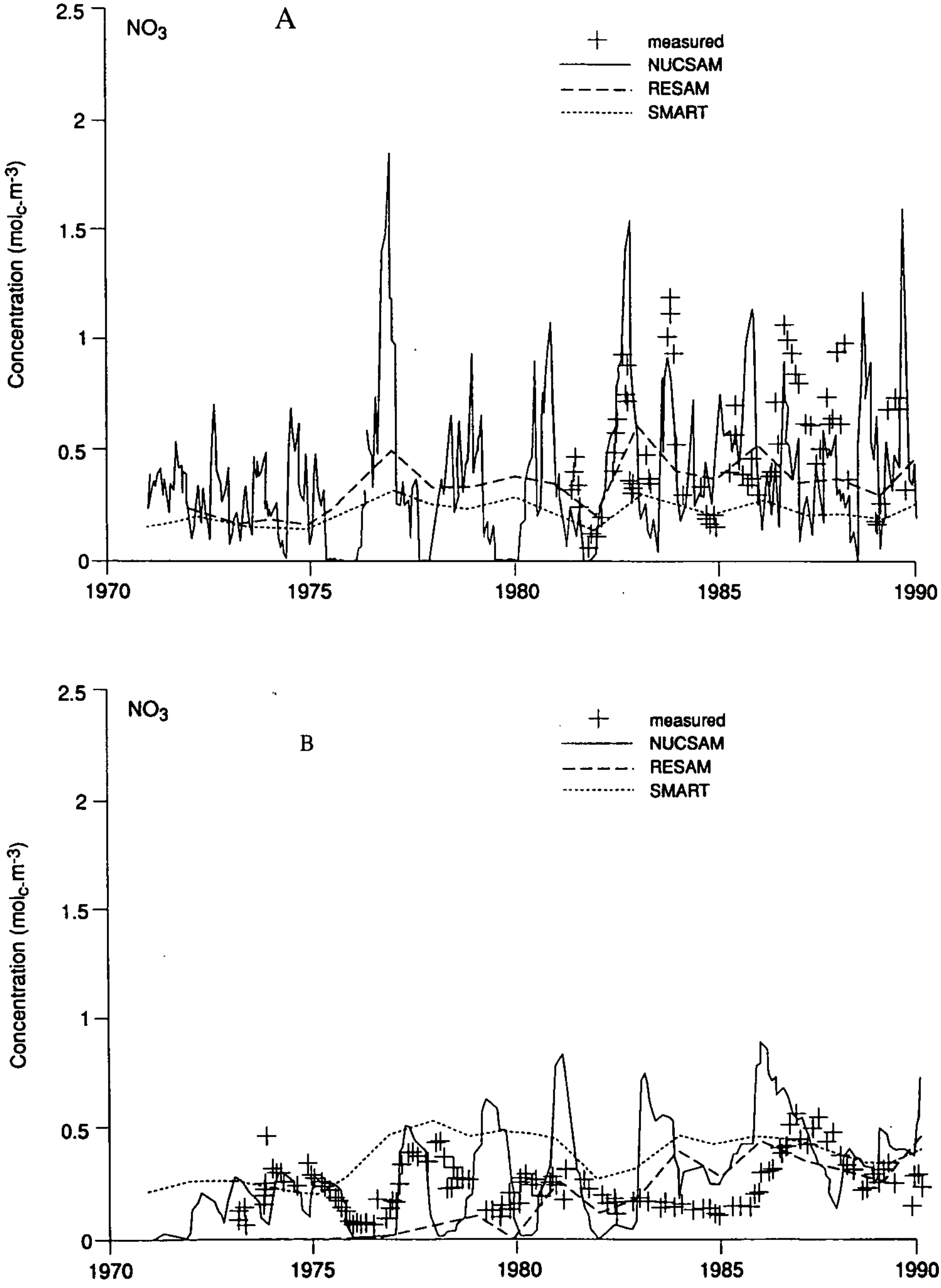


Figure 5 **Observed and simulated NO_3^- concentrations with SMART, RESAM and NUCSAM at the Solling site, Germany, at 10 cm depth (A) and 90 cm depth (B).**

and measured values during the period 1973-1989 (Van der Salm *et al.* 1995). Simulated and measured nitrate concentrations in the topsoil (10 cm) and subsoil (90 cm) are shown in Figure 5. The influence of the chosen temporal resolution on model performance can be seen most clearly for the simulated concentrations in the topsoil, which are strongly influenced by seasonal processes. Nitrate concentrations in the topsoil (Figure 5a) simulated with NUCSAM were in close agreement with the measurements, whereas RESAM could not accurately simulate the seasonal peaks in nitrate concentrations. SMART underestimated nitrate concentrations in the topsoil due to the neglect of nutrient cycling and mineralization.

To gain an insight into the reliability of the predictions of the model RESAM on a national scale, a comparison was made between results of 550 model simulations on the soil solid phase and soil solution chemistry in 1990 with measurements in 150 forest stands during the period March to May in the same year (De Vries *et al.* 1994). The tree species and soil types included in the field survey were similar to those included in the simulations. The comparison between simulated and observed nitrate concentrations shows a reasonable fit (Figure 6).

The SMART model has been applied on a European scale (De Vries *et al.*. 1995). At this scale no soil solution data were available for validation. However, an important aspect in the nitrogen dynamics in SMART is the relation between C/N ratio of the soil and N immobilisation. Some authors state that there is no such relationship because there is little correlation between observed nitrate concentrations and C/N ratios. However, the response is simulated with SMART (Figure 7), suggesting that SMART does at least produce consistent results.

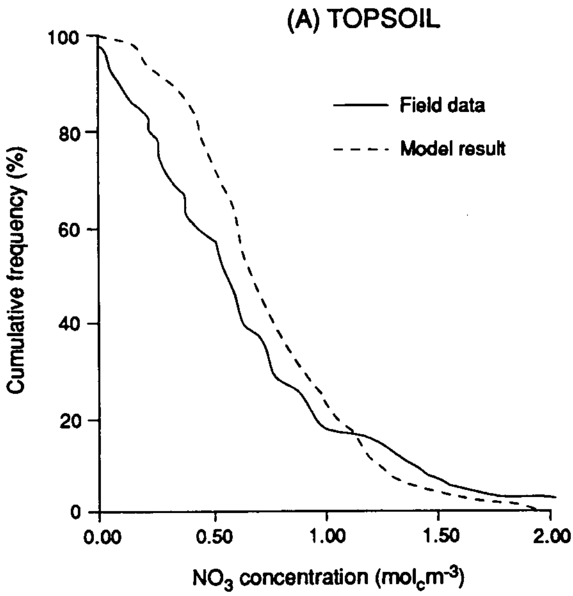
2.3 SAFE

2.3.1 **Approach**

The nitrogen chemistry of the dynamic SAFE model and its steady-state counterpart PROFILE is basically driven by forcing functions. In principle, these are N deposition and uptake, while cycling of N can be viewed as increased deposition and uptake. For SAFE it is vital to represent the dynamics in nitrogen chemistry, while PROFILE requires lumped information incorporating the whole forest rotation. Nitrogen and Base Cation cycling is important for the chemical dynamics of forest ecosystems. At present, the nutrient dynamics caused by an increase in mineralization after forest harvest are not modelled in a mechanistic way driven by fundamental processes such as the rate of photosynthesis and/or microbial activity. Instead, a separate module has been developed to support the parameterisation of SAFE with respect to deposition, uptake, and cycling. This module is used to scale these processes with respect to amplitude and time in a systematic manner.

Model inputs:

- stand history,
- present biomass in different compartments,
- nutrient content in tree compartments,
- present bulk deposition and throughfall.



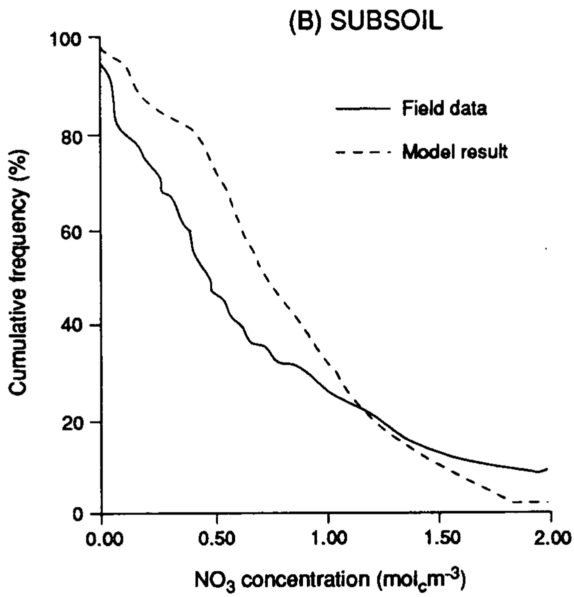


Figure 6 **Frequency distribution of field data and model predictions with RESAM of the NO_3^- concentrations of the topsoil (A) and subsoil (B) of forest stands in the Netherlands.**

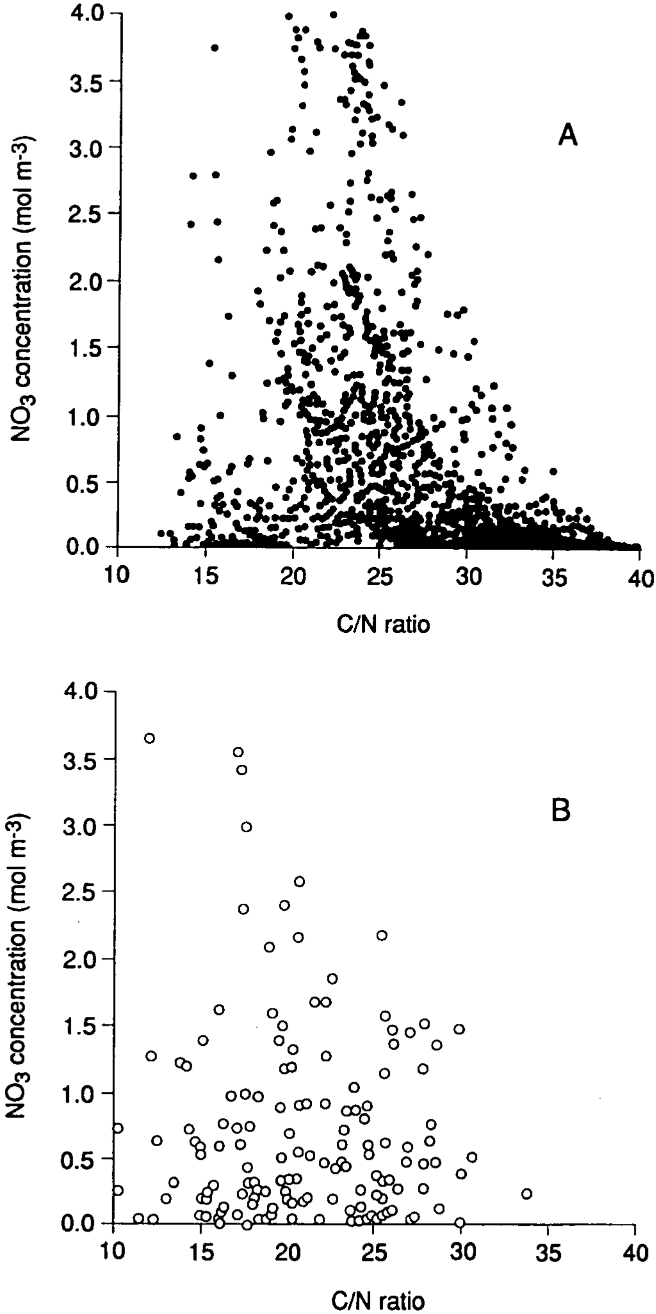


Figure 7 **Relationships between dissolved NO_3^- concentration and C/N ratio of the soil as predicted in European forest soils with SMART (A) and measured in 150 Dutch forest soils (B).**

Scaling functions are:

- historic deposition patterns (from RAINS and EMEP),
- logistic growth curves, including growth of foliar biomass,
- rate of mineralisation of litterfall.

Model outputs are:

- time series of total deposition both of marine and anthropogenic, wet and dry,
- time series of gross uptake.

This module allows the user to determine whether nitrogen is supplied by deposition and mineralisation of litter only, or if other nitrogen sources can be utilised. Another option that the user has regards nitrogen immobilisation. As this is not driven by internal parameters such as C/N, immobilisation has to be specified as being either complete, zero, a fixed maximum value, or a fraction of free nitrogen in solution.

In summary, this model fills an important gap in model parameterisation, but it cannot be used to predict N-saturation based on actual biogeochemical processes. N saturation may indeed suddenly appear in SAFE runs when this parameterisation procedure is applied, for example, in situations where N immobilisation is limited. The procedure implemented in this support module does, however, result in consistent parameterisation of SAFE which reduces the risk of incompatible assumptions. The only mechanistic element included in these models is nitrification which is kinetically controlled in PROFILE. In SAFE, nitrification is assumed to be complete, which is equivalent to assuming that there is no ammonium leaching from the top soil horizon. This also means that all N deposition is regarded as acidity, and all N uptake as a neutralising process.

SAFE is presently undergoing major revisions and extensions. The simple structure with only three state variables is being modified to include all major cations and anions and their associated biogeochemistry. It remains to be seen if this will improve the possibilities of predicting the major responses of terrestial ecosystems to atmospheric deposition of nitrogen and other substances.

3 General discussion

It is clear that dynamic models are the only tool available to extrapolate coupled sulphur and nitrogen dynamics, both in terms of further acidification and the potential for recovery, and are therefore important for the design of effective abatement strategies. Given an agreed empirical critical load, dynamic models calculate the length of time required to achieve the desired chemistry. Similarly, models provide a means of assessing the response of a system given agreed reductions in anthropogenic deposition, and the consequence of not achieving critical load.

Dynamic models are of importance in evaluating the significance of interacting processes such as land-use change and deposition. In many European systems fast growing commercial afforestation has a crop rotation length of approximately fifty to seventy years. In some of the dynamic modelling studies, this same time period is used for the calculation of critical loads. Therefore, such calculations must incorporate temporal changes during the life of the forest, and dynamic models provide a tool with which to detail changes in water use, cation uptake by the biomass, and patterns of canopy interception.

It is also important to have a mechanism with which to investigate the relative importance of deposition of different nitrogen species. For an accurate assessment of the impact of changing patterns of nitrogen deposition, models must be able to evaluate the relative effects of both NH_x and NO_x species and their accompanying counter-ions.

Finally, development of new modelling approaches must be compatible with the acquisition of appropriate field response and process based information. Dynamic models have the potential to be utilised at a range of spatial scales from the catchment to region to national, but the applicability of the modelling output at these different spatial levels is limited by the quality of the data available.

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5 References

Aber, J.D., Nadelhoffer, K.J.,Streudler, P. and Mellilo, J.M. (1991) Factors controlling nitrogen cycling and nitrogen saturation in temperate forest ecosystems. *Ecological Applications*, 1, 303-315.

Agren, G.I. (1983) Model analysis of some consequences of acid precipitation on forest growth. In: Ecological effects of acid deposition, Swedish National Environment Protection Board, Stockholm, Report PM 1636, pp. 233-244.

Cosby, B.J., Hornberger, G.M., Wright, R.F. and Galloway, J.N. (1985) Modelling the effects of acid deposition: assessment of a lumped parameter model of soil water and stream water chemistry. *Water Resources Research*, 21, 51-63.

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

Dise, N. and Wright, R.F. (Eds) (1992) The NITREX Project (Nitrogen Saturation Experiments). Commission of the European Communities, Brussels, Ecosystems Research Report No.2, pp 101.

De Vries, W., Posch, M. and Kamari, J. (1989) Simulation of the long-term soil response to acid deposition in various buffer ranges. *Water, Air, and Soil Pollution*, 48, 349-390.

De Vries, W., Kros, J. and Van der Salm, C. (1994) The long-term impact of three emission-deposition scenarios on Dutch forest soils. *Water, Air, and Soil Pollution*, 75, 1-35.

De Vries, W., Kros, J., Van der Slam, C. and Voogd, J.C.H. (1991) Effects on soils. In: Heij, G.J. and Schneider, T. (Eds.) Acidification in the Netherlands. Elsevier, Amsterdam, pp. 169-179.

De Vries, W., Posch, M., Oja, T., Van Oene, H., Kros, J., Warfvinge, P. and Arp, P.A (1995) Modelling critical loads for the Solling Spruce Site *Ecological Modelling*, (in press).

Frogner, T., Wright, R.F., Cosby, B.J. and Esser, J. (1994) Maps of critical load exceedance for sulphur and nitrogen to forest soils in Norway. NIVA Report No. O-91147, Oslo, Norway.

Grennfelt, P. and Thornelof, E (Eds) (1992) Critical Loads for Nitrogen - a workshop report. Nordic Council of Ministers, Nord,41.

Groenenberg, J.E., Kros, J., Van der Salm, C. and De Vries, W. (1995) Application of the NUCSAM model to the Solling Spruce Site. *Ecological Modelling*, (in press).

Jenkins, A. and Shaw, C. (1992) Comparison of empirical and dynamic model approaches for surface waters. In: Hornung, M. and Skeffington, R.A. (Eds.) Critical loads: concept and applications. ITE Symposium No. 28, pp. 109-114.

Mohren, G.M.J., Jorritsma, I.I.M., Florax, J.P.G.G.M., Bartelink, H.H. and Van den Veen, J.R. (1991) FORGRO 3.0. A basic forest growth model: model documentation and listing. Research Institute for Forest and Urban Ecology Report 524, Wageningen, The Netherlands, pp. 137.

Van Dam, D. (1992) NITREX model NIICE. In: Dise, N. and Wright, R.F. (Eds.) NITREX- Nitrogen Saturation Experiments. CEC Report No. 2, pp.101

Van der Salm, C., Kros, J., Groenenberg, J.E., De Vries, W. and Reinds, G.J. (1995) Validation of soil acidification models with different degrees of process aggregation in an intensively monitored spruce site. In: Trudgill, S. (Ed.) Solute modelling in catchment systems (submitted)

Van Grinsven, J.J.M., Van Minnen, J.G. and Van Heerden, C. (1991) Effects of growth of Douglas Fir. In: Heij, G.J. and Schneider, T. (Eds.) Acidification in the Netherlands, Elsevier, Amsterdam, pp. 180-190.

CHAPTER 7
CRITICAL LOADS OF NITROGEN FOR MARINE ECOSYSTEMS

DISCUSSION GROUP REPORT

The numbers at the end of each paragraph apply to Sections 6.3.1 to 6.3.5 of the Mapping Manual (TFM, 1993).

The conclusions from Lökeberg were reviewed, and are found to be still valid.

Even though much research on marine eutrophication and modelling of physical and biological processes has been performed since the Lökeberg meeting, not much progress has been achieved in terms of critical loads for the marine environment. Only a few countries have until now actively taken part in the critical levels work.

- There seems to be agreement among marine scientists that the critical load concept can be applied to some well defined geographical areas. Among these areas are: the Baltic Sea, Skagerrak/Kattegat coastal waters and some coastal waters in the Southern North Sea and fjords. (*Section 6.3.1*)

- It now seems possible to establish empirical nitrogen concentration-effects relationships for these areas. (*Section 6.3.1*)

- The definition of eutrophication is still not generally accepted and will have to be reviewed. (*Section 6.3.2*)

- The main nutrients/parameters of interest are nitrogen and phosphorous. Carbon and silicate may be of interest. (*Section 6.3.3*)

- Empirical relationships on increased N concentrations and effects (in terms of increased chlorophyll a, or changes in planktonic and/or benthic communities) should be established for the areas where such data exist. Data from Denmark, Sweden, Norway and The Netherlands are currently being summarized and should be published shortly. (*Section 6.3.3*)

- Critical N concentrations/critical levels should be based on empirical data at the present time. (*Section 6.3.3*)

- It seems premature to establish a link between critical N concentration and critical load. Reasons are partly that we still are not able to quantify the different diffuse (non-point) sources for N or the relative contribution of atmospheric N deposition (and N leaching?) to land run-off from agricultural areas.

General remarks

- An example of a simple empirical model approach is described by Sverdrup and Barkman in the following keynote paper.
- The definition of critical loads for N of Sverdrup and Barkman (this volume) could be useful.
- The work on modelling is improving. Models are now being developed to include both physical and biological processes.
- Some parties to the OSPARCOM are now in favour of adopting a dose-response approach for the North Sea abatement strategies.
- Further work is necessary within the the Convention on Long-Range Transboundary Air Pollution (LRTAP) in order to establish a fluid communication with other international actions assessing the effects of Nitrogen deposition on the marine environment in particular areas of the UNECE. Other International Maritime Conventions with which a future Convention on LRTAP action should be coordinated include the:
  - Oslo and Paris Conventions for the North Atlantic and North Sea (OSPARCOM, 1993)
  - Helsinki Conventions for the Baltic Sea (HELCOM, 1991, 1994)
  - Barcelona Convention and the Mediterranean Action Plan (Ederman et al., 1994)
- It must be noted however that the terrestrial inputs of nitrogen to the Mediterranean coastal waters are very large compared with direct atmospheric deposition.
- Toxic algal blooms and benthic community effects are probably the two main short term effects of marine eutrophication in the Mediterranean.

References

Ederman L., Soudine A., Subbotin S., Dedkova I., Afinogenova O., Cheshuikina T. and Pavlovskaya L. (1994) Assessment of airborne pollution of the Mediterranean Sea by sulphur and nitrogen compounds and heavy metals in 1991. Mediterranean Action Plan (MAP) Technical Report Series No. 85. UNEP/WMO, P.O. Box 18019, Athens, Greece. 304 pp. [ISBN 92-807-1438-4].

HELCOM (1991) Airborne pollution load to the Baltic Sea 1986-1990. Baltic Sea Environment Proceedings No. 39, Baltic Marine Environment Protection Commission, Helsinki Commission, Katajanokanlaituri 6B, FIN-00160 Helsinki. [ISSN 0357-2994].

HELCOM (1994) Intergovernmental activities in the framework of the Helsinki Convention 1974-1994. Baltic Sea Environment Proceedings No. 56, Baltic Marine Environment Protection Commission, Helsinki Commission, Katajanokanlaituri 6B, FIN-00160 Helsinki. [ISSN 0357-2994].

OSPARCOM (1993) North Sea quality status report 1993. North Sea Task Force, Olso and Paris Commissions. London. [ISBN 0 946956 32 4].

TFM (1993) Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded. Texte 25/93. TFM / CCE / UNECE. Task Force on Mapping, Uweltbundesampt (UBA), Bismarckplatz 1, W-1000 Berlin 33. Germany.

Keynote Paper - **Critical Loads of Nitrogen for Marine Ecosystems: Suggesting and Applying a Simple Method to the Bothnian Sea**

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Abstract

A simple empirically based mass balancing method is proposed for calculating critical loads of N for coastal waters, estuaries and the Baltic Sea using empirical concentration-response relationships. The proposed method relates observed change to present N concentrations, and N concentrations to N loads using load models. For defined areas, simple mass balance models can be used to obtain approximate values for the critical load. For the Bothnian Bay, a critical load for direct atmospheric deposition of N is approximately 0.9-4.1 kg ha^{-1} yr^{-1} sea surface, for the Bothnian Sea, 6.9-8.5 kg N ha^{-1} yr^{-1} in a calculated example. The range depends on how much N input is permitted from land. The critical nutrient nitrogen loads regardless of source, including what can be received from land or atmosphere, are 7.2 kg ha^{-1} yr^{-1} for the Bothnian Bay and 11 kg ha^{-1} yr^{-1} for the Bothnian Sea.

Introduction

Marine areas are sensitive to excess loads of nutrients. Most marine environments are N limited, but several coastal areas and estuarine environments are P limited. If both P and N loads are high, very drastic ecological changes may occur. Critical loads for N and P to marine areas are wanted for the 1996 N protocol, since N deposition to sea is considered significant. In the Baltic, 40% of the N load is estimated to arrive by air. A significant part of the N-load from land to the Baltic Sea is derived from deposition of air pollutants on land (0-25%).

Many aquatic ecosystems have a very delicate balance between many different trophic levels in the system. Changes in nutrient load or changes in nutrient ratios will be able to push the systems beyond ecosystems stability thresholds, causing establishments of relatively stable alternative states. Some of these alternative states may be highly undesireable in terms of structure, size or economic potential. The induced ecosystem changes may give rise to internal nutrient cycles, able to sustain the alternative states for considerable periods of time.

Over the time period 1968 to 1992, the phosphate concentration in the deeper waters of the Bothnian Sea rose from an average of 0.015 mg P l^{-1} to approximately 0.025 mg P l^{-1}. At the same time it increased from 0.001 mg P l^{-1} to 0.003 mg P l^{-1} in the Bothnian Bay. A large increase occurred for N concentrations after 1972 increasing in the Bothnian Sea from 0.03 mg N l^{-1} to 0.12 mg N l^{-1}, and in the Bothnian Bay from 0.05 mg N l^{-1} to 0.1 mg N l^{-1}. This implies that the N/P ratio rose from 2 to 3.5 in the Bothnian Sea, and from 50 to 33 in the Bothnian Bay. (Data were taken from Wulff *et al.* (1994) and Grimevall *et al.* (1994)). These changes have been linked with ecological changes in this area.

Definition

The critical load of nutrient nitrogen was defined by the International workshop on critical loads held at Skokloster, Sweden, in 1988. For marine ecosystems the definition has been modified to:

The maximum load of nitrogen and phosphorus that will not cause long term damage to ecosystem structure and function.

Phosphorus is included in the definition since it is intimately connected to how the systems are able to react to nitrogen input. For each ecosystem assessed, an indicator organism is chosen. A chemical limit is found for that indicator organism, and the chemical limit is entered into a chemical mass balance equation including all sinks and sources of acidity in the system. The chemical limit is applied to the solution concentration in the system. Thus ecosystems become connected through concentrations via mass balances to acid deposition.

Receptors

Ecosystems in estuarine areas and large constricted ocean areas: Gulf of Bothnia, Finnish Bay, Baltic Sea, Kattegatt and Aresund, Riga Bay, St. Petersburg Bay, large Norwegian fjords, Mediterranian Sea, White Sea, Black Sea, Caspian Sea, Adriatic Sea, Aegean Sea. Small estuaries and fjords along the seas just listed.
Ecosystems in marine areas open to larger oceans; North Sea, North Sea coast of Netherlands and Germany, Gulf of Biscay, Irish Sea.

Indicator species will be different types of phytoplankton and zooplankton. Indicator species of vertebrates may be stationary fish, or the spawning process of migratory fish.

Basic Philosophy

In the effort to calculate critical loads, the basic philosophy is to use the best available estimate. In the current absence of values and maps, even crude estimates are welcome. Thus we divide up the critical load estimation approach into several steps:

Empirical estimation

Simple mass balance model

Integrated ocean circulation and biodynamic model

Here we will suggest a simple empirical method to begin estimating the order of magnitude for the critical load. Other approaches must start in parallel in order to secure the credibility of the simpler estimates as well as be able to later produce more detailed and more accurate estimates.

The Empirical Model

The empirical marine model (TEMA) is suggested as the first step in establishing critical loads for marine areas. It is suggested that data available on N concentrations from coastal waters are used together with information of ecological change to generate empirical relationships between ecological change and N concentration.

In many estuarine waters, the N concentrations change drastically as one moves from land out to sea. This gradient is often caused by large N loads to the waterbody from land. Going out along the gradient, a change in the trophic status may be observed. Such sites should be used to generate empirical relationships.

Thus we divide up the procedure into two steps:

From ecological change to N concentration

From N concentration to N load

The relationships are stratified with respect to such factors as salinity, mixing conditions or N:P ratio, by limiting the correlations to large but limited marine areas.

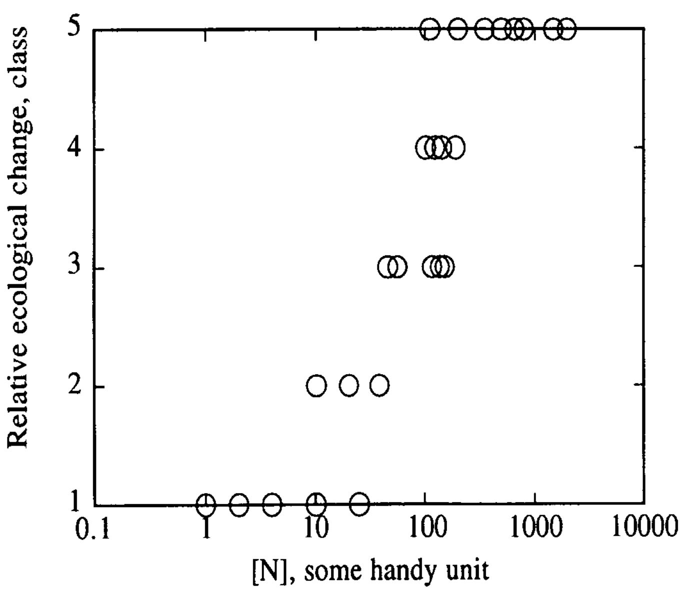

Figure 1 **Example of empirical relationship between relative ecological change and observed N concentration that can be derived from surveys in estuaries and confined marine areas. The data are hypothetical.**

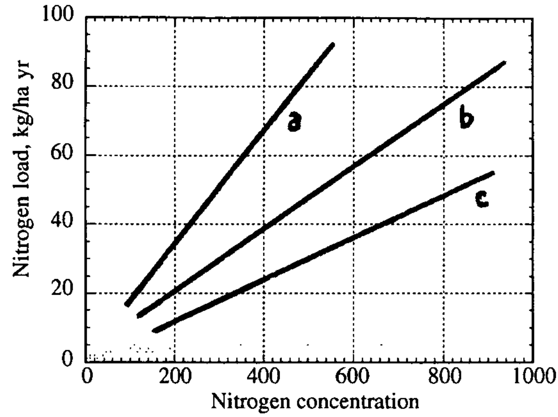

Figure 2 **Models, but also empirical relationships, can be used to draw simple load-concentration diagrams for different waters. The presented diagram is hypothetical.**

Models and empirical relationships are used to draw simple load-concentration diagrams for different waters. In confined marine areas, simple mass balance approaches can be used, but in more open marine systems such as the North Sea or open sea coastal waters, the construction of load-concentration nomograms using more sophisticated hydrological models, may be important. Data from several areas must be used to generate diagrams, since important parameters are omitted, such as water exchange and salinity. It is suggested that attempts are made to derive such diagrams for areas such as (1) the Northern Bothnian Gulf, (2) the Southern Bothnian Gulf down to Åland (3) Bay of Finland, (4) Southern Baltic Sea, (5) Kattegatt and Öresund, (6) Riga Bay, (7) Skagerak and (8) North Sea.

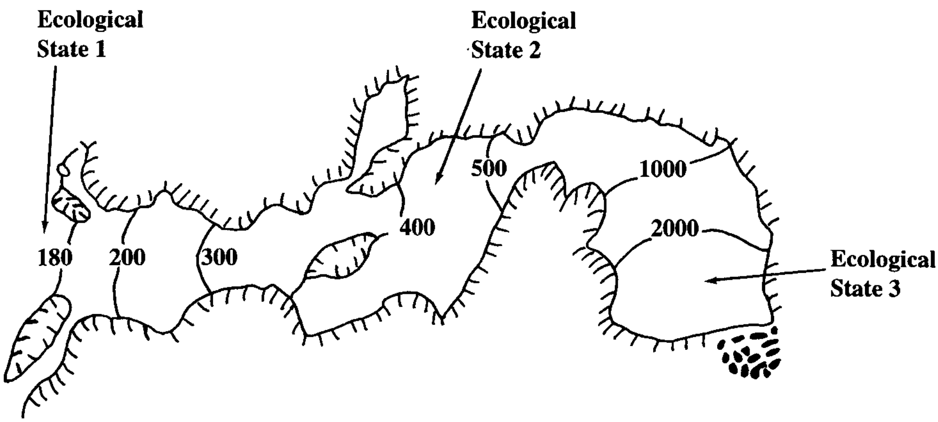

Figure 3 **Observed N concentration gradients in estuaries may be used to estimate eco logical change. Along the concentration gradient different ecological states can be observed. These can subsequently be plotted against concentration, to obtain a concentration-effect diagram.**

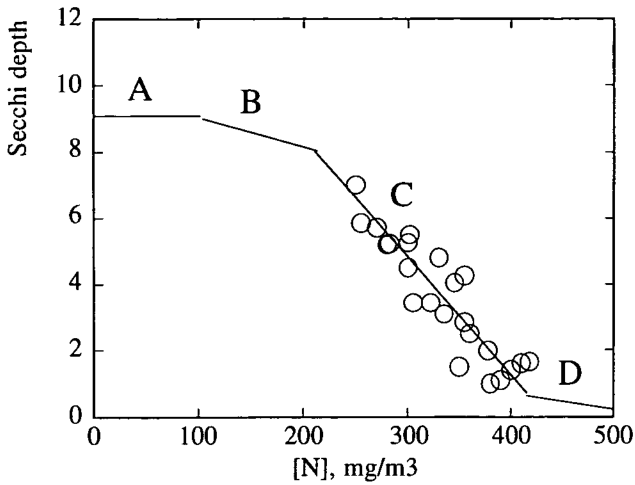

Figure 4 **Relationship between total N concentration and secchi depth using an example from Wallin (1993). Data from 22 stations in the Baltic Sea. Ecological change is quantified using secchi depth, analog with eutrophication of freshwater lakes. Secchi depth is sometimes used as an indicator of environmental quality.**

{Example area; The Bothnian Sea and Bay between Sweden and Finland.}

In the case of the Mediterranian, the Black Sea, the White Sea and the Caspian Sea, the methodology would be to subdivide them into different basins in much the same way as we have attempted for the Baltic Sea.

The Bothnian Sea and Bay

In his 1991 thesis from the Ecology Institute of Stockholm University, Wallin studied the sensitivity of the Baltic Sea using data from 23 stations. The stations were spread around the Baltic from Helsinki to Southern Sweden. He was not using local gradients, but rather differences within the Baltic. Wallin states that the relationship between load and ecological change is mostly correlated with the following factors:

Chloropyll-a concentration

Secchi depth

Nutrient flux between bottom sediments and the water body. Nutrient refluxes are very important

Oxygen saturation

These factors are related to state variables or properties of the sea water such as those affecting the relationship between N-concentration and ecological change:

N:P ratio

Salinity

Oxygen saturation

Iron concentration

Whereas other parameters affect the relationship between external N load and N concentration such as large scale water mixing conditions, salinity and nutrient reflux from bottom sediments. In the work of Wallin (1991), examples of relationships between chlorophyll-a and secchi depth are shown. He also relates secchi depth to N-concentration.

Wulff from the Ecology Institute at Stockholm University and colleagues (Wulff *et al.*, 1994) have suggested the use of simple mass balance calculations for the Baltic Sea, resembling the Simple Mass Balance method used for terrestrial systems (Sverdrup *et al.*,1990; Hettelingh, *et al.*, 1992).

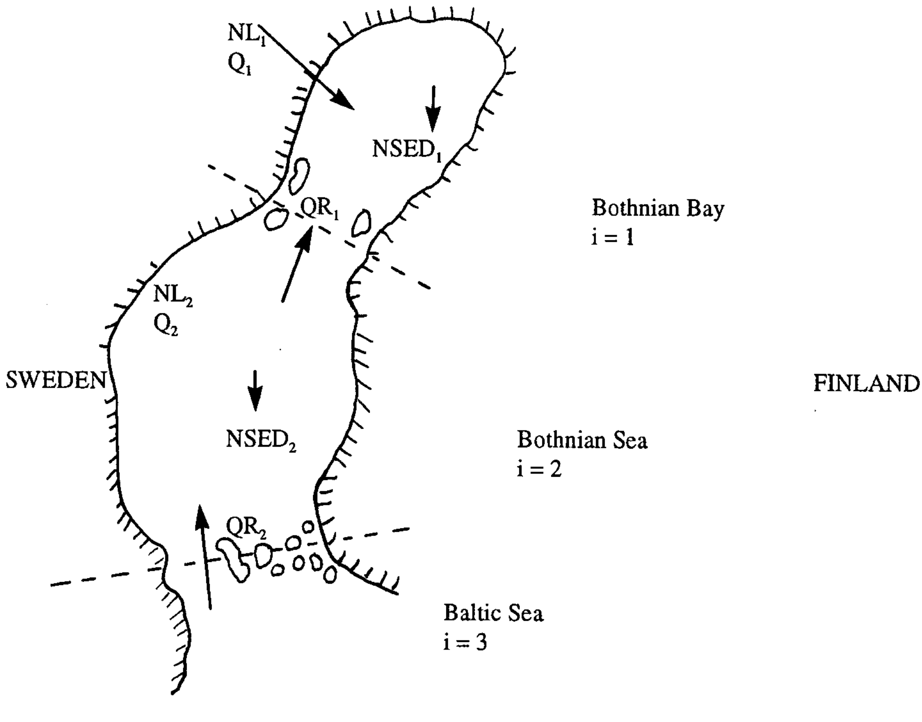

Figure 5 **Example area; The Bothnian Sea and Bay between Sweden and Finland.**

For the two areas shown in Figure 5. the following mass balances may be made for N:

$$Q_{R1}(N)_2 + NL_1 + NDEP_1 = NSED_1 + (Q_1 + Q_{R1}).(N)_1$$

$$Q_{R2}(N)_3 + (Q_1 + Q_{R1}).(N)_1 + NL_2 + NDEP_2 = NSED_2 + (Q_1 + Q_2 + Q_{R1}).(N)_2$$

where:

Q_i	Flow rate into basin *i*	$m^3\ yr^{-1}$
Q_{Ri}	Flow rate from basin *i*+1 to basin *i*	$m^3\ yr^{-1}$
$(N)_i$	N concentration in basin *i*	kmol N m^3
NL_i	Stream flux of N to basin *i*	kmol yr^{-1}
$NSED_i$	Sedimentation in basin *i*	kmol yr^{-1}
$NDEP_i$	Deposition flux of N to basin *i*	kmol yr^{-1}

The critical load for the inner basin (Bothian Bay) is then given by:

$$CLN_1 = NSED_1 - NL_2 + (Q_1 + Q_2).(N)_{\{crit,1\}} - Q_{R1}.(N)_{\{crit,2\}}$$

The outer basin (Bothian Sea) have a critical load equal to:

$$CLN_2 = NSED_2 - NL_2 + (Q_1 + Q_2 + Q_{R2}).(N)_{crit,2} - (Q_1 + Q_{R1}).(N)_{crit,1} - Q_{R1}.(N)_{crit,3}$$

where $(N)_{crit,i}$ is the critical N concentration in basin i as determined by the local empirical effect relationships. The critical concentration can be derived using effects and N concentrations direct or by plotting effect versus N concentration, stratified with respect to P concentration and N:P ratio.

From the time series of concentrations of N and P in the Baltic presented by other researchers (Grimevall *et al.*, 1994) a limit $(N)_{crit,i} = 0.03$ mg N l^{-1} can be suggested for both the Bothnian Sea ($i = 2$) and the Bothnian Bay ($i = 1$). We assumed that the concentration level observed in 1968, was equivalent to a level causing no adverse ecological effects due to excess N.

Using Fig. 3 would imply that we require a secchi depth of approximately 4 meter. Alternatively, we may assume that the N concentration was already elevated in 1968 and set a lower value, based on Fig.4, maybe at 0.02 mg N l^{-1}.

At the same time the critical limit for phosphorus $(P)_{crit,2} = 0.01$ mg P l^{-1}, and $(P)_{crit,1} = 0.002$ mg P l^{-1}. The critical load for phosphorus can be estimated applying the same logic as described here for N.

Wulff and colleagues have observed actual sedimentation rates which could be used. For the Bothnian Bay the sedimentation rate is suggested to be approximately 23,000 tons N annually. For 1991 they observed a sedimentation of 230,000 tons of N in the Bothnian Sea. These rates are probably dependent on the polluted state of the water, and too high for a situation relevant to the critical load. Increased nitrogen causes more primary plankton production, producing more material that can settle to the bottom, inceasing the sedimentation rate.

One first approximation could be to relate sedimentation rate to the N concentration. This would lead to an estimate of a net sedimentation rate at the critical concentration of 0.03 mg N l^{-1}, equal to:

$$\frac{230000 \text{ tons yr}^{-1} \times 0.03 \text{ mg N l}^{-1}}{0.1 \text{ mg l}^{-1}} = 69000 \text{ tons yr}^{-1}$$

The sedimentation rate for the Bothnian Bay is estimated assuming that a sedimentation rate as observed in 1968 would be applicable:

$$\frac{23000 \text{ tons yr}^{-1} \times 0.06 \text{ mg N l}^{-1}}{0.1 \text{ mg N l}^{-1}} = 14000 \text{ tons yr}^{-1}$$

Table 1: *Fluxes of nitrogen to the Bothnian Sea and Bay from land at present (1991).*

Source	Bothnian Sea	Bothnian Bay
Forest and farming	45,100 ton N yr^{-1}	44,200 ton N yr^{-1}
Point sources	7,100 ton N yr^{-1}	3,200 ton N yr^{-1}
Atmospheric from land	45,000 ton N yr^{-1}	14,000 ton N yr^{-1}
Sum	97,200 ton N yr^{-1}	61,400 ton N yr^{-1}
Atmospheric on sea	15,000 ton N yr^{-1}	3,000 ton N yr^{-1}
Sum	112,200 ton N yr^{-1}	64,400 ton N yr^{-1}

Table 2: *Fluxes of nitrogen to the Bothnian Sea and Bay from land used in the critical load calculation examples. The land load is dominated by effluents from forests and farm land, the effluents from farmland will be dominating.*

Source	Bothnian Sea	Bothnian Bay
Forest and farming	23,600 ton N yr^{-1}	22,100 ton N yr^{-1}
Point sources	3,500 ton N yr^{-1}	1,600 ton N yr^{-1}
Atmospheric from land	0 ton N yr^{-1}	0 ton N yr^{-1}
Sum	27,100 ton N yr^{-1}	23,700 ton N yr^{-1}

The land load (Table 1) today to the Bothnian Bay is 61,400 ton N yr^{-1}, where 15,000 ton N yr^{-1} is of atmospheric origin. The land load to the Bothnian Sea is 97,000 ton N yr^{-1}, where 45,000 ton N yr^{-1} is of atmospheric origin. 15,000 ton N yr^{-1} is assumed to fall directly into the water in the Bothnian Sea (20.8 kg N ha^{-1} yr^{-1}), in the Bothnian Bay only 3,000 ton N yr^{-1} is assumed to fall (8.5 kg N ha^{-1} yr^{-1}).

The net flux of N out of the Bothnian Sea is today 12,000 ton N yr^{-1}, the net N flux from the Bothnian Bay to the Bothnian Sea is 24,000 ton N yr^{-1}. Assuming that the critical load for N over land to take away the atmospheric element in river transport of N, would yield a net land load of 47,000 ton N yr^{-1} to the Bothnian Bay and 52,000 ton N yr^{-1} to the Bothnian Sea.

Fluxes of nitrogen i.e. to the Bothnian Sea and Bay from land used in the critical load calculations are shown in Table 2. The land load is dominated by effluents from forests and farmland, the effluents from farmland will be dominating.

Table 3: *Input data values used to calculate critical loads of nitrogen to the Bothnian Sea and Bay.*

Q_1	Flow rate into basin 1	360 $km^3\ yr^{-1}$
Q_2	Flow rate into basin 2	340 $km^3\ yr^{-1}$
Q_{R1}	Flow rate from basin 2 to basin 1	80 $km^3\ yr^{-1}$
Q_{R2}	Flow rate from basin 3 to basin 2	140 $km^3\ yr^{-1}$
$Q_1 + Q_2 + Q_{R2}$	Flow rate from basin 2 to basin 3	360+340+140 $km^3\ yr^{-1}$
$Q_1 + Q_{R1}$	Flow rate from basin 1 to basin 2	360+80 $km^3\ yr^{-1}$
$(N)_1$	N concentration in basin 1	0.03 g N m^{-3}
$(N)_2$	N concentration in basin 2	0.03 g N m^{-3}
$(N)_3$	N concentration in basin 3	0.03 g N m^{-3}
$NSED_1$	Sedimentation in basin 1 at critical load	14,000 ton N yr^{-1}
$NSED_2$	Sedimentation in basin 2, at critical load	69,000 ton N yr^{-1}
NL_1	Land load in basin 1 at CL	23,700 ton N yr^{-1}
NL_2	Land load in basin 2 at CL	27,100 ton N yr^{-1}

The flow rate from land to sea to the Bothnian Bay is estimated to 360 $km^3\ yr^{-1}$ giving an apparent residence time of 4 years, for the Bothnian Sea to 700 $km^3\ yr^{-1}$ giving an apparent residence time of 6 years. If refluxes are considered, the effective residence times will be shorter (Approximately 3 years and 5 years). The reflux is estimated to 80 $km^3\ yr^{-1}$ from the Bothnian Sea to the Bothnian Bay, and as 140 $km^3\ yr^{-1}$ from the Baltic to the Bothnian Sea. The data used in the calculation is shown in Table 3.

Critical Load for the Bothnian Sea and Bay

Using the expressions given above we calculate in our example, assuming the atmospheric part of the load from land to be zero, the effluent on land derived from agriculture and should be reduced to 50% the point sources reduced by 90%.

Keeping the limit in mg l^{-1} and the flow rate in $km^3 yr^{-1}$, yield fluxes in thousand tons yr^{-1}. Filling in the equations for basin 1, the Bothnian Bay, gives:

$$CL1 = 14 - 23.7 + (360 + 80)*0.03 - 80*0.03 = 3.1$$

The critical nutrient load of 3,100 ton N yr^{-1}, is equivalent to a critical load of 0.9 kg N $ha^{-1}\ yr^{-1}$ Bothnian Bay surface area. Reducing the N load from farms by 75% instead of 50%, will increase the critical load to 14,150 ton N yr^{-1}, equivalent to 4.1 kg N $ha^{-1}\ yr^{-1}$ for the Bothnian Sea surface area. The total critical load including any load from land or air is 25,200 ton N yr^{-1} equivalent to a critical load of 7.2 kg N $ha^{-1}\ yr^{-1}$ of the Bothnian Bay surface area.

The exceedance is at present 7.5-0.9 kg N ha^{-1} yr^{-1}, depending on the assumptions made concerning the land load. This implies that the atmospheric load must be reduced by 89%, 51% or 14%. The present land load of 61,400 ton N yr^{-1}, as well as the non-atmospheric part of the land load, 47,400 ton N yr^{-1}, are alone sufficient to exceed the critical load.

For the Bothnian Sea, we get the critical load:

$$CL2 = 69 - 27.1 + (360 + 340 + 140)*0.03 - (360 + 80)*0.03 -140*0.03 = 49.7$$

The critical nutrient load of 49,700 ton N yr^{-1}, corresponds to a critical nutrient load of 6.9 kg N ha^{-1} yr^{-1} Bothnian Sea surface area. Reducing the N load from land sources, and farms in particular by 75% instead of 50%, will increase the critical load to 61,250 ton N yr^{-1}, equivalent to 8.5 kg N ha^{-1} yr^{-1} for the Bothnian Sea surface area.

The total critical load including any load from land or air is 79,800 ton N yr^{-1} equivalent to a critical load of 11 kg N ha^{-1} yr^{-1} Bothnian Bay surface area. The present land load of 97,200 ton N yr^{-1}, is alone sufficient to exceed the critical load, the non-atmospheric part of 52,000 ton N yr^{-1} is not.

The atmospheric load is 20 kg N ha^{-1} yr^{-1} in the area, the exceedance is at present 13.5-9 kg N ha^{-1} yr^{-1}. This implies a required deposition reduction in the range of 65% to 45%.

We are aware of the fact that N is present in organic and inorganic form, and the two forms may have a different ecological effect. Still, we suggest as a first approximation that they are lumped into total N. Further research in parallel with the critical load calculations can develop concepts for critical loads which consider the different forms. We are also aware of the fact that we have smoothed local gradients in the mass balance approach, still we believe the assumption of deriving an approximate value this way is valid.

The shown example may seem very much simplified to some, and that is the intention. It is meant as a way of making approximate estimates of the critical load, until more accurate models and calculations can be made. All the data used in the calculation were approximate, and may be subject to future change. More elaborate calculations will require more time, and in the mean time approximate values are needed in the international negotiations. The international critical loads mapping programme has been specially designed to accomodate gradual updates of the critical loads values reported into the European Coordination Centre. This design allows for the use of approximate numbers which are repeatedly replaced by better numbers as better tools become available.

References

Hettelingh, Posch, de Vries, Bull and Sverdup (1992) Guidelines for the computation and mapping of nitrogen critical loads and exceedance in Europe. (Eds. Grennfelt and Thörnelöf) Nordic Council of Ministers, 287-303, Nord 1992: 41.

Grimevall, Johnsson, Cederlöf and Hilder (1994) Föroneningshoten mot Bottniska Viken. Vatten, **50**, 231-246.

Sverdup, H., de Vries and Henriksen (1990) Mapping critical loads, Nordic Council of Ministers, Miljorapport 1990: 15.

Wallin , M. (1991) Nutrient loading models for coastal waters. Ph.D Thesis, University of Upsala, Department of Geography. ISBN No. 91/506/0859 9 2

Wulff, Pertillä and Rahm (1994) Omsättning as närsalter i Bottniska Viken. Vatten, **50**, 220-230.

CHAPTER 8
CRITICAL LOADS OF NITROGEN: MAPPING OF ATMOSPHERIC INPUTS

Discussion Group Report

Introduction

The discussion group was convened during the workshop to provide a short review of the current status of the chapter on Mapping Deposition Loads and Concentrations in the Mapping Manual (TFM, 1993). In particular, the aim was to review current approaches for estimating inputs and compare them with the procedures advocated by the mapping manual.

The Mapping Manual description of atmospheric deposition loads has been greatly improved following the workshops at Lökeborg (Grennfelt and Thörnelöf, 1992) and Göteborg (Lövblad, Erisman and Fowler, 1993), but several important areas of the chapter require further modification to clarify the procedures. In particular, it is important to provide clearer, unambiguous guidance to the appropriate methods and sources of data to define inputs for the production of exceedance maps. The mapping of concentrations is relevant for estimating both atmospheric deposition and critical levels exceedance. Critical levels are not discussed further here, and the scientific basis for these has been published from UNECE workshops at Bad Harzburg (1988), Egham (Ashmore and Wilson, 1994) and Berne (Fuhrer and Achermann, 1994).

The following brief report of the Discussion Group provides recommendations of those areas in the current mapping manual chapter on deposition where future re-drafting is needed to provide clarification of methods and updating to meet new critical loads data requirements.

Recommendations

The following section identifies the aspects of the current mapping manual that require modifications. References refer to section numbers in the mapping manual.

Groups of chemical species are referenced according to the following definitions: SO_x, oxidised sulphur, includes: sulphur dioxide (SO_2) and sulphate (SO_4^{2-}); NO_y, oxidised nitrogen, includes: nitrogen dioxide (NO_2) and nitric oxide (NO) (collectively NO_x), nitric acid (HNO_3), nitrous acid (HNO_2) and nitrate (NO_3^-). NH_x, reduced nitrogen, includes: ammonia (NH_3) and ammonium (NH_4^+). Atmospheric inputs of the ionic species may occur via wet deposition, cloud water deposition or dry deposition of aerosols, while the other species are gases, with inputs occuring by dry deposition.

Mapped Items (section 2.2)

- Total AND Non-marine maps of SO_x deposition are required.

- Total AND Non-marine maps of base cation deposition are required.

These terms are necessary for the revised mass balance critical loads formulations, such as for peats, where total atmospheric inputs are used (see Working Group Report: Mass Balance Methods).

Methods of Mapping, their underlying assumptions and data requirements

General (section 2.3.1)

- The priority in methods to estimate atmospheric inputs needs to be clearly stated.

  For example, EMEP estimates are most suitable for country to country budgets ('Blame Matrices'), but, where air concentration monitoring data are available, inferential resistance modelling is the most appropriate method for modelling fine scale deposition estimates for comparison with critical loads and to quantify exceedance.

- The effect of spatial scale on critical load exceedance should be stated.

  The spatial scale of deposition and critical loads estimates will affect the resulting critical loads exceedance estimates. A common scale and approach should be used when comparing deposition and exceedances for individual countries.

- Land-use-specific atmospheric inputs should be provided

  Ecosystems of different species, management and roughness receive pollutants at different rates, it is necessary therefore to define atmospheric deposition in relation to land-use type. So called 'Filter factors' attempt to simulate the effect of atmospheric roughness but lack a rigorous basis and should be removed from the Manual Procedures. Other approaches which are well supported by the scientific literature and are discussed in the Goteborg report, provide a satisfactory alternative.

- Inferential methods should be applied for particle and fog deposition.

  Inputs of particles may be estimated using inferential resistance models and estimates of air concentrations of the particles. In the case of cloud water deposition, cloud immersion frequency is also necessary.

- Throughfall methods are valuable for sulpur inputs to forests, but are generally unsuitable for nitrogen deposition and for non-forest surfaces.

For deposition of nitrogen species (NO_y, NH_x) canopy interactions greatly complicate the quan tification of atmospheric inputs from throughfall methods. This is especially true at low deposition levels. The relationship between total nitrogen deposition and the sum of NO_3^- and NH_4^+ in throughfall is not expected to be linear.

Mapping Sulphur (SO_x) and Oxidised Nitrogen (NO_y) Deposition Loads (Sections 2.3.2, 2.3.3)

- More detail of the methods and guidance to those wishing to apply them should be included in the manual including references.

Mapping Reduced Nitrogen (NH_x) Deposition Loads (Section 2.3.4)

- Level II approaches are advocated which are known to be wrong, especially close to sources.

- Inputs of reduced nitrogen are land-use specific, and procedures which include these effects must be used.

  In addition to the variability in rates of deposition to different ecosystems exhibited for SO_x and NO_y, in exchange of NH_3, there may be both deposition and emission with different ecosys tems. This effect makes it essential that deposition estimates are defined for specific land-use types.

Mapping Base Cation Deposition Loads (Section 2.3.5)

- Revision is required to the text to clarify what can be done and how uncertain each of the methods are.

  This approach is also subject to large uncertainties and requires validation measurements. The manual needs a description of the approaches and further research is necessary to develop a new rigorous approach. The dry deposit ion of base cations is not monitored and yet is of considerable importance for exceedance estimates based on mass balance approaches. The best current approach estimates the air concentrations of base cations from rain chemistry measure ments and scavenging ratios and applies deposition velocities based on the scheme of Slinn (1983).

Mapping Ammonia Concentration Levels (Section 2.3.11)

More helpful text and references are needed to replace 'no methods advised'

General

A detailed description of the methods and their limitations may be found in the report of the Göteborg workshop held in 1992 (Lövblad *et al.* 1993). Developments in the requirements for critical loads data, and in the methods for assessing atmospheric deposition would justify a future meeting to revise this section.

References

Ashmore M.R. and Wilson R.B. (Eds.) (1994) *Critical levels of air pollutants in Europe.* Proceedings of the UNECE workshop, Egham 23-26 March 1992). Air Quality Division, Department of the Environment, London SW1P 3PY, UK.

Fuhrer J. and Achermann B. (Eds.) (1994) *Critical levels for ozone: a UNECE workshop report.* (Proceedings of the Bern workshop 1-4 November 1993). Federal Research Station for Agricultural Chemistry and Environmental Hygiene (FAC), CH-3097 Liebefeld-Bern, Switzerland. (ISSN -1013-154X).

Grennfelt P. and Thörnelöf E. (Eds.) (1992) *Critical loads for nitrogen: a workshop report.* (Proceedings of the Nordic Council / UNECE workshop, Lökeberg, 6-10 April 1992). Nord 1992:41. Nordic Council of Ministers, DK-1255, Copenhagen K. (ISBN 92 9120 121 9).

Lövblad G., Erisman J.W. and D. Fowler (Eds.) (1993) *Models and methods for the quantification of atmospheric input to ecosystems.* (Proceedings of the Nordic Council / EUROTRAC-BIATEX / UNECE EMEP workshop, Göteborg, 3-6 November 1992). Nord 1993:573. Nordic Council of Ministers, DK-1255 Copenhagen K. (ISBN 92 9120 253 3).

Slinn W.G.N. (1983) Predictions for particle deposition to vegetative surfaces. *Atmospheric Environment.* 16, 1785-1794.

TFM (1993) Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded. Texte 25/93. TFM / CCE / UNECE. Task Force on Mapping, Uweltbundesampt (UBA), Bismarckplatz 1, W-1000 Berlin 33. Germany.

Keynote Paper - Atmospheric Inputs of Nitrogen

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Introduction

The requirement for estimates of the terrestrial inputs of fixed nitrogen for Europe with a spatial resolution of between 5 km x 5 km and 50 km x 50 km places great demands on the monitoring information available and our knowledge of processes. It also provides a clear focus for the monitoring and process based research to provide both the input and an estimate of the uncertainty in inputs and its spatial variability.

For the estimates of critical loads exceedance maps for nitrogen, maps of the total inputs are of course essential. In this brief review the focus is on what is known, and can be mapped, what is not known because insufficient monitoring is in place, and lastly what is not known because the processes are not understood or for which key parameters are uncertain. In addition, because the principal applications of mapped data are to estimate critical loads exceedance, the issue of the scale dependence of deposition estimates and the extent to which current approaches can be extended to provide fine scale or land use specific resolution is considered qualitatively and to a limited extent quantitatively.

Wet Deposition

The monitoring networks for wet deposition provide good estimates of the precipitation weighted concentrations of major ions in simple terrain throughout Europe. There are areas of uncertainty, mainly as a consequence of complex terrain such as in the Alps and in the mountains of north and west Britain and Scandinavia. For relatively simple terrain the wet deposition estimates based on the product of a very spatially detailed precipitation field and the much less variable spatial pattern in weighted mean concentration are entirely adequate for the mapping procedures at 10 km x 10 km scale and upwards. The problems in wet deposition are due primarily to complex terrain. In such areas orographic effects modify the precipitation scavenging process by, for example, " seeder-feeder" effects and these lead to marked increases in wet deposition in the uplands of the UK and western Scandinavia. In the seeder-feeder effect, relatively clean precipitation from higher level cloud "washes" out more polluted cloudwater from lower level orographic (feeder) cloud. The mechanisms of orographic enhancement have been studied in detail and simple parameterization schemes have been developed to modify wet deposition maps for this effect over the UK. While intensive field studies have been made at a range of mountain locations to examine the processes and determine the variability in concentrations of major ions in precipitation, orographic cloud and wet deposition with altitude, these have generally been 2 dimensional transects through "simplified complex terrain". For example, trends in concentration and deposition have been observed along a transect over a simple ridge at Great Dun Fell in the Pennines of northern England. Larger mountains in more complex environments have been studied, as for example, on the western slopes of Ben More Assynt in Sutherland, north west Scotland. In all of the campaign studies, the effect of seeder-feeder enhancement in precipitation and concentration have been observed. The wide range of study sites for the field studies is illustrated in Figure 1 which shows the sites at which the measurements have been made. The individual profiles of concentration and deposition lie outside the scope of this brief review but have been simulated using models of the processes (Choularton *et al.* 1988). However a summary of the results of the campaigns (each of typically 4 to 6 weeks sampling of each precipitation event) and of the longer term monitoring of hill cloud and precipitation are given in Table 1.

The overall mean enhancement in concentration of scavenged feeder cloud/seeder rain of 2.3 for SO_4^{2-} from the table provides support for the simplifications made to model orographic enhancement of wet deposition at the UK scale. Such approaches are necessarily crude and do not allow for fine scale (<20 km) effects of orography on deposition patterns. The wet deposition maps provided with orographic enhancement are therefore restricted in spatial detail to a 20 km x 20 km grid and cannot be used to obtain the fine scale wet deposition inputs for exceedance estimates. (The specific effects of spatial scale averaging on exceedance estimates are considered later.)

It is a matter of importance and not a mere coincidence that the geographical areas in which complex terrain complicates the scavenging processes, and introduce extra uncertainty into input estimates, are also the areas of greatest critical levels exceedance for wet deposition.

Cloud Droplet Deposition

The importance of these processes for inputs to high altitude forests were little known or understood in the early years of acidic deposition research. The interest in processes at high elevation sites led to research on the processes of turbulent deposition of droplets onto natural surfaces (Dollard *et al.* 1983; Fowler *et al.* 1989; Gallagher *et al.* 1988). These studies demonstrated the dependence of deposition rates on droplet size and are illustrated in Figure 2 from the work of Gallagher *et al.* (1991) which showed that for the bulk of the liquid water in orographic cloud on windy uplands of Northern Europe the droplets are captured by the vegetation at rates similar to those of momentum, and can therefore be modelled from the relatively well known (and monitored) data on wind and roughness length.

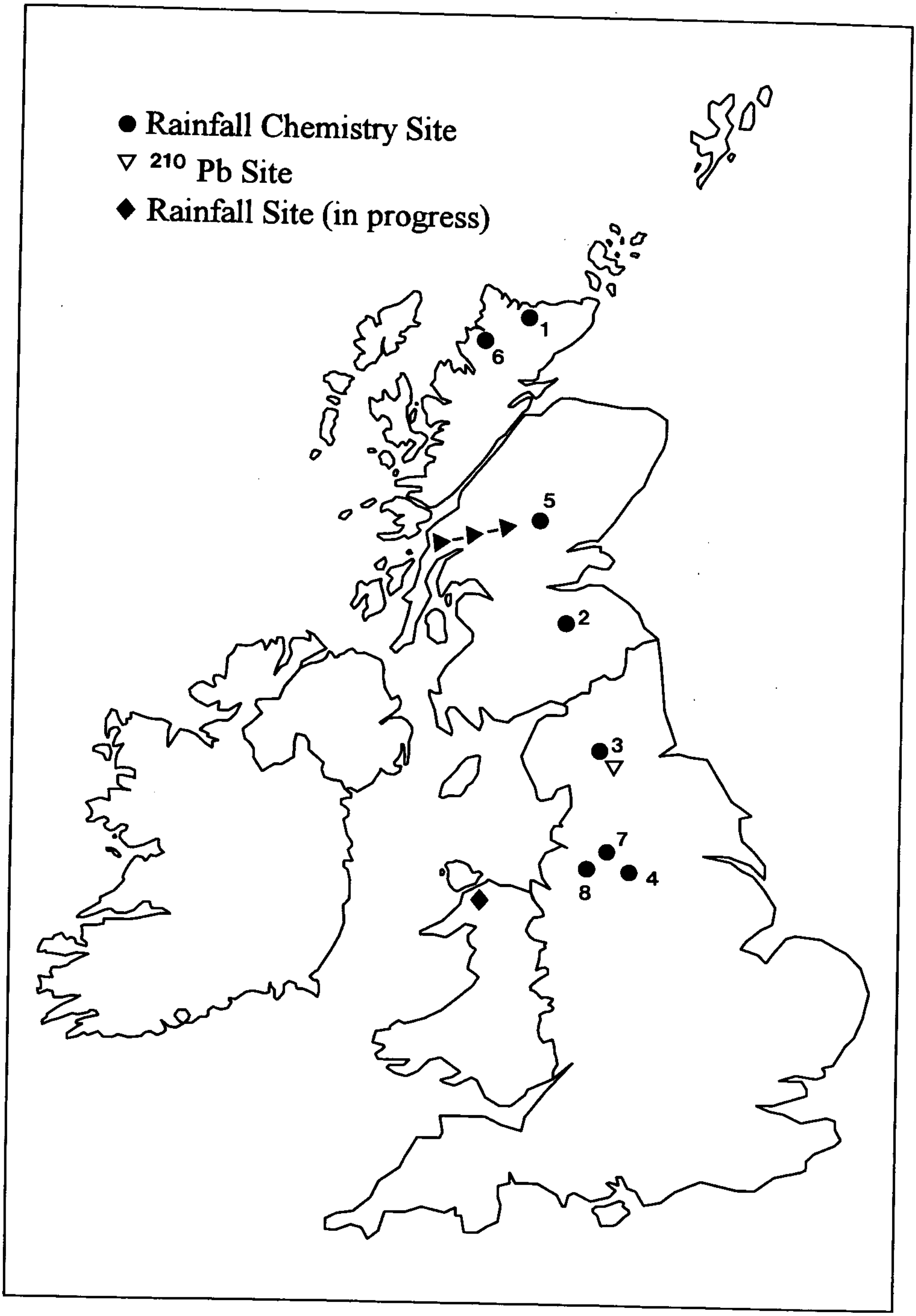

Figure 1 **Sites of orographic enhancement of wet deposition in the UK**

Table 1 Summary of results of long-term monitoring measurements and campaign studies

		Concentration ratio hill cloud/seeder rain			
Long-term measurements	Site No.	SO_4^{2-}	NO_3^-	NH_4^+	Cl^{-1}
Halladale	1	8	12.8	1.1	14.4
Dunslair	2	7.7	7.3	7	7
Great Dun Fell	3	4.51	9.72	4.05	5.68
Holme Moss	4	4.55	6.6	4.21	10.4
Short-term (campaign) measurements					
Glen Bruar	5	2.9	1.89	2.34	3.95
Assynt	6	3.56	1.53	-	3.35

		Concentration ratio scavenged feeder cloud/seeder rain			
Long-term measurements	Site No.				
Halladale	1	2.5	3.1	1.1	4.1
Great Dun Fell	3	1.46	2.8	4.6	1.58
Holme Moss	4	2.48	2.05	3.04	1.96
Short-term (campaign) measurements					
Great Dun Fell	3	3.3	3.5	4.7	4.4
Holme Moss (Saddleworth Moor)	7	1.98	2.33	2.15	1.97
Winter Hill	8	1.88	2.74	2.2	5.13
Very long-term measurements ^{210}Pb at Great Dun Fell 2.2					

To estimate the actual deposition rates of nitrogen in cloud droplets in the uplands from the above work, it is necessary to quantify the spatial variability in cloud frequency and the concentration of the NO_3^- and NH_4^+ in cloud. These quantities are not widely monitored in Europe, although there are 4 sites in Britain (Dunslair Heights, Great Dun Fell, Holm Moss and Halladale) for which more than 1 year of continuous monitoring of cloud and precipitation composition and frequency have been made. Likewise, there is a similar number of sites in Germany at moderate elevation for which similar data are available. These data show a consistent height dependence of the concentrations of major ions as illustrated in Figure 3.

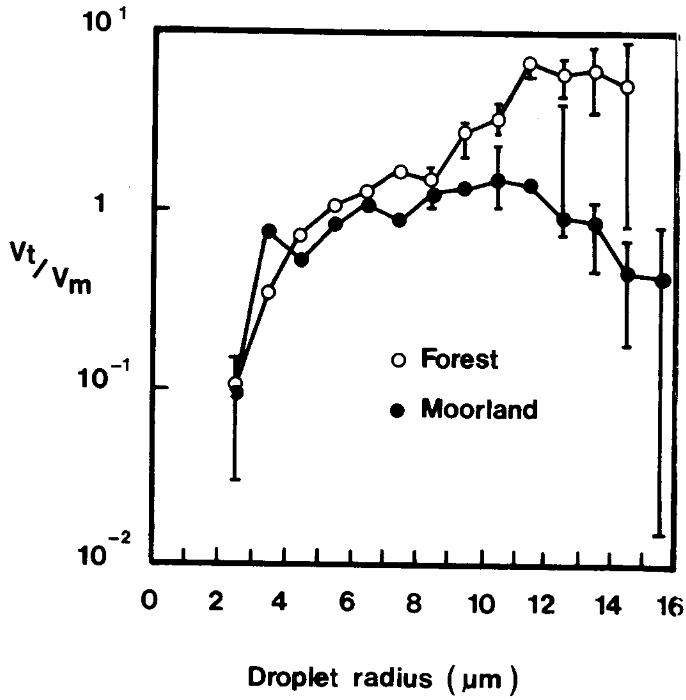

Figure 2 **Effects of droplet size in the range 2-15 m on the ratio of turbulent deposition velocity (V_t) of cloud droplets to that of momentum (V_m) (where $V_m = r_{am}^{-1}$ for forest and moorland from measurements by Gallagher *et al.* 1991).**

Extending the existing measurements to regional scales presents a problem, and to provide a consistent mapping approach for the UK, the relationship between concentrations of NO_3^- and NH_4^+ in hill cloud and those in precipitation have been used together with a map of cloud frequency (Weston, 1991) and wind velocity and land use data from national UK data bases. Estimates of the regional importance of cloud droplet deposition have not been made for other European countries.

A missing component of this area of work has been that of radiation fogs, which also contribute to the deposition budget. Key detailed fog chemistry measurements have been made in the Po Valley (Fuzzi *et al.* 1988). These have been extended to estimate fog water deposition, but only to a very limited extent and more clearly needs to be done in other countries. It may be much more important for the assessment of effects through exposure of vegetation to very large concentrations than as a contribution to the deposition budget.

Dry Deposition

Gases

The key nitrogen containing species are NO_2, NH_3 and HNO_3. Although other species including NO, HONO and PAN are exchanged at the surface, the rates of exchange are small and make only a minor contribution to ecosystem inputs. On continental and global scales the emission of NO from soils makes a major contribution to the oxidized nitrogen budget of the atmosphere and the approaches of Williams *et al.* (1992) provide a helpful if rather empirical method to estimate

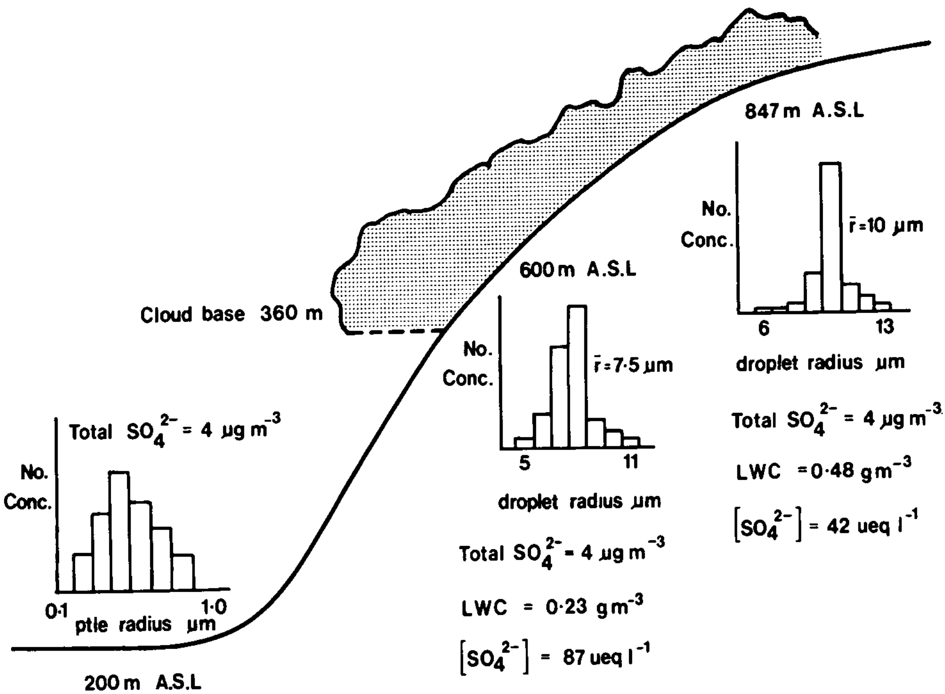

Figure 3 **An illustration of the changes at Great Dun Fell in particle size and ionic concen tration as aerosols are activated into cloud droplets and grow as they are advected up the hillside. (From Fowler *et al.* 1991)**

annual fluxes. Similar simplistic approaches by Skiba *et al.* (1992) for the UK suggest that this source contributes only *ca* 5% of the national emissions from all sources.

NO_2

A mechanistic basis for NO_2 deposition has developed from the large field measurement campaigns of the last 8 years and laboratory studies to investigate processes at the leaf level. The measurements of Hargreaves *et al.* (1992) for example show that for agricultural crops during the growing season, rates of NO_2 exchange are regulated by stomata, and no significant mesophyll (internal) resistance to deposition is present. Such measurements have been used to parameterize a big-leaf resistance model to qualify the spatial variability in NO_2 dry deposition
over regional scales (Duyzer & Fowler, 1994).

A major complication in the provision of field measurements of NO_2 dry deposition rates is chemical reaction in the lowest few metres of the atmosphere in which gradients in concentrations are used to infer fluxes. The reactions between NO, NO_2 and O_3 in the surface

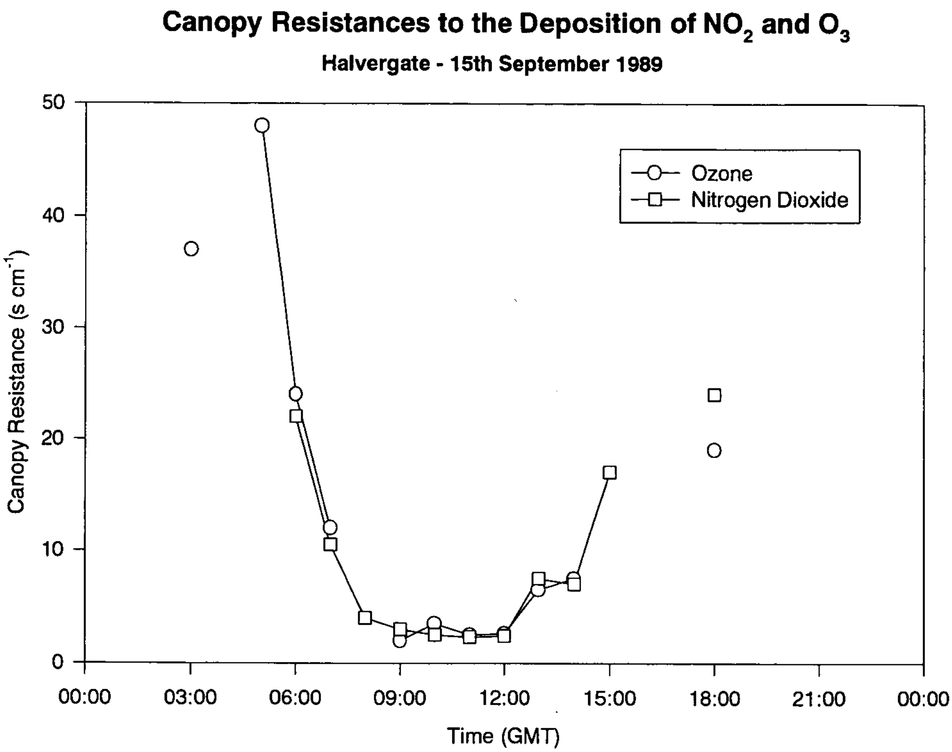

Figure 4 **Canopy resistances to the deposition of NO_2 and O_3 measured at Halvergate in East Anglia, September 1989.**

layers in the presence of upward fluxes of NO from soil microbial processes and NO_2 deposition to the ground, complicate the simple interpretation of fluxes from the gradients and require a more complex analysis. Such approaches have been developed by Kramm *et al.* (1991), Gao *et al.* (1991) and by Duyzer *et al.* (1995) among others. All approaches make considerable demands on the quality of field data and the more pragmatic approach of Duyzer *et al.* offers a realistic opportunity for making progress in the underlying science with the equipment currently available. The strong dependence of NO_2 uptake on stomatal opening lead to an assumption in

the modelling that leaf surface uptake is negligible. New measurements in winter conditions over moorland of NO_2 deposition show that small but significant rates of NO_2 deposition occur at times when stomata are closed (during cold winter weather over senescent moorland). An example of these recent NO_2 dry deposition data is shown in Figure 5.

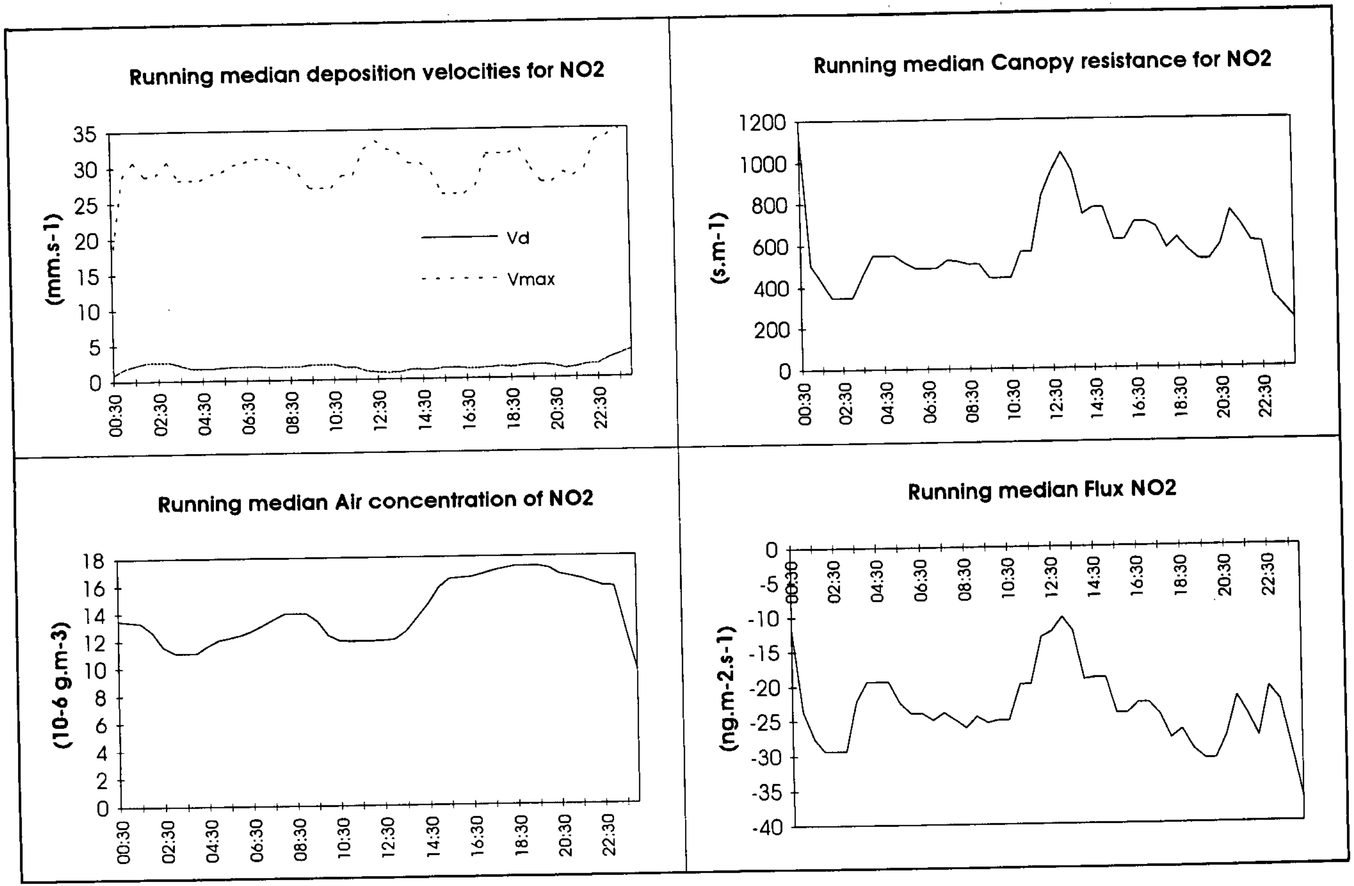

Figure 5 NO_2 measurements at Authencorth Moss, near Edinburgh (LIFE site) on 4 January 1995. a)Running median deposition velocities;b) Running median canopy resistance; c) Running median air concentration; d) Running median flux.

Application of the existing simple parameterization scheme for NO_2 deposition in the UK leads to an annual input flux of about 100 kt N. Unlike the similar modelling exercise for SO_2 the data are not supported by an NO_2 concentration and dry deposition network with dedicated rural monitoring stations and a number of continuous monitors. The concentration field has been provided by two intensive (6-month) campaigns of NO_2 diffusion tube measurements (1986 and 1991) throughout the UK. For the larger scale (European) estimates of concentration and dry deposition fields, the monitoring data are even more patchy, with excellent data for the Netherlands and parts of Germany and few available data for large areas of France and Spain. In the absence of monitoring data, the concentration fields provided by EMEP have generally been applied. The upgrading of NO_2 monitoring and development of a continuous flux measurement stations are clear priorities to assess oxidized N deposition in Europe.

HNO_3, HONO, PAN

The field data on deposition processes support the treatment of natural surfaces as perfect sinks for nitric acid (Muller *et al.* 1992). Climatological (or weather) data can be readily adapted with land use to map spatial variability in deposition rates. What is missing is an adequate measurement network for HNO_3 concentrations. The few monitoring stations available within the EMEP network and additional measurements from research studies show that in northern Europe, HNO_3 makes a minor contribution to oxidized nitrogen input, while in central and southern Europe, the contributions are much larger and require measurement networks to define concentration fields.

Similar arguments may be advanced for HONO and PAN, both of which are present in much larger concentrations in central and southern Europe than in western and northern Europe and Scandinavia. The measurement base is weak and is inadequate to support estimates of fluxes. For these two gases the exchange with natural surfaces also remains uncertain. Recent measurements by Kitto and Harrison (1992) show the evidence of HONO production at the ground from NO_2. However too little of the underlying mechanisms and its dependence on NO_2 concentration temperatures and other variables is available to predict annual fluxes and the importance of this process on regional scales.

NH_3

The exchange of ammonia over natural surfaces has now been studied in detail over moorland forest and agricultural land by Duyzer *et al.* (1992), Sutton *et al.* (1994) among others. The main features of the process are understood. The exchange of NH_3 is bidirectional, with emission fluxes from vegetation when ambient concentrations lie below a canopy compensation point, and fluxes are directed towards the canopy when ambient NH_3 concentrations exceed the canopy compensation point. The leaf surface sink uptake is a major factor competing with stomatal exchange to determine the net flux (Figure 6). The net NH_3 flux over natural vegetation is dominated by uptake, which at times approaches the maximum values possible (i.e. canopy resistance $r_c = 0$). However, even over these surfaces a canopy resistance is usually present with values in the range 5 to 30 s m^{-1}. These limit the deposition rates of NH_3 to semi-natural vegetation to 0.5 to 3 cm s^{-1} under most conditions. In some conditions, such surfaces may be seen to emit NH_3 to the atmosphere (Sutton *et al.*, 1994).

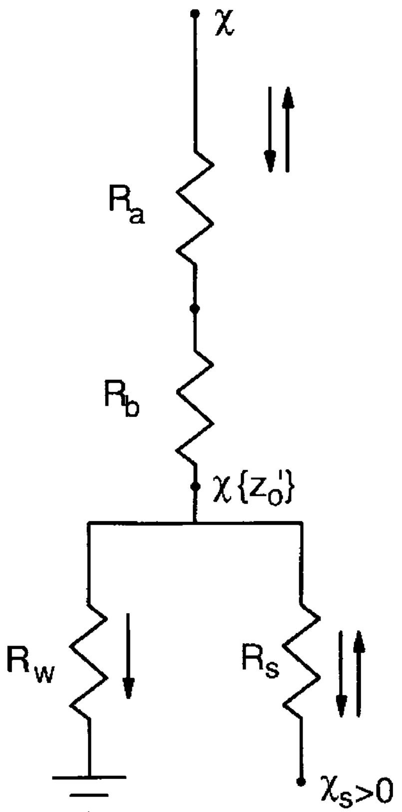

Figure 6 **Proposed resistance model of NH_3 exchange with plant canopies accounting for parallel deposition onto leaf surfaces (R_a) and exchange through stomata (R_s) with a stomatal compensation point (χ_s). The net canopy compensation point is given by $\chi\{z_o'\}$.**

For agricultural crops bidirectional exchange is a much more common feature of the data. As an example, Figure 7 shows data for measurements over a wheat crop which indicate a change in direction of the flux (from deposition to emission) when air concentration of NH_3 reaches about 1 µg NH_3 m^{-3}. A model simulating net canopy-atmosphere exchange using the canopy compensation point model χ_c

where $[\chi\{z\}/(R_a\{z\}+R_b)+\chi_S/R_S]\,\chi_c = [(R_a\{z\}+R_b)^{-1}+R_S^{-1}+R_W^{-1}]$

simulates the net exchange very well, assuming 100 µmolar NH_4^+ in intercellular fluids Figure 6 (Sutton & Fowler, 1993). These recent developments in simulating net exchange between the atmosphere and terrestrial ecosystem provide the basis for mapping annual reduced nitrogen inputs and regional scale. However, the spatial variability in several key parameters remains unknown; the temporal and spatial patterns in intercellular NH_4^+ concentration for agricultural crops, the temporal variability in leaf surface resistance and last and most important, the ambient ammonia concentrations are all required to complete this work. The approach of the Netherlands in providing a dedicated monitoring network for NH_3 concentrations provides the ideal model for other contries of Europe. Many European countries have no monitoring of NH_3 at all.

Within the UK, diffusion tube surveys by Atkins and Lee *et al.* (1992) in the UK for 1988 provide the current best national estimate of measured concentration field, but is subject to significant uncertainty, not least because there appears to be a significant over-estimate of actual NH_3 concentration using these methods. To provide the best estimate of reduced nitrogen deposition with current information, a correction factor for the diffusion tube data of 0.43 has been applied. The semi-natural vegetation has been assumed to be a consistent sink for NH_3 with a mean canopy resistance of 10 s m^{-1} and deposition velocities and fluxes have been calculated for measured wind and temperature fields and land class cover information to identify the roughness length (z_0) for each of the major land uses. The arable land is at present assumed to be ammonia neutral (that is that with reference to measured air concentration there is no net NH_3 exchange with the atmosphere over the year).

This is the area of nitrogen deposition that is most uncertain and in need of major improvement in mapping and synthesis of current data. The state of the measurements of NH_3 is similar in all other countries with the exception of the Netherlands where a monitoring network is in place. The modelling of net NH_3 exchange over regional scales is at a rudimentary stage everywhere.

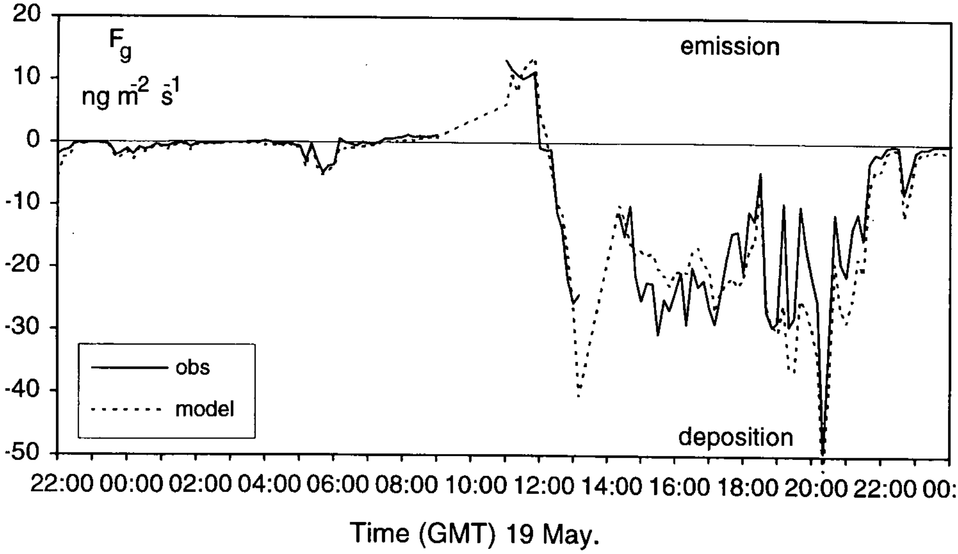

Figure 7 **Predicted flux of NH_3 compared with measurements. Model assumes 100 molar NH_4^+ in leaf tissues and R_w dependant on relative humidty. (Sutton & Fowler 1993)**

Particle Deposition

With the exception of cloud droplet deposition, the field of particle deposition has been largely ignored during the last 10 years. However, a number of issues and scientific developments have made it important to provide particle deposition inputs throughout Europe. First, base cation deposition is a very important contributor to the mass balance approach for critical loads estimates, and atmospheric base cations are deposited on forests at significant rates. Second, recent field data show that for aerodynamically rough vegetation, rates of (1 to 5 μm) particle deposition are in the range 7 to 15 mm s^{-1} (e.g. Wyers *et al.* 1995). Thus for forests and other aerodynamically rough surfaces the inputs of NH_4^+ and NO_3^- in particle form may be a significant contribution to the total input.

To provide the necessary parameters and fields for mapping, a low density network of monitoring stations for size resolved aerosol NO_3^- and NH_4^+ concentrations is necessary. The research required is essentially the size dependence of deposition velocities of aerosols over short ('grass'), medium height (e.g. cereals) and tall vegetation (forest) and urban areas. These data are also required to quantify the atmospheric life-times of aerosols by dry deposition, an issue of considerable importance in the current debate over aerosols and human health and aerosols and radiative effects.

The Effect of Deposition Grid Scale for Exceedance Estimates

The concentration fields, meteorological and land use input data used to obtain estimates of the annual input have an optimum grid scale, which varies with the different gaseous species and the mechanism of deposition. The choice of 20 km x 20 km for application in all maps is largely a consequence of the wet deposition data within which orographic enhancement has been calculated on rather simple assumptions to avoid the real complexity of orography which is currently beyond the modelling approaches that have been developed. Thus, for a 20 km x 20 km grid square in complex terrain, the actual deposition at a number of points at the same altitude may differ as a consequence of upwind topography.The average value mapped results from the enhancement of precipitation for the grid square over the interpolated coastal values. While these simplifications prevent the application of deposition data at a finer scale than 20 km x 20 km, it is possible for selected grid squares to estimate variability in actual 1 km x 1 km deposition that would be expected on the basis of current understanding of deposition processes. An exercise in quantifying these effects by Smith *et al.* (1995) showed that in the high rainfall areas of the UK, the 1 km x 1 km critical loads exceedance would be doubled by quantifying actual wet deposition variability with 20 km x 20 km grid square. Procedures to provide this detail for all grid squares have not been developed. The consequence of this finding is that current maps of critical load exceedance (for soils and acidification) underestimate the actual exceedance throughout the areas of complex terrain.

Conclusions

- Wet deposition. This is generally well estimated in simple topography with current networks, relative to dry deposition. For the UK, the network is currently at its minimum size for the mapping tasks and a few more monitoring stations are necessary.

- Uncertainty in complex terrain leads to under-estimation of deposition (and exceedance). There is a need to develop procedures to provide finer scale inputs and validate inputs and their spatial variability in complex terrain with measurements.

- Cloud deposition. Estimates appear satisfactory at a limited number of high altitude regions of northern Europe where monitoring underpins the modelling. The importance of radiation in fog to deposition is largely unknown.

- Dry deposition - NO_2. The theoretical basis for input estimates is good but there is very little validation and therefore large uncertainty and the network for NO_2 concentration measurement is not adequate. An NO_2 network and flux measurement facility is necessary.

- Dry deposition - NH_3. Most sensitive receptors show large rates of uptake and are therefore best estimated using small r_c (10-20 s m^{-1}). Uncertainty in input is very large. There is no current basis for net flux over agricultural areas. There is no network for NH_3 concentration measurement and the lifetime of NH_3 due to gas to particle conversion is also very uncertain. Much work is needed.

- Particle deposition. Particle deposition is important for forests, especially in areas remote from sources, and is poorly estimated. Rural concentration means of particle concentrations for NH_4^+, NO_3^- are not known. Process research (size dependence of aerosol Vg) and monitoring are both necessary.

- Land-use. Land-use specific inputs of nitrogen provide a more rigorous basis for Critical Loads exceedance mapping.

- Spatial scale of deposition estimates. The accumulated area of exceedance increases as the area used to estimate deposition decreases (for example, the change from 20 km x 20 km to 1 km x 1 km leads to a doubling of the area of exceedance).

- Complex terrain. The effects of complexity in the terrain due to hedgerows, small woodland patches and changes in roughness length on deposition rates remain unknown.

- Urban deposition. The deposition rates of NO_2, NH_3 and particle NO_3^- and NH_4^+ to urban surfaces are unknown and are needed to define inputs and the lifetime of pollutants in urban air.

References

Atkins, D.M.F. and Lee, D.S. (1992) The distribution of ammonia in the United Kingdom. AEA-EE-0330, AEA Technology, Harwell, Oxon.

Choularton, T.W., Gay, M.J., Jones, A., Fowler, D., Cape, J.N. and Leith, I.D. (1988) The influence of altitude on wet deposition.Comparisons between field measurements at Great Dun Fell and predictions of a seeder-feeder model. *Atmos. Environ.*, 22, 1363-1371.

Dollard, G.J., Unsworth, M.H. and Harvey, M.J. (1983) Pollutant transfer in upland regions by occult precipitation. *Nature*, 302, 241-243.

Duyzer, J.H., Verhagen, H.L.M., Westrate, J.H. and Bosveld, F.C. (1992) Measurement of the dry deposition flux of NH_3 on to coniferous forest. *Environ. Pollut.*, 75, 3-13.

Duyzer, J. H. and Fowler, D. (1994) Modelling land atmosphere exchange of gaseous oxides of nitrogen in Europe. *Tellus*, 46B, 353 - 372.

Duyzer, J.H., Deinum, G. and Baak, J. (1995) The interpretation of measurements of the surface-atmosphere exchange flux of nitrogen oxides; correction for chemical reactions. *Phil. Trans. Roy. Soc. Lond.* (In press).

Fowler, D., Baldocchi, D. and Duyzer, J.H. (1991) Inputs of trace gases, particles and cloud droplets to terrestrial surfaces. In: *Acidic deposition, its nature and impacts.* Proceedings of the Royal Society of Edinburgh Vol 97B, F.R. Last and R. Walling (Eds).

Fowler, D., Cape, J.N., Leith, I.D., Choularton, T.W., Gay, M.J. and Jones, A. (1988) The influence of altitude on rainfall composition at Great Dun Fell. *Atmos. Environ.*, 22, 1355-1362.

Fowler, D., Cape, J.N. and Unsworth, M.H. (1989) Deposition of atmospheric pollutants on forests. *Phil. Trans. R. Soc. Lond.*, B324, 247-265.

Fuzzi, S. (1988) Fog chemistry and deposition in the Po Valley, Italy. In: *Acid deposition at high elevation sites*, edited by M.H. Unsworth & D. Fowler, pp 443-452. Kluwer, Dordrecht.

Gallagher, M.W., Choularton, T.W., Morse, A.P. and Fowler, D. (1988) Measurements of the size dependence of cloud droplet deposition at a hill site. *Quart. J. Roy. Met. Soc.*, 114, 1291-1303.

Gallagher, M.W., Beswick, K., Choularton, T. W., Coe, H., Fowler, D. and Hargreaves, K.J. (1991) Measurements and modelling of cloud water deposition to moorland and forests. *Environ. Pollut.* 75, 97-107.

Gao, W., Wesely, M.L. and Lee, I.Y. (1991) A numerical study of the effects of air chemistry on fluxes of NO, NO_2 and O_3 near the surface *J. Geophys. Res.*, 96, D10, 18761-18769.

Hargreaves, K.J., Fowler, D., Storeton-West, R.L. and Duyzer, J.H. (1992) The exchange of nitric oxide, nitrogen dioxide and ozone between pasture and the atmosphere. *Environ. Pollut.* 75(1), 53-60.

Kramm, G., Muller, H., Fowler, D., Hofken, K.D., Meixner, F.X. and Schaller, E. (1991) A modified profile method for determining the vertical fluxes of NO, NO_2, ozone and HNO_3 in the atmospheric surface layer. *J Atmos. Chem.*, 13, 265-288.

Kitto, A.M.N. and Harrison, R.M. (1992) Nitrous and nitric acid measurements at sites in south-east England. *Atmos. Environ.*, 26A (2), 235-241.

Muller, H., Kramm, G., Meixner, F., Dollard, G.J., Fowler, D., and Possanzini, M. (1992) Determinations of HNO_3 dry deposition by modified Bowen ratio and aerodynamic profile techniques. *Tellus*, 45B, 346-367.

Skiba, U., Hargreaves, K.J., Fowler, D. and Smith, K.A. (1992) Fluxes of nitric and nitrous oxides from agricultural soils in a cool temperate climate. *Atmos. Environ.*, 26A(14), 2477-2488

Smith, R.I., Fowler, D. and Bull, K.R. (1995) Quantifying the scale dependence in estimates of wet and dry deposition and the implications for critical load exceedances. In: *Acid Rain Research - Do we have enough answers?* Proceedings of the s-Hertogenbosch conference, The Netherlands, October 1994, Elsevier Science, (In press).

Sutton, M.A., Asman, W.A.H. and Schjorring, J.K. (1994) Dry deposition of reduced nitrogen. *Tellus*, 46B, 225-273.

Sutton, M.A. and Fowler, D. (1993) A model for inferring bi-directional fluxes of ammonia over plant canopies. In: *Proceedings of the WMO conference on the measurement and modelling of atmospheric composition changes including pollutant transport.* WMO/GAW - 91. (Sofia 4-8 October 1993). World Meteorological Organization, Geneva. pp179-182.

Weston, K.J. (1991) Objectivity analysed cloud immersion frequencies for the United Kingdom. *Meteorl. Mag.*, 121, 108-111.

Williams, E.J., Guenther, A. and Fehsenfeld, F.C. (1992) An inventory of nitric oxide emissions from soils in the United States. *J. Geophys. Res.*, 97, 7511-7520.

Wyers G.P., Veltkamp A.C., Geusebroek M, Wayers A. and Moels J.J. (1995) Deposition of aerosol to coniferous forest. In: *Acid Rain Research: Do we have enough answers?* Proceedings of the s-Hertogensbosch Conference (9-11 October 1994). Elsevier Scientific, (in press).

CHAPTER 9
MAPPING AND MODELLING CRITICAL LOADS

Discussion Group Report

Introduction

The discussions of the group focused on two main areas: one, calculating and mapping critical loads for nitrogen using the combined critical loads function with fixed values of sulphur deposition; and two, the use of gap closure scenarios and the varying degrees of ecosystem protection which may result for different ecosystems or grid squares for any given scenario.

Mapping Critical Loads for Nitrogen

In principle, the current formulation of the mass balance model allows sulphur and nitrogen critical loads and exceedances to be considered simultaneously. However, using the combined critical loads function it is not easy to define percentiles which are required for the optimization models. In addition, there are now time constraints to produce maps of critical loads for nitrogen for the UNECE by August 1995.

The group therefore discussed a simplified approach to generating the required numbers and maps. Working on the assumption that the second sulphur protocol has been signed and will be implemented, sulphur deposition values could be set to those resulting from the implementation of the protocol. By using the combined critical loads function, the critical load for nitrogen could be computed for this or any other given sulphur deposition value. The equation below and Figure 1 illustrate the procedure that could be adopted.

$$CL(N|S_{dep}) = \begin{cases} CL_{min}(N) & \text{for } S_{dep} > CL_{max}(S) \\ CL_{min}(N) + \dfrac{CL_{max}(N) - CL_{min}(N)}{CL_{max}(S) - CL_{min}(S)}(CL_{max}(S) - S_{dep}) & \text{for } CL_{min}(S) \leq S_{dep} \leq CL_{max}(S) \\ CL_{max}(N) & \text{for } S_{dep} < CL_{min}(S) \end{cases}$$

where

$CL(N|S_{dep})$ = critical load for nitrogen for a given sulphur deposition
$CL_{min}(N)$ = minimum critical load for nitrogen
$CL_{max}(N)$ = maximum critical load for nitrogen (depending on acidifying and/or nutrient nitrogen)
$CL_{min}(S)$ = minimum critical load for sulphur
$CL_{max}(S)$ = maximum critical load for sulphur
S_{dep} = sulphur deposition

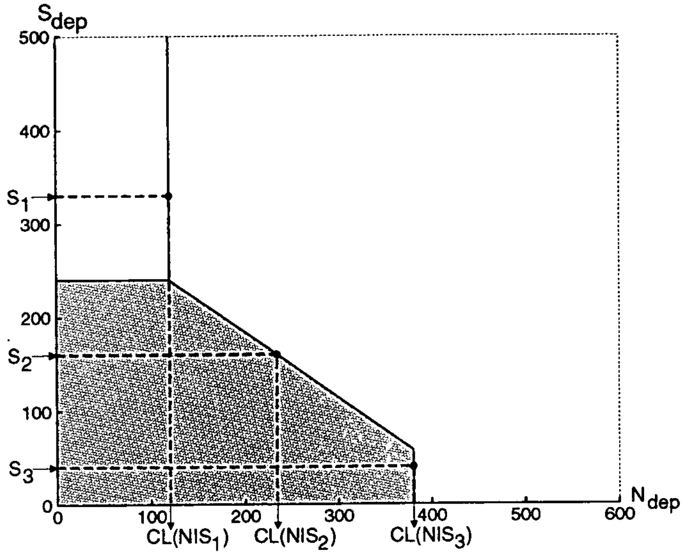

Figure 1 **Determining the critical load for nitrogen for three different cases of given sulphur deposition.**

Internationally the EMEP model could be used to estimate the sulphur deposition determined by the sulphur protocol. It is recognised that for some countries the deposition values may differ from national data and agreement needs to be reached as to which values should be used. However, it would be easy to modify the nitrogen critical loads for any given sulphur deposition values agreed upon. This approach has the advantage that it makes use of all the information provided to the CCE and the resulting critical loads for nitrogen can be used in the optimization models in the same way in which the critical loads for sulphur were utilised.

Ecosystem protection

The group discussed a number of issues related to reduction strategies and ecosystem protection. They concluded:-

- Current data do not indicate any time scales within which "harmful effects" will occur.
- The European data and maps may refer to different ecosystems in different countries and for some countries CCE data are used where no national data are available. It is important that land use information used for deriving critical loads and for transport modelling is consistent. For assessing target loads the ecosystem type may be important.
- Much of the work on reduction of sulphur emissions focused on a gap closure scenario which,

for each grid square, reduced deposition by 60% from its current level to the 5-percentile critical load value in order to obtain a target load. This results in some areas still exceeding critical loads where some ecosystems may be affected and others may be protected. By using gap closure the percentage of any ecosystem protected will depend on the cumulative distribution frequency of the ecosystem in any grid square. This may result in some countries achieving greater areas of protection than others. Figure 2 illustrates the different levels of protection that may occur for the same gap closure scenario.The group concluded that when setting target loads thought should be given to emphasize ecosystem protection over percentage reductions in exceedance.

- Some members of the group suggested it would be useful to be able to quantify the uncer tainty in the data provided to the CCE in some way, and to demonstrate how this may affect strategy.

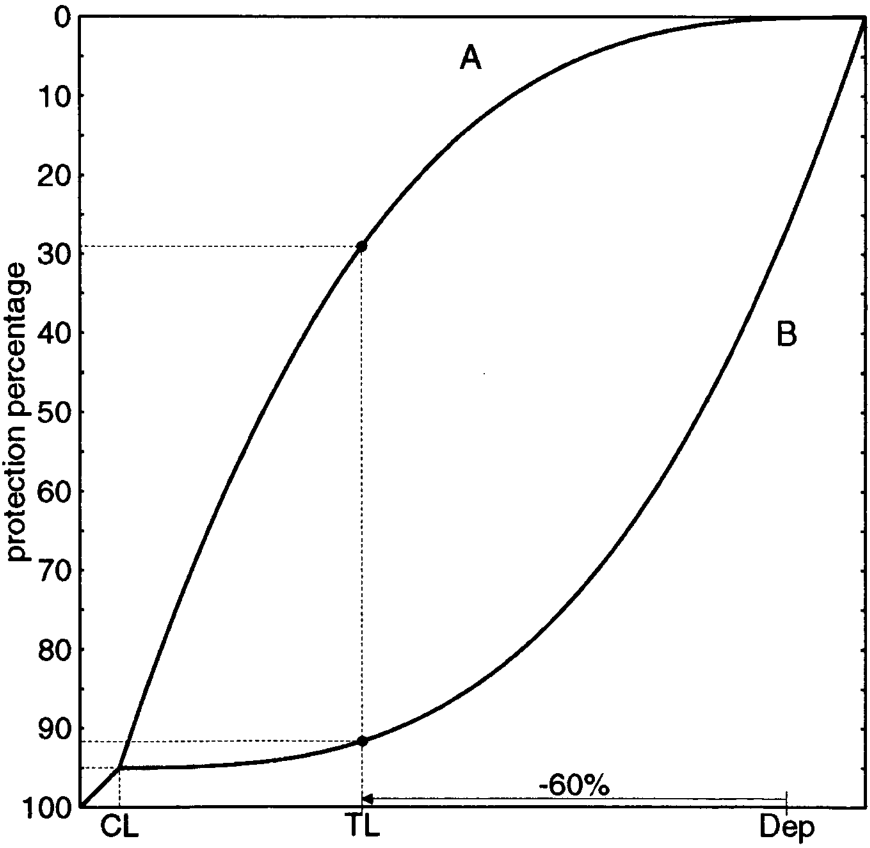

Figure 2 **Consequences of a 60% gap closure on the ecosystem protection level for 2 different cumulative distribution functions A and B.**

CHAPTER 10
CRITICAL LOADS OF NITROGEN: POSTERS

1 Fluxes of Inorganic and Organic Nitrogen for UK Hill Peats

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Leaching of nitrogen species from peats subjected to a range of pollution loads was quantified over 12 months in order to obtain data for estimating critical loads of N and acidity.

Duplicate peat turfs (460 mm x 320 mm x 150 mm depth) were collected from each of 9 moorland sites along a pollution gradient[1] (Sanger *et al.*, 1994). *Calluna vulgaris* was the dominant vegetation at all sites but the most polluted (Hatfield Moor, dominated by *Deschampsia flexuosa* and *Pteridium aquilinum*). The sites are also situated along a climatic gradient, from cool and wet in the north-west to relatively warm and dry in the south-east. The experiment aimed to eliminate the climatic effects by bringing peat microcosms from all sites to a controlled environment, so that rain composition was the only variable. The randomised microcosms were subjected twice weekly to site-appropriate simulated acid rain for 6 months to allow for settling in before the 12-month period for which results are presented here. The rain compositions were based on those at the nearby precipitation monitoring network sites, (RGAR, 1990). In the field, the sites received between 527 and 1262 mm rain per year (1986-88 average) (RGAR, 1990). In our experiments, every peat received the same rainfall, 1204 mm. Dry deposition was not taken into consideration, so generally the peats received smaller N pollution loads than they would in the field.

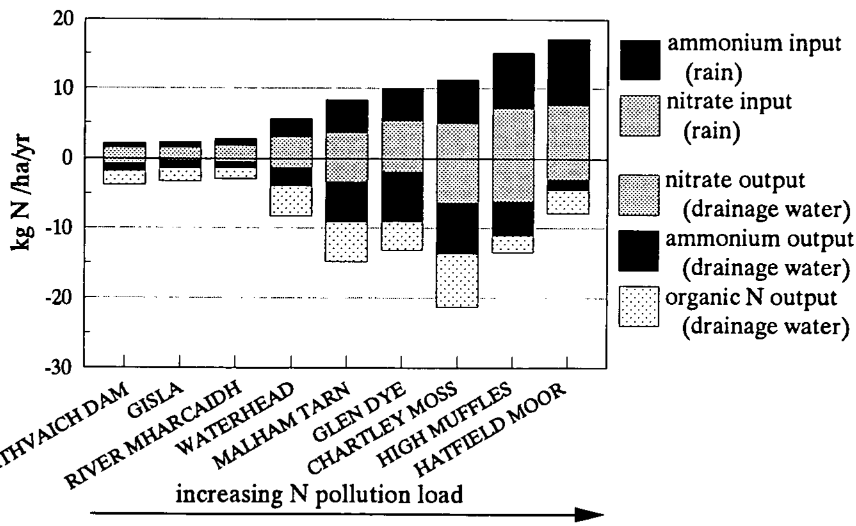

Figure 1 **Annual inputs and outputs of nitrogen species**

Leachate was collected weekly. Bulked leachate samples were analysed monthly for total N, nitrate-N and ammonium-N (Yesmin *et al.*, 1995). Organic N was determined by subtracting the values for nitrate-N and ammonium-N from those for total N.

The distribution of N species in simulated rain and drainage water is shown in Figure 1.

Results

1) When nitrate inputs were low to moderate, most of the nitrate was retained in the soil/plant system.

2) At higher nitrate inputs, nitrate uptake became relatively less important, and a large proportion of the nitrate input ended up in the drainage water.

3) For all sites except the two most polluted, ammonium leached out of the system exceeded ammonium deposition.

4) To a first approximation, soluble organic N leached out of the system increased linearly with N input. At higher N inputs, High Muffles and Hatfield Moor showed a quite different trend, with obvious organic N accumulation. This was reflected in the exceptionally low decomposition rate for *Calluina* litter at Hatfield Moor (with litter bags retaining 74±6.4% of their weight after 18 months embedded in the peat litter layer), resulting in a high hydraulic conductivity through clearly visible undecomposed plant remains.

   Nitrogen accumulation with increasing nitrogen deposition can be shown by the C:N ratio of the peats, which is negatively correlated with total N deposition (r^2=0.907, Figure 2).

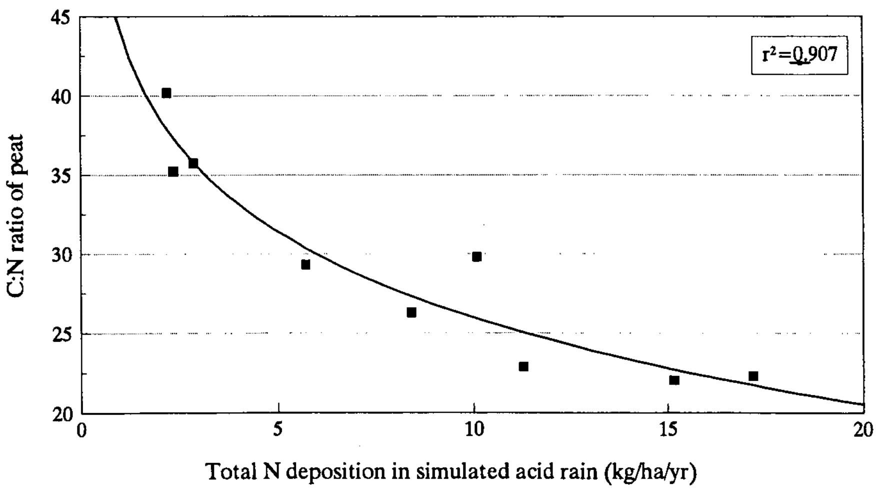

Figure 2 **Relationship between C:N ratio of peat at the end of the experiment and wet N deposition applied for 18 months.**

Nitrate flux is a measure of "net nitrification": if nitrate outputs exceed nitrate inputs, nitrification is occurring, and the nitrate flux gives a measure of nitrification which is corrected for biomass uptake. The results in Figure 1 suggest that net nitrification is not a significant process in these organic soils, with nitrate flux being negative in most cases. Furthermore, under conditions of low to moderate pollution, ammonium would not appear to contribute to soil acidification, since ammonium outputs exceed inputs.

Species diversity of the turfs at sampling time varied from 6-7 at Strathvaich Dam, Gisla, Mharcaidh, Waterhead and Glen Dye, through 5 at Malham Tarn and 4 at Chartley Moss and High Muffles, down to 2 species at Hatfield Moor. At the time of sampling, *Calluna* was found at all sites except Hatfield Moor. The high N deposition treatment resulted in loss of *Calluna* at Malham Tarn and Chartley Moss over 18 months, and at Hatfield Moor there was clear evidence in the plant remains in peat for *Calluna* loss in the field, before the start of the experiment. The *Calluna* loss may be a result of an observed sharp decline in mycorrhizal infection with increasing N deposition. High Muffles (which has a high mineral content relative to the other peats) retained its *Calluna*, but *Deschampsia* increased, from 0% at the start of the experiment, to 5% cover after 18 months.

The N flux results can be used to model N outputs from total N inputs. As can be seen from Figure 3, it is possible to approximate the amounts of the various N species in drainage water from total N inputs.

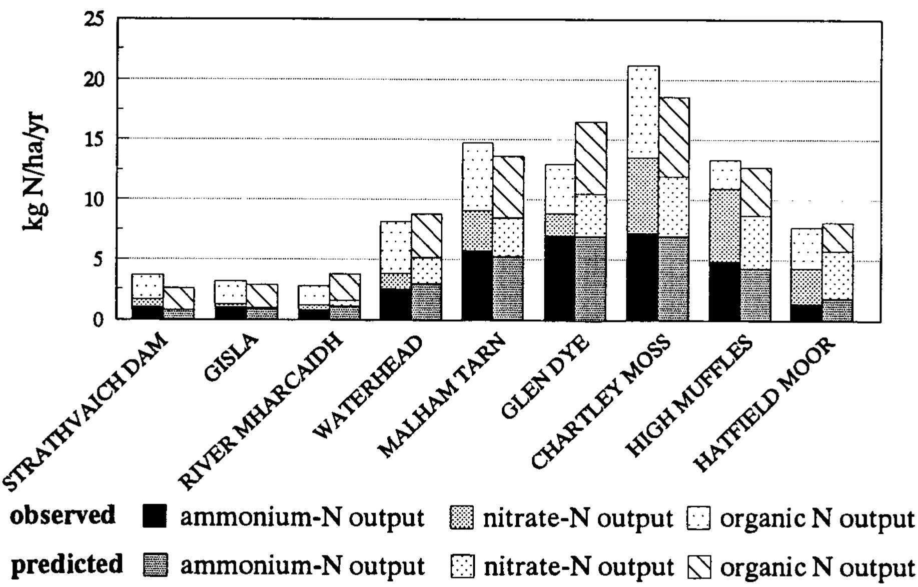

Figure 3 **Comparison of predicted N outputs with experimental results.**

To quantify exceedances of critical loads of acidity for organic soils, it is necessary to estimate the proportions of the deposited N species involved in acidification, and leached straight out of the plant/

soil system. It appears possible to make these estimates from simulated acid deposition experiments, for incorporation into critical load calculations.

In these experiments, peats from 9 sites along a pollution gradient were subjected to simulated acid rain of the same concentration as they received in the field, but with no dry deposition. Further experiments have been performed using peat from one site and varying the pollution load, including dry deposition. The data from these experiments is currently being analysed.

This work was funded by National Environment Council and the United Kingdom Department of the Environment. Ms. Yesmin's Ph. D. studentship was funded by the International Rotary Fund.

References

Sanger, L.J, Billet, M.F. and Cresser, M.S. (1994) *Chemistry and Ecology* 8, 249-264.

RGAR (1990) Third Report of theUnited Kingdom Review Group on Acid Rain *Acid Deposi tion in the United Kingdom 1986-88*. Warren Spring Laboratory.

Yesmin, L., Gammack, S.M., Sanger, L.J. and Cresser, M.S. (1995) Impact of atmospheric N deposition on inorganic- and organic-N outputs in water draining from peat. *The Science of the Total Environment* (in press).

2 Bioindicators of Norway Spruce Tree Tolerance to Natural and Anthropogenic Stress Impacts

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Introduction and Methods

Forest decline is a multifactorial complex problem caused by natural and anthropogenic global environmental changes (global warming, elevated CO_2 concentration, increased UV-B radiation, air pollution, natural and man-made soil acidification, etc.) imposed by acute climatic and pollution stress events. The long-term studies of atmospheric deposition, shoot and root responses to the recent stress events and regenerative capacity of individual trees could enable an assessment of the critical levels for atmospheric deposition and realistic predictions of tree response to single or synergistic environmental impacts (Cudlín and Šiffel, 1991).

To approach the variability of the whole Krkonoše Mts., one of the most beautiful and endangered national park in Europe, given by differences in elevation, slope exposure, nutrient availability and atmospheric deposition inputs, four permanent research plots (50 x 50 m) were established in 1986. During the last twenty years, spruce forest stands in Krkonoše Mts. have been subjected to increased synergistic action of natural and anthropogenic stress impacts, often exceeding their adaptation capacity.

Throughfall and bulk precipitation measurements, soil analyses, damage analysis of the spruce crowns and needles and monitoring of fungal fruitbody occurrence have been performed since the establishment of the plots. In addition, estimation of fine root and mycorrhiza growth potential in forest stands and of fine root and mycorrhiza development of spruce seedlings grown in soils from permanent plots (spruce seedling biotests) have been investigated since 1990 (Cudlín *et al.* 1994).

Results

The proportion of dead trees on permanent research plots ranged from 35 to 92% in 1993. In contrast, average defoliation of a sample of trees does not vary so dramatically (from 30 to 53%). A transformation of primary shoots to secondary shoots of higher orders (shoots originated from proventitious buds) seems to be a better indicator of assimilative organ response to chronic and acute stress impacts.

The highest percentage of non-mycorrhizal root tips was estimated on more polluted and damaged plots under both the field and greenhouse conditions. However, the fully developed mycorrhizas were less frequent on these plots only by spruce seedlings grown in the foil house.

Numbers of ectomycorrhizal fungal species on single permanent plots have changed very dramatically during the last 10 years. With the exception of the most damaged plots, the species diversity again increased during the last three years (in one plot from 0 to 10 species). The fruitbody production does not appear to correlate directly with the current tree health state.

Discussion

Data analysis confirmed the multivariate nature of Norway spruce stand response to natural and anthropogenic stress impacts and the necessity of long-term observations. Each forest ecosystem possesses its own strategy to counteract an adverse change in growth conditions.

Forest stands relatively protected from extreme climatic conditions and pollution inputs owing to their topographic situation, resist heavy damage for a long time. Gradually, a deterioration of some measurable characters (fructification and diversity of ectomycorrhizal fungi, growth characteristics of spruce seedlings grown in soil monoliths from plots under study) becomes observable. Subsequently, the health of the forest stand deteriorates rapidly (often in connection with a mass needle yellowing). In following years, there is frequently a regeneration of both above- (secondary shoot formation) and below-ground parts of the tree (fine root regeneration, adaptation of rhizospheric microflora including ectomycorrhizal fungi). These regeneration processes could be explained by periodical change of climate conditions in the mountains and slightly reduced atmospheric deposition in recent years. Some inner conditions of forest stands (e.g. increased quantity of litter for decomposition, reduced area of photosyntetically non-active needles), occurring after the end of progressive tree damage, could also promote regeneration processes, too.

The spruce forest stands, exposed to the higher environmental stress impact (because of their higher elevation, more exposed situation, adverse soil conditions or earlier beginning of stress action), can respond three ways:

1) similar course to that described above but without a long resistance phase, with more pronounced fluctuations of damage and regeneration phases around a higher level of forest ecosystem disturbance;

2) pronounced fluctuations of damage and regeneration phases around a relatively stable level of disturbance, arising in the first years after the beginning of increased stress impact;

3) pronounced fluctuations of damage and regeneration phases with gradually increasing disturbance of forest ecosystem, culminating in the forest stand dying.

Periodic investigations of needle damage, defoliation and soil quality (using the spruce seedling biotest) detect the changes in forest stand due to the fluctuating environmental impacts (including atmospheric deposition). This enables the precise critical levels for forest stands under different site conditions to be quantified.

The features of regeneration processes in the crown and fine roots and the mycorrhiza potential of the tree are sufficient indicators of the regeneration capacity of individual trees. The occurrence of secondary shoots can be considered as an indicator of stress exceedance. The definition of critical levels for these forests could be modified: *"the maximum concentration of a pollutant at which the forest stand is able to compensate for harmful effects by regeneration processes"*.

References

Cudlín P. and Šiffel P. (1992) Mutual responses of the assimilative apparatus and root system of Norway spruce in a mountain forest stressed by air pollution. In: Proc. Int. Seminar, Industrial Pollution Damage of Forest Ecosystems in ÈSFR, September 1991, Srní, Czechoslovakia, Federal Committee for Environment, Prague, p. 8-14.

Cudlín P., Chmelíková E. and Rauch O. (1994) Monitoring of mountain Norway spruce forest response to environmental impact in Krkonoše Mts. In: Proc. Int. Conf. IUCN & MAB, Moni toring and Management of National Parks, September 1993, Špindlerův Mlýn, Czech Repub lic (in press).

3 Nitrogen Input into Damaged Forest Stands in the Krusne Mts. (Erzgebirge), Czech Republic

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The Krusne Mts. (NW Bohemia) are known for heavy air pollution (part of the so called "Black Triangle"). Air pollution is a consequence of burning of locally mined lignite in the obsolete power plants, unequipped with desulphurization units. Approximately 40,000 ha of spruce forests died in the 1970s and 1980s. Most of the area was clearcut, limed, and reforested.

In order to assess the present day level of atmospheric deposition we perform bulk precipitation sampling and throughfall measurements in surviving forest stands. Rain is collected by PE funnels, snow is collected in buckets. Bulk precipitation samples are collected in duplicate, throughfall is collected as one composite sample per stand. Ammonium is measured spectrophotometrically, nitrate by ion chromatography.

The Nacetin spruce stand lies close to the German border (50° 35´ N, 13° 15´E). The 70 year old stand is relatively sheltered from the main emission area. Open field precipitation and throughfall (denoted as NET - 25 collectors) data are reported. Measurements started in 1991.

Over the observation period, a decrease in ammonia mass fluxes was observed (Table 1). Ammonium flux decreased in bulk precipitation as well as in throughfall. Net removal of ammonium (difference between throughfall and bulk precipitation fluxes) decreased in a similar pattern, in 1993 to 59%, in 1994 to 36 % of the 1992 value, respectively.

The nitrate load in bulk precipitation remains constant, but throughfall flux decreased relative to 1992 to less than 54 and 56 % in 1993 and 1994, respectively.

Throughfall flux of inorganic nitrogen in the Nacetin forest decreased from more than 2.19 kmoles N ha^{-1} yr^{-1} in 1992 to 1.39 kmoles N ha^{-1} yr^{-1} in 1994. Decreases of a similar relative magnitude were observed for all major compounds, e.g. sulphur, hydrogen ion, base cations, chloride and fluoride. Bulk precipitation load decreased only for ammonia.

Table 1 Annual mass fluxes of nitrogen measured at Nacetin 1992-1994. Units: moles ha^{-1} yr^{-1}.

Mass flux	Bulk precipitation		Throughfall		Net removal	
Compound	NH_4^+	NO_3^-	NH_4^+	NO_3^-	NH_4^+	NO_3^-
1992	411	352	918	1274	507	922
1993	347	362	645	864	298	502
1994	313	374	498	888	185	514

The Jezeri catchment (50° 33´ N, 13° 30´E) lies close to main emission sources; within a 20 km radius 750 000 tons of SO_2 and 190 000 tons of NO_x are produced annually. This catchment (2.61 km^2 has been studied since 1982. Nitrate export from the catchment varied between 205-575 mol ha^{-1} yr^{-1}, showing the high leaching from dying or dead spruce forests in the uppermost part of the catchment.

Since 1993 throughfall measurements has been conducted under the funding of the Czech Grant Agency. A comparison of throughfall fluxes at Nacetin and Jezeri is given in Figure 1. The left side of the figure shows the Nacetin spruce site, the centre shows deciduous stands within the Jezeri catchment and the right hand side shows the spruce stands within the Jezeri catchment of which JET and MHT are the most exposed.

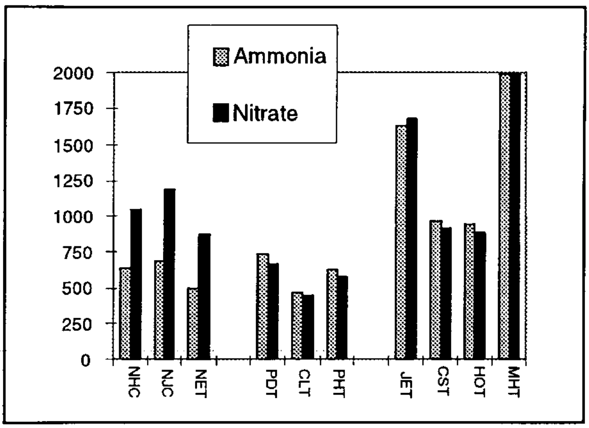

Figure 1 **Annual throughfall fluxes of of nitrogen measured at Nacetin (NET, NJC, NHC) and within the Jezeri catchment in 1994. PHT, CLT and PDT are deciduous stands, MHT, HOT, CST and JET are spruce stands within the Jezeri catchment.**
Units: moles ha^{-1} yr^{-1}.

Nitrogen throughfall fluxes in deciduous stands are lower than in any spruce stand. The highest fluxes were encountered in the most exposed, windy spruce sites in the polluted Jezeri catchment. Nacetin and Jezeri differ not only in the magnitude of throughfall fluxes, but also in the ratio between nitrate and ammonium flux in throughfall. Close to emmission sources both fluxes are almost equal (on equivalent basis), but ammonia forms only about 40% of the nitrate flux at Nacetin.

Decline of the ammonium flux at Nacetin (both in time and space, relative to 1992 data and relative to Jezeri catchment, respectively) may result from the changing patterns of air pollution in the region. Ammonium sulphate aerosol produced through reaction between SO_2 and NH_3 may be currently produced in smaller quantities due to the shutdown of some sources or may not be effectively transported outside the emission sources to the more distant Nacetin. Another uncertainty is gradual deterioration of the canopies of spruce stands, which may adversely affect the ability of spruce trees to capture gases or particles from the atmosphere. Direct measurements of NH_3 and ammonium and sulphate aerosol at Nacetin will start in 1995 to help elucidate this problem.

4 Dynamic Modelling and the Analysis of Critical S and N Loads

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The objective of this report is to demonstrate attempts to use the For SVA model (Arp and Oja, 1992) to assess the atmospheric N and S deposition loads critical for the maintenance of site quality and forest productivity, and how such loads might vary across the sites, and with tree harvesting. Also, the results are compared with critical S and N soil acidification loads as calculated by way of the steady-state mass balance (SMB) approach.

Derivation of Critical Loads with For SVA.

The absence of a decrease in biomass production was considered to be the main effects criterion. Accumulative wood biomass was checked within three consequent natural dominant tree/gap cycles. We also used the value of relative nutrient productivity as a more sensitive characteristic. Aluminum concentration in soil solution leaving the rooting zone (60 cm) and the Al:Ca ratio in the same were checked as additional soil acidification effects.

The Solling Spruce Site

is a Norway spruce plantation which is subjected to high S and N deposition loads. ForSVA was one of several other models used for calculating the critical S and N deposition loads at this site (de Vries *et al.* 1995). Two deposition scenarios are presented in Table 1 for estimating the effects of controlled S and N deposition:

1) proportional decrease in N and S deposition,

2) a decrease of S deposition by 75%, and an additional decrease of N deposition until critical loads (CL) are attained. For comparison, CL as calculated by SMB are also listed in Table 1.

The Nashwaak Experimental Watershed Project

(NEWP, west-central New Brunswick 46°18°N, 67°02°W, 200 to 480 m above sea level). The uplands at this location support old-growth tolerant hardwoods which include sugar maple, beech, sugar maple and scattered yellow birch. Atmospheric deposition rates and critical loads calculated with ForSVA for NEWP are listed in Table 2 (Oja and Arp, 1994). The ForSVA projections for biomass do not show a decline under current atmospheric S and N deposition rates. This means that net primary production at NEWP is not affected by current rates of S and N deposition. Harvesting, however, imposes an additional stress. In fact, ForSVA simulations suggest a gradual decline in biomass production with conventional harvesting under current S and N deposition rates, and an accelerated decline with whole-tree harvesting. Apparently, whole tree harvesting leads to a non-recoverable rate of N and K removal. With ForSVA, critical S and N loads were calculated for two scenarios.

Table 1 *Critical deposition rates (in eq ha^{-1} yr^{-1}) ensuring sustainable biomass production at the Solling spruce site (scenarios 2-4) compared with present S and N deposition rates and the SMB critical load scenarios. Also shown are the simulated Al concentration and molar Al/Ca ratio in the soil solution, both for constant and S proportional base cation deposition.*

				Decrease in base deposition		No decrease in base deposition	
Scenario	SO_x eq/ha/yr	NO_y	NH_4	Al eq/m^3	Al/CA eq/eq	Al eq^3	Al/Ca eq/eq
1. present	3731	1411	1473	1.0-1.5	7-8	1.0-1.5	7-8
2. proportional	1231	466	492	0.4-0.5	4-5	0.2-0.3	1-2
3. realistic	1000	564	453	0.4-0.5	4-5	0.2-0.3	1-2
4. SMB	1193	432	455	0.4	4	0.2-0.3	1-2

Scenario 1:

Increase atmospheric S and N deposition rates until the ForSVA simulations show a declining trend in forest biomass; also, increase S and N deposition rates such that current proportions between S and N are maintained, adjust H ion deposition according to S and N deposition, but keep base cation deposition rates fixed at current levels.

Scenario 2:

Keep S deposition rates at current levels, but increase atmospheric N deposition rates until the critical loads are reached.

Northern Tolerant Hardwood sites Turkey Lakes (Ontario), Huntington Forest (New York State) and Harp Lake (Ontario)

The ForSVA model was used to calculate the critical S and N deposition loads for the three other Northern Tolerant Hardwood sites in North America (Table 3, Oja and Arp, 1995) The corresponding values for Al concentrations, and the ratio of Al to base equivalents in the soil leachates Huntington Forestare shown as well. The critical load effects criteria were set at 0.2 eq m^{-3} (New York State) and for the Al concentration, or 1.5 eq m^{-3} for the Al/base ratio in the forest soil leachates. It appears that the 0 biomass loss criterion (or biomass sustainability criterion) meets the other two effects criteria at all three sites.

Table 2 Atmospheric deposition rates at NEWP and the critical S and N deposition rates at this location; data for 1993 represent the averages at Holtville and Canterbury, New Brunswick.

	H_2O mm yr^{-1}	Ca^{2+}	Mg^{2+}	K^+	Na^+	Cl^- eq ha^{-1} yr^{-1}	NO_3^-	NH_4^+	SO_4^{2-}	H^+
Total deposition, 1972-81	1324	205	53	51	184	178	180	140	521	345
Total (estimate), 1993	1262	92	63	10	239	257	180	68	284	299
Critical loads; Scenario 1	1324	205	53	51	184	178	270	210	780	516
Critical loads; Scenario 2	1324	205	53	51	184	178	430	390	521	467

Table 3 Present deposition rates (Pr) and critical loads (CL) at Turkey Lakes (Ontario), Huntington Forest (New York State) and Harp Lake (Ontario) for SO_4^{2-} and NO_3^- deposition, assuming that the current ratio of nitrate to sulfate deposition is not changing. Also provided are corresponding deposition rates for NH_4^+, H^+, bivalent base cations and Al^{x+} concentrations, Al/Ca and Al/total bases (Ca+Mg+K+Na) ratios in the leachates.

Site	SO_4^{2-}	NO_3^-	NH_4^+ deposition eq ha^{-1} yr^{-1}	H^+	Ca^{2+}	Mg^{2+} eq m^{-3}	Al^{x+}	Al/Ca in soil percolate eq/eq	Al/bases
TL Pr	760	460	211	479	186	52			
CL	460	280	190	340	186	52	0.12	0.6	0.4
HL Pr	1350	454	270	990	342	51			
CL	670	230	160	500	240	38	0.02	0.1	0.1
HF Pr	488	417	150	622	102	21			
CL	440	370	130	550	102	21	0.10	1.4	0.8

Conclusion

With ForSVA, it is possible to evaluate the relation between different effects criteria and the associated critical loads for upland forest sites, to compare the results with actual field observations regarding the chemical composition of forest soil leachates (especially Al and base cation concentration, as well as Al/BC ratios), and examine all of this in terms of forest biomass production and nutrient cycling.

Acknowledgments

The work was supported by Env. Canada, and is part of the NSERC-supported CEA-CPPA-Forestry Canada - Env. Canada Partnership Project on S and N loads for forest soils.

References

Arp, P.A. and Oja, T. (1992) Acid Sulfate/Nitrate Loading of Forest Soils: Forest Biomass and Nutrient Cycling Modelling. In: Grennfelt & Thörnelöf (Eds.) Critical Loads for Nitrogen - a workshop report. NORD 1992: 41, Copenhagen, Denmark, pp. 307-358.

de Vries, W., Posch, M., Oja, T., van Oene, H., Kros, J., Warfvinge, P. and Arp, P.A. (1995) Modelling critical loads for the Solling spruce site. submitted to Ecol. Modell.

Oja, T. and Arp. P.A. (1994) ForSVA - a tool to analyze patterns in nutrient cycling as affected by atmospheric deposition: a maritime example. Environment Canada Occasional paper series No. 3 (in press).

Oja, T. and Arp, P.A. (1995) A Forest Soil Vegetation Atmosphere Model (ForSVA), II: Northern Hardwoods. submitted to Ecol. Modell.

5 Exceedances of Acidity and Nutrient Nitrogen Critical Loads

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Regionalising the Areas of Exceedance defined by a Critical Loads Function (CLF)

A CLF for an ecosystem simply defines protection and exceedance for all ratios of sulphur and nitrogen deposition in relation to acidity and nutrient nitrogen critical loads. However, when the load is exceeded it is possible to relate exceedance to excess sulphur, nitrogen, or both, by examination of the relative deposition loads of sulphur and nitrogen. In this exercise we have defined a number of regions, described by the CLF parameters, which help indicate reduction strategies for sulphur and nitrogen deposition.

The Regions of Exceedance

Figure 1 shows seven regions for a CLF. They may be summarised as follows:

7 Area of protection - critical loads not exceeded.

6 Area of "options" - either sulphur or nitrogen reductions can offer protection.

5 Area where nitrogen deposition should be reduced - reduction of sulphur gives no benefits.

4 Area where nitrogen is a major contributor to exceedance - nitrogen must be reduced to pro vide the possibility of options.

3 Area where sulphur and nitrogen are major contributors to exceedance - both must be reduced before there are options.

2 Area where sulphur is a major contributor to exceedance - sulphur must be reduced to provide the possibility of options.

1 Area where sulphur deposition should be reduced - reduction of nitrogen gives no benefits.

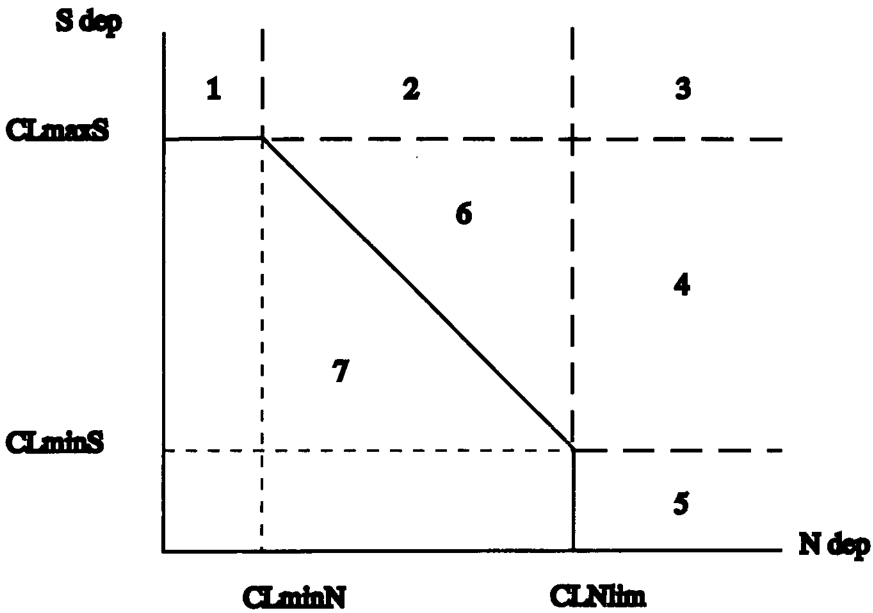
Mapping the Regions of Exceedance using Critical Loads and Deposition Data

Maps have been drawn for Great Britain using data provided by the Critical Loads Advisory Group (CLAG) which show the geographical distribution of different exceedance regions. They highlight the relative importance of sulphur and nitrogen controls for both current deposition levels and for those anticipated under the UNECE sulphur protocol in 2005 and 2010. For these maps:

(i) acidity critical loads were calculated using a modified mass balance equation;
(ii) nutrient nitrogen critical loads were estimated with the steady state equation;
(iii) nitrogen deposition was based upon current deposition measurements;
(iv) current sulphur deposition was based upon measurements;
(v) future sulphur deposition was modelled using a UK dispersion model HARM 7.2

Conclusions

Using current (1989-91) deposition data it is evident that large parts of Britain require reductions in both sulphur and nitrogen emissions or nitrogen emissions alone before any increase in protection will occur. With the predicted decreases in sulphur emissions and deposition, areas where nitrogen is a major contributor to exceedance appear over much of the country. These maps show that nitrogen emissions must be reduced to prevent exceedance and in some areas further reduction of sulphur would give no additional benefits.



CLNlim = minimum (CLmaxN, CLnutN)

Figure 1 **Regions of exceedance defined by a Critical Loads Function (CLF).**

6 Critical Loads for Nutrient Nitrogen for Soil-Vegetation Systems.

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Introduction

Members of the UK Critical Loads Advisory Group (CLAG) have calculated critical loads for nutrient nitrogen to produce maps for Great Britain. The results of three methods, based upon the conclusions from the Lokeberg workshop are described below. Two of these methods use the empirical approach and the other the steady state equation ("mass balance") for nitrogen saturation.

Empirical Approach using Species Distribution Data

The CLAG vegetation sub-group have identified plant species indicative of the vegetation types described at the Lökeberg workshop. The occurrence of these vegetation types have been defined by the National Vegetation Classification (NVC) species lists and species distribution maps from the Biological Records Centre (BRC). For each vegetation type, where the number of species recorded was above a selected minimum value, the appropriate critical load was applied. Since ranges of critical loads were agreed at Lökeberg, separate maps showing the lower, upper and mid-range value for that vegetation can be produced.

For national data, maps for individual vegetation types were combined by selecting the lowest critical load value for a grid square. Three combined maps were generated, one for each set of range values. These maps show significant differences resulting from the choice of the upper, lower or mid-range critical loads.

Empirical Approach using the ITE Land Cover Map

The ITE land cover map, derived from Landsat satellite imagery, identifies several types of natural and semi-natural vegetation. Studies at ITE Bangor have related these classes to nine of the vegetation types identified at the Lökeberg workshop. A map showing the critical load values for areas where natural vegetation predominates was originally produced for Wales. A national map has extended this to the include the rest of Great Britain. Again, the choice of critical load value from the ranges given in the Lökeberg table has a significant effect upon the final map.

Modelled Critical Loads for Nutrient Nitrogen

The steady state equation reported at the Lökeberg workshop has been used for calculating critical loads for nitrogen by the CLAG soils sub-group. Appropriate default values for the various nitrogen processes have been applied to produce data for three vegetation types. Maps have been produced for grassland, heathland and woodland, assuming that each vegetation type covers the whole country. Using the ITE land cover map to identify the presence of these vegetation types in different parts of the country and applying the appropriate critical load values, a national map has been generated.

Conclusions

The empirical maps generated using the ITE land cover map to identify areas of natural vegetation are less sensitive (i.e. higher critical loads) than those using NVC and BRC data to define the Lokeberg vegetation types. This is probably due to the latter maps being based upon the most sensitive vegetation type within an area whereas the former are based upon dominant land cover. For both sets of maps a marked change in sensitivity to nitrogen deposition results from the choice of value from the ranges listed at Lökeberg. It is important to choose an appropriate value in order to obtain a reliable critical loads map.

The national map produced using the mass balance approach tends to identify low critical loads for vegetation which is not harvested (e.g. acid grassland). Consequently it is more similar to the empirical map derived from NVC and BRC data with the minimum value of the Lokeberg ranges mapped.

The maps generated were based on the critical load values and methods reported at the Lökeberg workshop held in April 1992. All are likely to be revised following the discussions at the Grange workshop.

7 Nitrogen from Mountains to Fjords - Presentation of a Research Programme

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The Programme

To approach the problems connected to increased nitrogen inputs and to the leaching of nitrogen from Norwegian ecosystems, an inter-institutional five-year project: "Nitrogen from mountains to fjords" was established in 1992. The institutions involved are The Norwegian Institute for Water Research (NIVA), Center for Soil and Environmental Research (JORDFORSK), The Norwegian Forest Research Institute (NISK) and The Norwegian Institute for Air Research (NILU). The project considers both the atmospheric and terrestrial contributions of nitrogen to freshwaters and the sea, and the transformations of nitrogen in soil and freshwater.

Two river basins have been selected for this study, one with atmospheric nitrogen as anticipated dominant N-source (Bjerkreim), the other with agricultural activities as the dominant N-source (Auli). Typical water chemistry for the two river outlets is given in Table 1. Representative subcatchments have been selected. For the Bjerkreim river basin in southwestern Norway this includes 2 heathland areas, 2 forested areas (natural and planted forest) and an agricultural area.

Table 1. Typical water chemistry for the outlets of the two selected river basins; Auli (A) and Bjerkreim (B).

River	pH	Ca	Mg	Na	K	Cl	SO_4	NO_3	TotN	TotP
		mg L^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}	mg l^{-1}
A	7,03	20,0	6,2	17,2	4,3	27,5	21,0	4,30	4,80	0.030
B	5,60	1,1	0,7	3,8	0,3	6,8	2,8	0,37	0,41	0.003

Retention of Atmospheric-derived N in Terrestrial and Aquatic Ecosystems

Very small seasonal and between stations variations in both NO_3-N and total N concentration have been found in the upper two thirds of the 693 km^2 Bjerkreim river basin.

Mean area-weighted total N concentation of the runoff was 310 g l^{-1} N in 1992; 75 % as NO_3-N and 25 % as organic N. Specific N transport was 10 kg ha^{-1} yr^{-1} in 1992. Total N deposition (both wet and dry and about equal amounts of NO_3-N and NH_4-N) was 14 kg ha^{-1} yr^{-1} in 1992. The ratio between N transport and N deposition of 0.7 indicates very low N retention in the upper part of the river basin. N deposited directly on a total of 35 km^2 lake surface in the upper two thirds of the river basin was 50 metric tons in 1992 and contributed 11 % to the N transport.

There was insignificant N retention in the 240 m deep, ultra-oligotrophic Lake Ørsdalsvatn, the largest lake in the catchment. The calculation was based on the N deposition on the lake surface and data from the two inlet streams and the outlet stream. The insignificant N retention in Lake Ørsdalsvatn is probably representative for the other lakes in the upper parts as well and is supposed to be due to very low total P concentration (2-3 g l^{-1} total P). The other lakes are not as large and deep as Lake Ørsdalsvatn, but are also ultra-oligotrophic (low P).

An important part of the programme is the continous monitoring of NO_3 and NH_4 in subcatchments. Almost unchanged concentrations of these N species (100-115 g l^{-1} NO_3^-N and < 10 g l^{-1} NH_4^-N) from the remote heather catchment Øygard creek during a period of rapid changes in water flow in September - October 1994 was found.

These preliminary results indicate that the low retention of N in the remote parts of the Bjerkreim river basin is a result of high specific runoff (90 l s^{-1} km^{-2}) and exceedance of N uptake capacity both in the terrestrial and aquatic ecosystem.

8 Long Term Effects of Ammonium Sulphate on a Lowland Dry Heath in Southern Britain

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There is widespread concern that increased atmospheric deposition of nitrogen is causing changes in the species composition of natural and semi-natural ecosystems. Increased nitrogen deposition is thought to have been an important factor in the conversion of lowland dry heaths (dominated by *Calluna vulgaris*) to grassland which has occurred in the Netherlands and in the Breckland heaths of East Anglia. A critical load of 15-20 kg N ha^{-1} yr^{-1} has been suggested for the conversion of lowland heathland to grassland, on the basis of a model of nitrogen cycling in heathlands (Bobbink *et al.* 1992), but there is a lack of long-term field experiments at inputs around this proposed load.

In order to test whether model-derived critical loads are soundly based, experimental additions of low levels of ammonium sulphate to a nitrogen-poor dry heathland were started in 1989. The field site is part of the Thursley Common National Nature Reserve, 45 km SW of London. The study area consists of an even aged (18 year old), virtual monoculture of *Calluna* growing on a Lower Greensand podzol and receives an estimated background nitrogen deposition of 15-20 kg N ha^{-1} yr^{-1}. Experimental nitrogen additions of 7.7 or 15.4 kg ha^{-1} yr^{-1} therefore provide total deposition rates only slightly in excess of the proposed critical loads.

There are four replicate blocks, each with four plots receiving one of four treatments. All plots are sprayed with 15 litres of solution on 42 occasions each year. The treatments comprise a control which receives an artificial rain solution, a low treatment receiving an additional 7.7 kg N ha^{-1} yr^{-1}, a high treatment receiving 15.4 kg N ha^{-1} yr^{-1} and an alternating treatment which receives either the control or the high nitrogen additions, in alternate years. Shoot growth, flower production, cover repetition, litter production as well as shoot and soil chemistry are measured regularly using standard techniques. In addition, relative growth rates of heather beetle larvae (*Lochmaea suturalis*) were determined on shoots collected from each of the plots in June 1994.

The most obvious effect of the ammonium sulphate applications has been a large increase in flowering, with twice as many flowers in the high nitrogen treatment as in the control. A significant increase in flowering was also seen in the low treatment in 1994. Flower production in the alternating treatment strongly reflected additions of ammonium sulphate, with greatly reduced flowering in years when ammonium sulphate additions were suspended (1992 and 1994).

Shoot growth was also stimulated by ammonium sulphate application, with growth commencing earlier and shoots reaching greater final lengths in plots receiving additional nitrogen in each of the past 5 years. As might be expected with such a stimulation in shoot growth, there has also been a significant increase in the height and density of the *Calluna* canopy in 1993 and 1994. Litter production increased slightly in plots receiving ammonium sulphate in 1991-92 and 1992-93. In 1993-94, this effect was statistically significant, with more than twice the amount of litter produced in the high treatment (96.2 g m^{-2}) as in the control (47.6 g m^{-2}).

Shoot nitrogen concentrations have remained low throughout this experiment (0.84-0.90% dry weight). There has been a general trend towards increasing shoot nitrogen with ammonium sulphate application, although so far this has not been significant. The additional nitrogen may be largely used for the production of extra shoot growth and flowers, preventing much change in tissue concentrations. Similar results were found with a study of relative growth rates of heather beetle larvae reared on shoots from the different treatments. No significant treatment differences were found, although beetle growth rates tended to increase with the level of ammonium sulphate application, and were significantly correlated with shoot nitrogen content.

Current models of the response of dry *Calluna* heathland to these loads of nitrogen suggest that accumulation of higher tissue nitrogen levels will occur and be accompanied by heightened sensitivity to stresses such as drought, frost and heather beetle attack; this will ultimately lead to canopy breakdown and replacement by grassland. This is the basis of the current critical load of 15-20 kg N ha^{-1} yr^{-1} for lowland dry heathland. To date, the application of nitrogen at deposition rates only slightly in excess of this critical load over five years has provided clear evidence of a large positive effect on shoot growth, flowering, litter production and canopy density of *Calluna*. So far, there has not been a significant increase in shoot nitrogen content, although recent data suggest there are incipient effects on heather beetle performance that are related to shoot nitrogen content. The observation of these responses at the very low application rates used in this study do appear to support the current proposals for critical loads for nitrogen.

Since nitrogen deposition might be expected to have a number of soil-mediated effects, we are planning to carry out more detailed investigations of soil chemistry and plant root systems in the near future. It is hoped that continuation of this field experiment will allow us to establish whether increased nitrogen deposition is causing significant effects on stress sensitivity of *Calluna* at this site.

References

Bobbink, R., Boxman, D., Fremstad, E., Heil, G., Houdijk, A. and Roelofs, J. (1992) Critical l oads for nitrogen eutrophication of terrestrial and wetland ecosystems based upon changes in vegetation and fauna. In: Grennfelt & Thörnelöf, E. (Eds.). Critical loads for nitrogen. Report from a workshop held at Lökeberg, Sweden, 6-10 April 1992, pp 111-159. Nord (Mijorapport) 41, Nordic Council of Ministers, Copenhagen.

9 Investigating Methods to Evaluate Critical Loads for Nitrogen

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Abstract

The Stockholm Environment Institute is carrying out an exercise to investigate methods for the calculation of critical loads for nitrogen and is developing consistent datasets across Europe to map these critical loads, using GIS techniques. This work forms part of a wider project, sponsored by the UK DoE, that also includes the calculation of nitrogen emissions in Europe and costs of abatement measures.

Due to the uncertainties in deriving critical loads for nitrogen it was decided that more than one method should be used. The two methods being investigated are:
i) a sensitivity method;
ii) a nitrogen saturation method.

The sensitivity method is an extension of the empirical method to determine critical loads for nitrogen. The empirical critical loads have only been suggested for vegetation types for which there is experimental evidence (Bobbink *et al.* 1992). The sensitivity approach classifies vegetation types into categories that have similar reactions to nitrogen deposition. The classification is based upon the characterization of the controlling factors determining vegetation response. It would seem that the vegetation types most likely to respond to inputs of nitrogen are those with low nitrogen availability, and, generally, of low nutrient status. The empirical critical loads for some vegetation types may then be extended to other vegetation types classified as having similar sensitivity, but for which there is little experimental evidence.

The nitrogen saturation concept has been developed for some time (Nilsson and Grennfelt, 1988; Skeffington, 1988; Tamm, 1991) and is defined as the accumulation in the ecosystem that gives rise to deleterious effects, such as excess nitrate leaching and nutrient imbalance, leading to growth effects and interactions with pests and pathogens. The critical load based on the saturation approach therefore attempts to limit the input of nitrogen to levels which will prevent these effects. Therefore, the calculation method sums the nitrogen outputs in terms of removal from the system by harvesting biomass and by denitrification and acceptable leaching. An acceptable nitrogen accumulation rate is also defined. Each of these parameters is being estimated and mapped. The calculation, using two different methods will give an insight into the range of critical loads that might apply in different parts of Europe.

Results and Discussion

The poster presented at the workshop showed an initial classification of vegetation types, example maps of the land use database produced for this project and the reclassified sensitivity map shown for Scandinavia (Figure 1). Preliminary critical load ranges were suggested for the different sensitivity classes. Removal rates of nitrogen from logged forests were calculated across yield ranges for certain species in different countries of Europe, and the dominant tree species map for EU countries was presented. Example maps required for the site class assessment (for the allocation of nitrogen removal rates within countries) were shown. Maps showing the C:N ratio (Figure 2) and organic matter contents applied to soil types of the EU were also displayed. The project is on-going and will be completed in 12 months.

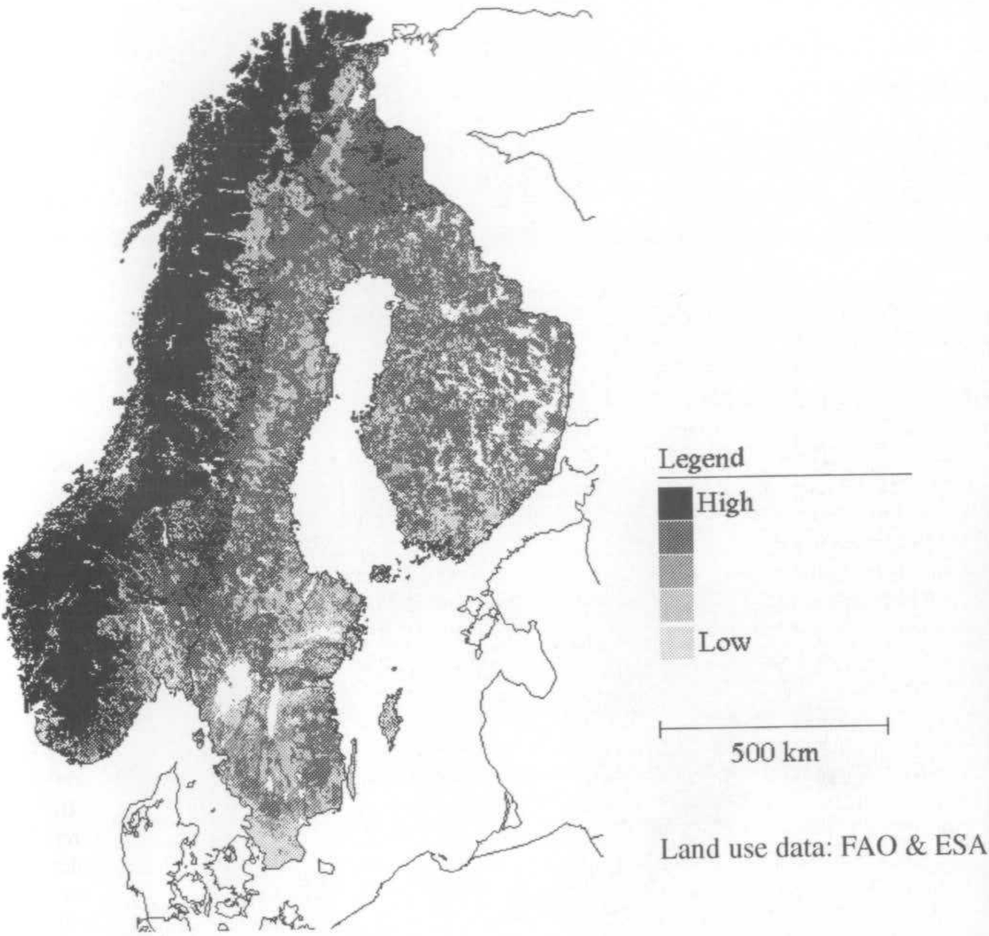

Figure 1 **A preliminary assessment of sensitivity of vegetation to nitrogen induced changes in composition shown for Nordic countries.**

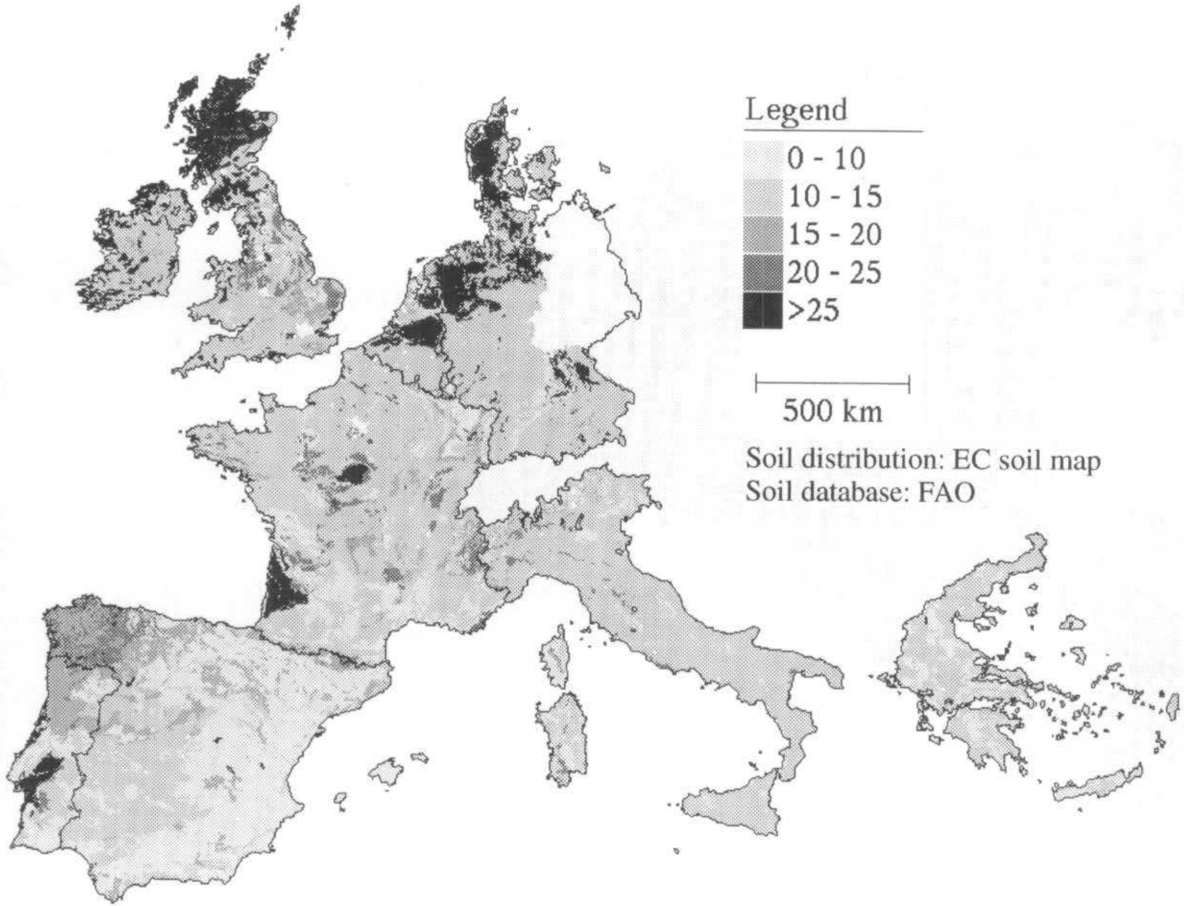

Figure 2 **A classification of soil types according to the C:N ratio in the A-horizon (Soil map source: the Digital Soil Map of the European Communities from CORINE).**

References

Bobbink, R., Boxman, D., Fremsted, 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 flora. In: *Critical Loads for Nitrogen* (ed. by P. Grennfelt and E. Thörnelöf), 111-60. Nordic Council of Ministers, Copenhagen.

Chadwick, M.J. and Kuylenstierna, J.C.I. (1991) The relative sensitivity of ecosystems in Europe to acidic depositions. A preliminary assessment of the sensitivity of aquaatic and terrestrial ecosystems. *Perspectives in Energy* **1**, 71-93.

Kuylenstierna, J.C.I. and Chadwicjk, M.J. (1992) The acidifying Input of Nitrogen Depositions to Ecosystems: A Preliminary Method to Allow Comparison to Critical Loads for Acidity. In: *Critical Loads for Nitrogen - a Workshop Report* (ed. by P. Grennfelt and E. Thörnelöf). Nordic Council of Ministers Report, Nord **1992:41**, 359-81.

Nilsson, J. and Grennfelt, P. (eds.) (1988) *Critical Loads for Sulphur and Nitrogen.* Miljörapport **1988:15**. Nordic Council of Ministers, Copenhagen.

Skeffington, R.A. (1988) Excess Nitrogen Deposition. *Environmental Pollution* **54** - Special Issue.

Tamm, C.O. (1991) *Nitrogen in Terrestrial Ecosystems. Questions of Productivity Vegetational Changes and Ecosystem Stability.* Ecological Studies **81**, Springer Verlag.

10 Critical S and N Loads and their Current Exceedances in Southern Ontario: Preliminary Results

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Introduction

This report presents results obtained from estimating how much S and N deposition can be tolerated by upland forest ecosystems without leading to non-acceptable levels of soil and water acidification and N eutrophication in Southern Ontario. Levels of atmospheric ion deposition pertaining to N and S are high in Southern Ontario where concerns about area-wide soil and water acidification and N eutrophication have been expressed for at least two decades. For upland forest systems, suggested tolerance criteria for NO_3 in soil solution are generally below 0.1 mmoles l^{-1} (Downing *et al.* 1993). The objective of this report is to evaluate the soil acidification and N eutrophication effects criteria for Southern Ontario, and to present the results.

Methods

Critical S and N load levels were obtained from existing information about soils, vegetation and atmospheric ion deposition for 12 locations across Southern Ontario (forest sites located within Provincial Parks or Conservation Areas). The evaluation was done by way of the Steady-State Mass Balance (SMB) approach (deVries 1991; Downing *et al.* 1993). This approach assumes that atmospherically deposited S and N compounds (SO_4, SO_2, NH_4, NO_3, NO_x) that are not taken up by the vegetation lead to soil acidification and/or N eutrophication. Locally, soil acidification is assumed to be modified by soil weathering and base cation uptake. Nutrient ions not taken up by the vegetation remain part of the soil complex or are subject to leaching. The whole process was evaluated for the steady state situation, i.e., the situation where the local forest stand structure is stable, and much of the on-site N retention depends on the ability of the tree vegetation to accumulate woody biomass. Relevant information about geological substrates (bedrock), soils, vegetation, weather, and atmospheric deposition was compiled from existing records (Table 1).

Results and Discussion

The results are shown in Table 1 by way of critical S and N loads and the related exceedances with respect to the [Al]/[BC] = 0.15 moles/moles and $[NO_3]$ = 0.1 m mole l^{-1} effects criteria. It appears that atmospheric S, N and Ca deposition rates vary across Southern Ontario, with highest deposition rates in the southwest corner (Wheatley), and the least deposition rates in the northeast corner (Wilberforce). The results show a clear north-south dichotomy for critical S and N loads and related exceedances, the latter being positive in the northern parts, and negative in the southern parts.

Results obtained from two Al/BC ratio criteria (ratios of Al/BC weathering rates and Al/BC soil solution concentrations) were similar to one another, and both criteria suggest exceedance of critical soil acidification levels in the northern parts for some of the assumed criterion values. For example, current S and N deposition rates are such that the critical level of [Al] = 0.2 mmoles l^{-1} (critical soil pH = 4) is not exceeded in any region. In addition, soil pH values may drop to below 4 in some instances when [Al]/[BC] is allowed to become equal to 1.5 region-wide. We note that high [BC] also means high [Al], which - in turn - means low soil pH. A prudent choice, therefore, would be to select the critical soil acidification effects criteria such that [Al] = 0.02 mmole l^{-1} (anticipated critical soil pH = 4.33), [Al]/[BC] = 0.15 and (or) Al/BC weathering rate ratios = 0.1. At these levels, all three effects criteria produced fairly similar results.

Table 1 *Average annual temperature precipitation, actual evapotranspiration rates, wet at mospheric deposition rates for N, S and base cations, soil weathering rates, critical soil acidification and N eutrophication loads, and their current exceedances, for 12 upland forest sites in Southern Ontario.*

	Temp.	Precip	AET[1]	N dep[2].	S dep[2].	BC dep[2].	Soil Weath[3].	Crit A load	Crit N load	S&N exc.	exc.
	°C	mm	mm					eq ha^{-1} yr^{-1}			
Wilberforce	4.7	824	414	458	452	131	130	388	497	521	-39
Bon Echo	4.8	904	475	575	569	194	163	474	558	670	17
Dorset	4.7	1040	512	660	583	170	215	547	639	696	21
Silent Lake	4.3	981	499	653	606	160	226	499	617	760	36
Mill Pond	4.8	920	501	647	635	378	418	987	555	295	92
Hilton Falls	5.5	914	476	843	844	796	873	1942	692	-254	151
Pinery	7.8	895	496	866	795	562	924	1840	485	-179	381
Sibbald's Point	7.0	882	467	681	622	433	951	1620	515	-317	166
Maple Keys	7.8	1029	536	942	809	431	1066	1738	615	13	327
Glen Mgt. Area	7.2	997	517	790	692	262	1107	1573	581	-91	209
Ferris	7.7	881	479	942	703	382	1630	2044	624	-399	318
Wheatley	8.0	918	515	805	842	475	2478	3220	860	-1573	-55

1: Estimated with a hydrological model (Arp and Yin, Can. J. For. Res., 1992).
2: Ontario Ministry of the Environment (1986). A. Neary (Ed.). ARB-93-86-Phyto.
3: From simple estimates involving soil mineralogy and clay content (deVries, 1991). Soil data from: Ontario Ministry of Environment and Energy. 1993. Ontario Soil Baseline Database.

Site	Location	Rock type	pH	Site	Location.	Rock type	pH
Wilberforce	Can. Shield, NE	Granite	4.7	Pinery	Lake Huron, W	Limestone	7.8
Bon Echo	Can. Shield, NE	Granite	4.8	Sibbald	Lake Simcoe, Ctr	Limestone	7.0
Dorset	Can. Shield, NW	Granite	4.7	Maple K.	Lake Huron, W	Limestone	7.8
Silent Lake	Can. Shield, NE	Granite	4.3	Glen Mgt	Georgian Bay, W	Shales	7.2
Mill Pond	Can. Shield, NE	Granite	4.8	Ferris	Lake Ontario, S	Limestone	7.7
Hilton Falls	Lake Ontario,Ctr	Shales	5.5	Wheatley	Lake Erie, SW	Limestone	8.0

Ctr: Centre; NE: northeast, etc. pH: subsoil pH. exc.: exceedance

To curb both soil acidification and potential N eutrophication, one should develop a strategy for controlling S and N deposition rates simultaneously. National and international conservation practices as well as improved air pollution emission standards and technologies would all help in systematic exceedance reductions. The need for N deposition control is particularly strong in the southern part of the study, in spite of the tentative conclusion that soil acidification is likely not a major concern for this area.

The results shown are preliminary for the following reasons: only 12 sites were considered; atmospheric deposition rates relate to wet fractions only; there was no allowance for denitrification; N immobilization or N fixation; soil weathering rates were based on the European soil weathering formulation; base cation deposition rates are assumed to be uncorrelated with S and N deposition; the area is treated as if uniformly forested. In fact, the southern portion of the region is highly fragmented, with forests broken up in small patch-sized woodlots.

In conclusion, forested areas in Southern Ontario appear to be affected by current rates of atmospheric S and N deposition. In the northern area, soil acidification exceedances are positive based on reasonable soil acidification effects criteria. In addition, some areas appear to be unable to absorb the incoming N deposition, leading to soil leachates with [NO3] >0.1 mmole l^{-1}.

References

de Vries, W. (1991) Methodologies for the assessment and mapping of critical loads and of the impact of abatement strategies on forest soils. DLO Winand Staring Centre, Wageningen (The Neth erlands), Report 46.

Downing, R. J., Hettelingh, J-P. and de Smet, P.A.M. (1993) Calculation and mapping of critical loads in Europe: Status Report 1993. RIVM Report N0. 259191003. Bilthoven, The Netherlands.

11 Estimates of Critical N Concentration/Load with respect to Eutrophication of Freshwater

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Introduction

Increased periphyton growth has been observed in the Nordic mountains. The cause may be elevated N deposition. N is also suspected to influence algal growth in lakes.

The work has been initiated and supported by the Swedish Environmental Protection Agency, through Dr. H. Staaf.

Materials and Methods

Data from a Lake survey in the winter of 1990, covering 986 statistically selected lakes analysed for N and P, about 170 Forest lakes, sampled at least three times per year, in late winter, late summer, and at "lowest pH-value", and data for running waters.

Nutrient Relationships

The effect of several nutrients on the biomass can be described as multiplicative (e.g. Smith (1982), Equation 1) or as a threshold effect.

Not only the biomass is affected by the nutrient conditions, both concentrations and load also influence the algal species composition. Several models for the estimation of chlorophyll *a* are based on P exclusively, few include N.

Smith (1982) published a model including total N and P.

$$\log c = 0.653 \log TP + 0.548 \log TN - 1.517 \quad (1)$$

where

c = chlorophyll, $\mu g\ l^{-1}$
TP = total P, $\mu g\ l^{-1}$
TN = total N, $\mu g\ l^{-1}$

An additional two equations will be evaluated in the near future.

Studies on Periphyton

During recent years increased occurrence of periphyton in natural waters has been reported. In Norway a nationwide survey (Lindstrøm, 1993) was made in 1992 showing that areas with increased growth coincide with those where the critical load for acid is exceeded. Increased N deposition has been recorded, but in addition, the summer temperatures were elevated and the deposition unusually high. Thus there are several possible explanations to the increased growth. In Sweden, Degerman *et al.* (1992) made a review of status and changes in Swedish montane waters and quoted several statements about increased presence of periphyton. They discuss various causes; diminished grazing by bottom fauna, pH preferences by the algae as well as increased leaching of N.

Evaluation of Swedish Water Chenistry Data

There is a large variation in the N:P quotients, which are highest during the winter and lowest during late summer (Fig. 1). The seasonal variation is more pronounced in the northern parts of Sweden, at Lilltjärnsbäcken in NW Jämtland (N63° 46'), and Lill-Fämtan in western Dalecarlia (N60° 50') the N:P quotients <7 during the vegetation period almost every year. As a contrast the quotient for the River Enningdalsälven (N58° 52') in a high deposition area is <7 only on two occasions.

N may thus be the most limiting substance during the summer, at least in northern waters.

Critical Concentration of Nitrogen Compounds of Water

Smith's equation (1) makes it possible to estimate the effect, as chlorophyll *a*, of various N concentrations while taking the P concentration into account. This value can be compared with:

- a scale of nutrient status (e.g. Vollenweider).
  chlorophyll a = 2.5 $\mu g\ l^{-1}$ is the limit between oligotrophy and mesotrophy.

- modified degree of perturbation (Swedish water quality criteria. SNV, 1991). The quality is classified as zero or insignificant perturbation when the chlorophyll *a* concentration is >1.5 times the natural. A prerequisite for the calculation of perturbation is that the background (natural) concentration for N can be estimated. Ahl (1994) calculates "the most likely upper limit for pristine nutrient condition" for nutrients based on the concentration of organic matter (CODMn)

$$ONp = 52.0 + 12.3\ CODMn \qquad (2)$$
$$TNp = 1.2\ ONp \qquad (3)$$

where

ONp = organic N, mg l^{-1}
TNp = total N, mg l^{-1}

TNp and measured total P were used for the estimation of the "natural" chlorophyll *a*.

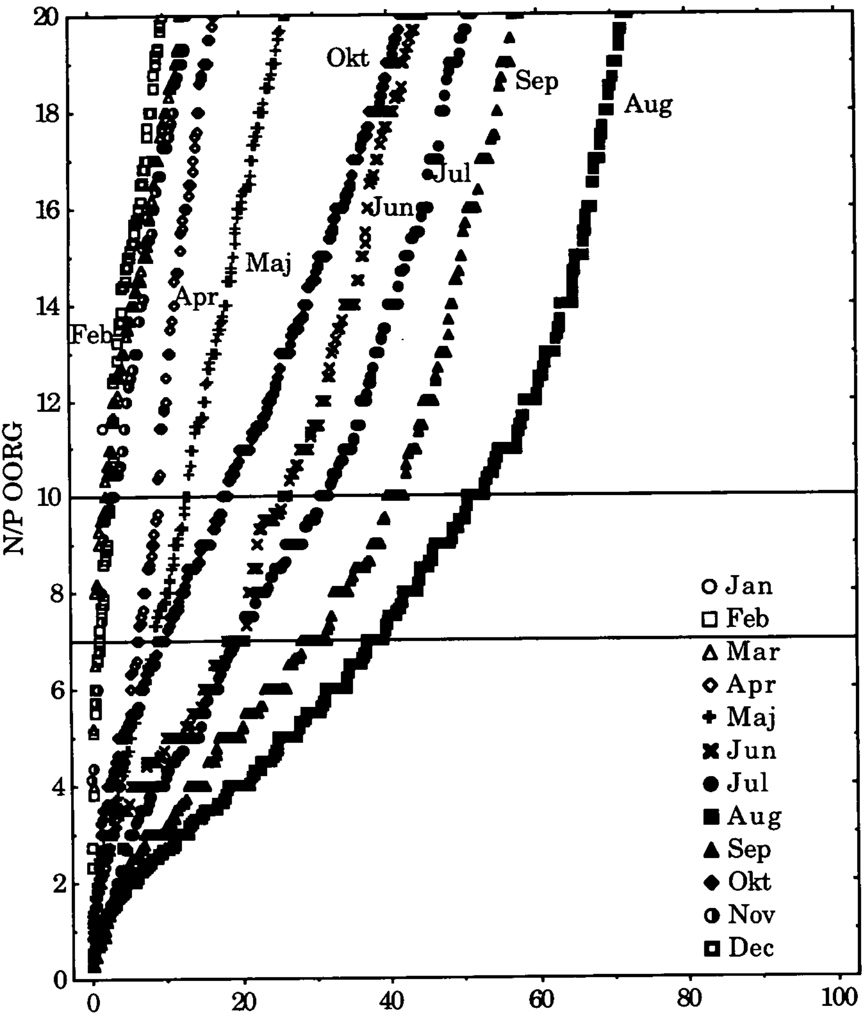

Figure 1 **Variation in N:P (inorg N/total P) for Swedish forest lakes**

The first alternative, using the Smith equation (1), has been applied to forest lakes in order to be able to quantify the possible effects of increased N.

For northern lakes the portion of observations indicating an oligotrophic state diminishes from 34% to only 2.2% when the increase is 400 μg l^{-1}. The precipitation in E Jämtland has an inorganic N concentration of about 300μg l^{-1}, thus such additions of N can influence the algal biomass.

In southern Sweden the concentrations in precipitation and surface water are elevated, therefore the calculations were instead made for reductions. The portion of observations which can be identified as oligothrophic is 12.5% under present conditions, it increases to 23% upon a diminution of the concentrations by 100 µg l^{-1}, and to as much as 57% if the diminution is allowed to be 400 µg l^{-1}. The latter value is about the same as the difference between the concentrations in deposition of S and N in Sweden.

The second alternative was used on the lake survey, and the results indicated that the limit for zero or insignificant perturbation is violated for about 58% of the nutrient poor lakes (TP >15 µg l^{-1}, n = 670). For lakes N of River Dalälven, the anthropogenic influence is estimated to affect 44% of the lakes (n = 359) studied, while as many as 74% of the southern lakes (n = 307) are affected. Data for forest lakes in Norrbotten county (n = 13) show that a change within the oligotrophic level on 10% of the occasions, allows for an additional N supply of only 15 µg l^{-1}.

Critical Load of Nitrogen Compounds

The N in deposition may be taken up by vegetation and become immobilised, but this may be neglected in mountainous areas or adjusted for using various models.

The critical load can be calculated from a critical concentration using a relevant value for runoff. Such calculations are usually made yearly, here the critical period is summer. This is adjusted for by taking the seasonal variation in N deposition into account.

The critical concentration for northern forest lakes was found to be 110 µg l^{-1} if clear perturbation is allowed for 5% of the measurements. In areas with no N eliminating processes this corresponds to a critical load ($CL(N)_{max}$) of 0.19 g m^{-2} year. These values are lower than those found for terrestrial biotopes.

Conclusions

Inorganic N is limiting for algae in certain oligotrophic waters, especially northern and mountainous areas. An estimate of the critical concentration in water can be made using the Smith equation for calculation of chlorophyll *a* and comparison with a perturbation factor.

Background N concentration according to Ahl can be used in high deposition areas.

Based on the 1990 lake survey 58% of the Swedish lakes are influenced with respect to eutrophication due to anthropogenic N deposition.

The critical concentration and load for oligotrophic lakes seems to be lower than those for many terrestrial biotopes.

References

Ahl, T. (1994) Regression statistics as a tool to evaluate excess (anthropogenic) phosphorus, nitrogen and organic matter in classification of Swedish freshwater quality. *Water Air Soil Pollut.* 74 :169-187.

Degerman, E., Engblom, E., Lingdell, P.-E., Melin, E. and Olofsson, E. (1992) Försurning i fjällen. *Inf. Sôtvattenslab. Drottningholm.* 1:1992

Lindstrøm, E. A. (1993) *Økende grønske i norske vassdrag. Resultat av en spørreundersøkelse.* (Increased growth in Norwegian running waters. In Norwegian. Results from interviews.). Projektnr E-92432, NIVA Rapport 2859.

SNV (1991) *Quality criteria for lakes and watercourses.* Swedish Environmental Protection Agency informs

Smith, V. (1982) The nitrogen and phosphorus dependence of algal biomass in lakes: An empirical and theoretical analysis. *Limnol. Oceanogr.* 27:1101c1112.

Vollenweider, R. A. and Kerekes, J. (1980) The loading concept as a basis for controlling eutrophication philosophy and preliminary results of the OECD Programme on eutrophication. *Prog. Water Technol.* 12:5-18

12 First Maps of Critical Loads of Nitrogen for Switzerland

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Critical Loads maps for nutrient nitrogen have been produced by applying both the Nitrogen Steady State Mass Balance (SMB) approach and the empirical approach, as proposed at the UNECE Workshop in Lökeberg in April 1992. First drafts of these maps were included in the Status Report 1993 of the Coordination Center for Effects. In addition, maps of Present Loads of nitrogen and corresponding exceedance maps have been computed on a 1x1 km^2 grid.

SMB Approach

The SMB approach is used for forests only. The following equation was applied at 11,863 receptor points [Units: kg N ha^{-1} yr^{-1}]:

$$CL_{nut} = N_u + N_i + N_{l(crit)} / (1 - f_{de})$$

where:

N_u	= 0.7 - 7	Uptake, derived from the long-term harvesting rate
N_i	= 2 - 3	Immobilization, depending on soil type
$N_{l(crit)}$	= 2 - 5	Critical leaching, depending on forest type and altitude
f_{de}	= 0.2 - 0.8	Denitrification rate, is determined according to the drainage conditions of the soil

The calculated critical load values are in the range of 5-25 kg N ha^{-1} yr^{-1}.

Empirical approach

Table 1 *Natural and semi-natural ecosystems used for the empirical approach, with critical load values assigned according to the ranges proposed at Lökeberg 1992 [kg N ha^{-1} yr^{-1}].*

ecosystems (vegetation communities)	critical load
Molinio-Pinion, Ononido-Pinion, Cytiso-Pinion	17
Quercion robori-petraeae, Quercion pubesc.-petraeae, Orno-Ostryon	15
Erico-Mugion, Erico-Pinion, Calluno-Pinion	10
Mesobromion, Andropogonetum gryllii	19
Molinion	25
Seslerio-Bromion, Festucion spadiceae, Caricion ferrugineae	12
Stipo-Poion xerophilae, Oxytropido-Elynion, Seslerion coeruleae	10
Littorellion	7
Scheuchzerietalia, Caricion fuscae, Caricion davallianae	25
Sphagnion fusci	7

The ecosystem types in table 1 are mapped on the basis of a national vegetation inventory with a spatial resolution of 1 x 1 km^2. When more than one ecosystem type exists within a grid-cell the most sensitive critical load value is chosen.

Present Loads Present loads are calculated as

wet deposition:	NO_3^- and NH_4^+
dry aerosol deposition:	NO_3^- and NH_4^+
dry gas deposition:	NO_2, HNO_3 and NH_3

Table 2 *Total present loads of NH_y and NO_y on forests and on open land in Switzerland for 1986-1990, total loads modelled by EMEP for 1986-1990 as well as total emissions from Switzerland for 1990.*

Item	Area [km^2]	NH_y-N [kt yr^{-1}]		NO_y-N [kt yr^{-1}]	Total N [kt yr^{-1}]
		NH_3-N	NH_4-N		
load on forest	11863	13.6	11.9	14.4	39.9
load on open land	30393	9.9	21.1	23.6	54.6
total loan on Switzerland (1 x 1 km^2 grid)	42256	56.5		38.0	94.5
total EMEP load (150 x 150 km^2 grid)	42256	51.6		32.6	84.2
total emissions from Switzerland	42256	52.5		56.0	108.2

The total national deposition values calculated on the basis of deposition measurements, concentration measurements, altitude dependencies, etc. correspond quite well with the total N depositions modelled by EMEP. But the highly resolved national data show a large variation from 6 to 60 kg N ha^{-1} yr^{-1} with an average of 22.3.

Conclusions

Two different maps of critical loads of N were produced by applying the SMB method and the empirical method as described in the UN ECE Manual on Mapping Critical Levels/Loads, which includes the findings of the Lökeberg Workshop. If the SMB method is applied with the upper bounds of the proposed ranges for immobilization, leaching and denitrification, the results for Switzerland show a cumulative frequency distribution of critical loads similar to the empirical approach. Exceedances of the critical loads were calculated by overlaying the critical load maps with a present load map on a 1 x 1 km^2 basis. The largest exceedances are found in the lower regions, where NO_2 and NH_3 concentrations are higher than in remote alpine regions.

13 An Empirical Statistical Model for the Prediction of Surface Water Critical Loads at the Catchment Scale

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The critical loads approach to surface water acidification is geared towards targeting emission control strategies at regional, national and international levels. Critical load maps have been developed at a resolution of 10 km^2 based on a sample taken from a single point. Catchment management decisions taken using data at this resolution will inevitably fail to account for the variations that exist at higher resolutions. A method is required, therefore, whereby critical loads can be estimated for given catchments without the need for costly field surveys.

Work is currently underway to produce an empirical statistical model which will enable key water chemistry determinands, and thus critical loads, to be predicted using a variety of readily available catchment parameters.

Preliminary analysis was undertaken using water chemistry and secondary catchment parameters for 1082 sites throughout UK. At this stage, catchment specific secondary data were not used as secondary data were not available in this form. Analysis was based on 1 km^2 resolution digital maps from ITE Monks Wood. Although this means that the 'catchment' variables do not relate directly to the contributing area, but to the 1 km grid square that contains the site, at this preliminary stage the primary concern was to assess the potental for more detailed analysis. Additionally, the secondary data variables were surrogates, in the absence of more readily available data, for soils, geology, and land use. These comprised soil critical load (based on the critical load of the dominant soil), site sensitivity (derived from a combination of soil type, geology and land use) and land classification. Additional data relating to sampling sites included S and N deposition, altitude and rainfall.

These data have been analysed using multivariate statistical techniques, redundancy analysis and ANOVA. The results are presented in Tables 1 and 2.

Table 1 Summarised Redundancy analysis results

Chemistry determinand	Variance explained by all catchment variables
All determinands	45%
Calcium concentration	64%
Nitrate concentration	32%

Table 2 Summarised ANOVA results

Chemistry determinand	Catchment variable	Variance explained by catchment variable
Calcium	Site sensitivity	45%
	Land classification	43%
	Soil critical load	40%
Nitrate	Site sensitivity	20%
	Land classification	23%
	Soil critical load	15%

Both analyses indicate that calcium concentrations are strongly related to the catchment parameters used here. From these relationships, given the coarseness of the data resolution, it is suggested that a catchment specific approach using more detailed data may yield a model which will, within certain confidence limits, enable the sulphur critical load at any site to be predicted using readily available catchment data. The next stage is to characterise catchments according to soil, geology, land use, topographical and geographical attributes at varying spatial resolutions. These attributes will then be used to calibrate the predictive model. A comparison can be made of the relative success of the model at varying data resolutions.

The relationships involving nitrate are much weaker. This reflects the complexity of the catchment/surface water process response system for N. Further work will be necessary to incorporate N into the model thus enabling the prediction of total acidity. To ensure the widespread applicability of the model it will be necessary to include a predictor variable which approximates the N term in the critical load for total acidity.

14 Calculating and Mapping of Critical Loads of Nitrogen: A case study for Poland

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Background

Studies on calculating and mapping of critical loads for acidification of natural ecosystems in Poland started in late 1990. This studies are performed by the Polish National Focal Center (NFC), a subsidiary body to the UN/ECE Coordination Center for Effects (CCE). The NFC has been established at the Institute for Ecology of Industrial Areas in Katowice. In the years 1991 - 1993 the National Focal Center has produced and submitted to the CCE critical loads maps of acidity and sulphur for forest soils and surface waters ecosystems. These maps have been integrated by the CCE into the European critical loads maps and formed a scientific basis for the negotiations of the Second Sulphur Protocol.

Currently the action of the CCE and NFCs is focused on ecological impact assessment of deposition of oxidized and reduced nitrogen species. According to the workplan established by the UN/ECE Working Group on Effects, by the end of 1994 preliminary European maps of critical loads of nitrogen are expected to be completed. On account of this the Polish National Focal Center produced a set of maps based on national data. These maps, presenting the necessary input data and critical loads and exceedances of acidifying and nutrifying nitrogen, together with the relevant data bases, have been sent to CCE, which is responsible for production of the European maps.

Calculations of critical loads of nitrogen for Poland have been based on commonly agreed methodology published in ìCalculation and Mapping of Critical Loads in Europeî, (Downing†*et al.* 1993), prepared by Coordination Center for Effects. The major input of the Polish National Focal Center into this calculation was the acquisition and adoption of the detailed, national data concerning nitrogen; uptake, immobilization, denitrification as well as nitrogen deposition data. All these calculations were performed for forest soils, observing the distinction between coniferous and deciduous forests.

To calculate nitrogen uptake, immobilization and denitrification coefficients, national data on forests age, branch to steams ratio and annual growth ratio were used. All these data were obtained from the Forest Management and Geodesy Office. In addition, concentrations of main elements in forest trees were taken from Mapping Critical Loads for Europe (Hettelingh *et al.*, 1991). In calculation of critical loads of nutrient nitrogen the dynamic denitrification fraction equation was applied to estimate the denitrification coefficient (Downing *et al.* 1993).

Results and Discussion

The resultant maps of all major input data and critical loads of acidifying and nutrifying nitrogen display the sensitivity of different Polish forest ecosystem types to total nitrogen deposition.

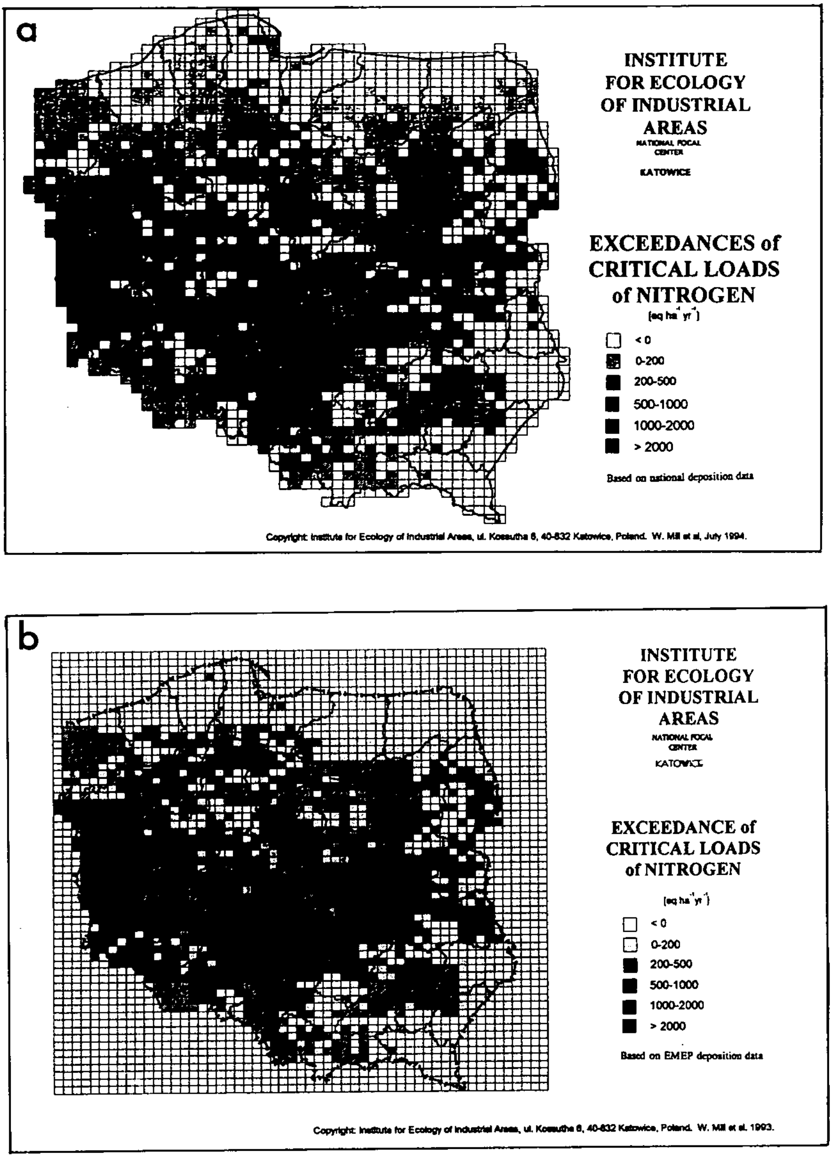

Figure 1 **Exceedance of critical loads of nitrogen in Poland in (a) 1994 and (b) 1993.**

For calculation of exceedances of the Critical Loads both EMEP and national deposition data were used. National deposition data were obtained from Warsaw University of Technology. Values of both these data are well correlated, however usage of these two spatial resolutions i.e. 150 km x 150 km and 30 km x 30 km respectively, show big differences in spatial distribution of calculated exceedances values, which are clearly reflected on the produced maps (Figure 1a, 1b). The relatively large size of EMEP grid cells results in the highest values of exceedances being spread over a 30 per cent larger area of the country, than when Polish deposition data are applied. It leads to a notable conclusion that there is a general need for implementation of as detailed as possible, national input data for all critical loads calculation within all parties to the Convention on Long-Range Transboundary Air Pollution.

References

Downing, R.J., Hettelingh, J-P. and de Smet P.A.M. (1993) Calculation and Mapping of Critical Loads in Europe. Status Report 1993. Coordination Centre for Effects. National Institute for Public Health and Environmental Protection, RIVM report no. 259101003, Bilthoven, The Netherlands.

Hettelingh, J-P. Downing, R.J. and de Smet, P.A.M. (1991) Mapping Critical Loads for Europe. CCE Technical Report. Coordination Centre for Effects. National Institute for Public Health and Environmental Protection, RIVM report no. 259101001, Bilthoven, The Netherlands.

15 Critical Loads of Nutrient Nitrogen for various Ecosystems of Russia

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On the basis of modified simple steady state mass-balance equations, the critical loads for nutrient and acidifying nitrogen as well as for sulphur and acidity have been calculated for various ecosystems of European and Asian parts of Russia. Due to the huge dimensions of the given area, all calculation and mapping procedures have been carried out using a geographical information system (GIS) with elements of a simplified expert-modelling system. The initial information consisted of geological, geochemical, geobotanic, landscape, soil, hydrochemical, biogeochemical and hydrological regionalization. In addition, for every elemental tax on the main links of biogeochemical cycles of N, S and **base cations** have been characterised quantitatively on the basis of available case studies. The grid cells were 1 grid x 1 grid (1.11 degree x 1.11 degree) for the Asian part (including the whole area of former USSR) and 1 grad x 0.5 grad for the European part of Russia.

The algorithm for computer calculations of the critical load for nutrient nitrogen ($CL_{ntr(N)}$) included the following equation:

$$CL_{ntr(N)} = {}^*N_u + {}^*N_i + {}^*N_{de} + {}^*N_{l(crit)} \tag{1}$$

where * means that each of the terms refers to the value at the actual total atmospheric deposition at a site. N_u and N_i are permissible nitrogen uptake by biomass and soil immobilization, N_{de} is permissible denitrif-ication and $N_{l(crit)}$ is permissible critical nitrate leaching.

Permissible atmospheric nitrogen uptake (${}^*N_u$) was given as:

$${}^*Nu = N_{upt} - N_u \tag{2}$$

where N_{upt} is annual accumulation of N in biomass and N_u is annual uptake of N from soil. N_{upt} was calculated accounting for the coefficient of annual biogeochemical turnover (C_b), the values of which varied from <0.1 up to >25. Annual N_u was calculated on a basis of nitrogen mineralizing capacity (NMC) of soils, which was determined experimentally or calculated using regression equations dependent on soil C:N ratios (Bashkin et al., 1993):

$$N_u = (NMC - N_i - N_{de})C_t \tag{3}$$

where

$N_i = 0.15$ NMC, if C:N<10
$N_i = 0.25$ NMC, if 10<C:N<14
$N_i = 0.30$ NMC, if 14<C:N<20
$N_i = 0.35$ NMC, if C:N>20
$N_{de} = 0.145$ NMC+6.477, if NMC>60 kg/ha/yr
$N_{de} = 0.145$ NMC+0.900, if NMC<10 kg/ha/yr
$N_{de} = 0.145$ NMC+0.605, if 10<NMC<60 kg/ha/yr

Permissible immobilization of atmospheric N deposition ($^*N_i$) was found as:

$$^*N_i = [(0.2\ NH_4 + 0.1\ NO_3)/C_b]C_t, \text{ if } C{:}N<10 \tag{4a}$$
$$^*N_i = [(0.3\ NH_4 + 0.2\ NO_3)/C_b]C_t, \text{ if } 10<C{:}N<14 \tag{4b}$$
$$^*N_i = [(0.35\ NH_4.0.25\ NO_3)/C_b]C_t, \text{ if } 14<C{:}N<20 \tag{4c}$$
$$^*N_i = [(0.4\ NH_4 + 0.3/C_b]C_t, \text{ if } C{:}N<20 \tag{4d}$$

where C_t is a hydrochemical coefficient expressed as the part of the year with mean daily temperature >5°C

Permissible denitrification from atmospheric deposition was found as ($^*N_{de}$):

$$^*N_{de} = (N_{de}/NMC)N_{td}\ C_t, \tag{5}$$

where N_{de}/NMC is the denitrification fraction, which depends on many features of soils and is calculated on a basis of experimental data and N_{td} equals total nitrogen deposition.

Finally, permissible critical leaching of atmospheric nitrogen ($^*N_{lcrit}$) was given as:

$$^*N_{l(crit)} = Q\ C_{Ncrit} \tag{6}$$

with Q being the annual surplus of precipitation (run off) and C_{Ncrit} the permissible nitrogen concentration in surface waters.

Critical loads for sulphur and acidity as well as exceedances for all studied parameters were calculated on the basis of the Mapping Manual (Task Force on Mapping 1993).

Using the above approaches it has been shown that, due to a permanent deficit of nitrogen as a nutrient, the critical loads for nitrogen in the majority of ecosystems are >500 eq $ha^{-1}\ yr^{-1}$. Only for pine forest ecosystems on sandy soils and oligotrophic tundra lakes are these values less than 200 eq $ha^{-1}\ yr^{-1}$. The biggest values (>2000 eq/ha/yr) have been calculated for Far East forests and Middle Asian dry steppe and desert areas, due to very rapid biogeochemical turnover of nitrogen in these ecosystems.

For most Russian ecosystems critical loads for sulphur are <500 eq $ha^{-1}\ yr^{-1}$ and only in calcerous soils of Middle Asia and Central Yakutia are the values >2000 eq $ha^{-1}\ yr^{-1}$. Consequently, taking into account acidifying effect of S and N depositions, ecosystems of middle Asia are **not sensitive** to acidification (CL values >2000 eq $ha^{-1}\ yr^{-1}$). Moderate **sensitivity** can be shown for ecosystems of boreal and sub-boreal north humid forests of European Russia, the Far East and South Siberia. In contrast, most of the area situated north of 60 grad. latitude and east of the River Enisey has the lowest values (<200 eq $ha^{-1}\ yr^{-1}$). These calculations have been used as the basis of various abatement strategy scenarios.

References

Bashkin V. N., Kozlov M.Ya. and Snakin V.V. (1993) Calculation and mapping of critical loads in Europe: Statur report 1993. CCE,RIVM, The Netherlands, p.102-112.

Task Force on Mapping (1993) Mapping critical levels/loads. Report 25/93. Umweltbundesamt, Bismarckplatz 1, Berlin.

16 A Provisional Empirical Nitrogen Critical Load Map for Terrestrial Ecosystems in Wales

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Ecosystem data were taken from a version of the ITE digital land cover map which records twenty five land cover types, derived from satellite data, giving the dominant land cover type for each twenty five metre cell of the British National Grid. For the purposes of this initial study, modal data at one kilometre resolution were used. The nitrogen critical loads for selected terrestrial ecosystems were taken from the classification developed at the Lokeberg workshop (Hettlingh *et al.* 1992). Nitrogen critical loads were assigned to ITE land cover types based on the nearest match between ITE land cover type and ecosystem descriptions. Because the land cover types are essentially broad structural categories and the ecosystem types for which critical loads have been derived are relatively narrow functional classes, the cross matching is fairly subjective. Alternative solutions could be considered, but, as a first attempt, critical loads were assigned as a mean of the reported range for each ecosystem in kgN ha^{-1} yr^{-1}, as shown in Table 1. The resulting map is shown in Figure 1.

Table 1 Land cover types and nitrogen critical loads.

Land cover value	Land cover description	Critical load	Ecosystem
5	Grass heath	13.5	Species-rich lowland heaths, acid grassland
7	Meadow/verge/semi-natural grassland	22.5	Calcareous species-rich grassland. Neutral-acid species-rich grassland
8	Marsh / rough grass	27.5	Mesotrophic fens
9	Moorland grass	12.5	Montane-sub alpine grassland
13	Dense shrub heath	18.5	Lowland wet & dry heath
15	Deciduous woodland	17.5	Acidic (managed) deciduous forest
16	Coniferous woodland	17.5	Acidic (managed) coniferous forest
25	Open shrub heath	18.5	Lowland dry & wet heath

Deposition data (wet + dry) for oxidised nitrogen (NO_x) and reduced nitrogen (NH_y) were imported as separate grid coverages, at twenty kilometre resolution. In order to produce the exceedance maps at the same resolution as the nitrogen critical load grid coverage, the deposition grid coverages were resampled to a one kilometre resolution. This meant that each component one kilometre cell was ascribed the same value as the original full twenty kilometre square and leads to the curious appearance of the exceedance maps (eg Figure 2). A total inorganic nitrogen deposition grid coverage was also produced by summing the NO_x and NH_y deposition grid coverages.

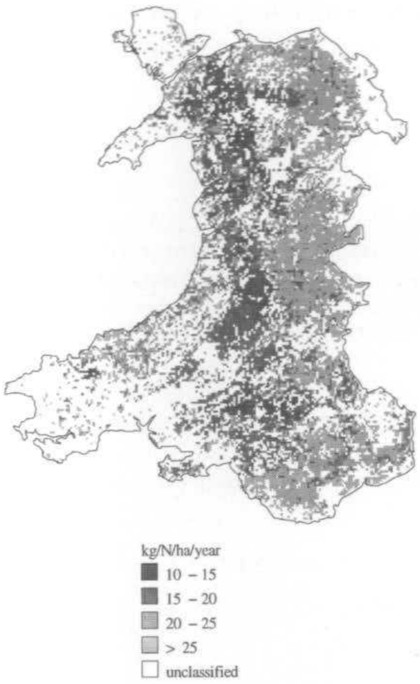

Figure 1 **Provisional Empirical Critical loads for nitrogen for terrestrial ecosystems**

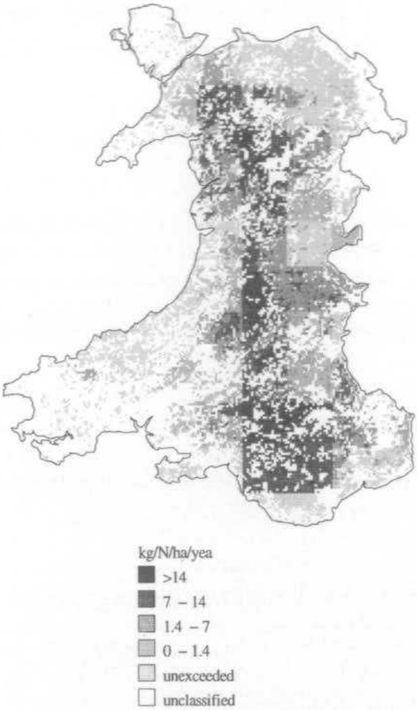

Figure 2 **Exceedance of Provisional Critical Loads for Nitrogen Total N deposition for terrestrial ecosystems**

Reference

Hettelingh, J-P, Posch, M., de Vries, W., Bull, K. and Sverdrup, H.U. (1992) Guidelines for the computation and mapping of nitrogen critical loads and exceedances in Europe. In: Critical Loads for Nitrogen - A workshop report, Eds. P Grennfelt & E Thornelof, Nord 1992:41, Nordic Council of Ministers, Copenhagen.

17 Critical Loads for Nitrogen to Avoid Eutrophication: Assessment of the Mass Balance Approach using the Aber Site, N.Wales.

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The nitrogen critical load to avoid eutrophication has been calculated for a 32 year old stand of Sitka spruce in N.Wales which has been intensively studied for 4 years. Default and measured values for the mass balance equation have been used to calculate a range of probable nitrogen critical load values which have then been compared to the range estimated from methods other than the mass balance approach.

The stand is planted on stagnopodzol soil and receives c. 16 kg N ha^{-1} yr^{-1} in throughfall + stemflow. Estimate of total inputs including dry deposition are c. 25kg N ha^{-1} yr^{-1} (D. Fowler, Pers. Comm.). There are indications that this site is nitrogen saturated as leaching losses are *ca.* 9 kg N ha^{-1} yr^{-1}. In addition, experimental addition of nitrate results in immediate and equivalent increases in nitrate leaching (Emmett *et al.*, 1995).

The mass balance equation used to calculate the nitrogen critical load CL(N) is the one defined in Grennfelt and Th^rnel^f (1992).

$$CL(N) = N_{up} + N_{de} + N_i + N_{l(crit)}$$

Nitrogen uptake (N_{up})

N_{up} was calculated using either (i) default values for nutrient to nitrogen ratios for bolewood, (ii) nutrient to nitrogen ratios recorded in the Aber stand (Table 1) or (iii) removal of N in present-day bolewood.

Table 1 *Nutrient to nitrogen ratios in Sitka spruce at Aber and default values.*

Ratio (%)	*Picea sitchensis*[1] (Bong(Carr.)	Default values[2]
Ca:N	0.80	1.2 - 1.5
Mg:N	0.30	0.2
K:N	0.95	0.7
P:N	0.13	0.08 - 0.1

[1] determined from five harvested trees at the Aber site.
[2] from Grennfelt and Th^rnel^f (1992).

For approaches (i) and (ii), weathering of nutrients was calculated using the PROFILE model to be 15, 124 and 31 eq ha^{-1} yr^{-1} for calcium, magnesium and potassium respectively. Atmospheric inputs of phosphorus were estimated from bulk precipitation. For base cations, inputs in bulk precipitation were multiplied by a scaling factor to account for aerosol and dust inputs. This scaling factor was calculated from the ratio of inputs of sodium in throughfall + stemflow to that in bulk precipitation. Resulting N_{up} values using the above three approaches were 2.10, 1.10 and 3.80 kg N ha^{-1} yr^{-1}. The low values of N_{up} using the nutrient to nitrogen ratio methods were a result of insufficient present-day inputs of phosphorus to ensure sustainable removal of biomass without depletion of the soil store. However, fertiliser application of phosphorus and potassium is frequently used in UK forestry and will ensure continued production. Assuming application of fertiliser will be carried out, N_{up} estimated from nitrogen removal in bolewood is lower than N_{up} calculated using calcium or magnesium to nitrogen ratios. As the minimum N_{up} calculated should be applied, approach (iii) should be used in CL(N) calculations at this site.

Denitrification (N_{de})

Denitrification at critical load was estimated using three approaches outlined by Grennfelt and Th^rnel^f (1992) (i) default values for aerated soils, (ii) Ineson and Sverdrup (1992) and (iii) de Vries *et al.* (1992). This resulted in estimates of 1, 2.4 and 5.2 kg N ha^{-1} yr^{-1} respectively. The moisture factor (f_m) in approach (ii) was set to 0.5 for moist soil. Using the de Vries *et al.* approach, f_{de} was set to 0.5 for sand with gleyic features. Studies in the field indicate that the lower end of this range (1-2 kgN ha^{-1} yr^{-1}) may be more applicable to the site.

Nitrogen immobilisation (N_i) and Nitrogen leaching (N_l)

The default range for N_i is 0 - 3 kg N ha^{-1} yr^{-1}. If N_i is estimated using the total N in soil store and years since last glaciation (11 500 yrs), a value of 2.1 kgN ha^{-1} yr^{-1} is computed.

Critical N leaching has been set to 4 kg N ha^{-1} yr^{-1} (default range 2 - 4 kg N ha^{-1} yr^{-1}) as this is similar to acid grassland leaching losses in the uplands of Wales.

Critical Load for Nitrogen (CL(N))

Comparison of CL(N) using the above values provides a range of values from 7.6 kg N ha^{-1} yr^{-1} using the mid-range default values to 12.3 kg N ha^{-1} yr^{-1} using nitrogen removal in bolewood for N_{up}, 2.1 kg N ha^{-1} yr^{-1} for N_i, 4 kg N ha^{-1} yr^{-1} for N_l and the Ineson and Sverdrup equation to calculate denitrification (2.4 kg N ha^{-1} yr^{-1}). The CL(N) to avoid eutrophication in managed acidic coniferous forest has been estimated by methods other than the steady state mass balance to be 15 - 20 kg N ha^{-1} yr^{-1} (Hettelingh *et al.*, 1992). These values are greater than those calculated here, however N_l in particular may be underestimated in the mass balance approach. Both ranges are below present-day inputs to Aber and thus are in agreement with the experimental data which indicates that this site is nitrogen saturated (Emmett *et al.*, 1995).

References

Emmett, B A., Brittain, A., Hughes, S., Görres, J., Kennedy, V., Norris, D., Rafarel, R., Reynolds, B. and Stevens, P. (1995). Nitrogen additions ($NaNO_3$ and NH_4NO_3) at Aber forest, Wales: I. Re sponse of throughfall and soil water chemistry. Forest Ecology and Management 71:45-59.

Grennfelt, P. and Thörnelöf, E. (1992). Critical Loads for Nitrogen - a report from a workshop held at Lokeberg, Sweden, 6-10 April 1992, Nord 1992:41, pp. 17.

Hettelingh, J., Posch, M., de Vries, W., Bull, K. and Sverdrup, H.U. (1992). Guidelines for the computation and mapping of nitrogen critical loads and exceedances in Europe. In: Critical Loads for Nitrogen - a report from a workshop held at Lokeberg, Sweden, 6-10 April 1992, Nord 1992:41, pp. 287-301.

Acknowledgements

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CHAPTER 11
CRITICAL LOADS OF NITROGEN: CONCLUSIONS AND RECOMMENDATIONS

The UNECE Expert Workshop held at Grange over Sands On 24 - 26 October 1994 addressed three main objectives

(1) to update the Lökeberg Report

(2) to produce appropriate revisions to the Nitrogen Critical Loads Chapter in the Task Force on Mapping Manual

(3) to review and provide an updated scientific basis for the mapping and modelling of critical loads for use in forthcoming discussions on the Second Protocol on Nitrogen Oxides.

The proceedings of the Grange Workshop published in this report demonstrate how successfully these objectives have been achieved and what gaps in knowledge still exist.
Several recommendations were also made during the course of the meeting:

(1) The meeting unanimously agreed that the Working Group on Effects request the Task Force on Mapping to prepare a new mapping manual no later than 1996. This recommendation arose because of the many changes that have taken place over the last two years making it difficult to maintain a consistency throughout the present mapping manual.

(2) The workshop felt that the present chapter on deposition in the mapping manual does not now reflect current understanding of atmospheric processes. It was suggested that EMEP and the WG Effects should be asked to jointly convene a group to revise this chapter.

(3) At present the mapping manual defines a non-marine approach for the calculation of deposition and critical loads. Some reservation was expressed at the Grange meeting that sea salts may need to be included for some areas and that further work should be done by those countries likely to be affected by its effects.

(4) The Mass Balance working group identified an urgent need for a literature review to extract the necessary information to quantify the values of the various parameters in the mass balance equations. The workshop recommended that the UNECE Secretariat be approached to see whether funding could be made available for this exercise. Without this information critical loads estimates, especially those which will be used for the gap filling exercise by the CCE (Co-ordination Centre for Effects), will continue to be uncertain.

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ANNEX 3

NITREX: Effects of Enhanced N Deposition to a Spruce Forest in Denmark -Implication for Critical Loads

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Materials and Methods

The fate and effects of increased N deposition were investigated by experimental manipulation in a mature Norway spruce plantation at Klosterhede, Western Denmark. Ambient N deposition was 23 kg N $ha^{-1} yr^{-1}$. Addition of 35 kg N $ha^{-1} yr^{1}$ as NH_4NO_3 to a 500 m^2 plot was carried out by handspraying monthly simulating a 2.5 increase of N deposition. The addition plot was compared to 3 parallel control plots. For soil solution sampling there were, however, 5 sets of samplers within the control plots.

The forest stand is second generation after heathland with 860 trees/ha and a basal area of $29m^2$. The soil type is a Typic Haplorthod (podzol). The soil is coarse sand with low base saturation. An organic layer of 7 cm with C:N ratio 33 has developed during the current rotation. Further experimental details are found in Gundersen & Rasmussen (1995). Here results from the pre-treatment period and the first 2.5 years of N addition are presented.

Results and discussion

Tree response

The N treatment caused no detectable response on diameter growth, shoot length or needle weight during the first two growing seasons. But for both growing seasons tree growth was determined by very low water availability (drought) in the spring. Further there was no change observed in the N content of the needles (Figure 1a) although the foliage was considered N deficient. However for K there was a significant decrease in the content of the needles compared to the controls (Figure 1b). The K:N ratio decreased from 0.6 before the treatment to 0.35 after treatment during two growing seasons. A K:N ratio below 0.3-0.4 may be considered as critical for the nutritional balance (Nihlgård, 1990). The decreased K content in the needles may result from an increased competition between NH_4^+ and K^+ in root uptake (Boxman *et al.* 1991), since the soil water concentrations of NH_4^+ after N addition were increased in the organic layer where most of the fine roots are present. In a complementary experiment in the Netherlands, where N deposition was decreased by a roof installation, an improvment of the K/N ratio and other nutrient ratios were found in the first years of treatment (Boxman *et al.* 1995).

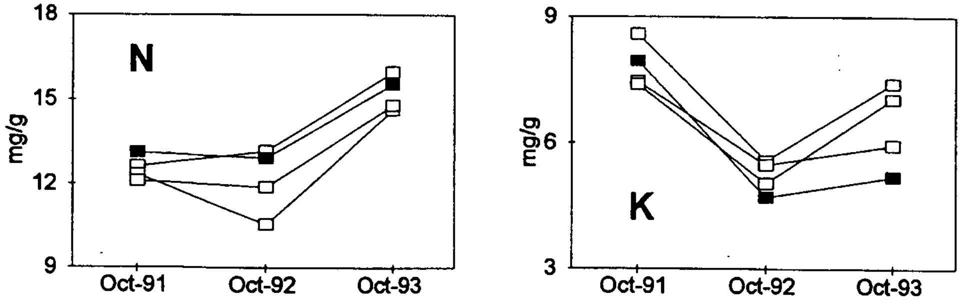

Figure 1 **Content of (a) nitrogen and (b) potassium in current year needles (composite sample from 5 randomly selected trees) on the N addition plot ( ) and 3 control plots ( ).**

Decomposition and Mineralization Response

Needle litter decomposition was studied in litter bags in the N addition plot and in one of the control plots (2 plots x 5 sets x 15 replicates). The litter bags were incubated from the start of the treatment and retrieved after 3, 6, 9, 12, 24 months. Weight loss and N content were determined.

The weight loss was not affected by the N treatment at any incubation time, but the N content of the remaining litter was increased on the N addition plot. The weight loss increased with

i) increased distance to the nearest trunk,
ii) increased moss cover, and
iii) to some extent by a low position in old plough furrows.

Therefore most of the variabilty of the decomposition was due to differences in moisture conditions. In a Sitka spruce forest Emmett *et al.* (1995) also found that decomposition did not respond to a similar increase of N deposition.

Mineralization was studied after 14 months (second year of treatment) by a sequential *in situ* incubation technique (*e.g.* Tietema *et al.* 1990) in the N addition plot and in one control plot. Two intact soil cores (10 cm diameter) of the organic layer were sampled side by side in three replicates per plot. One soil core from each pair was taken directly to the laboratory for determination of the initial conditions, the other core was closed on top and left in the field for incubation for one month (3 months during winter). In the laboratory the cores were separated into L+F layer and H layer. From each layer 10 g fresh soil were used for determination of water content and 10 g (2 replicates) were extracted in 1 M KCl for determination of NH_4^+ and NO_3^- concentrations. The accumulation of NH_4^+ and NO_3^+ in the incubated cores was considered an estimate of net ammonification and net nitrification rates, respectively.

The results for the NH_4^+ accumulation (Figure 2) showed no clear effect of the treatment. However, a possible treatment effect may be hidden by a within plot variability and by a number of obscure results, which are currently rechecked. The method is restricted by the large number of operations behind each data point. However, the calculated ammonification flux for a whole year increased from 15 kg N $ha^{-1} yr^{-1}$ in the control plot to 25 kg N $ha^{-1} yr^{-1}$ in the N addition plot. A drought in the spring 1993 inhibited the mineralization (Figure 2).

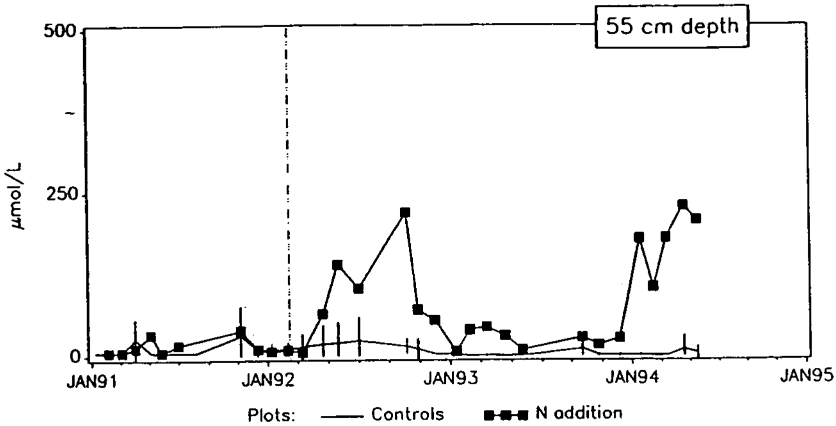

Figure 2 **Nitrate concentration in soil water at 55 cm depth in the N addition plot (1 set of samplers;) and in the control plots (5 sets of samplers; mean ± std).**

The soil was not accumulating NO_3^- (not nitrifying) except for a 'hot spot' in the N addition plot. It can not be concluded whether this was a result of the treatment or if this type of hot spot was present in the control plot as well.

Soil solution response

The soil solution chemistry responded promptly to the N application (Figure 3). Nitrate concentrations increased at all depths, and NO_3^- leaching increased from 0.3 to 2.3 kg N $ha^{-1} yr^{-1}$. Nitrate leached during winter and spring when the water transport was at the highest. Despite the increased NO_3^- leaching, 92% of the NO_3^- input was retained. The NH_4^+ input was completely retained within the system, but soil water concentrations of NH_4^+ were increased below the organic layer and at 15 cm depth on the addition plot. An increase of exchangeable NH_4^+ could account for 20% of the added NH_4^+ on the addition plot. No changes in concentrations of other major ions were detectable due to the N addition.

References

Boxman, A.W., Krabbendam, H., Bellemakers, M.J.S. and Roelofs, J.G.M. (1991) Effects of ammonium and aluminum on the development and nutrition of *Pinus nigra* in hydroculture. *Environ. Pollut.* 73: 119-136.

Boxman, A.W., Van Dam, D., Van Dijk, H.F.G., Hofervorst, R.F. and Koopmans, C.J. (1995) Ecosystem response to reduced nitrogen and sulphur input into two coniferous forest stands in the Netherlands. *For. Ecol. Manage.* In press.

Emmett, B.A., Brittian, A., Hughes, S. and Kennedy, V. (1995) Nitrogen addition ($NaNO_3$ and NH_4NO_3) at Aber forest, Wales: II. Response of trees and soil nitrogen transformations. *For Ecol. Manage.* In press.

Gundersen, P. and Rasmussen, L. (1995) Nitrogen mobility in a nitrogen limited forest at Klosterhede, Denmark, examined by NH_4NO_3 addition. *For. Ecol. Manage.* In press.
Nihlgård, B. (1990) The vitality of forest trees in Sweden - stress symptoms and causal relations (in Swedish with English summary). In: Liljelund, L.-E. (ed) *Vitality fertilization -present knowl edge and research needs.* Swedish Enviromental Protection Agency, report 3813, 45-70.
Tietema, A., Duysings, J.J.H.M., Verstraten, J.M. and Westerveld, J.W. (1990) Estimation of actual nitrification rates in an acid forest soil. In: Harrison, A.F., Ineson, P. & Heal, O.W. (eds), *Nutrient cycling in terrestrial ecosystems*, Elsevier Applied Science, London, 190-197.

ECOFEE: Element Cycling and Output-fluxes in Forest Ecosystems in Europe

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Background

Due to continued and increased concern on the effects of acidification, the nitrogen deposition, and the climate changes in European forests the Nordic Council of Ministers (NMR) has initiated a project to update and expand the data base "Evaluation of Nitrogen and Sulphur Fluxes" (ENSF) (Haus *et al.*, 1989; Rose-Siebert, 1989) on element fluxes in European forest ecosystems. The Danish Forest and Landscape Research Institute is gathering this information and preparing a data base.

The aim is to compile yearly element budgets including internal processes like litterfall, pool sizes (soil and vegetation), and other ecosystem characteristics (nutrient status, vegetation and soil parameters, climate) from all major forest ecosystem research sites (control plots only) in Europe. The emphasis is on plot-scale studies including plots within studied catchments. Catchment-scale data bases already exists within the ENCORE network and the ICP-Integrated Monitoring network.

The data compilation will be used to evaluate the input-output relations, time trends, geographical trends etc. The following questions will be addressed:

- Has enhanced N deposition increased the internal cycling of N?
- Which site and ecosystem characteristics may regulate the output fluxes of nitrate?
- Is the magnitude of the internal cycling elements correlated to the leaching loss?

This information may be valuable in the discussion of methods to estimate critical loads and in the evaluation of critical loads maps.

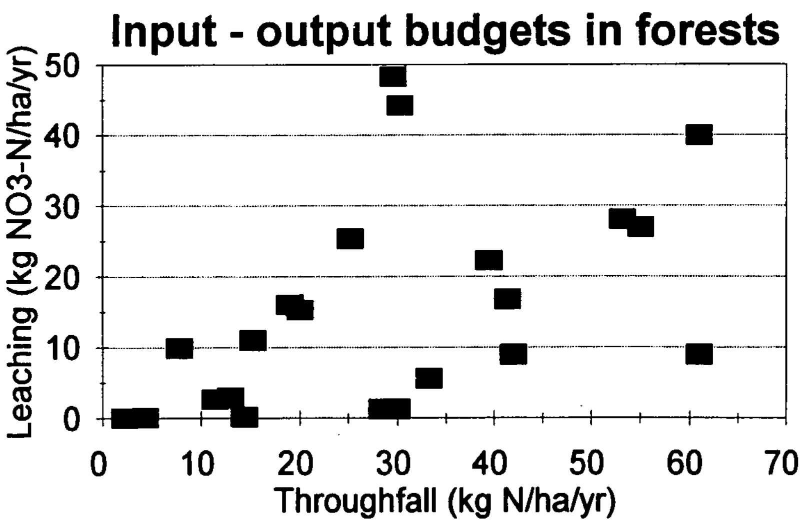

Figure 1: Results from c. 25 forest ecosystems which up till now is included in ECOFEE.

Mapping and modelling of critical loads for nitrogen

Within the United Nations Economic commission for Europe (UNECE) the Convention on Long Range Transboundary Air Pollution is currently working to update the Existing Protocol for Control of Emissions of Nitrogen Oxides. It is planned that a revised protocol should be based on the critical loads approach, in a similar way to that adopted by the second Sulphur Protocol, signed in Oslo in June 1994. Since environmental impacts of nitrogen are a consequence of both nitrogen oxides and ammonia emissions, a new protocol would need to consider the critical loads for total nitrogen, including the impacts of both oxidized and reduced nitrogen.

With this motivation, a UNECE Expert workshop was held at Grange-over-Sands on 24-26 October 1994 under the auspices of the Convention's Working Group on Effects (WGE). The workshop addressed three main objectives: (1) to update the findings reported in the proceedings of the workshop held at Lökeberg, Sweden in April 1992; (2) to produce appropriate revisions to the Nitrogen Critical Loads chapter in the Mapping Manual of the WGE Task Force on Mapping; (3) to review and provide an updated scientific basis for the mapping and modelling of critical loads for use in the forthcoming discussion on the Second Nitrogen Oxides Protocol.

Three main working groups at the workshop reviewed the situation for terrestrial and freshwater ecosystems using empirical mass balance approaches. Five further discussion groups also reported on the sections in the Mapping Manual relating to: atmospheric deposition, treatment of total acidity and sea salts in the mass balance approach, dynamic modelling, marine ecosystems, and mapping issues. Together with background papers and posters, and the workshop recommendations, the output from each of these groups form the substance of this report.