Institute of
Terrestrial
Ecology

Critical loads: concept and applications

Natural Environment Research Council

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Ecology

Critical loads: concept and applications

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Introduction

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The concept of a 'critical load' is coming to dominate European legislation on air pollution. A critical load can be defined as 'the maximum deposition of a given compound which will not cause long-term harmful effects on ecosystem structure and function, according to present knowledge'. It is intended, in other words, to be a threshold deposition which ecosystems can tolerate without damage. One aim of the pollution control policies of many countries is now to reduce the deposition of acidifying pollutants below their critical loads. To enable this to be possible, the scientific community has to be able to define well-supported critical loads for any European ecosystem. As an attempt to codify a vast amount of ecological knowledge and use it as a rational means of pollution control, the critical loads approach is an ambitious undertaking.

Maps of critical loads for various receptors have now been published for the whole of Europe by several bodies on a variety of scales. In the UK, the Department of the Environment has published maps of critical loads on scales down to 1 km. The concept is being used by the UK government as a guide for its policies on pollution control, and by European governments under the auspices of the United Nations Economic Commission for Europe as a basis for negotiations for a new protocol on limiting long-range trans-boundary sulphur pollution. The concept, clearly, is of great political importance.

The development of the concept and production of the maps have taken place with remarkable speed, generally within working parties whose composition has been determined by the governments of the countries concerned. There has been little opportunity for the ecological community as a whole to participate in these discussions, or for those who have been involved to present the results of their deliberations to their scientific peers. To remedy this situation somewhat, we organised a Conference open to all interested parties, and invited contributions from both scientists and those whose job it is to develop and implement environmental policies. The Conference was held at Grange-over-Sands on 12–14 February 1992, under the auspices of the British Ecological Society Industrial Ecology Group and the Natural Environment Research Council (NERC). The National Power/PowerGen Joint Environmental Programme provided some sponsorship.

The Conference generated some new ideas on critical loads, showed that there was a variety of views on the validity of the concept as it stood, and sparked off some controversies. This volume documents the proceedings of the Conference, both written papers provided by the speakers and reports of workshop discussions on specific questions. We hope it will be of use in further development of the critical load concept, and of interest to anyone who wishes to understand the progress made so far.

The development of the critical loads concept

Acidification research: evaluation and policy applications

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The United Kingdom government is deeply committed to the support of environmental research into the mechanisms of the atmospheric transport and deposition of acidity, in order to understand the impacts of that acidity on soils, surface waters, forests, crops and the built environment, and the consequences for fishery status, freshwater and soil ecosystems. The United Kingdom collaborates fully with international research programmes within the framework of the United Nations Economic Commission for Europe (UN-ECE) Convention on Long Range Trans-boundary Air Pollution, with these aims in mind.

International action agreed so far on acidic depositions has taken little account of the complexity of their impacts. It has focused on the abatement of emissions and applied the straightforward instruments of negotiated national percentage reductions and the best economically feasible abatement techniques to major sources. These tools have the advantages of simplicity and intelligibility to policy-makers, regulators and industry, which are important at a stage when we are still a long way from achieving our environmental objectives.

They are, however, relatively blunt instruments. Even over their present planned lifetime they risk entailing some wastage of resources on action which will not secure environmental improvement. Looking ahead to the next stages of an international abatement programme, we must build our actions on scientific consensus about the nature and distribution of the damage being done by acid deposition, and the links between observed deposition patterns and sources of emissions.

Within the framework of the UN-ECE Convention, we have begun the search for more subtle and sensitive ways of tackling the problems of environmental acidification. Member States have agreed to adopt a rational approach based on the underpinning environmental science, for the revision of the NO_x SO_2 and VOC protocols. This is the critical loads approach.

Last year, the 34 nations of the ECE region agreed at the Bergen Conference on Sustainable Development that:

- strategies for the abatement of SO_2 and NO_x emissions should be designed in the most cost-effective way, and
- the concept of critical loads should serve as a guideline to formulate these strategies where science has provided the necessary information.

For a deposited pollutant, we define the critical load as the maximum deposition that a target ecosystem can withstand without long-term damage occurring. The critical load is thus one point in the damage-response curve for a particular pollutant/target combination, and marks the point where, with increasing deposition load, damage becomes discernible. The critical load for a pollutant/ target combination is not a universal constant; it varies spatially depending on a range of meteorological, ecological, biological, geological and hydrological factors. These variations can be understood, estimated and mapped to show their distribution both nationally and internationally.

The UK Department of the Environment is committed to the mapping of critical loads and levels for a whole range of pollutant/target combinations. It has established the UK Critical Loads Mapping Centre at ITE Monks Wood, and the staff there have begun in earnest to work with many other research institutes to collect the mapping data. This National Focal Centre will assist the Coordination Centre for Effects which has been established at the National Institute of Public Health and Environmental Protection (RIVM), Bilthoven, in The Netherlands, to assemble the maps from each European country and to produce the European critical loads and levels map for the complete range of pollutant/target combinations.

The critical loads approach is based on a comparison or overlaying of maps - maps of critical loads with those of deposition loads. These two sets of maps are essential components of the critical loads approach. The accurate mapping of deposition loads requires the folding together of several sets of environmental data. Ambient and precipitation concentration data have been provided for a wide range of pollutants for the UK from the national primary and secondary precipitation networks operated by the Warren Spring Laboratory. Detailed information on the mechanisms of dry deposition, occult deposition and seeder-feeder enhanced wet deposition has been assembled at ITE's Edinburgh Research Station. Together, concentrations and data on mechanisms have been employed to provide a comprehensive and quantitative assessment of deposition inputs of all acidic pollutants on a 20 km grid across the UK.

A further essential component of the critical loads approach is the means by which deposition loads of each pollutant can be attributed to the sources of their emission into the atmosphere. The long-term weather patterns bring different deposition loads to each point in Europe, from each of the major source areas. These weather patterns are tracked by the European Monitoring and Evaluation Programme (EMEP) Meteorological Synthesising Centre (West) of the Norwegian Meteorological Institute. Using the EMEP models, it is able to provide estimates of how the deposition in one country can be attributed to the emissions in all the other European countries. Such studies provide an important input both to the critical loads approach and to the UN-ECE Convention. National modelling can be used to increase the spatial resolution of long-range transport models, but spatial resolution is ultimately limited by our understanding of the vagaries of European weather conditions and of the atmospheric physical and chemical processes involved.

Once the European critical load maps are available, they can be overlain with their corresponding deposition load maps to produce critical loads exceedance maps. Long-range transport models can then be used to provide a country attribution to the source of any exceedances. This information will be vital to the ongoing policy formulation process in the international context. Each Member country will be able to compare its commitment to emission reductions in the future with the implications of the critical loads approach.

The UK is contributing vigorously to the critical loads approach through the mapping exercises, the environmental studies that underpin them, and the investigation of the driving deposition mechanisms which lead to both pollutant removal and ecosystem contamination. The Department of the Environment has established a series of independent review groups to assess the significance of many of the scientific issues, and to advise and recommend how the environmental science is to be advanced. Last year saw the publication of the third report from the Review Group on Acid Rain and the second report from the Photochemical Oxidants Review Group. In addition, the Critical Loads Advisory Group has made significant progress in assessing the effects of air pollution and acidic deposition in the United Kingdom. Critical load maps for the acidification of soils have been published, and the mapping of the sensitivities of freshwater ecosystems is well advanced.

Maps have also been prepared to show how current estimated levels of acid depositions and sulphur depositions compare with critical loads for soils. Where depositions exceed critical loads, long-term environmental deterioration is likely to be occurring. There is a good correspondence between the areas of exceedance shown and areas where acidification damage has been reported, particularly in the upland part of Wales, Cumbria, Galloway and the Scottish Highlands.

This mapping work, and the similar work which has been carried out in other UN-ECE countries, provides an invaluable new input into the domestic and international policy-making process. However, on its own, it cannot determine policy, for a number of reasons.

First, damage by acid depositions is not limited to the acidification of soils and freshwaters and the consequent effects on ecosystems. There are also effects on forests and on buildings to be considered. While intensive research continues, we have not yet identified dose-response relationships for these effects with sufficient confidence to be able to establish critical loads, and to map them. Neither, even in the case of soils and freshwaters, are we yet as clear as we need to be about the roles of nitrogen and ammonia in relation to that of sulphur.

Second, a policy of bringing acid depositions down to critical loads in respect of all effects, in all parts of the UK or the UN-ECE region as a whole, would be practicable, if at all, only in the very long term. Priorities, therefore, need to be identified and interim targets set. This process will require the application of economists' as well as scientists' tools, to help evaluate damage and to compare the costs and benefits of abatement, though decisions must at the end of the day be a matter for political judgement. Before the economists can be of much assistance to us, however, more of the scientific groundwork will need to have been laid.

Third, it is only to be expected that individual countries' domestic targets for abatement will prove difficult to reconcile with each other, because emissions are differently distributed internationally from the damage to which they give rise. There will be severe challenges for the international negotiating machinery, which we are already beginning to encounter.

For the UK, the foundations of present policy, as regards stationary sources, are:

- the European Community Large Combustion Plants Directive, with its requirements for major percentage reductions in SO_2 and NO_x emissions from existing power stations and other boilers, and stringent emission standards for new plant;
- the NO_x protocol to the UN-ECE Convention, requiring total national emissions to be reduced to 1987 levels by 1994;

- the obligation laid on operators of industrial processes by Part I of the Environmental Protection Act 1990, itself derived from the EC Air Framework Directive, to use best-available techniques not entailing excessive cost (BATNEEC) for reducing harmful emissions and minimising their impact.

In addition, the United Kingdom has played a leading role in securing EC agreement to new Directives on emissions from cars and heavy diesel vehicles, which will secure major reductions in (particularly) NO_X emissions from this sector.

The environmental effects of these policies can be forecast only within wide margins of uncertainty. However, projections based on conservative assumptions suggest that they should enable the proportion of the UK surface area exposed to depositions in excess of critical loads for soil acidification to be reduced to around 8% by the year 2005. As regards acidification of freshwaters, only 4% of the surface area of Scotland will remain unprotected.

These existing policies do not take specific account of the varying relationships between present deposition levels and critical loads in different parts of the UK. However, the mapping work which has now been done will help to facilitate their detailed application. In particular, the determination by the regulatory authorities of BATNEEC at individual plant level, and the consideration of any proposals for major plant modification, will be assisted by knowledge of the likely effect on soils and freshwaters of acid emissions from the plant in question.

All the maps of critical and target soils have been submitted to the UN-ECE, and other UN-ECE countries have been preparing similar maps of their own. They will provide the basis for discussion of the proposed new UN-ECE agreement to reduce emissions of sulphur dioxide, currently being negotiated.

Future progress with the UN-ECE Convention on the Long Range Trans-boundary of Air Pollution and the revision of NO_x, SO_2 and VOC protocols will rest in very large measure on the shared confidence within Europe in the knowledge of the underpinning environmental science. The critical loads approach should provide an important policy focus within the international scientific community to set environmentally based targets for future co-ordinated emission control programmes.

Development of the critical loads concept and the UN-ECE mapping programme

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INTRODUCTION AND THE DEFINITION OF CRITICAL LOADS

The critical loads approach to controlling acidic emissions has attracted increasing interest in recent years. International activities aimed at reducing sulphur and nitrogen emissions (especially those within the United Nations Economic Commission for Europe (UN-ECE)) are looking to critical loads as a practical means of linking emission controls with environmental benefits.

The basic concept of critical loads is a simple one - the threshold at which a pollutant load causes harm to the environment. However, much thought has been given to turning this simple idea into scientifically acceptable criteria. Bull (1991) has discussed the problem in detail in relation to the efforts made by various specialist groups to find acceptable definitions for different sensitive receptors. However, a starting point for many recent deliberations has been the definition which Nilsson and Grennfelt (1988) adopted for a UN-ECE workshop in 1988. They described the critical load as:

'a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge'.

For this definition, much discussion has focused upon specifying appropriate sensitive elements of the environment and defining the meaning of significant harmful effects. This has not always proved easy.

When sufficient is known of the spatial pattern of sensitivity, maps of critical loads may be drawn which provide an opportunity to assess potential areas of damage in relation to known sources of pollution. In this way, it may be possible to calculate if critical load values are exceeded in an area, and thus to identify the likelihood of damage. This assessment may be done using current (usually measured) or future (modelled) pollution deposition loads. Maps showing where critical loads are exceeded are usually called exceedance maps.

The critical loads concept may be used to consider the recovery of damaged areas of the environment following a reduction in pollutant levels. However, it is necessary to consider the recovery process in more detail and to take account of time-dependent processes which may inhibit immediate recovery. It should be stressed that the pattern of recovery may be very different from that when the damage occurs.

In addition to the term critical load, it is important to draw the reader's attention to the terms critical level and target load. A critical level is analogous to a critical load but refers to a threshold of damage for gaseous pollutants acting directly upon (usually) vegetation species. A target load has been defined by Henriksen and Brakke (1988) as the pollutant 'load determined by political agreement'. Such a target may take account of social, economic or other constraints. It may be greater than the critical load and thus allow a degree of damage, or it may incorporate a safety factor and be less than the critical load value.

THE EVOLUTION OF THE CRITICAL LOADS CONCEPT

While the precise source of the origin of the term critical load is unknown, ideas regarding the response of aquatic ecosystems to certain loadings of pollutants were being considered in Scandinavia in the 1970s (Almer *et al.* 1978), and, towards the end of that decade, the concept of an acceptable pollutant load was discussed in Canada. The term critical load was first used at the Stockholm Conference in 1982, and the concept was promoted by the Scandinavian countries at workshops in the years that followed (Nilsson 1986; Nilsson & Grennfelt 1988).

Towards the end of the 1980s, the UN-ECE, under the Convention on Long Range Trans-boundary Air Pollution, adopted the critical loads concept as a potentially useful approach for future consideration of the abatement of nitrogen and sulphur. Following this adoption, a series of national and international activities were initiated which have worked towards applying the concept to future abatement strategies.

THE UNITED KINGDOM CRITICAL LOADS ADVISORY GROUP (CLAG)

When it was becoming clear that international abatement was beginning to focus upon the critical loads approach, the UK Department of the

Environment set up a small advisory group to provide expert advice on the application of critical loads in the UK. This group has subsequently grown to include more than 20 scientists from a wide range of organisations and areas of expertise. Subgroups of the CLAG, in particular those for freshwater systems and soils, have been actively calculating values for critical loads for the UK and using these values to draw critical load maps in collaboration with the Mapping Centre at ITE Monks Wood. The results of the efforts of the CLAG are described elsewhere in this volume and have been outlined by Bull *et al.* (1991).

The UK government's commitment to the critical loads concept became clear with the publication of the White Paper *This common inheritance* (Department of the Environment 1990). In this Paper, critical loads are one of the essential building blocks in a strategy for pollution abatement (Figure 1). Following the development of critical load maps, an iterative process of monitoring, assessing damage, planning abatement (setting targets), implementing controls, and remonitoring is envisaged. In this way, pollution levels should continue to decrease with the objective of achieving reductions below the critical loads. A similar iterative process may also be expected in the generation of critical load values and critical load maps. Current maps can only be based upon the knowledge available at present. This situation may change in the future and may result in changed critical load values for areas of Britain.

THE UN-ECE PROGRAMME

International activities within the UN-ECE have focused upon the revision of protocols for sulphur and, more recently, nitrogen under the Convention on Long Range Trans-boundary Air Pollution. The existing sulphur protocol (the so-called '30% Club' which has aimed at reductions of sulphur emissions by 30%) is due for revision in 1993, and most critical loads activities have been directed towards this timetable.

Under the Convention, a series of Working Groups and Task Forces have been set up to deal with specific aspects of international negotiations (Table 1). In addition to the European Monitoring and Evaluation Programme (EMEP), which is responsible for drawing up emission inventories and mapping pollutant levels for Europe, there are two other groups of significance.

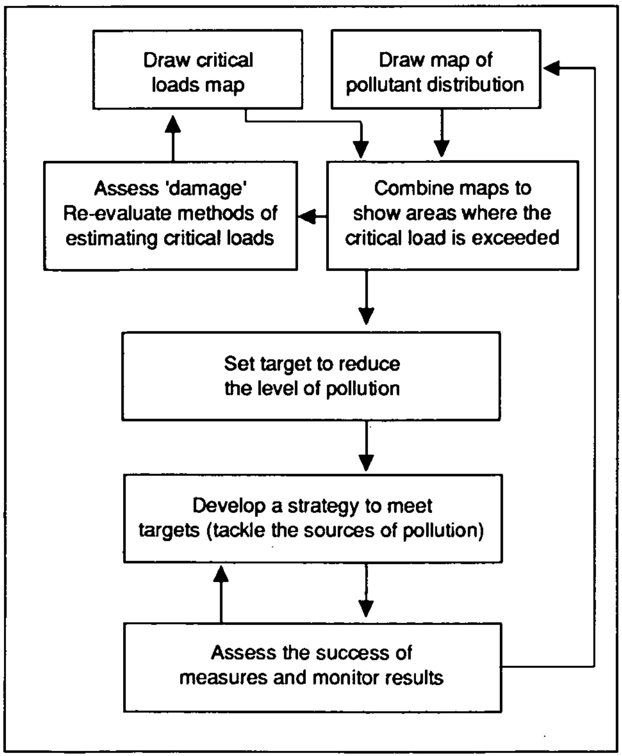

Figure 1. Flow diagram showing iterative re-assessment of critical loads, targets and strategies (based on Figure in Department of the Environment 1990)

Table 1. Structure of the relevant bodies of the UN-ECE Convention on Long Range Trans-boundary Air Pollution

Executive body of the Convention

- European Monitoring and Evaluation Programme (EMEP)
- Working Group on Effects
    - Task Force on Mapping
    - International co-operative programmes
- Working Group on Strategies
    - Task Force on Integrated Assessment Modelling

The Working Group on Effects (WGE) is responsible for considering the environmental impacts of pollutants and, in this role, has promoted a Task Force on Mapping which has the objective of drawing critical load maps for the whole of Europe. The WGE (which is chaired by the UK) is also responsible for the organisation of a number of international co-operative programmes which are considering impacts on specific parts of the environment, such as crops, forests, freshwaters.

The Working Group on Strategies (WGS), formerly the Working Group on Abatement Strategies, is responsible for developing strategies for pollution abatement. It is considerating economic instruments and the application of emission/deposition models which incorporate cost functions and environmental impact assessment using critical load maps. These integrated assessment models are being developed by the Task Force on Integrated Assessment Modelling (TFIAM), which reports to the WGS.

EUROPEAN CRITICAL LOAD MAPS

The development of methods for mapping critical loads and levels at the European scale has been the remit of the Task Force on Mapping. This Task Force has built upon a series of UN-ECE workshops at which methods for the calculation and mapping of critical loads and levels for different sensitive environmental receptors were discussed and agreed.

The approach adopted by the Task Force on Mapping for mapping critical loads and areas of exceedance has been discussed by Bull (1992) and Hettelingh, Downing and de Smet (1991). In general, it is dependent upon using known biological effects to derive 'critical chemical criteria' which can then be equated with pollution load in a numerical way, usually by some form of equilibrium model.

The responsibility for generating national critical load maps rests with the individual countries concerned, but it is recommended that they use agreed methods and procedures. To co-ordinate activities and collate national data sets for the European map, an international Coordination Centre for Effects (CCE) has been set up in The Netherlands. This Centre is responsible for generating European data for the Task Forces and Working Groups. To ensure that a complete map of critical loads for Europe is available, the CCE is responsible for filling the gaps in the map with appropriate data where there are no national submissions.

European maps of critical loads of acidity and of sulphur have now been compiled by the CCE and reported to the executive body of the Convention (Hettelingh *et al.* 1991). These maps are playing an important role in assessing environmental benefits arising from various deposition scenarios defined by TFIAM modelling activities. The work is aimed at the development of abatement strategies for a revised sulphur protocol in 1993.

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Problems with the critical loads approach: a view from industry

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IS THE CRITICAL LOADS APPROACH A GOOD THING?

The critical loads approach is potentially a great advance on some previous methods aimed at rational pollution control. It takes account of the ecological effects of pollutants, and thus can draw on the enormous efforts expended on studying them. It should, then, be superior to alternatives which do not take effects into account. One such is the arbitrary standards approach, exemplified by the European Community standard for pesticides in drinking waters, in which there is a limit of 0.1 $\mu g\ l^{-1}$ for each pesticide regardless of toxicity. Another is the blanket percentage approach, exemplified by the '30% Club', in which participant countries agreed to reduce SO_2 emissions by 30% of those in an arbitrary base year. It may seem self-evident, to an audience of scientists interested in pollution effects, that taking ecological effects into account is an improvement. To others this may not be so clear, and it is worth briefly considering why.

In their book *Acid politics*, Boehmer-Christiansen and Skea (1991) contrast British and German attitudes to, and definitions of, 'pollution'. In British usage, 'pollution' is something which has a deleterious effect on the environment. The corresponding German word 'Verschmutzung' (making dirty), together with the word for pollutant (or harmful substance) 'Schadstoffe', encourage the idea that the mere presence in the environment of a foreign substance constitutes pollution. In this view there is no need for research into effects: the aim should be to eliminate these substances. The means for doing so is also clear: it is to reduce emissions by fitting control technology (the 'best available technology' (BAT) approach). Thus, to control acid deposition, all sources of SO_2 and NO_x must be fitted with appropriate removal technology (such as flue gas desulphurisation and N reduction for stationary sources, catalytic reduction for motor vehicles) as soon as possible. Allied to this view is another concept derived from German thinking: the 'precautionary principle'. In its strongest form, this can be used to justify the maximum emission control which is technically achievable, on the grounds that emissions *may* do some damage. Again, there is no real need for effects research.

So what is the role of the critical loads approach? The UK government has a much more pragmatic view of the precautionary principle: 'where there are significant risks of damage to the environment, the government will be prepared to take precautionary action to limit the use of potentially dangerous materials, or the spread of potentially dangerous pollutants, even where scientific knowledge is not conclusive, if the balance of likely costs and benefits justifies it' (Department of the Environment (DoE) 1990). Note that this statement requires an assessment of the risk of damage and the costs and benefits of a given control strategy. The critical loads approach attempts to quantify the risks and benefits in ecological terms. In practice, all governments take costs and benefits into account, even when BAT is the official policy. Flue gas desulphurisation is not fitted to domestic central heating systems, neither are power plant operators obliged to replace their existing control systems if another offering 1% better performance is developed. In these cases, the costs clearly outweigh the benefits, while cheaper measures, such as restricting the sulphur content of domestic heating oil, might well be adopted.

Using the critical loads approach then, politicians could obtain answers to such questions as: 'what would it cost to reduce deposition to the critical load everywhere', or 'what would it cost to protect 95% of all soils against anthropogenic acidification', or 'given that we are prepared to spend £x million on pollution control, what is the most effective way to spend it'. So, the critical loads approach enables us to optimise the use of resources available for environmental protection, and to present to decision-makers and the public the effects of different policy options in a readily comprehensible manner. It must therefore be counted, potentially, as a 'good thing'.

CRITICAL LOADS IN PRACTICE: ARE THEY BASED ON GOOD SCIENCE?

The UK government's White Paper (DoE 1990; 1.16) stated the necessity '...in environmental decisions as elsewhere, to look at all the facts and likely consequences of actions on the basis of the best scientific evidence available. Precipitate action on the basis of inadequate evidence is the wrong response'. But how do we decide what is the best scientific evidence, and, indeed, what constitutes a 'fact'? In complex matters on the frontiers of research, such as the ecological effects of pollutants, respected scientists often disagree with each other on both the facts and their interpretation, and all agree there are substantial areas of ignorance which need investigation. Nevertheless, it is often possible to reach a tentative consensus which all but a few support. For instance, there is now a consensus that acid deposition has acidified soils, streams and lakes in sensitive parts of Europe, though there are still eminent dissenters from this view (eg Rosenqvist 1990; Krug 1989). Such a consensus was not possible at the beginning of the 1980s: it has been brought about by research.

How can the process of obtaining a tentative consensus, which can be taken as the 'best scientific evidence available', be expedited? I believe the way is to apply the same procedures and conventions which are responsible for much of scientific progress, but particularly:

- open publication of all relevant results and methods;
- open debate in scientific meetings and conferences;
- peer review of all results used for regulatory purposes;
- open access to data used in environmental analyses;
- explicit methodology which can be repeated by others from publicly available information;
- formulation of environmental ideas and models as hypotheses to be tested against existing or future data;
- critical evaluation of such ideas and models, by further research if necessary.

These processes take time, and political imperatives may, of course, require answers more rapidly. Provisional answers should then be supplied, but the range of uncertainty involved should be made clear, as should the source of the answer. Is it derived from one person, a select group, a workshop? It cannot be taken as the view of the scientific community, and hence the best available scientific information, unless the above criteria are satisfied.

The development of the critical loads approach in Europe since 1986 does not unfortunately meet the above criteria. The concept was derived from Canadian ideas, but its present form dates from a working group of seven scientists, all from the Nordic countries, who produced a discussion paper on the concept (Nilsson 1986). This paper, in turn, was presented to an 'invitation only' workshop of a further 27 scientists, 12 of whom came from outside Fennoscandia. The resulting publication (Nilsson 1986) contained many valuable ideas and was explicitly designed to 'stimulate further research and assessments'. After this promising start, things began to go awry. The United Nations Economic Commission for Europe (UN-ECE) became interested in the concept, and the next workshop, at Skokloster in Sweden, was held under the joint auspices of UN-ECE and the Nordic Council of Ministers. UN-ECE Member bodies were invited to send delegations to the workshop, producing a total attendance of 72, 56 of whom had not attended the previous workshop. Scientists who were not nominated by the UN-ECE Member bodies, no matter how expert, were not permitted to attend. The resulting publication (Nilsson & Grennfelt 1988) quantified and developed the previous ideas.

Though progress was being made, there was no opportunity for the wider scientific community to participate in the development of ideas on critical loads, though many wished to do so. Few of the ideas developed at these workshops had passed the test of publication in peer reviewed scientific journals.

After the Skokloster workshop, UN-ECE took over the development of the critical load concept almost completely. The style of working of such bodies as UN-ECE is appropriate for international negotiation, where some secrecy is necessary and productive, but not for the development of scientific ideas. The Working Group on Abatement Strategies of the executive body for the Convention on Long Range Trans-boundary Air Pollution of UN-ECE set up Task Forces, National Focal Centres, and an overall Co-ordination Centre in The Netherlands. A critical load mapping manual was apparently produced. Many scientists across Europe were drawn into these activities, and it was occasionally possible for outsiders to obtain copies of some of the documentation from them. Some individuals did start to publish their ideas in the peer reviewed literature (eg De Vries 1988; Sverdrup 1988; Henriksen & Brakke 1988; Schulze *et al.* 1989), but the relationship of these papers to the official methodology was unclear. Such activities are a long way from the open debate which is necessary to produce best available scientific information. In contrast, the Stockholm Environment Institute ran a parallel critical loads programme, which was openly published and in which the methodology was clearly described (Kuylenstierna & Chadwick 1989).

Eventually, the results of the UN-ECE activity emerged into the open in the form of a report from the Coordination Centre, dated July 1991, but not derestricted until November (Hettelingh, Downing & de Smet 1991). This report was not presented as a document for discussion by the wider scientific community, which at last had access to it, but as a *fait accompli*, and there was no suggestion that there would be better access to future activities. A parallel effort on critical levels has so far produced no open information at all.

In the UK, the Department of the Environment was advised by the Critical Loads Advisory Group (CLAG), as described by Bull (pp 8–10). Two reports to the DoE from CLAG have been published, and DoE (1991) has published a popular exposition containing critical and target load maps for the UK. In none of these documents is the methodology explicit in the sense that the maps they contain could be reproduced by someone with access to the data, neither is it available openly elsewhere. Moreover, both DoE (1991) and the CLAG report on critical loads for soils contain the same mistake: many 10 km grid squares are shown in which the critical load for sulphur is exceeded, whereas the critical load for acidity is not. This result is not possible if the critical load is partitioned between sulphur and nitrogen in the way the reports state. It is not possible to tell which map is correct. Peer review might have avoided this mistake, and the inevitable doubts which then arise in the reader's mind about the other maps.

In contrast to the approach adopted by UN-ECE, the US National Acid Precipitation Assessment Program (NAPAP) went to great lengths to ensure openness and exposure to the whole scientific community in its ten-year programme of research into acid rain. All research projects were peer reviewed before approval and at intermediate stages by independent experts. The 27 final reports were reviewed by three independent reviewers, with particular stress on credibility; the authors were allowed to respond, and their response and the document as a whole were submitted to a fourth reviewer, who presented his findings at an open international conference. Comments from the conference were incorporated in the final versions of the report, which was again independently reviewed to ensure it took them into account. Degrees of uncertainty on statements in the text were explicitly identified by means of a star system. The resulting documents (eg Irving 1991) thus possess a good deal of credibility. NAPAP also illustrates some disadvantages of a consensus approach. Too much resource was devoted to the peer review process, and political decisions about emission reduction were taken before the NAPAP assessment was available. Nevertheless, the assessment will stand as a firm basis for future decision-making when the issues are revisited, as they inevitably will be.

I demonstrated above that the procedures used to derive critical loads are far removed from the optimum way to produce the best available scientific information for decision-making on environmental matters. But does it matter? Many talented people were involved in the process, and the results clearly have some value. Are there major scientific criticisms to be made of the results? Some of the contributors to this volume clearly think so, and I add a few more below. It is also worth remembering the profound policy implications which may flow from the critical load maps. At present, it seems that it is going to be rather difficult in the UK to obtain permission for new developments, or even to continue existing facilities, where the critical load is currently exceeded and the facility emits acidifying pollutants. This restriction applies to most of Scotland, Northern Ireland, north-west England and Wales: indeed, anywhere in the UK where it rains a lot. The lives of thousands of people in these areas could be profoundly affected by the economic consequences, and therefore it behoves the scientific community to do its utmost to make the underlying science as sound as possible. In addition, the economic interests at stake raise the possibility of legal challenge, alien and unproductive territory for science, which should be avoided at all costs.

IS THE CRITICAL LOAD A THRESHOLD?

In both popular and scientific accounts of critical loads, the key concept being transmitted is that of a threshold. If deposition is above the threshold, things are deteriorating, and the greater the difference, the greater the concern, because the deterioration will be greater or faster, or both. Conversely, if deposition is below the critical load, there are no problems. Do the published methods for calculating critical loads allow one to paint this simple picture? The problems are best illustrated by an example.

Skeffington and Brown (1992) presented a scenario analysis showing the influence of various degrees of reduction of acid deposition on a hypothetical catchment. The MAGIC model (Model for Acidification of Groundwater in Catchments) (Cosby *et al.* 1985) was used for the simulations. Catchment parameters were chosen to represent a sensitive geology (base cation weathering rate 11.3 meq m^{-2} yr^{-1}) with fairly high rainfall (1500 mm) and runoff (1360 mm), but average conditions otherwise. The deposition scenario represents estimated UK acid deposition up to 1985, with various options thereafter (see Figure 1). Figures 2 and 3 show the response of soil base saturation and drainage water pH respectively. Are these responses what would be expected from the calculated critical loads?

The steady-state mass balance method, as described

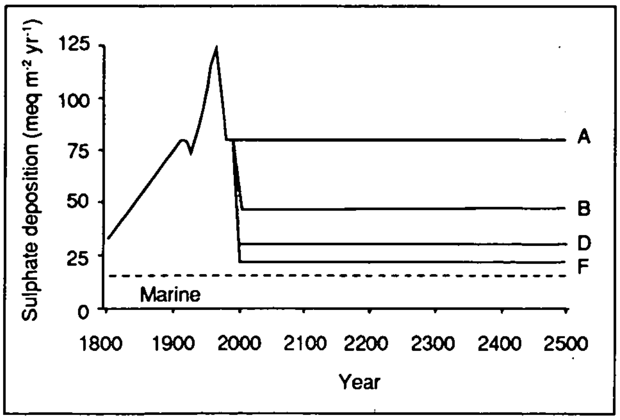

Figure 1. Sulphate deposition scenarios used in catchment simulations by the MAGIC model (Cosby *et al.* 1985). The pattern resembles UK SO$_2$ emissions from 1800 to 1993, followed by four scenarios: A, no further change; B, 60% reduction on 1980 values by the year 2003; D, 80% reduction on 1980 values by 2003; F, 90% reduction on 1980 values by 2003. Scenario numbers are from Skeffington and Brown (1992). Total SO$_4^{2-}$ is plotted: the constant marine component is also shown

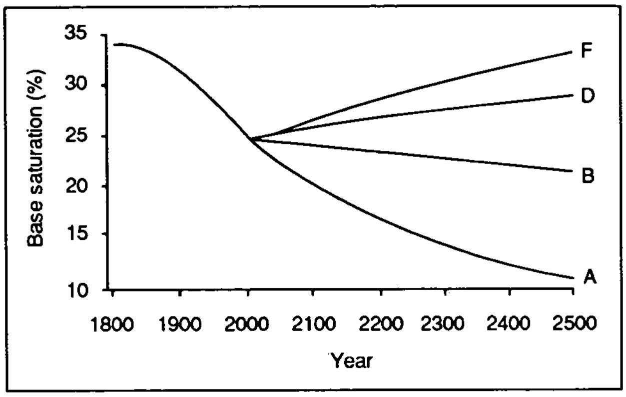

Figure 2. The response of soil base saturation to the scenarios described in Figure 1

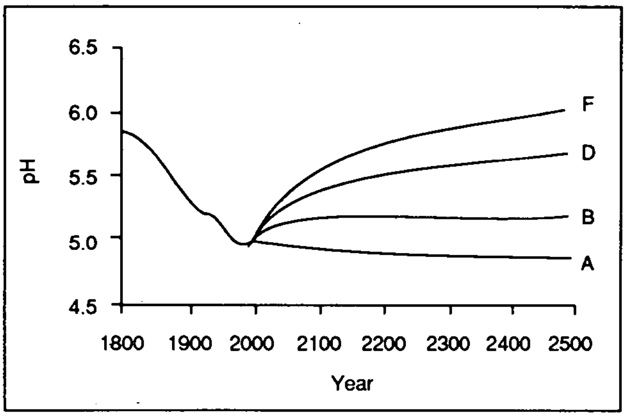

Figure 3. The response of drainage stream pH to the scenarios described in Figure 1

by Sverdrup, De Vries and Henriksen (1990) and Hettelingh *et al.* (1991), involves the following equation:

$$CL_{acidity} = BC_w - BC_u - ANC_l - AC_n \qquad (1)$$

where CL = critical load; BC_w = base cation weathering; BC_u = base cation uptake; ANC_l = permitted alkalinity leaching; and AC_n = acidity generated from soil N transformations.

In the model catchment, BC_u is set to zero; BC_w = 11.3 meq m^{-2} yr^{-1}; and AC_n = -4.5 meq m^{-2} yr^{-1}. ANC_l is an 'objective function', ie it is defined according to the organism or ecosystem component it is designed to protect. Hettelingh *et al.* (1991) propose critical chemical values for alkalinity leaching of -300 μeq l^{-1} for the protection of forest soils, and +20 μeq l^{-1} for the protection of freshwaters. ANC_l for the model catchment then becomes -408 meq m^{-2} yr^{-1} for forest soils, and +27.2 meq m^{-2} yr^{-1} for waters. Table 1 includes the resulting critical loads.

The steady-state water chemistry method provides another method for determining the critical load for surface waters. One equation (Sverdrup *et al.* 1990) is:

$$CL_{acidity} = (BC^* - ANC_l) \,.\, Q$$

where BC* is the concentration of non-marine base cations in a surface water, and Q is the annual runoff volume in mm. The critical loads for this method are also shown in Table 1, for two values of ANC_l: 20 μeq l^{-1}, as used by Hettelingh *et al.* (1991); and 0, as used in the UK implementation. Table 2 shows the final rate of acid deposition in four of the scenarios modelled by Skeffington and Brown (1992), with summarised outcomes for soil base saturation and stream pH. Figures 2 and 3 show the timecourses of these responses.

Table 1. Calculated critical loads for model catchment

Method	To protect	Critical load (meq m^{-2} yr^{-1})
Steady-state mass balance	Forest soils	424
	Freshwaters	-11.4
	Status quo	*ca* 20
Steady-state water chemistry	ANC = 20 μeq l^{-1}	-21.1
	ANC = 0 μeq l^{-1}	6.1

Table 2 . Rates of acid deposition and outcomes for model catchment

Scenario* (see Figure 1)	Final acid deposition (meq m^{-2} yr^{-1})	Outcome Base saturation	Stream pH
A	60	↓	↓
B	26.6	↓	↑ then ↓
D	10	↑	↑
F	1.8	↑	↑

*Scenario letters follow Skeffington and Brown (1992)

The results can be used to make a number of points.

1. It is still possible to have deterioration even if the critical load is not exceeded. The critical load for the protection of forest soil at 424 meq m^{-2} yr^{-1} is well above that of any scenario, yet Figures 2 and 3 show progressive deterioration of base saturation in scenarios A and B with depositions of 60 and 27 meq m^{-2} yr^{-1} respectively. The lesson here is that it is necessary to be absolutely clear what is being calculated. The critical load is so high because the moderately high water flux through this particular catchment implies that a very large acid generation is needed to reach the critical chemical value. Here, the UN-ECE criteria have deviated considerably from those adopted at the Skokloster workshop (without anyone noticing?), which stated that 'the reserves of exchangeable basic cations should not be depleted' (Nilsson & Grennfelt 1988). The UN-ECE criteria imply that it is acceptable to deplete the reserves of basic cations, provided the critical chemical value is never attained. It is a criterion to protect trees rather than forest soils. To return to the Skokloster criteria, the quantity ANC_l in equation 1 needs to be set to the steady-state output of ANC when base saturation has stabilised. This figure is roughly 4.2 meq m^{-2} yr^{-1} in this particular instance, resulting in a critical load shown as '*status quo*' in Table 1.
2. Conversely, conditions can improve even if the

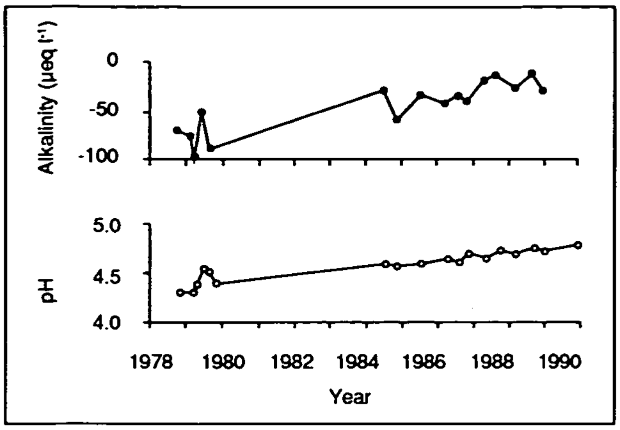

Figure 4. Alkalinity and pH improvement in Loch Enoch, SW Scotland, in spite of the critcal load being exceeded by a large amount (redrawn from Wright & Haus 1991)

critical load is exceeded. Scenario D has a deposition greater than any of the water chemistry critical loads, and yet both pH and base saturation improve steadily. But the critical chemical value is never attained. Figure 4 shows that something similar may be happening in practice; pH and alkalinity of Loch Enoch in south-west Scotland are steadily improving, yet the UK critical load maps for waters show that the critical load for the Loch is exceeded by more than 100 meq m^{-2} yr^{-1}, and will still be exceeded in the year 2005 after a 60% emission reduction.

3. Critical loads can be negative, depending on the critical chemical value adopted as criterion. This particular low weathering rate catchment is incapable of producing water of an alkalinity of 20 μeq l^{-1}, even in the absence of acid deposition. Critical loads are often presented diagrammatically as in line A - B in Figure 5, implying that there is always a value for deposition when the load is not exceeded. A fairer representation would be C - D, with some loads negative.
4. Land use can affect critical loads. Among other things, it affects the term BC$_u$ in equation 1, and will tend to push the line in Figure 5 still further to the left (E - F). An interesting point to consider is whether acidifying land uses (ie farming and forestry) should always have priority over atmospheric deposition in taking up the critical load.
5. The calculated critical loads agree well with the predictions of the MAGIC model. For example, scenario D has a deposition of 10 meq m^{-2} yr^{-1}, and the critical load for zero ANC is 6.1. MAGIC predicts zero ANC is not attained, at least for 500 years. Scenario F has a deposition of 1.8 meq m^{-2} yr^{-1}, which is thus less than the critical load. Zero ANC is attained after 190 years. This result is reassuring, though MAGIC and the critical load models are based on the same principles.

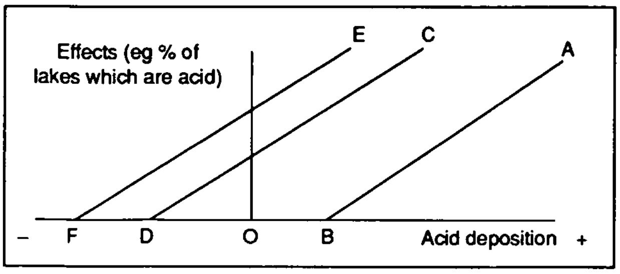

Figure 5. Schematic relationship between acid deposition, ecological effects and the critical load. A - B, as conventionally presented, showing a critical load (O - B) at which no sites are affected; C - D, a more realistic representation, showing that the most sensitive sites can have a negative critical load (O - D); E - F, if land use effects are included, the proportion of sites with negative critical loads increases

To conclude, the critical load as at present formulated does not behave intuitively like a threshold concept. It is necessary to be very clear about the objective to be attained, and how fast one wishes to achieve it. This is the theme of the next section.

WHAT IS THE DESIRED OBJECTIVE OF SETTING A CRITICAL LOAD?

In setting environmental quality standards, which the critical load is meant to be, it is essential to be able to define the end-point which it is desired to reach. Action can then be clearly directed towards this goal, and progress can be monitored. Figure 6 illustrates schematically that the end-point of the critical loads approach is not currently well defined.

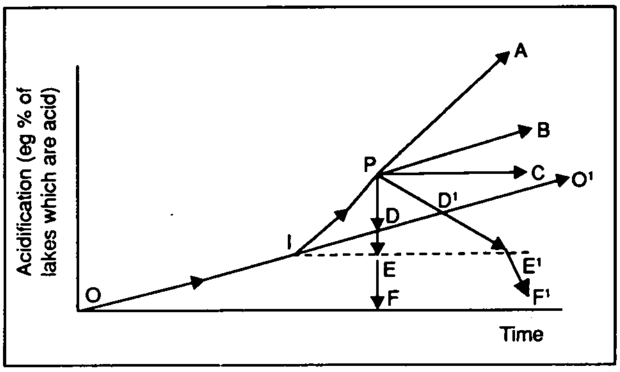

Figure 6. The objective of setting a critical load is unclear, given that, from the Industrial Revolution (I), the natural rate of acidification (O - O^1) has been accelerated up to the present (P), and no action could lead to further acidification (P - A). Is it to reduce to the natural rate again (P - B), to stabilise (P - C), to recover to the state in the absence of intervention by man (P - D), to a pre-industrial state (P - E), to a pristine state (P - F)? How fast does recovery need to be (P - D^1, P - E^1, P - F^1)?

The classic limnological view (eg Pennington 1984) is that post-glacial lake acidification is a natural process in wet areas, exacerbated by such human activities as forest clearance. This process is represented in Figure 6 by O - O^1. (The gradients and proportions in Figure 6 are illustrative only, and not meant to represent the relative proportions of natural and anthropogenic acidification.) At point I, the Industrial Revolution increased the rate of acidification, until we reach the present at point P. If emissions are not reduced, there will be further acidification (P - A). Here, the critical loads approach should guide us as to the degree of emission reduction necessary, but it cannot do so as its aim is not clear. Is it to reduce the rate of acidification to the natural rate (P - B), or to maintain the *status quo* at P (P - C), or to reduce acidification to what it would have been without man's intervention (P - D), or to a pre-industrial state (P - E), or even to an immediate post-glacial state (P - F)? And, if the latter three, on what timescale is it necessary to do it (P - D^1, E^1, F^1)? In discussion at the workshop, the general view appeared to be that for soils the aim should be to preserve the *status quo* and accept post-Industrial Revolution acidification. For waters, the aim appeared to be to restore to a pre-industrial state, which most participants did not distinguish from a post-glacial state. More discussion of these aims would clarify matters, especially as the current UN-ECE methodology for forest soils does not, in fact, maintain the *status quo* (see above).

SOME SCIENTIFIC UNCERTAINTIES

Emission control policies must be based on current understanding, but at the same time it is important to recognise that there are significant scientific uncertainties which may considerably alter both the critical loads and, indeed, the rates of deposition which are used to calculate exceedances. These factors should be the subject of continuing research. A few are discussed briefly below.

Altitude enhancement of deposition

Deposition to the relatively high-altitude areas of the UK is thought to be enhanced by a mechanism known as the seeder-feeder process (see UK Review Group on Acid Rain (UKRGAR) 1990). Clean rain falling from a high-level frontal cloud scavenges pollutants from a low-level cap cloud, resulting in enhancement of deposition in areas prone to hill fog. The official UK deposition estimates include a contribution from this mechanism, resulting in an enhancement of wet deposition relative to the uncorrected figures by over 60% in places. These places are generally those with low critical loads, resulting in high exceedance values. Though the seeder-feeder mechanism appears well established for the relatively unpolluted westerly airflows, its relevance to the easterly flows which supply the most polluted air masses to these areas is unclear (UKRGAR 1990). More work is thus needed to define deposition, as well as the critical load.

The filter factor

The UN-ECE methodology modifies deposition calculated by the European Monitoring and Evaluation Programme model, according to the proportions of land occupied by open ground, deciduous forest and coniferous forest. Different 'filter factors' are applied to each of these types, it being assumed for instance that coniferous forests are 1.8 times as efficient as deciduous forests at capturing SO_2. This parameterisation ignores current knowledge of dry deposition (Fowler, Duyzer & Baldocchi 1991), which suggests trees are no more efficient than low vegetation at capturing SO_2. Enhanced S deposition under trees may instead be due to co-deposition with NH_3 (McLeod *et al.* 1990), and thus be dependent on the presence of fairly local sources of NH_3, which could change the assumed deposition pattern over Europe considerably.

De-acidifying process

The major de-acidification process considered in the critical loads methodology is mineral weathering. It is clear from this volume (see Wilson, pp 79–89 and Sverdrup, Warfvinge & Jönsson, pp 48–61) that much is still controversial in the quantification of this process. Other de-acidification processes exist. Denitrification also removes acidity permanently from the system, and has not been well studied. Ineson (1990) reported rates of around 3 kg N ha^{-1} yr^{-1} in a forest soil, rising to 10-40 kg N ha^{-1} yr^{-1} in two years after clearfelling. Such values could, therefore, be added to any critical load for nitrogen. Nitrogen immobilisation, sulphate adsorption and sulphate reduction are de-acidifying processes which, although they store acidity in soils and sediments, represent a considerable capacity to remove acidity in the medium term. None of these processes are reliably quantified in the present scheme.

Critical chemical values

Critical chemical values for aquatic organisms are fairly well researched, although they are still being expressed in alkalinity terms instead of concentrations directly relevant to living organisms (Skeffington & Brown 1992). For trees, however, critical chemical values must be highly uncertain. Even for well-studied species like Norway spruce *(Picea abies)*, there is controversy over the concentration of Al which is toxic to roots, or whether Al/Ca ratios are important (see Roberts, Skeffington & Blank 1989). For most species, there is no information at all. Research on these aspects is continuing, but needs to be fed into the critical loads process.

Peat soils

Inspection of the UK critical load maps for soils (DoE 1991) reveals that the smallest critical loads tend to be associated with peats. Different techniques need to be used for peat than for mineral soils, because of the absence of a weathering term in equation 1. Only some of the methodology has been published (Skiba & Cresser 1989), but C Smith (pers. comm.) describes how it is based on the assumption of a uniform pre-industrial peat in equilibrium with rain of pH 5 and current calcium deposition. The deposition required to cause a downward shift of 0.2 pH units from this hypothetical situation is calculated as the critical load. This approach is interesting, but begs a large number of questions. Among them are: why 0.2 pH units; was peat ever in equilibrium with pH 5 rain; is it in equilibrium with modern rain; what would happen to peat pH if it was and if deposition was reduced; what is the effect of the different criterion compared with mineral soils (ie acceptable reduction from a hypothetical pristine state compared with maintenance of the *status quo*), and so on. Research into these questions would be both fruitful and interesting.

Validation

Scientific uncertainty is bound to exist, but one way it can be reduced is by paying more attention to validation. Opportunities for validation are often neglected. For instance, Figure 4 seems to show that Loch Enoch is approaching the critical chemical value of zero alkalinity in spite of deposition being well in excess of the critical load. Is the critical load then too low? Are there special circumstances which obviate this conclusion? Has this trend continued? Is a trend visible at other sites with long-term monitoring data? Can the trend be modelled using the standard acidification models? A critical approach to critical loads would be asking these questions, and others. The impression given to politicians and the public that critical loads have been defined once-and-for-all is unfortunate, because it loses us the ability to learn, monitor and modify as scientific knowledge advances.

CONCLUSIONS

Much good work has gone into defining critical loads. It would be a pity if this effort were jeopardised by the deficiencies in the process by which they have been set, and the resulting defects in the critical load values themselves. Work on critical loads should rejoin the mainstream of science and be subject to full and open scientific scrutiny. This work is time-consuming and may occasionally be uncomfortable. It has the immense advantage that the full resources of the scientific community can then be brought to bear on the problem, mistakes spotted and rectified, and a genuine consensus developed. It provides protection against political agenda taking over. The benefits for environmental protection will be well worthwhile.

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Critical loads for vegetation

Problems in determining critical loads for nitrogen in semi-natural vegetation in Britain

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POLLUTION CLIMATE

During the last two centuries, the pollution climate of Britain has changed considerably. Prior to the Industrial Revolution, major atmospheric pollution was associated with the smelting of metal ores, notably of galena, and this resulted in emissions of lead and sulphur dioxide in regions such as the northern Pennines which appear somewhat remote from industrial pollution sources today (see, for example, Raistrick & Jennings 1965). This industry had largely disappeared by the last few decades of the 19th century, but had been replaced by 'the superabundance of factory smoke' (Grindon 1859) producing 'the great smoke cloud of the north of England' (Wilson 1900). Evidence for the existence of this 'cloud' can be detected almost universally in the surface layers of ombrotrophic peat deposits in northern England, Wales and southern Scotland (see, for example, Livett, Lee & Tallis 1979) and in lake sediment cores (see, for example, Battarbee 1984). Ecological effects have included the acidification of freshwaters (Battarbee 1984) and the disappearance of sensitive species (see, for example, Grindon 1859; Tallis 1964). Coal smoke pollution including, most importantly, sulphur dioxide probably reached its peak during the First World War (Ferguson & Lee 1983), but remained high (for at least a century in many parts) until after the Clean Air Acts of the 1950s. During the last four decades, the mean annual concentrations of sulphur dioxide in at least the major towns have fallen by more than an order of magnitude, and are probably everywhere below the levels at which phytotoxic effects can be demonstrated by experimentation. However, changes in fuel usage and combustion technology have led to a marked increase in NO_x in the atmosphere, and this may have resulted in as much as a four-fold increase in nitrate deposition in parts of northern England during the last 100 years (see, for example, Lee *et al.* 1987). Changes in NH_x deposition are more difficult to estimate, but this form of deposition associated with intensive livestock production has been invoked as the cause of major ecological changes in Dutch heathlands (see, for example, Heil & Diemont 1983). The increase in atmospheric NO_x concentration has also resulted in the appearance of tropospheric ozone as a major atmospheric pollutant in Britain in the last few decades (see Photochemical Oxidants Review Group 1987). However, with the possible exception of ozone, alone or in combination with other phytotoxic gases, there is little evidence that gaseous pollutants alone are adversely affecting the growth of plants in semi-natural communities in Britain today.

SEMI-NATURAL VEGETATION IN BRITAIN

The British Isles have been subjected to human occupation for many thousands of years, and, since the Industrial Revolution, the population density has increased markedly. The end result of these factors is that very little or no vegetation exists which has not been affected by man or his grazing animals. The term semi-natural vegetation is usually restricted to those plant communities which have apparently been little altered by human activity. A further complication is that land use has changed (eg in terms of stocking densities) many times in the past, and the extent to which these semi-natural communities have changed and are changing in response is not easy to assess. Change may be induced both by human activity and by natural climatic fluctuations, facts which are perhaps most easily demonstrated in ombrotrophic peatlands by both stratigraphy and the pollen record (see, for example, Godwin 1975). These peat deposits demonstrate that, although some climatic or biotically induced changes may produce rapid responses in plant communities, ecological change may be extremely slow when measured against human lifespan. Thus, it may be difficult to detect change, or to establish a baseline against which change can be measured. Where change can be measured, it may also be difficult to distinguish between natural cyclical processes (eg gap generation and species response) and change induced by other agencies.

Direct evidence that the long history of atmospheric pollution has had major effects on semi-natural plant communities in Britain is rather fragmentary. Perhaps the best evidence for large-scale vegetation change comes from the extensive southern Pennine blanket mires. The disappearance of *Sphagnum* species from these mires is correlated with the appearance of soot in the peat deposits (Tallis 1964), and is probably the result of their sensitivity to sulphur pollution (Lee *et al.* 1987). The apparent sensitivity of lichen species to atmospheric pollution was first noted by Grindon (1859). The disappearance of many sensitive species from regions subjected to appreciable SO_2 pollution and the re-appearance of some species as this form of pollution has declined (see, for example, Seaward 1982) are evidence of the extreme sensitivity of many cryptogams to changes in aerial deposition. Although there is little evidence of direct effects of pollutants on vascular plants in these communities, the disappearance of cryptogams may have affected the competitive relationships between the higher plants, inducing further vegetation change. As many semi-natural communities are dominated by long-lived perennials, vegetation change may still be occurring in response to changes in sulphur pollution. A major difficulty in detecting such change is the general absence of detailed vegetation recording from permanent quadrats. Thus, it may be extremely difficult to establish a baseline against which the effects of the present and potential future pollution climates can be measured.

NITROGEN AS AN ECOLOGICAL FACTOR

Nitrogen is the element derived from the soil which plants require in largest amounts (commonly between 1-4% on a dry weight basis). In many boreal ecosystems, the supply of nitrogen from the soil limits plant growth and distribution (see, for example, Lee, Harmer & Ignaciuk 1983). Many plants are, however, largely independent of the soil supply of nitrogen. Those which have symbiotic associations with nitrogen-fixing bacteria or cyanobacteria may be at a competitive advantage in soils with extremely limited nitrogen supply. Also, many bryophyte species are largely dependent on wet and dry deposition of combined nitrogen from the atmosphere for growth, and these include many *Sphagnum* species which are the dominant peat-forming plants of ombrotrophic mire ecosystems. The ability of these plants to sequester largely the atmospheric nitrogen supply may be an important part of their dominance mechanism (Lee & Woodin 1988).

Evidence that nitrogen availability is limiting plant growth in British semi-natural vegetation is rather fragmentary. There is evidence that the growth of annual strandline plants on coastal dunes may be strongly limited by nitrogen supply (Lee *et al.* 1983), and Willis and Yemm (1961) demonstrated a major growth response to nitrogen in sand dune plants. Heather moorlands in Scotland and Wales may also be strongly limited by nitrogen supply (see, for example, Miller 1979; Lee, Caporn & Read 1992), but it is questionable whether heather moorlands intensively managed for grouse can be considered in the strictest sense as semi-natural. The regular burning regime leads to large losses of nitrogen from the ecosystem, which may induce nitrogen deficiency. Many observations in semi-natural grasslands have demonstrated that nitrogen is not usually the primary limiting element, and that phosphorus deficiency may be more common. However, this view of grassland soil limiting factors presupposes that past increases in nitrogen supply as the result of changes in the pollution climate have had little effect on plant growth.

DETECTING RESPONSES TO THE ATMOSPHERIC NITROGEN SUPPLY

The earliest responses to changes in the nitrogen pollution climate might be expected to be observed in cryptogams which contain the most pollution-sensitive species. Although growth is difficult to measure in many of these species, their ability to accumulate elements from the atmosphere has been used widely to study responses to some forms of pollution (see, for example, Rühling & Tyler 1968). *Racomitrium lanuginosum* forms extensive heaths on mountain summits where it may dominate, and is also a major component of the vegetation of drier hummocks on blanket mires. Baddeley, Thompson and Lee (1993) showed a marked gradient of tissue nitrogen concentration in the stem tips of this moss in Britain. Nitrogen concentrations varied by up to an order of magnitude, with the highest concentrations in moss in northern England close to major conurbations and the lowest in north-west Scotland. This concentration range broadly reflects the variation in atmospheric deposition (UK Review Group on Acid Rain 1990), suggesting that the two are causally correlated. These workers also showed a marked increase this century in the nitrogen concentration of the moss (using herbarium samples), and that transplantation of the moss into districts with high atmospheric nitrogen deposition resulted in a rapid increase in nitrogen concentration. Changes in total tissue nitrogen concentrations in relation to atmospheric nitrogen deposition have also been demonstrated by Pitcairn, Fowler and Grace (1991). These workers and Lee *et al.* (1992) showed that such changes could also be detected in vascular plants.

Growth responses to changes in the atmospheric nitrogen supply have been reported in transplant experiments and in experiments where the atmospheric supply has been increased artificially (see, for example, Press, Woodin & Lee 1986), but convincing evidence of changes in plant growth resulting from 'natural' increases in atmospheric nitrogen deposition is lacking from the UK. However, changes in species distribution have been correlated in southern Sweden and The Netherlands with increased nitrogen deposition (Falkengren-Grerup 1990; Heil & Diemont 1983).

Attempts to demonstrate physiological responses to increased nitrogen deposition have largely concentrated on nitrate assimilation. The substrate-inducible enzyme nitrate reductase provides a measure of nitrate assimilation. Woodin, Press and Lee (1985) showed that in 'unpolluted' atmospheres there was a very close association between wet-deposited nitrate and nitrate reductase activity in *Sphagnum fuscum*, each deposition event inducing enzyme activity. In polluted districts, however, nitrate reductase activity was not inducible in *Sphagnum cuspidatum*, and transplants of the moss into polluted districts rapidly lost their ability to induce activity (see, for example, Lee *et al.* 1987). Thus, the loss of nitrate reductase inducibility may represent a physiological measure of atmospheric nitrogen 'overloading'. However, although this has been demonstrated in other bryophytes (Baddeley 1991; E Sellers, pers. comm.), there is no indication from field perturbation experiments that the same is true for higher plants (Högbom 1992; E Sellers, pers. comm.).

In moss-dominated ecosystems, much of the atmospheric nitrogen supply is probably sequestered by the moss carpet (see, for example, Lee & Woodin 1988). However, in most other communities, although some of the atmospheric nitrogen supply is absorbed in the plant canopy, much of it eventually enters the soil. The potential effects of an increased nitrogen supply on the soil biota are very large. Even within moss mats, an increase in the nitrogen content of decaying stems may accelerate decomposition (Baddeley *et al.* 1993) and thereby reduce mat thickness, with large potential ecological effects (Lee & Woodin 1988). Decomposition processes *within* the soil may be accelerated if the nitrogen supply is increased. Morecroft, Sellers and Lee (1993) showed that an experimentally enhanced nitrogen supply (by 14 g m^{-2} yr^{-1}) resulted in a 25% stimulation of nitrogen mineralisation in both a calcareous and an acidic grassland soil. Thus, the atmospheric supply may both directly and indirectly increase the availability of nitrogen for plant growth. Increased mineralisation has also been associated with the effects of nitrogen deposition on Dutch heathlands (Berendse & Aerts 1984; Berendse *et al.* 1987), leading to the replacement of ericaceous shrubs by grasses.

It is also possible that atmospheric nitrogen deposition may have a detrimental effect on soil micro-organisms. A decline in the macromycete flora in The Netherlands has been associated with high nitrogen deposition (Arnolds 1988), and there are also reports of acidic deposition (including nitrogen deposition) reducing mycorrhizal infection (Gorissen, Joosten & Jansen 1991; Jansen, Dighton & Bresser 1988). However, Lee *et al.* (1992) showed no adverse effect of enhanced nitrogen deposition on mycorrhizal infection in heather *(Calluna vulgaris)*, and at some times of the year these workers have since demonstrated a stimulation of infection. Where nitrogen deposition leads to appreciable soil acidification, then there may be marked detrimental effects on at least some components of the soil biota, but clear evidence for the importance of this phenomenon is difficult to obtain. There is still much to be learnt about the effects of nitrogen deposition on soil processes in semi-natural ecosystems, and this knowledge may be fundamental to the understanding of any major ecological changes observed.

The large changes attributed to nitrogen deposition in Dutch heathlands highlight the potential importance of interactions in causing ecological effects. Thus, the replacement of heather by grasses may result from nitrogen deposition enhancing the population growth of the heather beetle *(Lochmaea suturalis)* (Berdowski & Zeilinga 1987), leading to large-scale defoliation of heather and to grass dominance. Increased supplies of nitrogenous solutes in plant tissues as the result of nitrogen deposition may potentially have a large effect on the population growth of insect herbivores, eg by increasing growth rate, reducing feeding time (thus reducing exposure to parasitism and predation), and increasing fecundity. This potential effect represents a major cause of change in semi-natural ecosystems. However, interactions of nitrogen deposition with the physical environment may be equally important. It has been suggested that nitrogen deposition leads to increased frost and drought sensitivity in heather (see, for example, Van der Eerden *et al.* 1991). In a field nitrogen enhancement experiment in north Wales, however, Lee *et al.* (1992) found that nitrogen increased the frost tolerance of heather. They also demonstrated that the interaction between nitrogen supply and frost tolerance might change over time. Thus, frost tolerance might increase in the autumn and winter in response to enhanced nitrogen supply, but, if the latter caused earlier growth in the spring, then the frost sensitivity might be markedly increased. Pollutant nitrogen deposition to plants may also increase their vulnerability to drought injury. Anecdotal evidence suggests this is true in field heather following ammonium sulphate application, but mechanistic understanding of such effect is incomplete. The effect has been attributed to the reduction in relative sizes of roots in relation to leaves, in which case the demand for water exceeds the supply. However, this hypothesis assumes that leaves do not adjust their use of water, which is principally lost in transpiration, to match its availability. In fact, an increasing body of evidence suggests that roots sense the extent of soil water reserves and send a signal which effects a partial closure of stomata and a consequent drop in water loss (Zhang & Davies 1989). Therefore, a more likely explanation of the effect of nitrogen pollutants on drought tolerance is that they cause an alteration in the normal regulation of stomatal water loss. Changes in stomatal sensitivity to several factors (CO_2, light, abscissic acid) were found in leaves of grasses and trees after exposure to NO_2 and SO_2 (Wright 1988; Lucas 1990; Atkinson, Wookey & Mansfield 1991). Other forms of toxic nitrogen compounds, such as ammonia and foliar-deposited ammonium ions, may well have similar effects on stomata. Much more evidence of the interaction between nitrogen supply and frost and drought sensitivity is required from long-term field experiments before the ecological importance of the phenomenon can be assessed, but frost and drought are major determinants of plant distribution in Britain.

CRITICAL LOADS FOR NITROGEN DEPOSITION

The importance of nitrogen as a plant nutrient is strongly suggestive that there is no threshold below which an enhanced atmospheric nitrogen deposition will not influence ecological processes. Thus, it may not be possible to identify a critical load for nitrogen deposition to semi-natural ecosystems. Rather, there may be a continuum of change induced in response to different rates of atmospheric deposition, and, because any anthropogenic change induced in these ecosystems may be said to be harmful, any current deposition rate may be above the critical load.

Target loads, what might be termed a damage limitation approach, may also be fraught with difficulty. First, there is the problem of deciding whether or not these loads should be assessed on the most sensitive ecosystems in a particular region, and whether or not they should be assessed on the most sensitive processes or organisms within an ecosystem. Second, there is the problem of detecting change in ecosystems which can be attributed to a particular nitrogen load. Third, there is the problem that variation in the proportion of NH_x to NO_x deposition may produce different ecological effects for the same total nitrogen load. For example, NH_x absorption in the plant canopy may cause problems of pH regulation in leaves, because primary assimilation of ammonia is usually confined to the roots, whereas many plants reduce nitrate in their shoots.

Of these problems, the detection of change is the most difficult. There are relatively few long-term measurements of vegetation change in Britain utilising permanent quadrats. Even fewer of these have detailed measurements of all vegetation components; for example, the most sensitive components, the lichens and bryophytes, are often not adequately recorded. A further complication is that variation in recorder 'efficiency' can lead to difficulties of interpretation in long-term data sets. There remains the fact that any changes observed in recent decades may not reflect changes in the nitrogen load, but rather a response to other changes in the pollution climate during this century. Alterations in land management in both uplands and lowlands are also strongly implicated in vegetation change. Change in semi-natural ecosystems will for the most part be slow, and, once set in train, may be difficult to reverse, emphasising the difficulty of determining whether there has already been change in British semi-natural ecosystems as the result of past increases in atmospheric nitrogen deposition.

It is doubtful whether the correlation approach can be regarded as a satisfactory method for setting critical or target loads. In this method, for example, some ecological attribute (thought to result from atmospheric nitrogen deposition) is correlated with the modelled deposition, and an appropriate load to minimise damage is derived. Perhaps the simplest test is correlating plant total tissue nitrogen concentrations with deposition, but, even if these concentrations can be related directly to nitrogen deposition, there are problems in defining an appropriate concentration to minimise damage.

There is probably no substitute for long-term perturbation experiments designed, at least to some extent, to mimic deposition processes. The vast majority of nitrogen addition experiments have involved a large single dose of nitrogen fertilizer, bearing no relation to deposition loads. Such experiments have their value, but there is an urgent need for other experiments where smaller doses are applied at regular intervals (weekly/monthly) over several years, combined with regular monitoring of vegetation and soil responses. A number of such experiments are under way in British upland and lowland heaths *(Calluna)* and in limestone and acidic grasslands. These experiments have already been running from two to four years, and a number of early responses to the range of nitrogen deposition treatments have been observed. The value of these experiments will increase with time, and should provide a firmer basis from which to apply the critical loads approach.

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The example of calcicolous grasslands and nitrogen

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RESPONSE IN A COMMUNITY CONTEXT

In most semi-natural vegetation, plants respond to shifts in the regime of nutrients and pollutants, in a community context, in association with the other plant species with which they grow. Even if a species has a simple strategic response to such changes, this must necessarily be played out in competition with its associates in the variety of vegetation types in which the species is found.

Such variety can be considerable. Yorkshire fog (*Holcus lanatus*), for example, a grass that is very responsive to increases in nitrogen, has been recorded in over 90 plant communities in Britain (Rodwell 1991), each a different assemblage of species providing a particular context for plant growth and reproduction. These include sea-cliff grasslands, chalk down pastures, neglected fens, rank sub-scrub and broadleaved plantations: a group of very varied vegetation types. Also, common birdsfoot-trefoil (*Lotus corniculatus*), a low-growing herb which is readily overwhelmed by the lush growth of mesophytes like Yorkshire fog, is found in 45 different plant communities, where it may succumb to enrichment of the soils among some quite contrasting assemblages.

THE CHARACTER AND DISTRIBUTION OF VULNERABLE VEGETATION

Characterising such suites of plant communities has been made simpler by the completion of the National Vegetation Classification (NVC), the first systematic and comprehensive account of Britain's vegetation types from all natural, semi-natural and major artificial habitats (Rodwell 1991). Using this scheme, for example, it is possible to identify those plant communities which are in some way dependent upon the maintenance of impoverished soil and groundwater conditions, and so are likely to be affected by nitrogen enrichment. Mapping the range of such communities, grouped together in broadly defined categories like 'calcicolous grasslands' and 'bogs and wet heaths', can thus give a general indication of the distribution of surviving stands of vulnerable semi-natural vegetation, a valuable enhancement of map data derived from soil and species distributions (Davison & Barnes 1991).

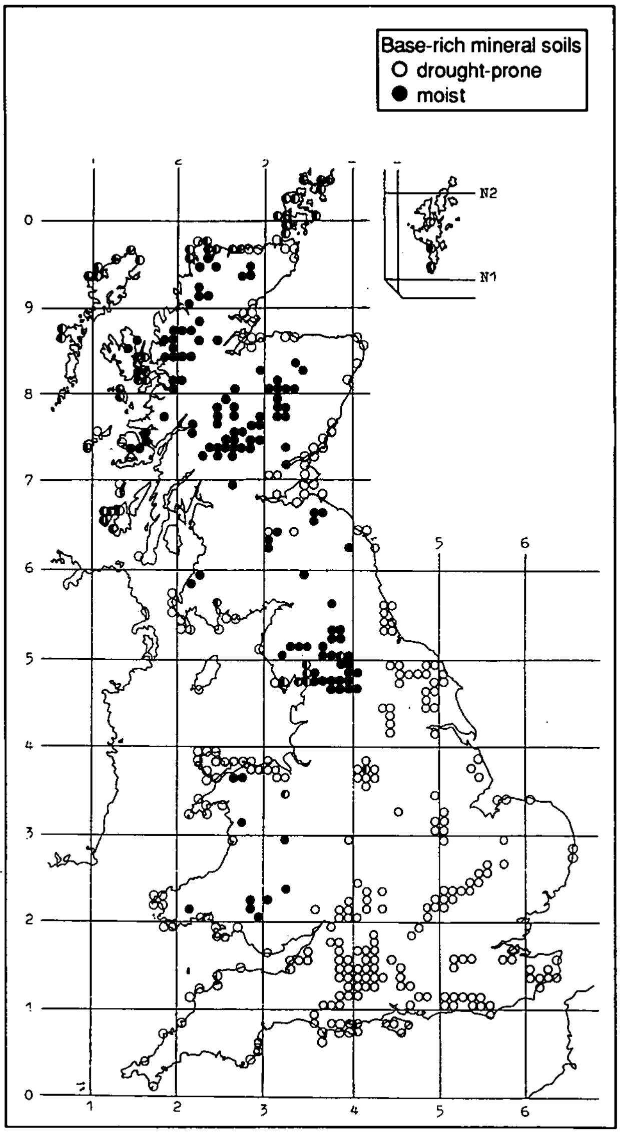

Figure 1. Calcicolous grasslands and dwarf-herb vegetation of drought-prone and moist base-rich mineral soils

Figures 1-5 show such vegetation maps prepared from distribution data for five broad groupings of plant communities, held in the UK vegetation data base at the University of Lancaster:

- calcicolous grasslands and related dwarf-herb vegetation of impoverished base-rich mineral soils (17 communities);
- calcifugous grasslands and dry heaths of impoverished base-poor mineral soils (26 communities);

- unimproved pastures and meadows, fen meadows and tall-herb ledge vegetation of more nutrient-poor neutral mineral soils (10 communities);
- calcicolous flushes and fens of wet and impoverished base-rich peats and peaty mineral soils (11 communities);
- bogs, wet heaths and soligenous mire vegetation of wet and impoverished base-poor peats and peaty mineral soils (27 communities).

The maps for calcicolous and calcifugous grasslands also make a distinction, using hollow and solid circles, between swards characteristic of drought-prone and moist soils respectively.

QUANTIFYING THE PATTERN OF EXPOSURE

By superimposing nitrogen deposition contours on to such maps, it is possible to quantify crudely the patterns of exposure of the different vegetation types to various levels of nitrogen. Figure 6 shows the contours for total annual nitrogen deposition as published by Williams *et al.* (1989), and Table 1 gives the percentage distribution of the vegetation types among the various deposition zones. It can thus be seen that 33% of our calcicolous grasslands on drought-prone base-rich mineral soils lie within the highest deposition zone, but only 11% of the bogs, wet heaths and soligenous mires on wet base-poor peats and peaty mineral soils.

The detailed NVC community accounts (Rodwell 1991) can then help define the particular character of the vulnerable resources in each deposition zone, and provide some indication of their value for conservation, amenity or productive agriculture. For example, the bogs and wet heaths within the highest deposition zone represent a small proportion of the total of this resource throughout Britain as a whole, and include many stretches of impoverished vegetation with little conservation or landscape value. However, they also include some precious remnants of lowland valley bog and raised mire, vegetation which is already under threat from draining practices and which provides a locus for such declining

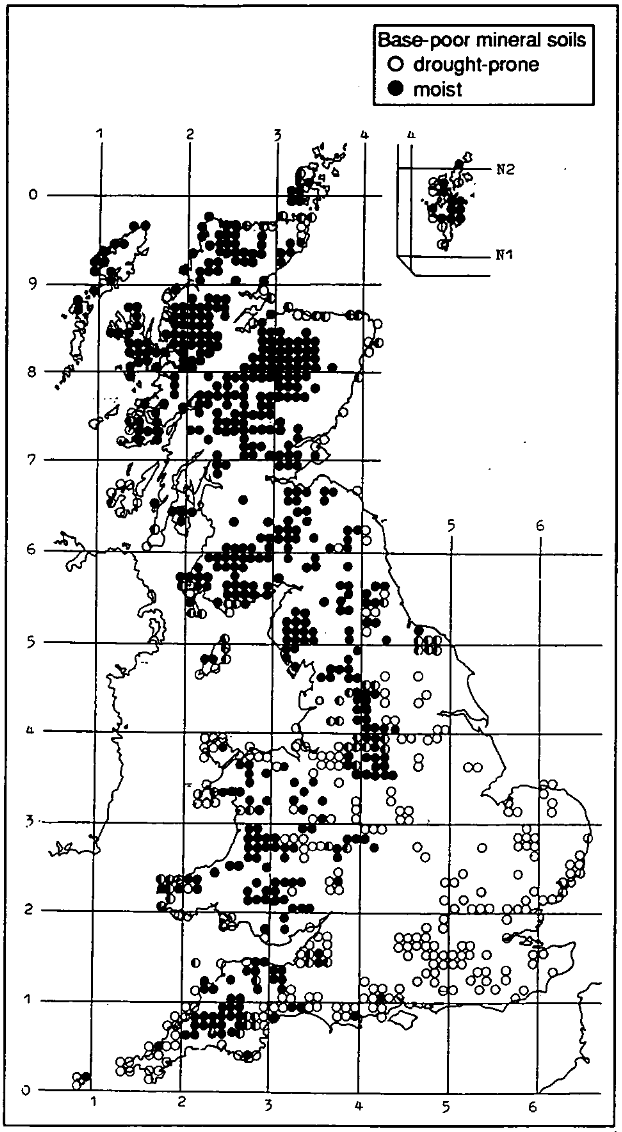

Figure 2. Calcifuge grasslands and dry heaths of drought-prone and moist base-poor mineral soils

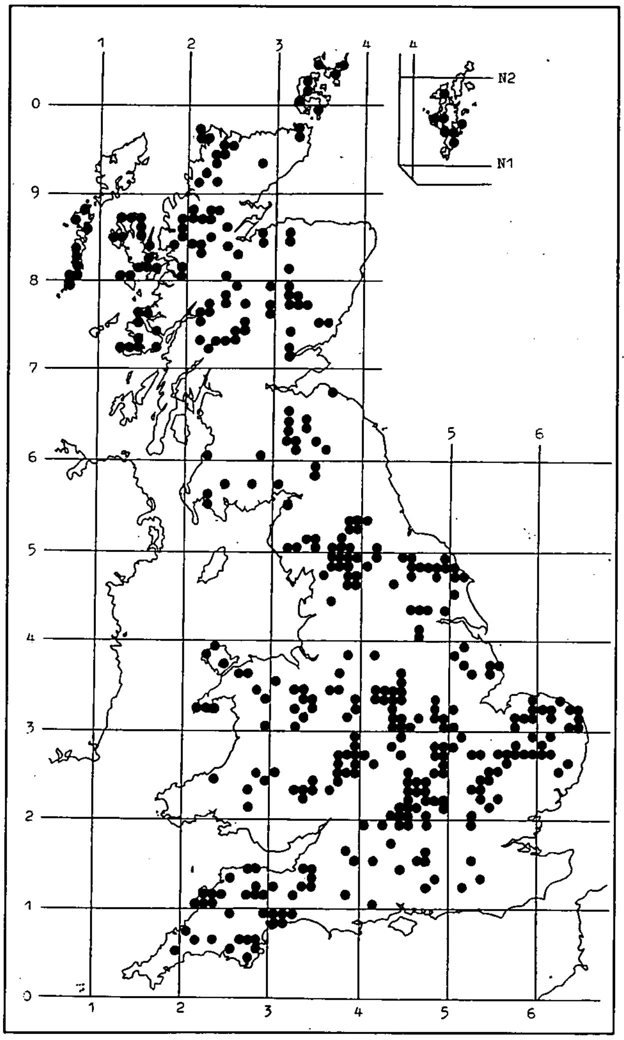

Figure 3. Unimproved pastures and meadows, fen meadows and tall-herb ledge vegetation of circumneutral mineral soils

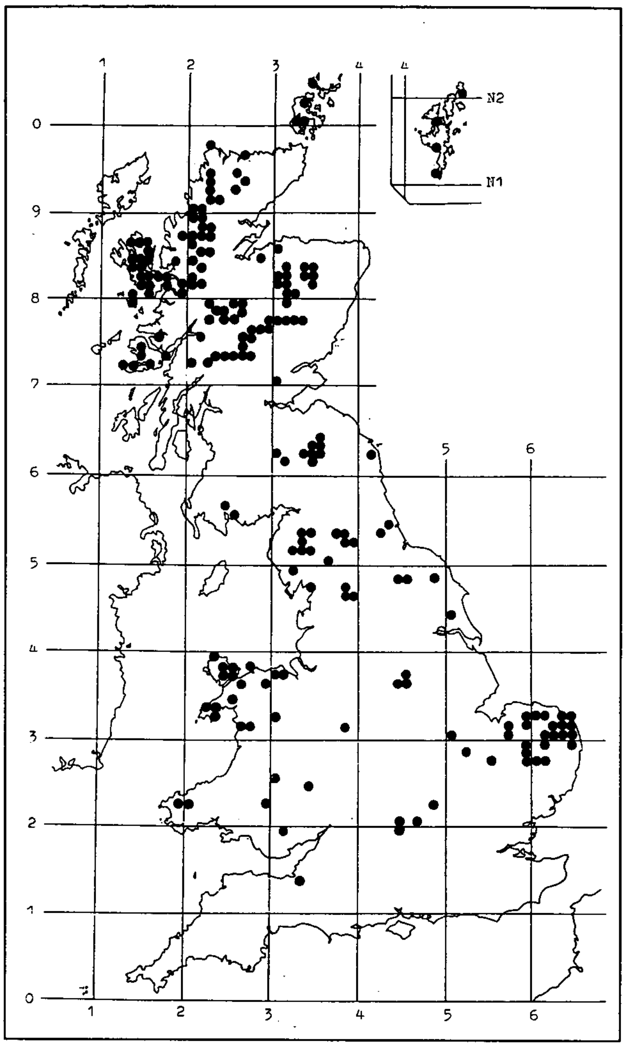

Figure 4. Calcicolous flushes and fen vegetation of base-rich peats and peaty mineral soils

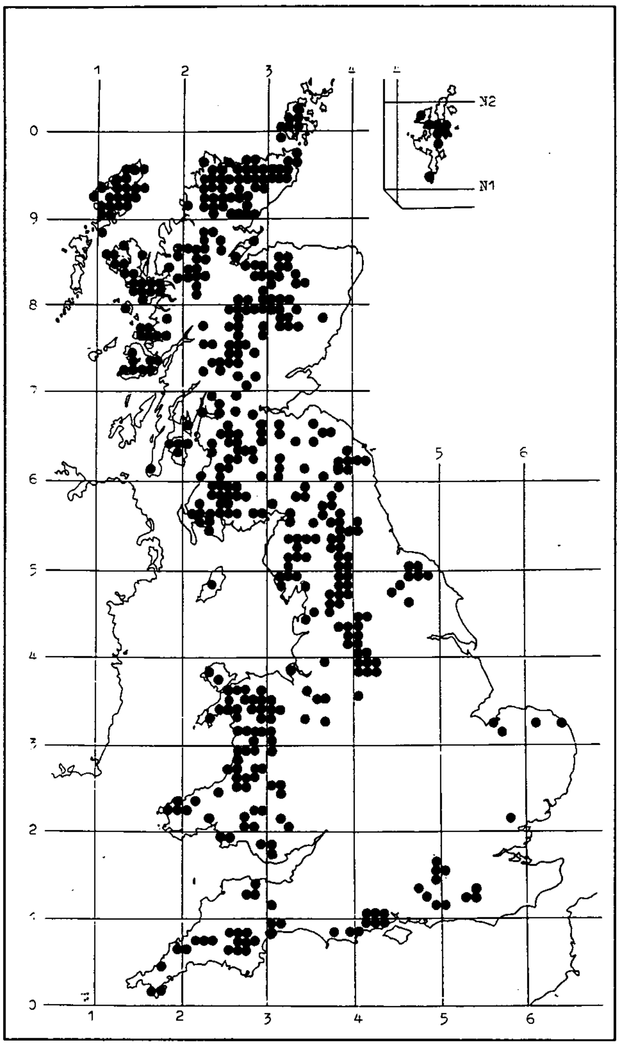

Figure 5. Bog and wet heath vegetation of base-poor peats and peaty mineral soils

Table 1. Percentage distribution of major vegetation types in different nitrogen deposition zones

Major vegetation type	Total annual nitrogen deposition (kg ha^{-1}) <8	8-12	12-16	16-20	>20
Calcicolous grasslands and dwarf-herb vegetation of drought-prone base-rich mineral soils	4	13	19	31	33
Calcicolous grasslands and dwarf-herb vegetation of moist base-rich mineral soils	14	53	14	15	4
All calcicolous grasslands and dwarf-herb vegetation of base-rich mineral soils	8	27	17	25	23
Calcifugous grasslands and heaths of drought-prone base-poor mineral soils	2	7	26	27	38
Calcifugous grasslands and heaths of moist base-poor mineral soils	12	33	31	12	12
All calcifugous grasslands and heaths of base-poor mineral soils	8	24	29	18	21
Unimproved pastures and meadows, fen meadows and tall-herb ledges of neutral mineral soils	7	19	14	19	41
Calcicolous flushes and fen vegetation of base-rich peats and peaty mineral soils	13	38	14	15	20
Bogs and wet heath vegetation of base-poor peats and peaty mineral soils	12	31	27	19	11

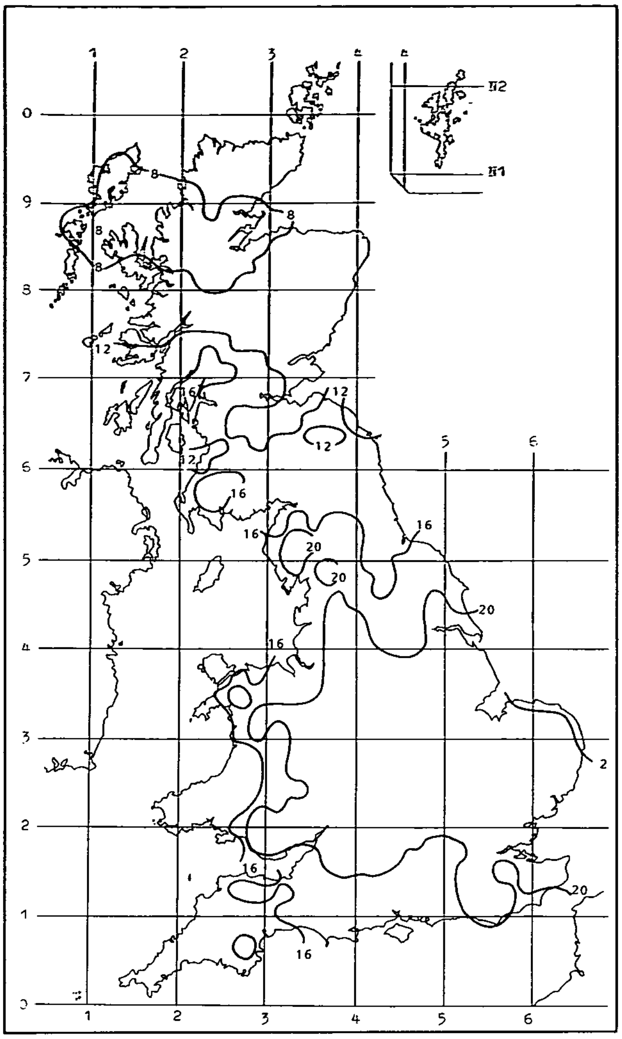

Figure 6. Total annual nitrogen deposition (kg ha^{-1})

species as brown beak-sedge (*Rhynchospora fusca*), marsh clubmoss (*Lycopodium inundata*), bog orchid (*Hammarbya paludosa*), and marsh rosemary (*Andromeda polifolia*).

PREDICTING PARTICULAR IMPACTS OF NITROGEN ENRICHMENT

The NVC can also help predict the impact of nitrogen enrichment on vulnerable kinds of vegetation. One possible response to enhanced nitrogen deposition is that some potentially aggressive plant, already present in the vegetation in small amounts, might expand its cover, with consequent adjustments in the representation of the associated plant species. On the Chalk of Limburg in The Netherlands, for example, much attention has focused on the increase in recent years of tor grass (*Brachypodium pinnatum*), a bulky plant whose expansion has ousted smaller companions to produce rank and impoverished swards. This spread has been attributed, at least in part, to enhanced nitrogen deposition from the atmosphere (Bobbink 1989; Bobbink & Willems 1987, 1988; Bobbink, Bik & Willems 1988). If such deposition were to be repeated here, we can predict its effect by comparing the existing floras of British grasslands which have very little tor grass, those in which it is a frequent but low-cover plant, and those where it is already abundant (Table 2).

Table 2. Changes in the frequency of species in calcicolous grasslands with increase in frequency and cover of tor grass *(Brachypodium pinnatum)*

Plant name	Grassland type CG2a	CG4a	CG4b
Sheep's fescue *(Festuca ovina)*	V	IV	V
Carnation grass *(Carex flacca)*	V	IV	IV
Birdsfoot-trefoil *(Lotus corniculatus)*	IV	III	IV
Salad burnet *(Sanguisorba minor)*	V	III	IV
Harebell *(Campanula rotundifolia)*	III	III	IV
Purging flax *(Linum catharticum)*	III	III	III
Common rockrose *(Helianthemum nummularium)*	III	IV	III
Quaking grass *(Briza media)*	IV	III	III
Mouse-ear hawkweed *(Hieracium pilosella)*	IV	III	III
Wild thyme *(Thymus praecox)*	IV	IV	II
Meadow oat *(Avenula pratensis)*	IV	IV	
Crested hair-grass *(Koeleria macrantha)*	V	III	I
Stemless thistle *(Cirsium acaule)*	IV	III	I
Spring sedge *(Carex caryophyllea)*	III	III	I
Squinancy wort *(Asperula cynanchica)*	III	II	
Eyebright *(Euphrasia officinalis* agg.*)*	II	II	I
Cock's-foot *(Dactylis glomerata)*	II	II	I
Hairy violet *(Viola hirta)*	II	II	I
Small scabious *(Scabiosa columbaria)*	III	I	I
Common milkwort *(Polygala vulgaris)*	II	I	I
Dropwort *(Filipendula vulgaris)*	II	I	I
Horse-shoe vetch *(Hippocrepis comosa)*	II	I	
Burnet saxifrage *(Pimpinella saxifraga)*	II	I	
Bulbous buttercup *(Ranunculus bulbosus)*	II	I	
Chalk milkwort *(Polygala calcarea)*	I		
Bastard toadflax *(Thesium humifusum)*	I		
Field fleawort *(Senecio integrifolius)*	I		
Gentian *(Gentianella anglica)*	I		
Tor grass *(Brachypodium pinnatum)*	I	V	V
		Rise in frequency and cover of tor grass	

CG2a Fescue/oat grassland, *Cirsium/Asperula* subcommunity
CG4a Tor grass grassland, *Avenula/Thymus* subcommunity
CG4b Tor grass grassland, *Centaurea/Leontodon* subcommunity

Roman numerals I-V indicate the frequency of species in the samples of each vegetation type:
I = 1-20% frequency, II = 21-40%, III = 41-60%, IV = 61-80%, and V = 81-100%

In other situations, nitrogen enrichment through agricultural improvement of soils using manures and fertilizers has converted diverse and unproductive turf into more species-poor and luxuriant swards. Adjacent fields on the southern English chalk downs, for example, differing only in the fact that one has been grazed and manured by cattle rather than sheep or otherwise fertilized, can show the kinds of contrasts in species composition illustrated in the left-hand and central columns of Table 3.

Table 3. Shifts in species composition with the eutrophication of calcicolous grassland

	Grassland type		
Plant name	CG2a	CG2c	MG6c
Ribwort *(Plantago lanceolata)*	IV (1-7)	V (1-6)	IV (3-4)
Sheep's fescue *(Festuca ovina)*	V (1-9)	V (2-9)	I (7)
Carnation-grass *(Carex flacca)*	V (1-8)	V (1-8)	
Salad burnet *(Sanguisorba minor)*	V (1-8)	IV (1-8)	
Crested hair-grass *(Koeleria macrantha)*	V (1-6)	V (1-4)	
Quaking grass *(Briza media)*	IV (1-9)	V (1-5)	
Birdsfoot-trefoil *(Lotus corniculatus)*	IV (1-6)	V (1-6)	
Meadow oat *(Avenula pratensis)*	IV (1-7)	IV (1-7)	
Rough hawkbit (*Leontodon hispidus)*	IV (1-7)	IV (1-5)	
Purging flax *(Linum catharticum)*	III (1-4)	IV (1-3)	
Mouse-ear hawkweed *(Hieracium pilosella)*	IV (1-7)	III (1-5)	
Small scabious *(Scabiosa columbaria)*	III (1-5)	IV (1-5)	
Wild thyme *(Thymus praecox)*	IV (1-7)	III (1-7)	
Common rockrose *(Helianthemum nummularium)*	III (1-8)	II (1-4)	
Bulbous buttercup *(Ranunculus bulbosus)*	II (1-4)	II (1-3)	
Upright brome *(Bromus erectus)*	II (1-4)	II (1-4)	
Dropwort *(Filipendula vulgaris)*	II (1-6)	II (1-5)	
Stemless thistle *(Cirsium acaule)*	IV (1-7)	II (1-7)	
Squinancy wort *(Asperula cynanchica)*	III (1-7)	I (1-3)	
Horse-shoe vetch *(Hippocrepis comosa)*	II (1-6)	I (3)	
Gentian *(Gentianella amarella)*	II (1-4)	I (1-3)	
Common milkwort *(Polygala vulgaris)*	II (1-4)	I (1)	
Yorkshire fog *(Holcus lanatus)*	I (1-5)	IV (1-6)	IV (2-5)
White clover *(Trifolium repens)*	I (1-4)	IV (1-7)	V (2-4)
Cock's-foot *(Dactylis glomerata)*	II (1-5)	III (1-6)	IV (2-7)
Crested dog's-tail *(Cynosurus cristatus)*	I (1-5)	III (1-6)	IV (2-8)
Yellow oat *(Trisetum flavescens)*	I (1-5)	III (1-5)	V (3-4)
Rye-grass *(Lolium perenne)*	I (1)	I (1-3)	V (4-7)
Red fescue *(Festuca rubra)*	I (2-9)	I (1-5)	IV (4-6)
Common bent-grass *(Agrostis capillaris)*	I (2-4)	II (1-7)	IV (4-8)
Common mouse-ear chickweed *(Cerastium fontanum)*	I (2-3)	I (1)	IV (1-3)
Timothy *(Phleum pratense bertolonii)*	I (1-2)	II (1-4)	V (2-5)
Smooth-stalked meadow-grass *(Poa pratensis)*	I (1-3)	I (1-3)	III (1-5)
Creeping thistle *(Cirsium arvense)*	I (1-6)	I (1-3)	III (3-4)
Daisy *(Bellis perennis)*	I (1-4)	I (1-3)	II (2-4)
Dandelion *(Taraxacum officinale)*	I (1-3)	I (1-3)	II (1-3)
Rough-stalked meadow-grass *(Poa trivialis)*			II (1-3)
Number of samples	343	167	10
Number of species/sample	30 (7-45)	30(16-44)	14(11-20)

CG2a Fescue/oat grassland, *Cirsium/Asperula* subcommunity
CG2c Fescue/oat grassland, *Holcus/Trifolium* subcommunity
MG6c Lolio/Cynosuretum, Trisetum subcommunity

Roman numerals I-V indicate the frequency of species in the samples of each vegetation type: I = 1-20% frequency, II = 21-40%, III = 41-60%, IV = 61-80%, and V = 81-100%
Arabic numerals give a measure of the abundance of species among the samples of each vegetation type using the Domin scale: 1 = less than 4% cover, 10 = 91-100% cover

On base-rich soils where the supply of phosphate was not too limiting, eg on brown calcareous earths with a mull humus regime, rather than on harsh rendziniform soils, similar changes might be expected with enhanced deposition of atmospheric nitrogen. Signs to watch for then would be increased frequency and cover of such plants as Yorkshire fog, white clover (*Trifolium repens*), cock's-foot (*Dactylis glomerata*), crested dog's-tail (*Cynosurus cristatus*) and yellow oat (*Trisetum flavescens*), and a reduction in the abundance of light-demanding mat-formers, some rosette plants, and smaller pauciennials needing gaps in the turf.

A third possibility might be expected where nitrogen enrichment was affecting calcicolous grasslands on very shallow rendzinas prone to drought in summer, a situation already widespread on limestones in the warmer and drier south-east of Britain (Figure 1), but likely to become more frequent and severe with the projected climatic changes. In such circumstances, we might expect a loss of drought-prone perennials from the swards and the periodic patchy prominence of weed communities, with coarse nitrophilous species like stinging nettle (*Urtica dioica*), burdocks (*Arctium* spp.) and mullein (*Verbascum thapsus*).

ANTICIPATING COMPLICATIONS

We may need to think, then, of a variety of possible responses to nitrogen enrichment, even among a restricted range of vegetation types like calcicolous grasslands. The NVC can also help anticipate further complications in these responses, caused for example by interactions between enrichment and soil acidification. It is often supposed that such acidification is unlikely to occur on soils so well buffered by the continual release of lime from the underlying calcareous parent materials. However, the NVC can characterise some rather precise situations where acidification might occur beneath calcicolous swards and where attention could be focused on the impact of forms of nitrogen likely to enhance this process.

Within the high nitrogen deposition zones, for example, there are extensive deposits of loess on limestones which do not impair the free-draining character of the soils, but which effectively muffle the overwhelming influence of the bedrock on the profile, certainly at the soil surface. Where such soils have developed in areas of higher rainfall, the tendency to surface leaching is enhanced, a feature very well seen on the Wiltshire chalk where loess occurs widely with annual precipitation approaching 1000 mm. The distinctive character of the grasslands developed in this area was first described by Wells (1975) and emerges clearly in the broader context of the NVC, marked by the occurrence of such species as betony (*Stachys officinalis*), saw-wort (*Serratula tinctoria*) and devil's-bit scabious (*Succisa pratensis*), all of these being plants indicative of a pH transitional to mildly acidic. In fact, these swards are also seen to be distinctly mesotrophic already, a feature which might be attributed to their being generally grazed by cattle as opposed to sheep. Such a vegetation type would thus be well suited to pursuing this particular problem.

THE NEED TO MONITOR

The use of the NVC as a framework for predicting and monitoring responses of this kind related to nitrogen deposition is being developed in a research project of the UK Department of the Environment, under its Critical Loads and Climate Change Programmes. A national network of 54 monitoring stations is being established, with 129 permanently marked plots of 12 m x12 m located in representative stands of the various types of calcicolous grasslands characterised by the NVC and distributed across the full range of deposition zones (Figure 7). The first round of recording is now well advanced, with frequency data for all vascular plants, bryophytes and macrolichens being collected from 30 quadrats, 50 cm x 50 cm, positioned on a stratified systematic grid within each of the large plots (Rich *et al.* 1991, 1992). The NVC expert system MATCH (Malloch 1992) is being used to provide a precise profile of the vegetation in each plot at the start of monitoring and to track any shifts in species composition at each subsequent recording.

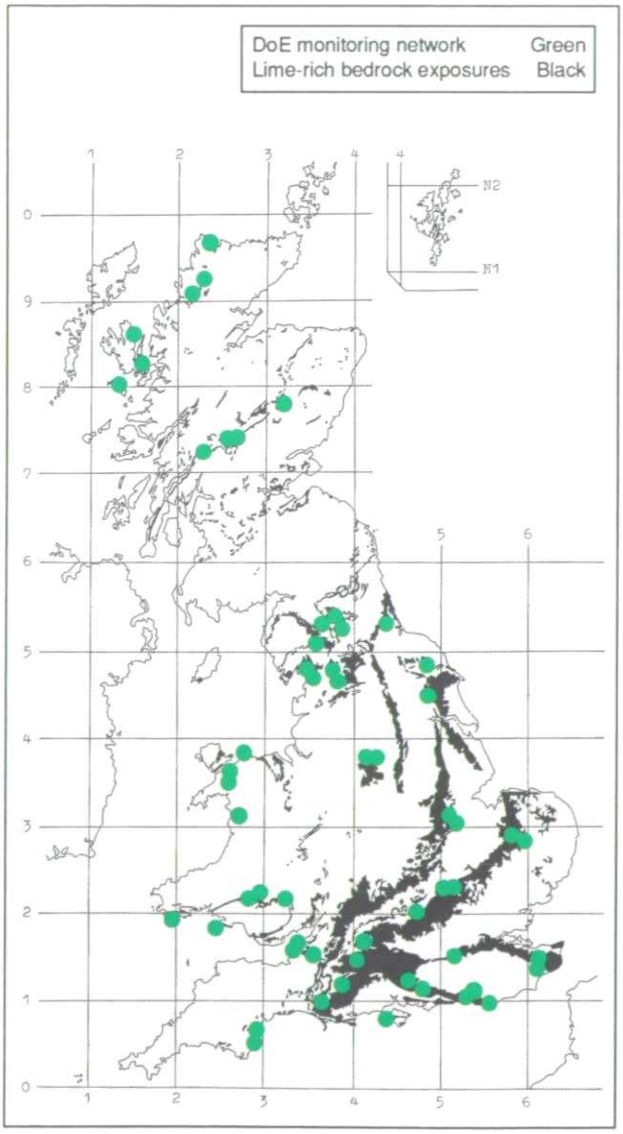

Figure 7. Department of the Environment monitoring network for calcicolous grasslands

OTHER REQUIREMENTS

Insofar as it is possible, monitoring plots have been located at sites with some security of ownership and stability of land management. With calcicolous grasslands, the particular concern is to limit variations in sward composition caused by changes in grazing treatments, because these could confound or overwhelm responses to a shifting nitrogen regime. Indeed, there is some suggestion that expansion of the relatively unpalatable tor grass in calcicolous pastures is influenced as much by the present use by farmers of 'choosy' stock, rather than the earlier less discriminating breeds, as it is by enhanced nitrogen deposition (see During & Willems 1984; Bobbink & Willems 1988). We urgently need a parallel investigation of the responses of the range of British swards to various grazing regimes using exclosures with, if possible, some contrasting kinds of stock, so as to isolate pollution effects and study interactions between these various influential factors.

Another important requirement is for experimental manipulations of the nitrogen regime on this same range of swards using a variety of forms of nitrogen. We know from the Rothamsted Park Grass Experiment (Brenchley 1958; Pigott 1982) just how informative such manipulations can be for understanding more widespread changes in semi-natural grasslands, yet we have hardly begun to investigate just how much nitrogen is required, and in what form, to produce the sorts of shifts in sward composition described above. The possibility of using widely accepted baseline definitions of grasslands, such as those provided by the NVC, would give this kind of experimentation a much firmer foundation.

More widely, we need to accept the fact that critical loads of pollutants should be investigated and defined in relation to vegetation and soils together. For too long we have treated these systems in isolation: the NVC shows just how subtly vegetation relates to climate and soils together.

Finally, we should ask a simple question about critical loads. Calculating the cost of exceedance becomes a little more achievable using the kinds of predictive model described above, but a particular response to a critical load of, say, nitrogen might be perceived as deleterious by one group of interests but beneficial by another. With the eutrophication of calcicolous grasslands, for example, often followed through to produce the kind of sward summarised in the right-hand column of Table 3, the loss in conservation value with the reduction in species diversity is only too apparent. For agriculture, however, exceeding the critical load results in a highly productive, luxuriant herbage, a gain by any grazier's standard. Critical then, but critical for whom?

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Critical loads of acidity and sulphur for soils

The provisional map of critical loads of acidity for the soils of Great Britain

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BACKGROUND

The provisional critical load map for the soils of Great Britain (Figure 1) has been developed using principles and proposals developed at two workshops, held in 1986 and 1988. The first workshop suggested that critical loads for soils should be set to prevent chemical changes which would lead to long-term harmful effects to the soil/plant system (Nilsson 1986). It also suggested that such chemical changes could be prevented, if acid inputs did not exceed internal (within the soil) alkalinity production, essentially the production of base cations by mineral weathering. The main factor determining the rates of weathering in soils is soil mineralogy. The 1988 workshop, at Skokloster in Sweden, divided soil materials into five classes, defined on the basis of their dominant weatherable minerals (Nilsson & Grennfelt 1988). Critical loads were then assigned to these classes, according to the amount of acidity which would be neutralised by base cation production, by weathering from the relevant minerals. It was also proposed that the critical load for a given soil could be adjusted, within the range of values suggested for each class of soil material, by applying modifying factors. Thus, if one of the 'decrease' factors applies, eg high precipitation, the value of the critical load is used which is at the lower limit of the values suggested for the given soil material class; if an 'increase' factor applies, then the upper value is used.

Given the excellent soil maps and accompanying data bases available for Britain, it was felt that sufficient information might be available for map units on existing, published, maps to be allocated to one of the soil material classes defined at the Skokloster workshop; the map units could then be assigned a critical load. If this allocation were possible, it would provide a relatively rapid method for producing a provisional critical load map for British soils. The approach was first tested in two exploratory studies.

EXPLORATORY STUDIES

In late 1988, an exploratory study was carried out in Wales to see if sufficient data were available to allow the soils of two test areas to be allocated to one of the five classes defined at Skokloster and then to produce a critical load map for soils. The two test areas were located in part of north-west Wales, including Snowdonia and the country around Llyn Brianne. The 1:250 000 soil map of Wales and its supporting memoirs and data bases were used as the basis for the exercise. The map units on this map are soil associations, which can contain a range of soil

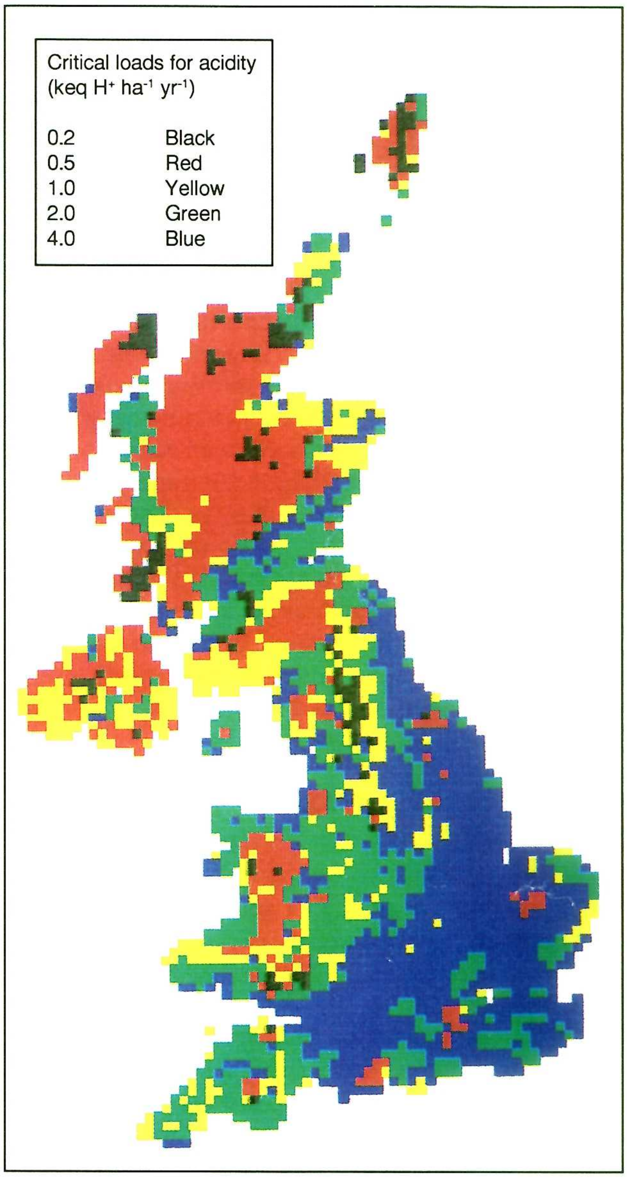

Figure 1. Critical loads of acidity for soils in Great Britain

types with different chemical and mineralogical properties, and hence critical loads. In the trial, map units were allocated to one of the critical load classes proposed at Skokloster on the basis of the mineralogy of the dominant soil type (series) within each map unit, thus converting the soil maps to critical load maps. The resulting maps of critical loads were then overlaid with deposition data, derived from a national monitoring programme carried out by the Welsh Water Authority, to produce an exceedance map. The exercise showed that the approach based on the Skokloster classes was feasible but that some modification of the Skokloster classes would be necessary to include a wider range of minerals. It also highlighted the fact that organic soils could not be allocated to one of the Skokloster classes as they contain very few weatherable minerals.

The approach was tested further in 1989 using a test area stretching from the Cumbrian coast to the coast of County Durham and north Yorkshire. This area included a wider range of soils than the Welsh sites. The north of England study also considered (i) the possible impact of land use on critical loads of soils, in particular the addition of lime, and (ii) the importance of including total deposition when producing exceedance maps. As in the Welsh example, the study suggested that the approach based on the Skokloster classes could be adapted to encompass a wide range of soils, with suitable, relatively minor, modifications to the mineralogical criteria used to define the five classes. The land use-related work confirmed that, if allowance were made for the addition of lime, the critical load of large areas of soil would be increased, albeit in the short term. Inclusion of total deposition, as opposed to wet deposition only, greatly increased the area of exceedance, and underlined the need for comprehensive deposition data.

THE GREAT BRITAIN MAP

Beginning in late 1989, the approach developed in the two trial studies was built upon to provide a critical load map of acidity for soils for the whole of Great Britain; the work was carried out as a collaborative exercise by ITE, the Soil Survey and Land Research Centre, the Macaulay Land Use Research Institute and the University of Aberdeen. As in the trial studies, the 1:250 000 soil maps were used as the base, with the map units on them allocated to one of the five Skokloster classes, plus a class of peat soils, on the basis of the dominant soil series within the map unit.

In Scotland, the map units could be allocated to one of the classes using essentially the same approach as in the two exploratory studies. The soil associations used on the 1:250 000 scale soil maps of Scotland comprise groupings of soils developed on a given parent material. Information on the mineralogy of the parent materials could, therefore, be used to allocate the soils of a given association to a soil material class, and hence to assign a critical load. The initial, mineralogically based allocation was then modified, where necessary, using information on soil depth, texture and drainage, as suggested at the Skokloster workshop. Thus, where a map unit was dominated by shallow soils on steep slopes, the sensitivity class based on mineralogy was increased by one class. Conversely, in units dominated by deep soils or soils inundated regularly the class was decreased by one (Langan & Wilson, pp 40–47).

Many of the soils in England and Wales, but especially in south-east England, are developed on reworked materials. These soils seldom contain the primary minerals used in the definition of the soil material classes at Skokloster; however, they commonly contain large amounts of clay minerals. A modified schema was, therefore, developed which incorporated texture and secondary minerals into the definition of the soil material classes (Loveland, pp 48-53).

The trial exercises had essentially converted the soil map to a critical load map using the actual boundaries on the map to delimit areas with differing critical loads. The magnitude of the GB exercise enforced a different approach based on machine-readable data. One km square cells of the Ordnance Survey's National Grid of Great Britain were used as the basic units to build the map. Each 1 km square grid was allocated to one of the six classes on the basis of the dominant soil unit within the square. The resultant data bases for England and Wales, and for Scotland, were then forwarded to the UK Mapping Centre where they were merged to produce a critical load map for Britain, based on 1 km square cells.

As noted above, a major weakness with the Skokloster classification from the GB point of view was that it did not consider peats. As a result, the exploratory study in the north of England and the initial GB map had large areas of land in a sixth class of 'peat soils', to which no critical load values were assigned. An approach for assigning critical loads for dystrophic peats was eventually developed based on work at the University of Aberdeen. It began with an examination of the relationship between peat acidity in Scotland, or base cation depletion, and current inputs of acidic deposition. The study showed a broad relationship between current peat pH and current rainfall pH. Later laboratory studies assessed the impact of acidic inputs on peat pH by equilibrating peat samples with solutions representing a spectrum of rainfall chemistries. The work demonstrated that both H^+ and Ca^{++} inputs had to be taken into account when calculating critical loads for dystrophic peats.

The resultant method of calculating the critical load uses the relationships derived from the equilibration studies plus data on H^+ and Ca^+ deposition at pristine conditions and the concept of an acceptable pH shift. This latter was set at 0.2 pH units and the critical load was calculated as that input of H^+ and Ca^{++} which would not result in a maximum reduction in the pH of the given area of peat of 0.2 pH units. Using this approach, a critical load was calculated for each 1 km square in GB in which dystrophic peat was the dominant soil (Cresser, Smith & Sanger, pp 34–39). Eutrophic peats were assigned to a critical load class primarily on the basis of data for lowland peats presented by Burton and Hodgson (1987), and taking into account land use and management practices and, for soligenous mires, the surrounding geology.

The GB map was further modified to make allowance for the impact of land use (liming). It was assumed that lime would be added, if necessary in all areas where arable or intensive grassland farmland dominated. The ITE land classification was used to predict those 1 km squares where these forms of agriculture dominated. In those squares, the critical load was increased by one class.

The resultant map (Figure 1) shows that soils with small critical loads are widespread in the west and north of Britain, areas which are dominated by relatively shallow soils derived from acid, base-poor rocks. In contrast, the soils of much of the south and east of England have large critical loads; many of these soils are formed in thick calcareous and/or clay-rich glacial deposits or in materials derived from calcareous rocks. The more acidic soils in the south and east which are under intensive agriculture are also limed regularly.

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Critical loads for peat soils

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INTRODUCTION

The acidification of peat soils has generally received much less attention than that of mineral soils, primarily perhaps because such soils, and their associated drainage waters, are often already very acidic. The application of the critical loads concept to soils hinges around the ability of biogeochemical weathering to replenish, on a sustainable basis, base cations leached as a consequence of acidifying pollutant deposition. For peats more than *ca* 500 mm in depth, with negligible weatherable mineral contents, mineral weathering is unlikely to play a significant role in amelioration of acid deposition effects. Thus, applying the same concept to peats that has been applied to mineral soils, peats might be expected to be highly sensitive to acidification.

EVIDENCE FOR PEAT ACIDIFICATION

The relationship between acid deposition and the pH of upland hill peats was investigated at the University of Aberdeen in the late 1980s, using both laboratory-based simulation experiments (Skiba & Cresser 1989) and field observations (Skiba *et al.* 1989). Both studies provided conclusive evidence for acidification.

The simulation investigation involved subjecting peats collected from two different sites in Scotland to repeated stir-and-centrifuge equilibrations with artificial precipitation (Skiba & Cresser 1989). The solute composition of the artificial precipitation was based upon long-term mean precipitation chemistry for a site in north-east Scotland (Edwards 1984). Four simulated precipitation solutions were prepared, covering the pH range 3.5-5.0 in steps of 0.5 of a pH unit. Acidification was achieved with a 2:1 mixture (on an equivalents basis) of H_2SO_4/HNO_3. In total, the peats were subjected to 1944 mm of 'rain' in 54 mm events. Drainage water was analysed at regular intervals, and sufficient replicates were used from the outset to allow replicated destructive sampling of peats for chemical analysis at regular intervals.

Typical results from these experiments are shown in Figure 1. Both the peat and its associated drainage water are acidified when equilibrated with rain at pH 3.5, but the pH rises for both peat and water when equilibrated at pH 4.0. The pH increases are greater

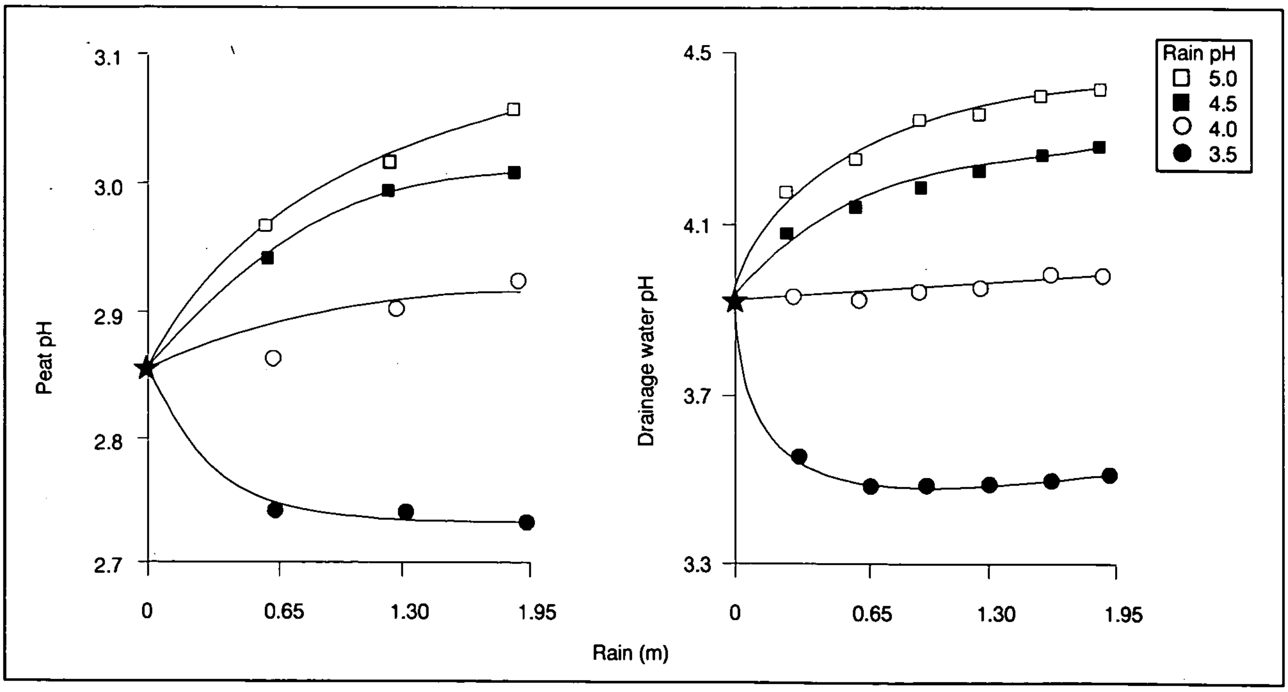

Figure 1. Effect of simulated rainfall equilibration pH upon the pH of peat from Glen Dye (left) and upon associated drainage water pH (right)

at pH 4.5, and greater still at pH 5.0. The pH changes of the peats were associated with corresponding changes in the base cation contents (Skiba & Cresser 1989).

Replotting the results shown in Figure 1 on a logarithmic scale, and assuming an annual precipitation of 2000 mm allow estimates of the effect of rain pH upon peat drainage water pH over 100 years (Skiba & Cresser 1989). Compared with what would be expected for pH 5.0 rain, a reasonable estimate for rain prior to the Industrial Revolution (UK Review Group on Acid Rain 1990), it appears that the drainage water has acidified by 1.02 and 1.12 pH units for peats from Glen Dye in Grampian and from the more polluted central belt of Scotland, respectively. These pH shifts are a consequence of both the peat acidification and the mobile anion effect. They are close to the values of 1-1.2 pH units found from palaeoecological reconstructions for lakes in south-west Scotland by Battarbee *et al.* (1985). Unfortunately, it is only possible to compare results for drainage waters, but the closeness of the agreement between results obtained by the two approaches is encouraging.

Evidence from field observations for peat acidification by acid deposition may be found in a comparison of the modelled wet-plus-dry acid deposition at 123 sites throughout Scotland with the peat pH and base saturation at the same sites (Skiba *et al.* 1989). The most acid peats (pH as $CaCl_2$ paste ≤ 3.0) and peats with the lowest base saturation (BS $\leq$ 10%) occurred where acid deposition exceeded 0.8 kg H^+ ha^{-1} yr^{-1}. Correlations between acid deposition flux and peat pH or BS were significant. The data used in the study were from the Scottish National Soil Inventory, set up by the Macaulay Land Use Research Institute in Aberdeen.

More recently, the authors have examined the chemical properties of heather moorland peat samples from along a pollution gradient between Gisla in the far north-west of Scotland (the Western Isles) and Pitsford in the south of England (Figure 2). Peat pH and exchangeable calcium and magnesium declined with increasing precipitation acidity. The relationships for the two base cations are shown in Figure 3. Several other marked chemical changes were observed, but these will be discussed later.

QUANTIFYING CRITICAL LOADS FOR DYSTROPHIC PEATS

The critical load of acid deposition for a soil is defined in general terms as the annual load which will not damage the soil system (Nilsson 1986; Nilsson & Grennfelt 1988). The concept of damage to soils is usually envisaged in terms of the production of an unacceptable (in terms of adverse effects upon plant growth) calcium/aluminium ratio in the soil solution, although other indicative parameters are sometimes employed (see, for example, Bull *et al.* 1992). This measure is not appropriate for peats, because concentrations of exchangeable aluminium in peats are often very low. However, critical loads for peats may be defined in terms of the acid deposition loads which would cause a specified pH reduction compared with pristine conditions.

The concept of defining a critical load with respect to pristine conditions, rather than existing soil conditions, is necessary because most peats may be regarded as being more or less in equilibrium with the precipitation they have experienced over recent years. Thus, to define the critical load with respect to current peat conditions would result in values being fixed at current deposition rates. Setting lower values implies partial recovery towards pristine condition values.

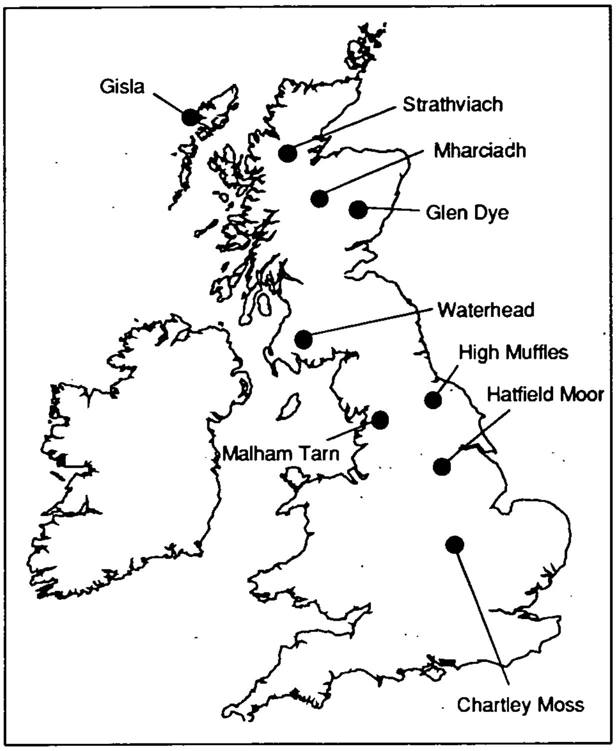

Figure 2. UK Precipitation Composition Monitoring Network sites used to provide samples of peat along a pollution gradient

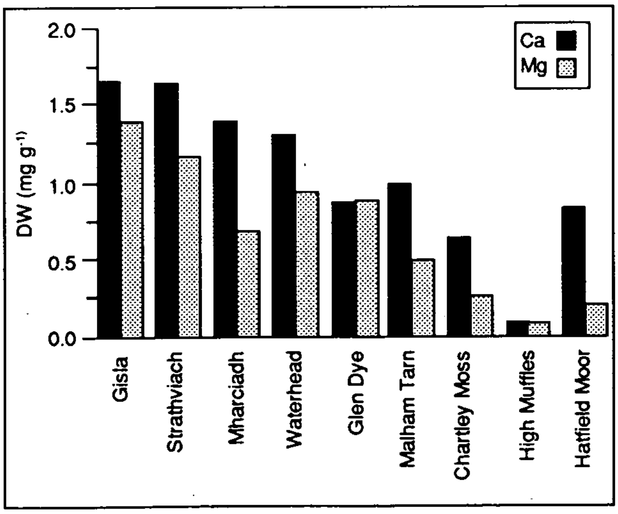

Figure 3. Effect of increasing pollution upon the exchangeable calcium and magnesium contents of peat

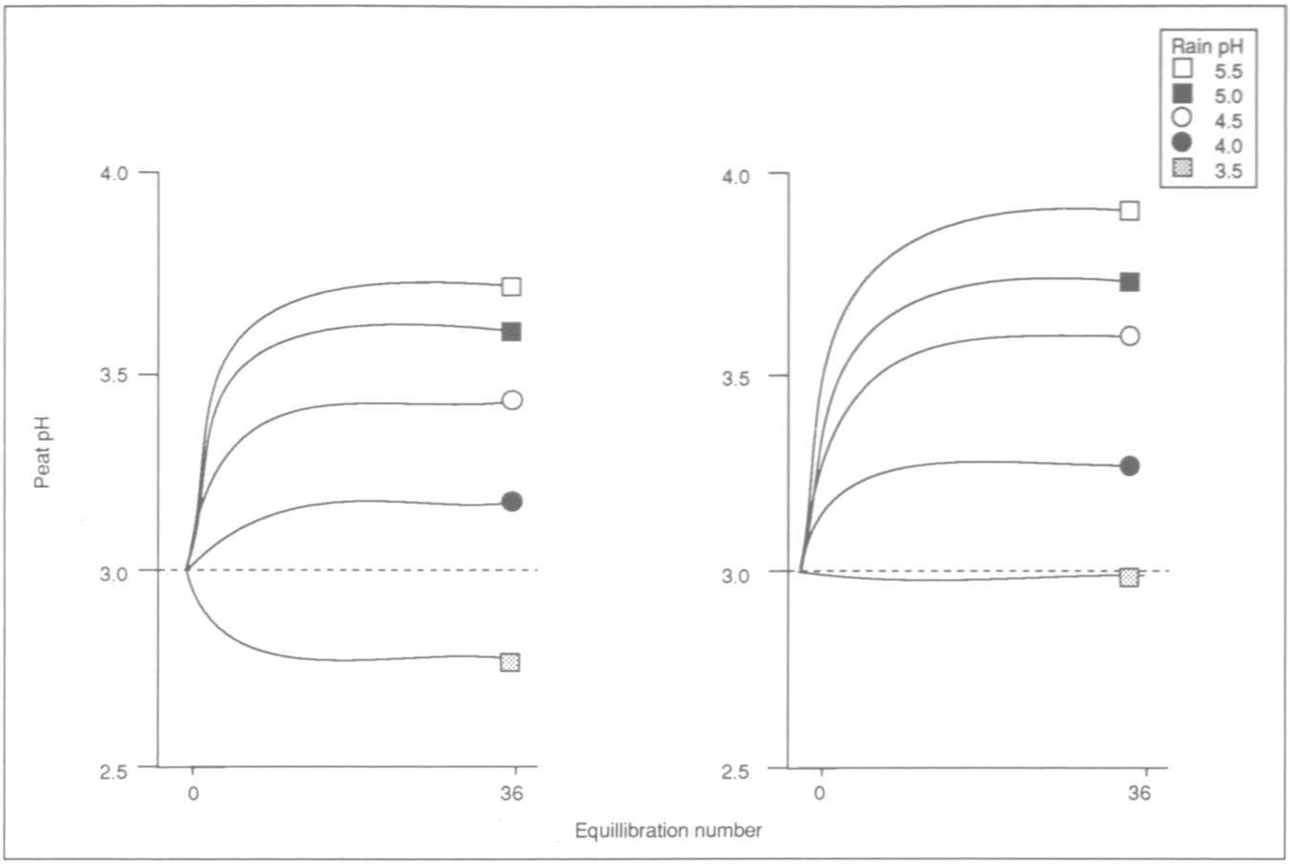

Figure 4. Effect of calcium concentration upon the equilibration pH of peat over a range of simulated rainfall pH values. Calcium concentration was 0.012 meq l^{-1} (left) or 0.060 meq l^{-1} (right)

The early simulation experiments clearly indicated that peat pH depends upon ion exchange competition between H$^+$ and base cations, especially calcium and magnesium. The equilibration experiments were repeated over a wider range of pH conditions (pH 3.5-5.5) and over a range of calcium and magnesium concentrations chosen to encompass the conditions likely to be encountered in the UK. Calcium concentration was found to be particularly important, but magnesium had less effect upon pH (Smith, Cresser & Mitchell 1993). Figure 4 gives an indication of why calcium concentration needs to be taken into account. Before proceeding any further, however, it is also necessary to decide how dry-deposited acid fluxes should be taken into account.

An interesting feature of the results in Figures 1 and 4 is that they show apparent recovery when peat is equilibrated with simulated rain at pH 4.0, even though the peat in the field might be regarded as having equilibrated with rain with a mean pH value of around 4.6. This evidence suggests that dry-deposited acid flux and possibly evapotranspiration concentration effects also need to be taken into consideration. If the equilibrating acid concentration is based upon total deposited acid divided by runoff, a new 'effective pH' value may be calculated. Figure 5 is a map of effective rain pH for Great Britain, based upon these principles. The 'effective pH' of rain at Glen Dye is about 3.9, which explains why pH is

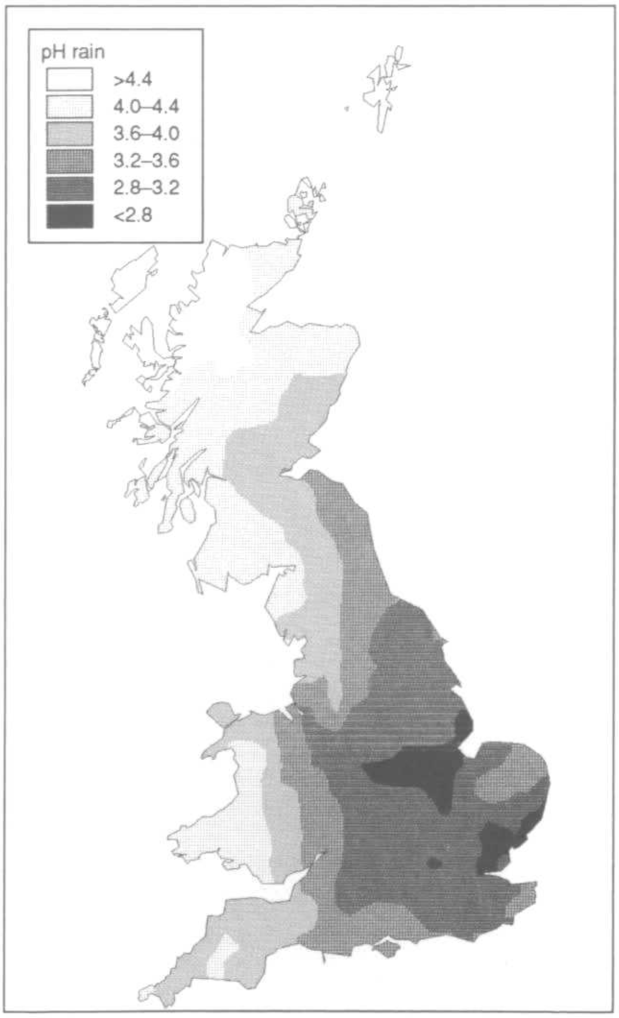

Figure 5. Map of the effective pH (as defined in the text) of rainfall in Great Britain

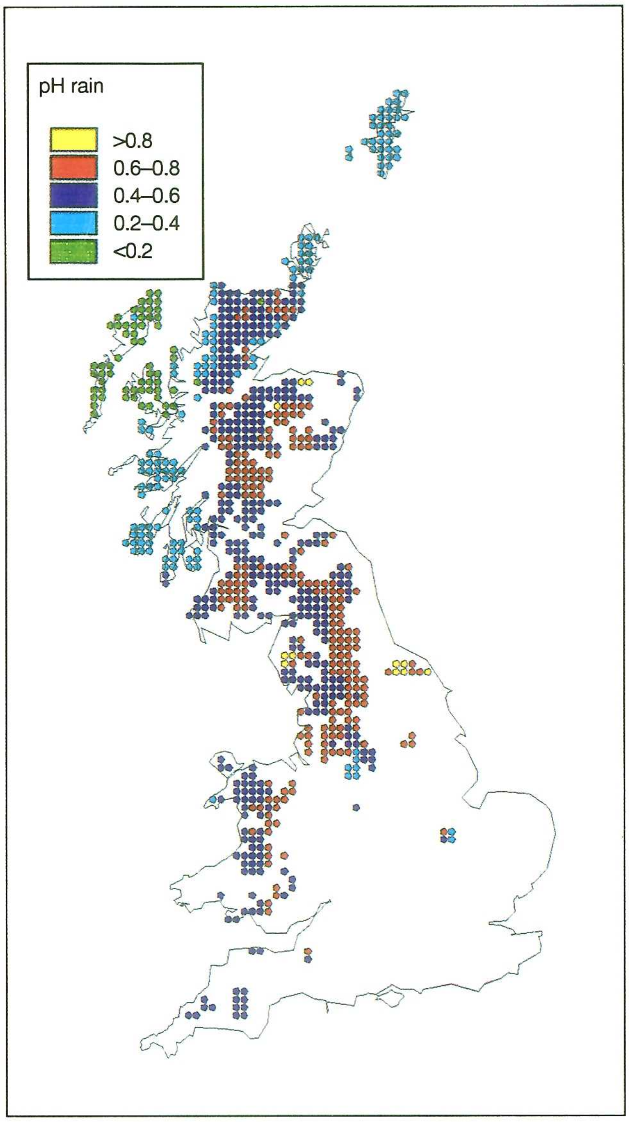

Figure 6. Map of predicted pH shifts of peat in Great Britain from pristine conditions

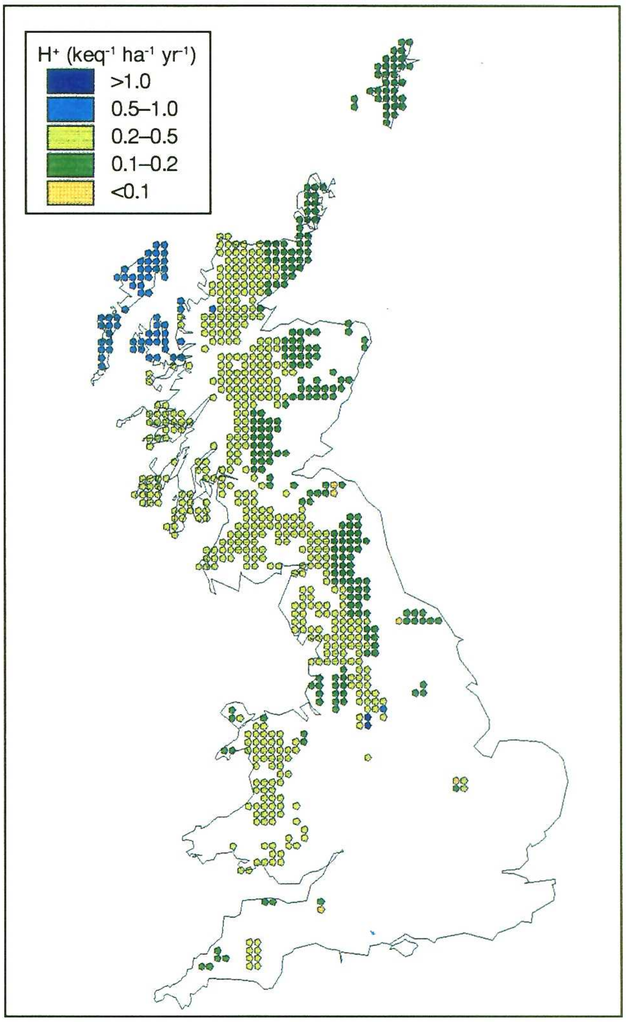

Figure 7. Critical load map for peat, based upon an acceptable peat acidification value of 0.2 pH units

improved when Glen Dye peat is re-equilibrated with simulated rain at pH 4.0. It is reasonable to suppose that effective calcium concentration should also be calculated by allowing for evapotranspiration, although in this instance no adequate dry deposition data are available.

Effective pH and effective calcium concentration have been calculated for each 10 km square in Great Britain containing a significant amount of peat. These values, and the results of the simulation experiments, have then been used to predict:

- the pH of hill peat in each square
- the pH value which the peat in each square would have had if the rainfall had an effective pH of 5.0
- the predicted current extent of acidification
- the acid load which would cause acidification by 0.2 pH units compared with 'pristine conditions' - the latter values constitute the critical load for each square.

Figures 6 and 7 respectively show the predicted degree of acidification of peat and the critical loads, if it is assumed that acidification by more than 0.2 pH units is unacceptable.

In some respects, defining critical loads with respect to a pH shift of 0.2 units is rather arbitrary. When peat from Glen Dye was limed with calcium hydroxide and thoroughly mixed to raise the pH back to the estimated pristine value, the growth of heather *(Calluna vulgaris)* was increased by 40% in a 6-month pot experiment (M S Cresser & R Sanyi, unpublished). Under field conditions, the growth suppression might be different. Moreover, the 40% reduction was for a pH shift of *ca* 0.5 pH units. More work needs to be done to establish links between peat acidification and the performance of the biota sustained by peat.

The critical loads estimated as described above were assigned to the sensitivity classes used to prepare the UK Department of the Environment critical load maps described elsewhere in this volume. Individual values were put in the next higher sensitivity class, which of necessity errs on the side of caution.

COMPARISON OF MEASURED AND PREDICTED pH VALUES

If the application of the simulation technique is a valid approach for estimating dystrophic peat pH, there

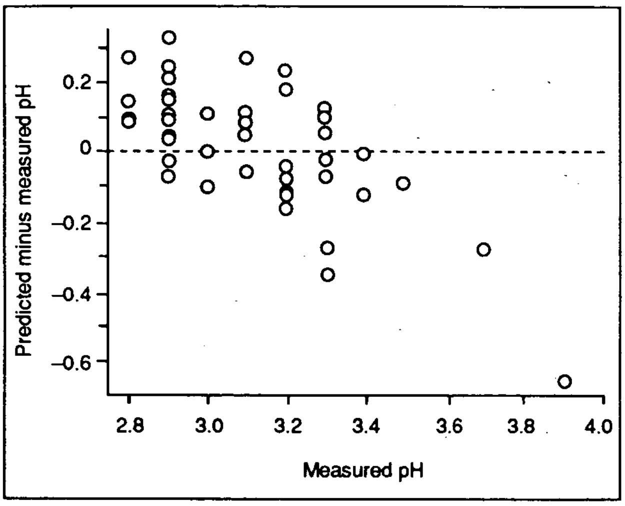

Figure 8. The relationship between the predicted and measured peat pH values

should be good agreement between the predicted peat pH values for Scotland, and the values in the Scottish National Soil Inventory data set. Figure 8 shows the agreement. Although it is generally satisfactory, there is a small systematic error, with the most polluted sites being up to 0.2 pH units more acidic than predicted. Part of the difference may be due to methodological differences for pH measurement, although tests showed these to be very small. Another possible contributing factor is the strong seasonal trends in deposition acidity (Skiba & Cresser 1989). These trends could conceivably be aggravated by elevated concentrations during early snowmelt. Additionally, ammonia and ammonium deposition effects may need to be taken into consideration. These aspects are currently being studied. When the difference between predicted and measured pH values was mapped, no systematic geographical trends emerged.

CLASSIFICATION OF EUTROPHIC PEATS

The classification of eutrophic peats obviously had to be based upon different criteria from that of dystrophic peats, and had to be possible using readily available data. A classification system was developed based upon mire type, with modification for probable mire nutrient status.

To identify eutrophic peats, Soil Survey maps were used in conjunction with data in *Lowland peat in England and Wales* (Burton & Hodgson 1987). This valuable work provided details of peat type (topogenous, soligenous or ombrogenous mire) in England and Wales. For most sites, soil pH values and site descriptions were also provided. Less information was available for Scotland. To facilitate integration into the overall GB soils maps, it was decided to use a five-point sensitivity scale, class 5 being the most sensitive. Soligenous mires were allocated to a Skokloster class of 3, and topogenous mires to a class of 4. Both can be upgraded by one class if developed in a base-rich parent material or influenced by base-rich groundwater. Drained peats in coastal areas were placed in the most sensitive class, because of the risk of generation of sulphuric acid from pyrite oxidation.

OTHER CONSEQUENCES OF ACID DEPOSITION TO PEATS

Apart from the effects of acid deposition upon the pH and exchangeable base cation contents of dystrophic peats mentioned earlier, several other chemical changes were observed in the peats collected from along the GB pollution gradient (Figure 2). There was a very obvious increase in exchangeable ammonium with increasing acidifying pollution deposition, and a decrease in extractable nitrate, suggesting inhibition of nitrification. There were substantial increases in organic-C, organic-N and organic-S leaching with increasing deposited acidity, although the relative importance of organic-N to total N flux declined with increasing acidity. The leachate chemistry and microbiological behaviour of peat turves from the sites in Figure 2 are still being investigated.

A further consequence of the peat acidification appears to be sustained nitrate leaching into rivers. Black *et al.* (1993) have studied changes in solute composition at 250 m intervals along the lengths of several rivers draining heather moorland in north-east Scotland. Wherever water draining from peat made a substantial contribution to the total discharge, they found elevated nitrate concentrations in river water, not only in winter but also in early summer. This finding suggests that the biota are unable to utilise the excessive atmospheric N inputs, possibly as a consequence of the severe acidification. There can be little doubt that in these ecosystems the critical load has been exceeded considerably for some time.

CONCLUSIONS

Simulation experiments allow the prediction of peat pH for dystrophic peats, provided that effective precipitation pH and calcium concentrations are known. They also allow the estimation of peat pH under pristine conditions, and the acid load at any specified site which would cause a pH shift downwards of 0.2 pH units compared to pristine conditions. The latter acid load values may be used as critical loads.

For eutrophic peats, a simple classification system has been developed based upon the probable base status of the mire.

Field observations of the pH and base cation status of peats are in reasonable agreement with predicted values. The small differences which occur, and the effects on other properties of peats, are still under investigation.

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The application of Skokloster critical load classes to the soils of Scotland

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INTRODUCTION

Soils provide the medium for plant growth and also act to modify the chemistry of drainage waters with respect to incident precipitation. The impact of acid deposition on soils is, therefore, of considerable concern in determining the sensitivity of natural ecosystems to acidification. Where cations are replaced at a rate less than that at which they are lost, then soil acidification will occur. Inputs of strong acids from atmospheric deposition may accelerate soil acidification and result in the enhanced leaching of calcium and magnesium, leading to plant nutrient deficiencies and imbalances and high concentrations of aluminium and other toxic metals.

The sensitivity of a soil to acidification is primarily determined by the balance between the loss of base cations following leaching and uptake into vegetation against the supply of base cations from the weathering of soil minerals. The rate of chemical weathering within the soil and soil parent material, therefore, largely determines a soil's sensitivity to acidification. Large areas of Scotland have soils derived from nutrient-poor materials and are situated in high-rainfall areas. In such conditions, the susceptibility to enhanced acidification from atmospheric deposition is considerable. One method of quantitatively estimating the sensitivity of an ecosystem to acidification is to use the critical load approach.

A critical load can be defined as 'the highest deposition of acidifying compounds that will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function' (Nilsson & Grennfelt 1988).

At the workshop on critical loads in Skokloster, Sweden (Nilsson & Grennfelt 1988), five critical load classes for forest soils were proposed (Table 1). These classes were based on the dominant mineralogy of the soil parent material because of its primary influence on the rate of chemical weathering. The most sensitive class (1) is composed of soils derived from highly siliceous parent materials such as quartzite and K-felspar-rich granite, and the least sensitive (5) of soils is derived from parent materials with a free carbonate content (marls, limestones, etc). Between these extremes are classes for soils derived from plagioclase-rich granites/gneisses (2), granodiorites, greywackes, schist, etc (3), and gabbro and basalt (4). The minerals thought to control base cation weathering rates in each of the classes are also given in Table 1. In addition to mineralogy, however, the critical load of a soil may be influenced by a number of modifying factors, such as soil texture, soil drainage, type of vegetation, or land management practices. These various modifiers may increase or decrease the critical load of the soil. The modifiers considered & their effect on critical loads are given in Table 2.

Table 1. Mineralogical and petrological classification of soil material and critical loads (source: Nilsson & Grennfelt 1988)

Skokloster class	Minerals controlling weathering	Parent rock	Critical load (keq H^+ ha^{-1} yr^{-1})
1	Quartz, K-felspar	Granite Quartzite	<0.2
2	Muscovite Plagioclase Biotite (<5%)	Granite Gneiss	0.2-0.5
3	Biotite Amphibole (<5%)	Granodiorite Greywacke Schist Gabbro	0.5-1.0
4	Pyroxene Epidote Olivine (<5%)	Gabbro Basalt	1.0-2.0
5	Carbonates	Limestone Marl	>2

Table 2. Modifiers influencing critical load to soils (source: Nilsson & Grennfelt 1988)

Factor	Decreasing critical load	Increasing critical load
Vegetation	Coniferous	Deciduous
Elevation/slope	High	Low
Soil texture	Coarse-sandy	Fine
Soil drainage	Free	Confined
Soil/till depth	Shallow	Thick
Soil sulphate adsorption capacity	Low	High
Base cation deposition	Low	High

Sverdrup, De Vries and Henriksen (1990) suggested that critical loads for soils can be calculated at three levels of complexity. At 'level 0', critical loads are assigned to soils using existing data. Where input/output budget data are available, critical loads may be calculated using a 'level 1' approach by mass balance calculation or the application of a steady-state model, such as PROFILE (Warfvinge & Sverdrup 1992). Where detailed information on biogeochemical fluxes and on soil properties is available, a 'level 2' approach may be used with dynamic models such as MAGIC (Cosby *et al.* 1985). However, at a regional scale, the input data for these two latter approaches are not always available for sufficient sites to produce national maps. The present work, therefore, builds on the Skokloster ('level 0') approach, in a similar manner to that described by Langan and Hornung (1992), where existing soil survey data were used to assign critical loads to soils in northern England. In the present study, it has also been possible to calculate a critical load using the 'level 1' mass balance approach for a small number of sites.

For mapping purposes, Scottish soils have been categorised according to their parent material (soil association). The association provides a particularly appropriate unit for assessing the critical load of soils according to the Skokloster classes. The area investigated in this work is shown on the seven Soil Survey maps at the 1:250 000 scale which cover the entire area of Scotland (Soil Survey 1984), the relevant associations being described in the accompanying handbooks.

Having derived and calculated a critical load for soils, it is possible to map areas where atmospheric deposition of acidity is in excess of the soil's critical load and to produce an exceedance map. The value used for the critical load for each of the classes in this work is the upper value proposed at Skokloster (column 4, Table 1).

METHODS

Creation of the critical load map

The application of the Skokloster classes to Scottish soils was undertaken in three phases (Figure 1). In phase one, each of the 118 soil associations mapped in Scotland was assigned to one of the five critical load classes, based as far as possible on the mineralogy of the dominant parent material. The data were obtained from Soil Survey handbooks, Soil Memoirs and from the Scottish National Soil Inventory, set up by the Macaulay Land Use Research Institute. Within each association, there is considerable variation in the soil morphology which will affect the soil's critical load. To refine the original mineralogically based critical load for the soil associations, by taking into account modifying factors (Table 2), phase two of the work reviewed each of the 580 soil units identified in the 1:250 000 soil map of Scotland. Soil units are spatially restricted in occurrence and describe soils with common physical attributes and land use; thus, for the critical load work they provide a suitable grouping to which the modifiers, taken from Table 2, can be applied.

The attributes most likely to modify the sensitivity assigned on the basis of parent material were considered to be till depth, texture, soil depth and slope. The main factor considered to increase a soil's sensitivity was a combination of thin, poorly developed (ranker) soils on steep slopes, frequently described as very stony, rocky or bouldery.

In summary, the soil association was used to derive the initial, mineralogically based allocation of the soil to a critical load class. The soil map unit (on the 1:250 000 soil maps) was then used to identify any relevant modifying factors (cf Table 2) which could increase or decrease the critical load.

Organic peat soils have been mapped separately as their chemistry is not influenced by mineral weathering (peat is defined as an organic soil >30 cm in thickness). Work on a surrogate measure of critical loads for these organic soils is currently being undertaken at the University of Aberdeen (Cresser, Smith & Sanger, pp 34-39).

A major influence in decreasing the sensitivity of a soil is that of land use, which has been treated separately so that it can be assessed more readily (phase three). Soils which are under agricultural management are less susceptible to acidification because of their management and the application of lime. Such practices will act to maintain the base status of the soil at a level which can sustain economic crop yields. Therefore, where a soil unit was described in the Soil Survey notes as arable or permanent pasture, the critical load was decreased by one class.

Calculation of critical loads for selected soils

Bain (1990) and Bain *et al.* (1990) have calculated the long-term chemical weathering rates for various upland soils across Scotland. These weathering rates were used together with the mass balance equation

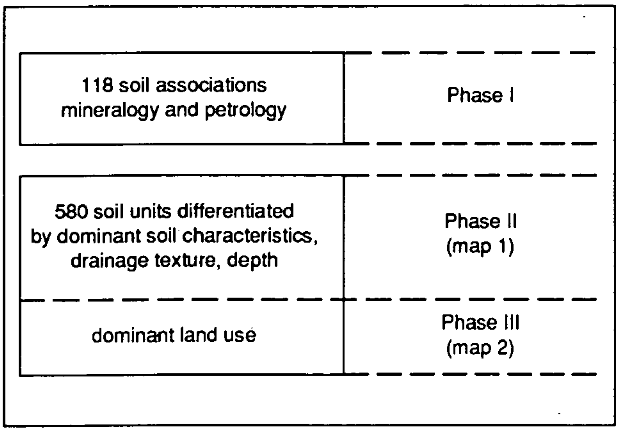

Figure 1. The three phases of application to mapping soil critical loads in Scotland

(1) (after Sverdrup *et al.* 1990) to calculate critical loads:

$$CL_{acidity} = BC_w - BC_u - NTOT_u - ANC_l \qquad (1)$$

where BC_w = base cations from weathering; BC_u = base cation uptake; $NTOT_u$ = total vegetation uptake of N; and ANC_l = alkalinity leaching.

Critical load exceedance for acidity was calculated using the deposition data from the United Kingdom Review Group on Acid Rain (UKRGAR) (1990). Current (1987-88) areas of exceedance were calculated from the sum of non-marine sulphur and nitrogen deposition components (dry, occult and seeder-feeder enhanced rainfall), subtracted from which were the neutralising effect of deposited non-marine calcium and magnesium for each 20 km x 20 km grid square. A detailed account of the methods of calculation and a full description of the data are given in UKRGAR (1990) and Fowler *et al.* (1993).

Maps showing the distribution of the critical load classes for acidity across Scotland have been produced by plotting the dominant soil unit in each 1 km square, whilst areas of critical load exceedance have been mapped at 20 km x 20 km, the resolution of the deposition data. The two critical load maps all use the Skokloster classes which are expressed in terms of keq H^+ ha^{-1} yr^{-1}. Organic peat soils have been mapped as purple, other classes as specified at Skokloster (column 1, Table 1). Peats are mapped as purple, other classes as specified. Exceedance is plotted as the percentage of 1 km soil critical loads exceeded within each deposition grid square.

RESULTS

Of the 118 associations identified, there are 17 which individually cover more than 1% of Scotland and, together, these account for 80.7% of the total area of Scottish soils. Figure 2 shows the distribution of the five Skokloster critical load classes for acidity as applied to the soils of Scotland. The map was constructed on the basis of the influence of parent material, soil development and topography. In general, the distribution of critical load classes for Scottish soils reflects a combination of underlying geology and topographic relief. The most sensitive soils are thin and occur on steep slopes in upland areas on parent materials with few weatherable minerals. The least sensitive soils are typically found in low-lying areas on gentle slopes over relatively easily weathered parent materials.

The most sensitive soils (ie class 1) occur sporadically, the most extensive area being on the island of Jura where the soils are predominantly derived from quartzite. Other small areas of the base-deficient class 1 soils are associated with the granites of the Grampian Highlands and in Galloway in south-west Scotland. There is also an extensive linear outcrop of these soils related to an outcrop of Cambrian quartzite running parallel to the coast of north-west Scotland.

Class 2 is the most extensive of the Skokloster classes and represents much of Scotland on both sides of the midland valley. These areas are dominated by acid soils of low base saturation and are derived from base-deficient slow-weathering rocks. To the south of the midland valley, the soils are derived from lightly metamorphosed shales, grits, flagstones and greywackes of Ordovician and Silurian age. To the north of the Highland boundary fault, and extending as far as the Outer Hebrides and the Shetland Isles, there are very large areas where the soils are derived from metamorphic assemblages of schists, gneisses and granulites of pre-Cambrian age.

Most of the midland valley, the Border country, large areas of north-east Scotland, Mull, Skye, the Orkney islands and north-east Caithness fall into the critical load classes 3 and 4. These are areas which often have deep well-developed soils on gentle slopes, developed on thick glacial till, or are associated with base-rich, easily weatherable parent materials, such as calcareous or base-rich sandstones, flagstones or shales, basaltic lavas, gabbros, etc.

The least sensitive, class 5, soils are rather restricted spatially and occur most noticeably on marine, alluvial, calcareous sediments, or are associated with carbonate-rich parent materials. The latter occur between the Highland boundary fault and the Great Glen (Dalradian and Moinian meta-limestones) and in the north-west Highlands (Cambrian limestone). Soils on shell sand occur in north-east Scotland and in the Outer Hebrides.

Figure 3 shows the distribution of critical load classes for acidity when the land use modifier is applied. The largest difference between Figures 2 and 3 is the reduced sensitivity of the soils in the midland valley, the Buchan area of north-east Scotland and much of the Border country. These areas are predominantly agricultural where soil management and lime application will reduce the sensitivity of soils to acidification.

The results from the application of the mass balance equation (Table 3) compare favourably with those using the derivation of critical loads from soil characteristics in the Skokloster approach. Of the four site calculations, three are within the class range allocated for the soils and the fourth suggests a slightly higher critical load. These initial comparisons are encouraging, and suggest that more work on the mass balance approach should be undertaken. However, the variation of critical load within each class does indicate that the setting of the appropriate value for exceedance is vital. This issue can only be assessed by further research on 'damage' to biological indicators in relation to deposition loading and exceedance.

Figure 4 shows those areas of Scotland for which current levels (1987-88) of acidity exceed the soils' critical load. The map indicates that in most parts of Scotland there are soils where the critical load is

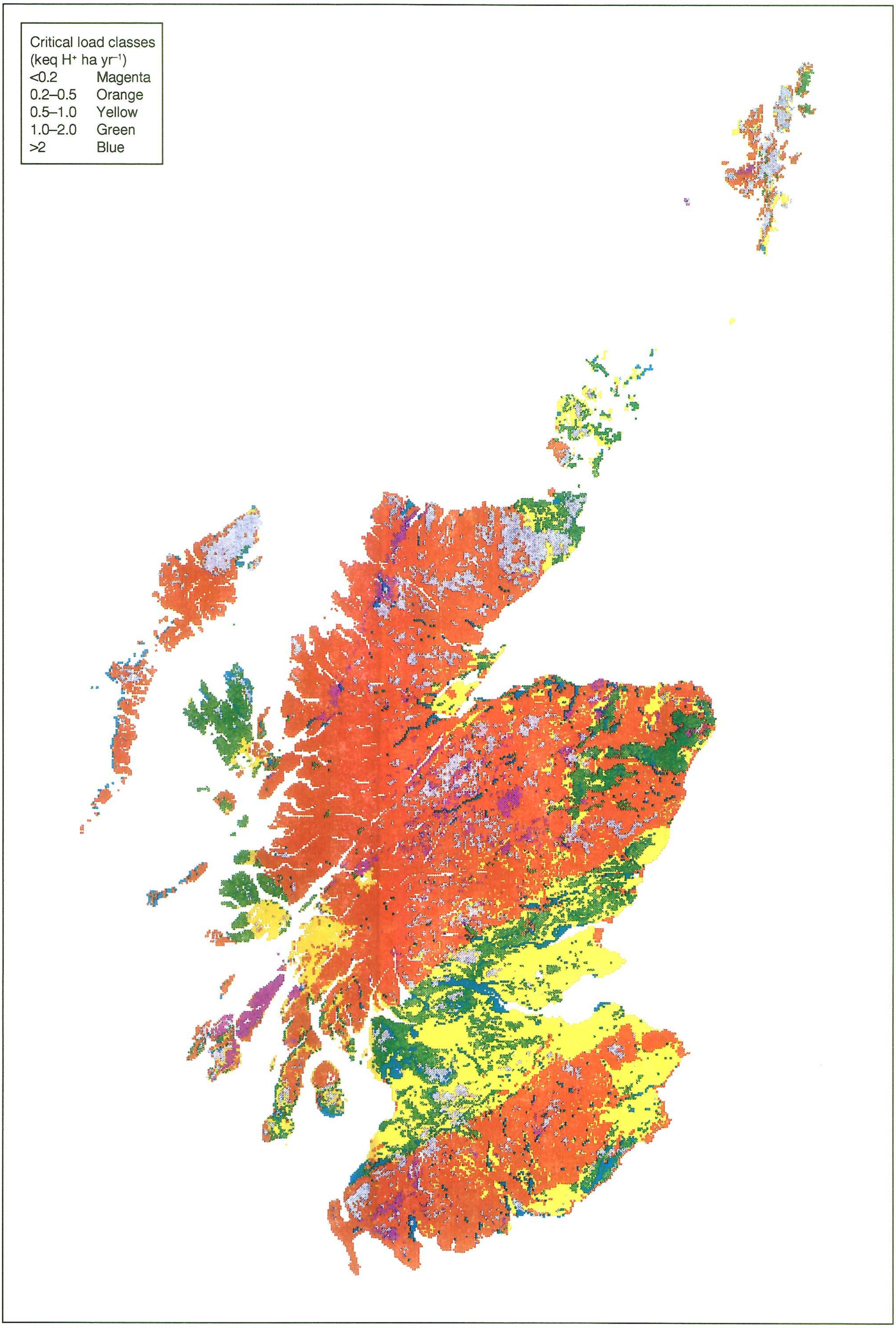

Figure 2. The geographical distribution of the Skokloster critical load classes of acidity to the soils of Scotland based on parent material, soils and topography

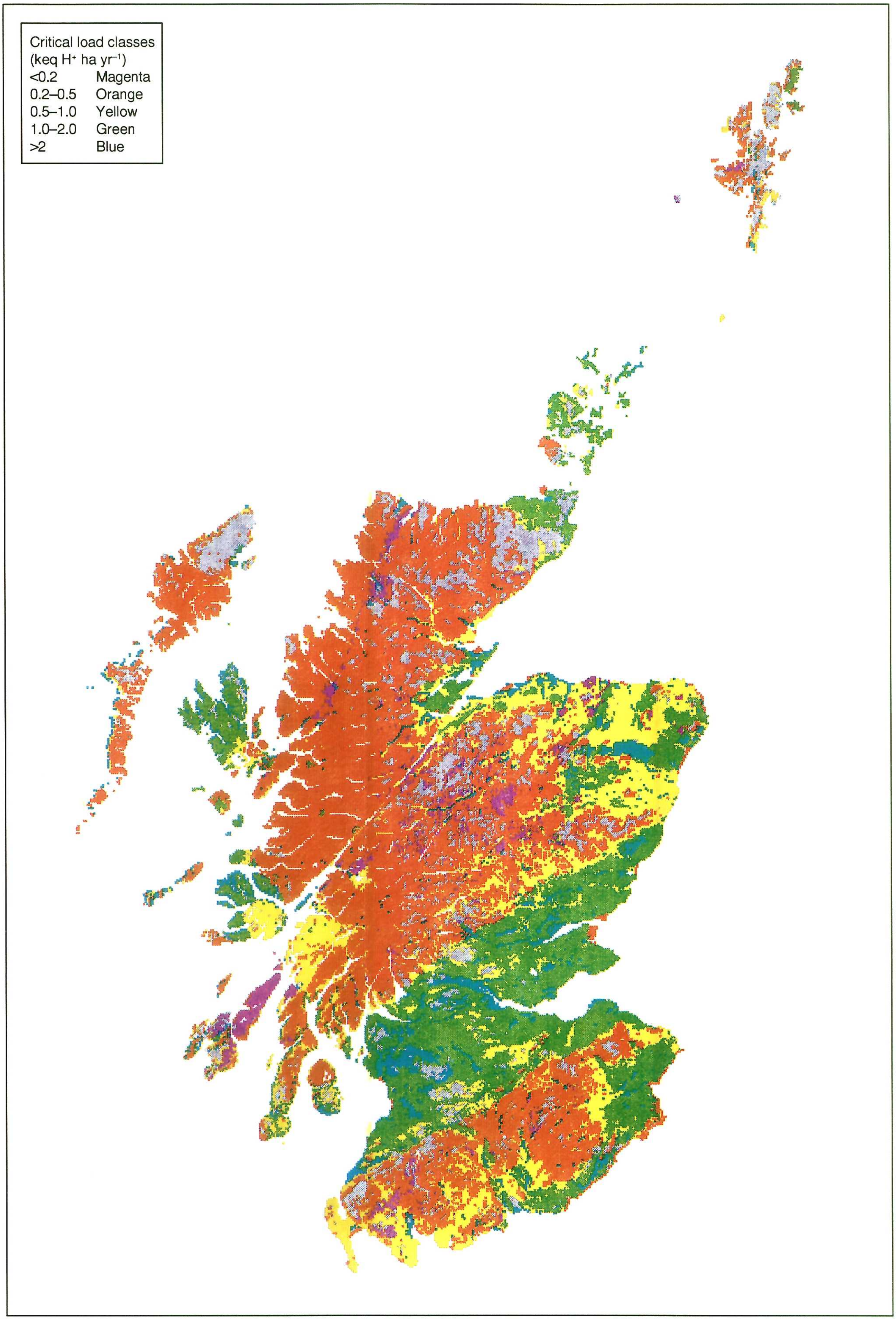

Figure 3: The geographical distribution of the Skokloster critical load classes of acidity to the soils of Scotland based on parent material, soils, topography and land use

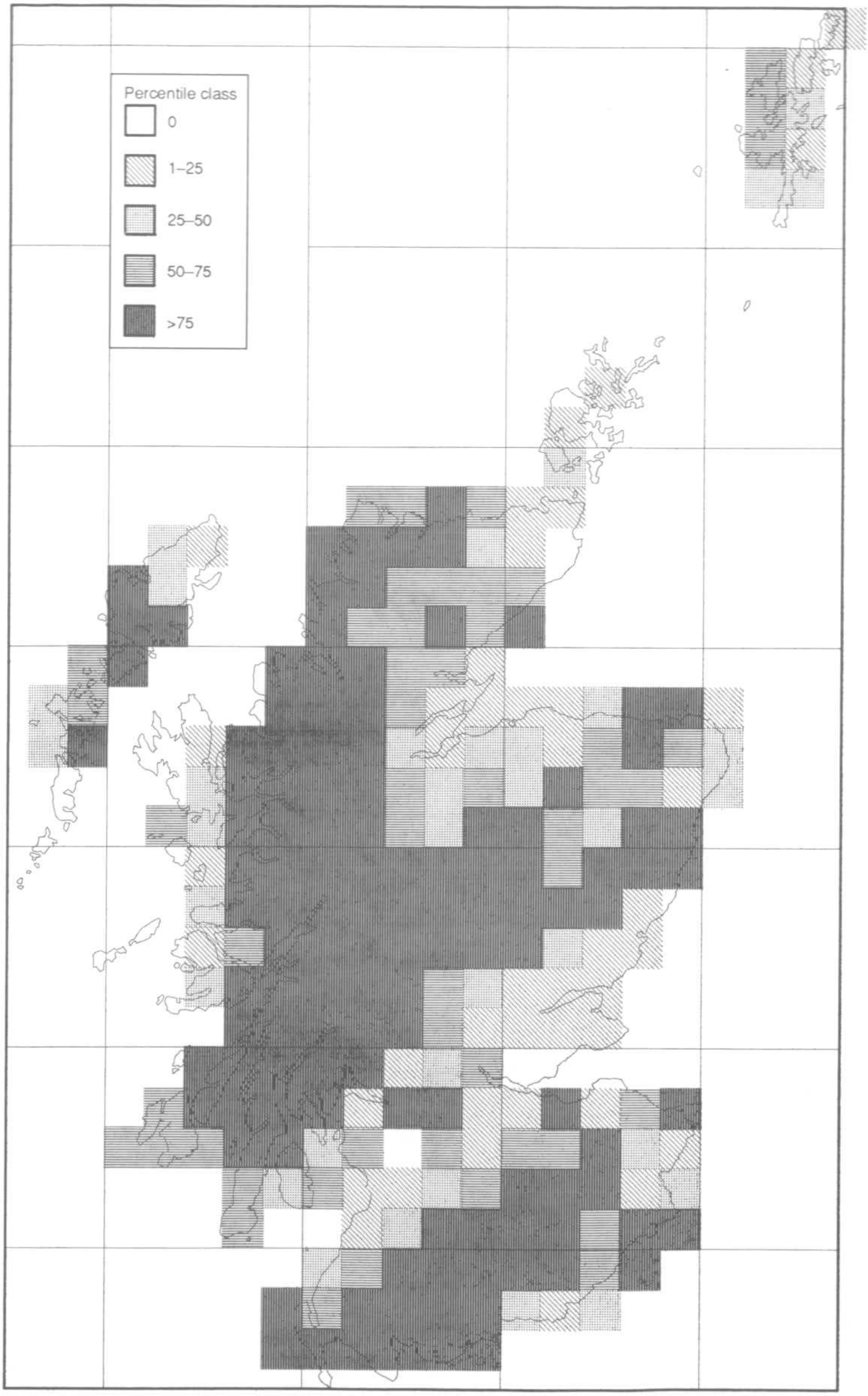

Figure 4. Areas of critical load exceedance for acidity based on the percentage exceedance of 1 km soil classes within each 20 km square grid

Table 3. Comparison of weathering rates of base cations and critical loads calculated from equation 1 with those mapped using the Skokloster approach

Site name	Weathering rate (keq ha^{-1} yr^{-1})	Critical load: mass balance (keq ha^{-1} yr^{-1})	Critical load: Skokloster (keq ha^{-1} yr^{-1})
Sourhope	0.5	0.7	0.5 to 1.0
Glensaugh	0.6	0.5	0.2 to 0.5
Mharcaidh	0.3	0.2	0.2 to 0.5
Chon	0.7	0.6	0.2 to 0.5

exceeded. The map is, of course, strongly influenced by the occurrence of small areas of the most sensitive soils (classes 1 and 2). For example, the 20 km x 20 km grid square covering the Mull of Galloway in south-west Scotland is dominated by class 3 soils for which the critical load is not exceeded, but the occurrence of 4 km^2 of more sensitive soils leads to the entire grid square being shaded. Similar examples can be found across the rest of the country. The map indicates areas where exceedance (or damage) may be occurring in a 20 km square, but not the areal extent within it.

The areas which show a high percentage of exceedance are centred on the uplands. These are the areas which are resistant to weathering (hence forming upland areas) and also receive higher amounts of deposition through orographic rainfall and seeder-feeder type enhanced deposition (Fowler *et al.* 1993). These areas of widespread critical load exceedance are coincident with the areas where acidification research fieldwork programmes have already shown problems of acidification, eg the Grampian Highlands, the Trossachs region to the north-west of Glasgow (Ferrier & Harriman 1990), and the Loch Dee project in south-west Scotland (Welsh & Burns 1987). Away from these areas the percentage exceedance of the soils decreases, although there are few grids containing no exceeded soils.

CONCLUSIONS

The critical load work presented provides a national assessment of Scottish soils which either may have already been affected, or are likely to be at risk from acidification as a result of acidic inputs from the atmosphere. The areas of exceedance which should, in principle, be showing adverse effects of this acidification are concentrated in the west of Scotland and extend east into the Cairngorms. Current fieldwork programmes investigating areas where there are known acidification problems in Scotland coincide with the geographical distribution of the more sensitive critical load classes identified here. In particular, the Loch Ard area in the south-west Highlands near Glasgow and the Loch Dee area in Galloway in south-west Scotland are both clearly situated in areas of exceedance. Further acidification effects might be anticipated in parts of the central and western areas of the southern uplands.

Whilst the critical load maps do not lend themselves to the interpretation of dose-response at the individual catchment scale, they do provide an indication of impacts. Not included in the present work schedule is the use of scenario-type analysis in which, for example, the large-scale effect of regional afforestation may be investigated. This initial analysis of critical loads for soils has been useful in identifying areas where further work is needed. Particular emphasis should now be given to the link between critical load, exceedance and biological effect.

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The classification of the soils of England and Wales on the basis of mineralogy and weathering – the Skokloster approach

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INTRODUCTION

Long-term buffering against soil acidification is, essentially, a function of soil mineralogy through cation release as a result of weathering. Mineralogy is, in its turn, a reflection of soil parent material. From this standpoint, four kinds of mineralogically mediated buffering reactions can be recognised:

i. dissolution of carbonate;
ii. release of base cations, such as calcium and magnesium from mineral surfaces or exchange sites;
iii. release of aluminium (and possibly iron) from exchange sites or from dissolution of secondary aluminium minerals;
iv. mineral dissolution (silicate weathering).

The relative contributions of these processes are, of course, dependent on numerous factors, some of which are poorly understood. A major controlling factor over the longer term is the reaction, or weathering, of the carbonate and silicate minerals. Sverdrup and Warfvinge (1988a) proposed a classification of soil mineralogy into reaction classes based on the weathering rate of the constituent minerals. A similar classification of soil mineralogy, on the basis of weathering rates, was used to calculate critical loads of soils for sulphur and nitrogen; this method was formulated at a workshop held at Skokloster in Sweden (Nilsson & Grennfelt 1988), and this type of classification will be referred to as a Skokloster classification in this paper. The classification developed at Skokloster was essentially based on the weathering rates of primary silicate minerals which dominate in soils derived from igneous rocks. Sverdrup and Warfvinge (United Nations Economic Commission for Europe (UN-ECE) 1990) extended the classification developed at Skokloster by adding additional minerals, including a number characteristic of metamorphic rocks and some secondary minerals. (It should be noted that in the original classification proposed at the Skokloster workshop the most rapidly weathering class of minerals was designated as class 5, while in the classification proposed by Sverdrup and Warfvinge (UN-ECE 1990) the most reactive minerals were designated as class 1, cf Table 1.) The approach implicit in the Skokloster classification and in the work of Sverdrup and Warfvinge was adopted as the basis of the development of a provisional critical load map for the soils of Great Britain. The work has been carried out under the auspices of the soils subgroup of the UK Department of the Environment's Critical Loads Advisory Group (CLAG).

THE SKOKLOSTER CLASSIFICATION

The starting point was the classification of Sverdrup and Warfvinge given in the UN-ECE *Draft manual on methodologies and criteria for mapping critical levels/loads* (UN-ECE 1990), which is reproduced as Table 1.

Table 1. Original Skokloster classification, giving approximate weathering rates (kmol(+) km^{-2} yr^{-1}) expressed as basic cations dissolved from primary and secondary minerals (pH 5 of soil solution) (source: Sverdrup & Warfvinge 1988a; UN-ECE 1990)

Mineral class	Average soil mineral class content 100%	30%	3%	0.3%
1 Very fast: carbonates	2500	1500	1000	300
2 Fast: olivine, garnet, anorthite, diopside, nephelihe, jadeite	1500	1000	300	30
3 Intermediate: enstatite, augite, hypersthene, chlorite, hornblende, biotite, glaucophane, epidote, zoisite	1000	300	30	3
4 Slow: albite, illite, oligoclase, labradorite	60	20	2	-
5 Very slow: K-felspar, muscovite mica, montmorillonite, vermiculite	30	10	1	-
6 Inert: quartz, rutile, anatase, gibbsite, koalinite	10	1	-	-

As noted above, the classes and mineralogies shown are oriented strongly towards the geology of igneous and metamorphic terrains, such as Fennoscandia

(Sverdrup & Warfvinge 1988a). In northern Europe, such terrain is dominated by coarse-textured soils in which the mineralogy reflects that of the parent material. However, this classification makes no allowance for:

- organic soils (peats)
- more strongly weathered soils
- soil texture, especially the influence of clay content.

The Skokloster classification also assumes that the soils are at pH 5. This is probably a reasonable value for managed forests or low-input agriculture, but is too low for the intensively managed situation prevailing over most of lowland England and Wales, where control of soil pH, by the addition of lime, to pH values between 6 and 6.5 (at least in the upper part of the soil profile) is common.

For these reasons, the original Skokloster classification was modified for use in England and Wales, and this modification is discussed below.

MODIFICATION OF THE SKOKLOSTER CLASSIFICATION

Organic soils (peats)

These were defined according to Clayden and Hollis (1984). Peat soils cannot be classified by mineralogy as they consist almost entirely of organic remains in various stages of decomposition. They were initially placed, therefore, in a separate Skokloster class. A method of assigning critical loads to peats is described later by Cresser, Smith and Sanger (pp 34-39).

Mineral soils

It was not thought meaningful to differentiate, in terms of soil chemical behaviour, between classes 5 and 6 proposed by Sverdrup and Warfvinge (UN-ECE 1990), ie very slow weathering and inert, respectively. Therefore, they were combined into one class.

Most soil parent materials in England and Wales are described in terms of lithology or rock type, rather than mineralogical composition. In igneous and metamorphic rocks, however, there is a good relationship between the two concepts (Clayden & Hollis 1984). Soils developed from these two suites of rocks were assigned to classes on the basis of broad petrological relationships, which reflect the increase in the proportion of more readily weatherable minerals with increase in basic character:

Acid rocks, eg granite, granitic gneiss	Class 4 (in Table 1)
Intermediate rocks	Class 3
Basic rocks, eg basalt, dolerite, gabbro	Class 2

The relationship between mineralogy and rock type in sedimentary rocks, which are much the most extensive rock types in England and Wales, is less clear. The coarser-grained rocks are usually dominated by quartz (inert, class 5, according to Skokloster), but may contain weatherable minerals. Information is sparse on the amounts and kinds of the latter minerals in soils derived from sedimentary rocks. A review of the literature (Loveland 1984, unpublished) suggested that, on a whole soil basis, the content of weatherable minerals of a grain size greater than about 20 µm, ie those which can be reliably identified by optical microscopy, is less than 1%.

Some soils, especially those derived from fissile materials such as shales and mudstones, contain considerable quantities of coarse fragments, the mineralogy of which reflects that of the parent rock. These contrasting factors were taken into account when assessing the mineralogical classification of soil associations.

A further, very extensive, group of soil parent materials in England and Wales contains significant amounts of clay-sized material (ie particles <2 µm in diameter). The mineralogy of this material varies, but in England and Wales can be classified into three main groups (Loveland 1984):

i. chloritic
ii. micaceous
iii. expansible (dominated by an interstratified mica-smectite)

The first group fits well into Skokloster class 3 (intermediate weathering), as shown in Table 1. The others present problems. Soil micas in England and Wales form a continuous series between muscovite (unweathered) and illite (weathered) (Avery & Bullock 1977; Loveland 1984). Illite is potassium-depleted and more hydrated than muscovite (Brindley & Brown 1980). The Skokloster system (Table 1) places these minerals in different classes, which seemed inappropriate for the UK; thus, the micaceous minerals were grouped together as class 4.

The expansible minerals are noteworthy for their large content of exchangeable cations, most commonly calcium, but also magnesium, potassium and sodium. The proportion of exchange sites populated by these ions rises with increase in pH. Above about pH 7, these minerals are usually regarded as fully saturated with base cations. Between about pH 6.5 and pH 7, ie slightly acidic, the degree of replacement of these base cations by acidic species, mostly hydrogen ions, is thought to be negligible. The total amounts of these exchangeable ions within the soil is controlled largely by the clay content. Therefore, soil particle size distribution (also described, incorrectly, as soil texture) plays an important role in the ability of a soil to buffer against acid inputs. The following textural modifiers were introduced, therefore, based on the textural classes used in England and Wales (Hodgson 1974), and were applied to the initial classification of the soil materials on mineralogy.

Particle size class	Skokloster class
Sand, loamy sand, sandy loam, sandy silt loam	Class 4
Clay loam, sandy clay loam, silt loam	Class 3
Clay, silty clay, sandy clay	Class 2

The effect of particle size becomes particularly marked in the gley major soil groups (major groups 7 and 8 in Clayden & Hollis 1984), which are particularly extensive in the English lowlands south and east of a line drawn from The Wash to the Bristol Channel. In these soils, clay contents are frequently >35%, and often >50%. In this situation, the particle size class modifier frequently moves the soils two Skokloster classes in the direction of decreasing sensitivity.

The influence of pH has been mentioned above, where it is greater than the Skokloster norm of pH 5. Whilst many soils in England and Wales fit this pattern, either from natural causes or because pH is maintained above this value by agricultural practices, some soils are below pH 5. No modifier was introduced for soils between pH 4.5 and pH 5. Where the soil was known to be below this value, and is often below pH 4, the initial classification on the basis of mineralogy was reduced by one unit. In practice, this applied to the very acid, coarser-textured soils of the uplands.

Soil associations dominated by very shallow soils, ie soils markedly less than 50 cm depth, also often contain extensive areas of bare rock or scree. Shallowness will generally make a soil more susceptible to acidification than a deeper soil on otherwise similar parent material. Thus, shallow, non-calcareous soils fall into classes 4 and 5.

Associations dominated by soils containing carbonate (whether the calcium or magnesium form) above 50 cm depth were invariably placed in class 1, irrespective of depth of soil to rock or unweathered parent material.

Reddish soils, ie those containing significant amounts of iron oxides or oxy-hydroxides, sometimes have smaller cation exchange capacities than otherwise similar materials (Loveland 1984), probably because of the blocking of some exchange sites. It is, however, difficult to quantify the magnitude of this effect as it is easily masked by a relatively small change in clay content. Although reddish soils might have a somewhat poorer ability to buffer acid inputs than their non-red counterparts, the present state of knowledge does not permit the introduction of a Skokloster modifier based on this property.

Soil drainage status does not form part of the Skokloster classification system. However, soil solutions will pass more rapidly through well-drained soils than through more slowly drained soils. The latter, therefore, could be seen as having a longer residence factor, which is likely to permit a more complete reaction between acid species in the soil solution and any buffering species in the soil than is likely in a rapidly draining soil. Well-drained soils were regarded, therefore, as more susceptible to acidification than poorly drained soils. The effect of these changes is shown in Table 2.

Table 2. Modified Skokloster classification (source: Sverdrup & Warfvinge in UN-ECE 1990)

Mineral class		
1	Very fast weathering	carbonates
2	Fast weathering	pyroxene, epidote, olivine (<5%)
3	Intermediate weathering	biotite (>5%), amphibole (<5%), chlorite
4	Slow weathering	biotite (<5%), muscovite/ illite, plagioclase, vermiculite, smectite, interstratified clay minerals
5	Inert	quartz, K-felspar, kaolinite
6	Organic	peats

SOURCES OF SOIL MINERALOGICAL INFORMATION FOR ENGLAND AND WALES

The information is patchy both in terms of geographical cover and the range of mineralogy examined. The position is summarised by Loveland (1984). The literature was re-examined during the course of this project (Loveland, unpublished), but few significant additions were found. The Soil Survey and Land Research Centre also had unpublished information on the mineralogy of the soils of England and Wales, and this was utilised. The dominant mineralogy of each soil association was assessed from these sources.

THE SOIL BASE MAP FOR ENGLAND AND WALES

The national soil map of England and Wales is published in six sheets at a scale of 1:250 000 (Soil Survey 1983). The smallest area of land which can be portrayed as an identifiable unit, eg by colouring, at that scale is about 0.25 km^2 in extent. The map shows 296 separations (map units), each of which represents a geographical soil association. The latter is identified by the name of the most extensive soil series within the association, ie the dominant soil series. A soil series is distinguished from other soils by possession of a particular combination of relatively permanent physico-chemical properties, such as particle size distribution, and the sequence of soil layers to a depth of about 1 m (or less, if rock is encountered at a shallower depth). A soil series is named after the geographical location where the type example was first identified, eg Andover Series. Although the geological parent material exercises considerable influence on basic soil properties, soil series are not defined or delineated solely in terms of parent material. Thus, soils derived from different parent materials occur within the same soil association (and *vice versa*), albeit the variation in the kind of parent material within any one soil association is small. For example, acid igneous rocks and their associated soils are separated from basic igneous rocks, whilst soils derived from calcareous materials are separated on the basis of types of limestones and associated sediments.

The soils subgroup of CLAG agreed that, for the purposes of this research, 'soil' is the upper 50 cm of the surface material. This layer is most likely to receive most of the acid inputs. Thus, in considering soil properties a judgement was arrived at which integrated the properties of this depth of soil. This judgement represents a compromise, the validity of which requires further investigation.

APPLICATION OF THE PRINCIPLES

The principles outlined above were applied to each of the soil associations on the national soil map in turn. A guiding principle was to arrive at the most sensitive class, indicated by the mineralogy or the modifiers discussed above, given that the assumptions in the exercise were untested. The results are illustrated in Table 3, which shows part of the final compilation of the properties of all the soil associations. The digitised national soil map was broken down into squares, 1 km x 1 km in area, referenced to the south-west corner of the Ordnance Survey's National Grid of Great Britain. The dominant soil association (above) within each square was assigned automatically to its appropriately modified Skokloster class from a computer-held table. That is, the association was first assigned to a class on the basis of the soil mineralogy, and then this initial assignment was modified, where appropriate, using particle size, pH and depth. In effect, the dominant soil series in each association determined the class to which the association was assigned. It is important to realise that the dominant soil series within an association might not be the most sensitive soil within that 1 km square, because the most sensitive soil might occupy such a small area of land that it was not taken into account when formulating the association, or the area was not large enough to portray cartographically. Table 4 gives the proportion of soil associations within the modified Skokloster classes in terms of the soil cover of England and Wales. In UN-ECE terms (UN-ECE 1990), therefore, the map is a 'level 0' map.

Table 3. The soil associations of England and Wales and their classification according to a modified Skokloster scheme. A representative group of the map units corresponding to those of the 1:250 000 national soil map of England and Wales (source: Soil Survey 1983)

Map unit	Soil association	Geology	Mineral-ology	Initial Skokloster class	Texture class	Comment	Land use	Final Skokloster class	Extent (km^2/%)
22	Unripened gley soil	Marine alluvium	MS	4	C	Tidal	Salt marsh	3	319/0.21
311a	Revidge	Pal grt & sst	Mic/Q	5	S	Shallow /v. acid	MRG	5	103/0.06
311b	Skiddaw	Pal mdst & zst	Chl	3	ZL	ditto	MRG	4	138/0.09
311c	Wetton 1	Carb lst	Calc	1	-	ditto	PG	1	136/0.09
311d	Wetton 2	ditto	ditto	1	-	ditto	PG	1	162/0.10
311e	Bangor	Acid igneous	Mic/Fel	4	-	ditto	MRG	4	533/0.35
313a	Dunwell	Basic igneous	FM	4	L	ditto	ST	4	48/0.03
313b	Powys	Pal mdst & zst	Chl	3	L	Shallow	ST	4	253/0.16
313c	Crwbin	Carb lst	Calc	1	L	ditto	ST	1	406/0.26
341	Icknield	Chalk	Calc	1	L/Z	ditto	ST/A	1	390/0.25
342a	Upton 1	Chalk	Calc	1	Z	ditto	ST/A	1	764/0.50
342b	Upton 2	Chalk	Calc	1	Z	ditto	A/ST	1	67/0.04
342c	Wantage 1	Silty drift	Calc	1	Z	Shallow in part	A/ST	1	94/0.06
342d	Wantage 2	ditto	Calc	1	Z	Coarse in part	A	1	138/0.09

continued over

Table 3. continued

Map unit	Soil association	Geology	Mineral-ology	Initial Skokloster class	Texture class	Comment	Land use	Final Skokloster class	Extent (km^2/%)
343a	Elmton 1	Jur lst	Calc	1	FL	Shallow	A/ST	1	1351/0.89
343b	Elmton 2	Jur lst & Slst	Calc	1	FL	Shallow & brashy	A/ST	1	319/0.21
343c	Elmton 3	Jur lst & clay	Calc	1	FL/C	ditto	A/ST	1	214/0.14
343d	Sherborne	ditto	Calc	1	FL/C	ditto	A/ST	1	1122/0.74
343e	Marcham	Jur lst	Calc	1	CoL & FL	Shallow	A	1	75/0.04
343f	Newmarket 1	Chalky drift	Calc	1	S & CoL	Variable depth	A/F	1	251/0.16
343g	Newmarket 2	Chalky drift	Calc	1	CoL & S	ditto	A/ST	1	443/0.29
343h	Andover 1	Chalk	Calc	1	Z	Shallow	A/ST	1	2845/1.87
343i	Andover 2	Chalk/CwF	Calc	1	Z with C	ditto	A/ST	1	413/0.27
346	Reach	Chalky drift	Calc	1	FL	Shallow /humose	A	1	47/0.03
361	Sandwich	Dune sand	Calc	1	S	Flood often	R/F	1	343/0.22
372	Willingham	Lake marl	Calc	1	Z & C	Often humose	A	1	43/0.02
411a	Evesham 1	Jur C & lst	Calc	1	C	Brashy in part	ST/A	1	923/0.06
411b	Evesham 2	Jur/Cret clay	Calc	1	C	Poor drainage	A/ST	1	1119/0.73
411c	Evesham 3	ditto	Calc	1	C & FL	ditto	A/ST	1	809/0.53
411d	Hanslope	Chalky till	Calc	1	C	ditto	A/ST	1	3634/2.40
421a	Stow	Jur mdst	Mic	4	C & FZ/FL	Steep	ST/A/F	4	107/0.07
421b	Halstow	Carb shale	Mic/Chl	4/3	C/FL	Mineral varies	ST/A	3	278/0.18
431	Worcester	PT red mdst	Chl/Calc	1	C/FL	Slow drainage	ST/A	1	925/0.61
511a	Aberford	Mixed lst	Calc	1	FL	Brashy	A	1	1018/0.67

Abbreviations

1. Mineralogy (principal species)
Kaol=kaolinite; Mic=muscovite or illite; Q=quartz; Chl=chlorite; MS=mica-smectite; P=organic soil(peat); Calc=carbonates; FM=ferromagnesian minerals; Fel=felspar

2. Land use (for each association this is given in decreasing order of importance)
MRG=moorland and rough grazing; ST=stock rearing and permanent grassland; A=arable crops; W=woodland (dominantly deciduous); F=forestry (dominantly coniferous); R=recreation; H=heathland; B=bog

3. Particle size class
C=clay(ey); Z=silt(y); S=sand(y); L=loam(y); FL=fine loamy; CoL=coarse loamy

4. Stratigraphic divisions
PC=PreCambrian; LP=Lower Palaeozoic (Cambrian, Ordovician, Silurian); Pal=Palaeozoic; Carb=Carboniferous; Cret=Cretaceous; Mez=Mesozoic; Jur=Jurassic; Dev=Devonian; PT=Permo-Trias; Tert=Tertiary; CwF=Clay-with-Flints

5. Rock types
sst=sandstone; grt=gritstone; Bi=basic igneous rocks; Ai=acid igneous rocks; Int=intermediate igneous rocks; met=metamorphic rocks; lst=limestone; mdst=mudstone; zst=siltstone; G/F=glaciofluvial; G/L=glaciolacustrine; P=peat; all=alluvium
Particle size modifiers are also used, eg Slst=sandy limestone

NB Where two descriptors are given separated by '/', the first is dominant

Table 4. Proportions of soils in each Skokloster class

Class	Nos of km^2	Land area (%)
1	35 525	22.30
2	30 856	20.31
3	36 667	23.72
4	26 030	19.15
5	5 820	3.67
6	4 667	3.02
Disturbed soils	532	0.35

CONCLUSIONS

A modified Skokloster mineralogical classification has been used to produce a map at 1 km resolution of the soil associations of England and Wales. This map can be used as a basis for interpreting deposition loads of sulphur and nitrogen.

The exercise is based on limited mineralogical and soil data, although the information is the best available. No attempt was made to validate the assessments which underpin this exercise, in terms of actual soil reaction or soil variability, against measured or modelled data. A validation exercise which involves a thorough study of soil mineralogy, soil and atmospheric chemistry at a number of carefully selected sites is a natural and necessary progression from this exercise.

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Critical loads of acidity for forest soils, groundwater and first-order streams in Sweden

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CRITICAL LOAD CONCEPT

The critical load was defined by the international workshop on critical loads held at Skokloster, Sweden, in 1988, as:

> 'the maximum amount of sulphur and nitrogen deposition that will not cause long term damage to ecosystem structure and function'.

During the course of 1989, 29 nations in Europe agreed to have critical loads in their countries mapped by the end of 1991. In Europe, the work is co-ordinated through the Coordination Centre for Effects located at the Dutch Environmental Research and Protection Agency (RIVM) in Bilthoven, The Netherlands, by the Task Force on Mapping at Umweltbundesamt, Berlin, Federal Republic of Germany, on behalf of the United Nations Economic Commission on Europe. The critical load maps will be used as an input to the Working Group on Abatement Strategies (WGAS), for optimising the abatement strategies for the whole of Europe. For the mapping work, a manual has been developed as a guideline (Sverdrup, De Vries & Henriksen 1990). Critical loads are determined for a number of different receptors, such as:

- first-order streams
- forest soils
- groundwater

The critical load is not calculated for a lake or forest as a physical entity, but for the lake or forest as an ecosystem.

Effects of acid deposition on soils

The development of the chemical status of the soil in a catchment is determined by the amount and composition of the solution percolating through the soil. Acidification of soils, and consequently lakes and streams, is caused by the input of acidity to the catchments. This is the agent which is consuming alkalinity produced in the soil, and causing leaching of the cation ion exchange matrix, reducing soil base saturation. In soils, acidity is consumed by chemical weathering reactions with silicate minerals. The rate of neutralisation is dependent on the chemical and physical properties of the soil, as well as the chemical conditions. The weathering of the mineral matrix will be the major long-term source of alkalinity to neutralise acidity in the system, as well as the major supply of base cations to replace removed base cations, and an important factor for determining the chemical status of the soil. In calculating critical loads, all the sources of acidity in the ecosystem are balanced against all sources of alkalinity, and at steady state, when no acidification of the system is permitted, the exchange alkalinity term must be zero. The sources of acidity to the ecosystem are balanced by the only long-term source of alkalinity, the weathering of minerals in the soils in the catchment.

Effects of acid deposition on trees

A number of adverse effects from acidification may affect trees. Trees take nutrients from the soil in proportion to their availability in the soil solution, by adsorption to roots prior to plant uptake. In an acid soil, two things may occur. High concentrations of hydrogen and aluminium will cause less nutrients and more aluminium to adsorb to the roots, and the tree will have to overcome a 'counter-pressure' of aluminium to get nutrients, and, second, if aluminium and heavy metals released in the soil by acid conditions get taken up, they will be able to poison the plant.

Acidification will strip the ion exchange complex of base cations, making them temporarily available in larger amounts than the natural supply, and thereby participating in the depletion of the soil storage of nutrients. Under increased nitrogen deposition, there will be temporarily good conditions for increased growth, as long as acidification helps in increasing the supply of base cations by depleting the ion exchange reservoir. As the soil gets more acidified and the storage is depleted, both base cation availability and uptake efficiency decrease.

In assessing growth rate reductions and estimating the costs of forest production reductions due to acidification, the relationship between the (Ca+Mg)/Al ratio and tree growth is important. The data available have been derived for spruce (*Picea* spp.) and fir (*Abies* spp.) seedlings in pot experiments and *in situ* observations of growth

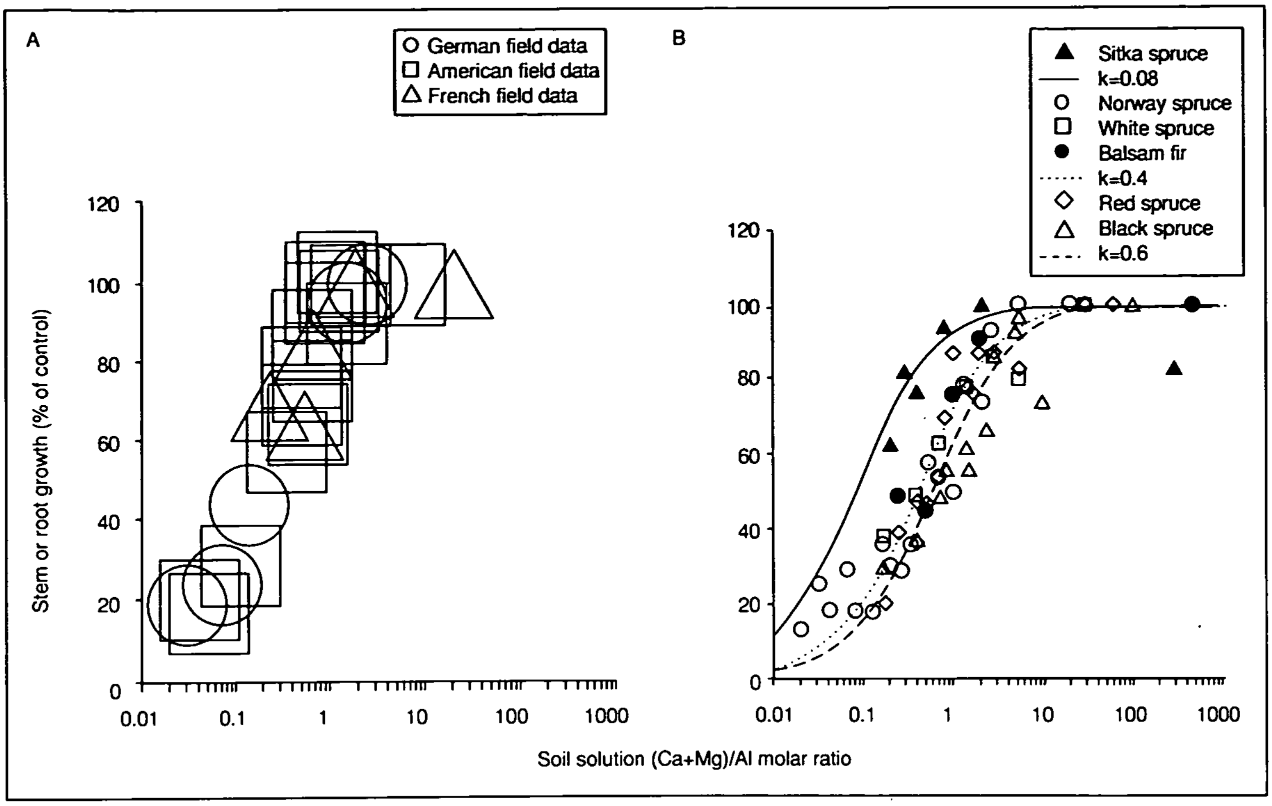

Figure 1. A, Field forest growth and B, root growth under laboratory conditions, in relation to the soil solution (Ca+Mg)/Al ratio, as derived from different literature sources

decline in Germany, North America and France. The connection between soil chemistry and forest growth decline is based on a re-evaluation of 200 bioassay studies in laboratory and field (Sverdrup, Kuylenstierna & Chadwick 1993).

The relationship between forest growth, acidification of the soil, the soil solution concentration of aluminium, base cations and the (Ca+Mg)/Al ratio, as derived from different literature sources, is shown in Figures 1 and 2. The data are either extracted from laboratory bioassays in Germany and Sweden (see Sverdrup, Warfvinge & Rosen 1992; Sverdrup *et al.* 1993) or from field studies in Germany, France and United States (Bonneau 1991; Becker 1991; Landmann 1991; Barnard 1990; Ulrich 1985). The net effect on the trees is that they grow slower, or in the worst case die, either because of nutrient deficiency or secondary causes affecting the weakened plant. In addition to these soil acidification effects, other environmental factors such as sulphur dioxide, nitrogen oxide and ozone fumigation, and climate effects such as drought and frost may contribute to the demise of trees.

Despite all the evidence, it is still sometimes disputed whether acidification affects trees directly; it is often claimed that Sweden's forests are producing more biomass than ever at the moment. For a large part of the Nordic countries, this statement may hold true for the time being. During the last 60 years, nitrogen deposition has increased sharply from 1-2 kg N ha^{-1} yr^{-1} to 20-30 kg ha^{-1} yr^{-1}. Trees in the boreal zone are accustomed by evolution to a deficiency situation

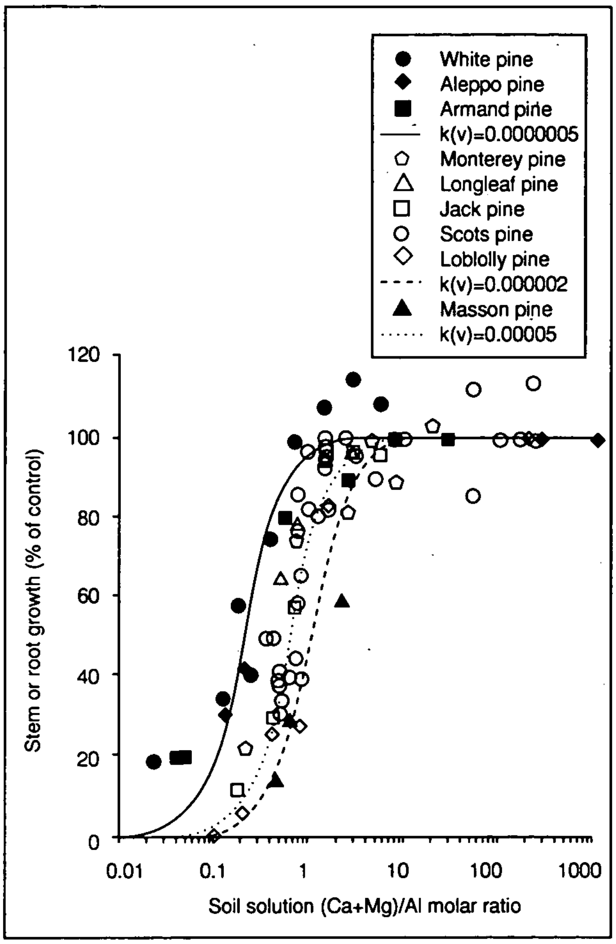

Figure 2. The relationship between forest growth, acidification of the soil, the soil solution concentration of aluminium, base cations and the (Ca+Mg)/Al ratio, as derived from different literature sources

for nitrogen, and hence nitrogen will greatly enhance tree growth. Tree growth in the Nordic countries has been strongly enhanced in this way. At the same time, acidification has helped to make base cations in the ion exchange reservoir available for the nitrogen-induced increased growth. If the weathering rate is insufficient to supply this increased demand for base cations, then the accelerated base cation uptake will rapidly deplete the forest reservoir of base cations. Figure 3 illustrates how base cations are initially made more available by acidification, but drop off sharply as the ion exchange complex becomes completely depleted. It also shows very clearly that good growth in the past cannot be just extrapolated into the future, as such simple models do not allow for predictions of sudden catastrophic changes in the system. For such events, explanatory prediction tools based on actual mechanisms are needed.

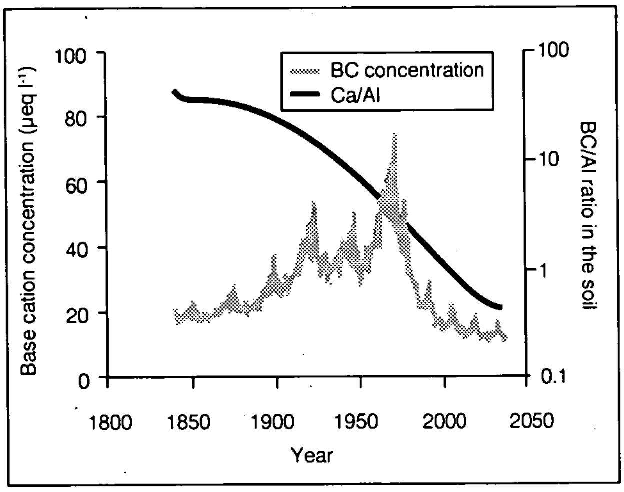

Figure 3. Typical development of base cation and BC/Al ratio over time in a podzolic forest soil receiving acidic deposition which exceeds the critical load

Receptors mapped

The present forest critical load map applies to all types of forests comprising Sweden's forest area of 267 000 km^2. For the forest assessment, each calculation point was area-weighted in relation to the absolute local density of sites and receptor density. For the surface water assessment, each point was also taken to represent a first-order stream. The result is related to streams rather than lakes as no sediment processes or lake-specific processes have been included. Each point is taken as representative of the groundwater at the location. The forest vegetation includes the southern Swedish deciduous forest, the coniferous forest and the northern birch (*Betula* spp.) forest.

METHODS

Introduction

The soil processes affecting major constituents in the natural soil are many. It is important to realise that it is not possible to assess single soil processes without taking all other significant processes quantitatively into account. The most important soil ecosystem processes for the control of base cation fluxes are chemical weathering, base cation deposition, vegetation net base cation uptake, hydrological transport, and ion exchange. Ion exchange can only be ignored when it can be proven that the system is at steady state, which requires a major long-term monitoring programme. In addition to the soil processes listed above, acidity deposition, nitrogen uptake and nitrification have a key influence over the above-mentioned processes, by modifying or partially controlling the chemical conditions in the soil. In addition, temporal imbalances may make biomass decomposition and vegetation recycling of base cations important.

The potential acidity in terms of deposition components is defined as the sum of total sulphate plus nitrate plus ammonium minus non-marine base cation deposition. The marine base cation deposition is considered to be acidity/alkalinity neutral. The total acidity input to the soil can, however, be larger than the deposition load alone. The total acidity input from external sources is the sum of atmospheric inputs plus the net effect of any type of land use or acidity-producing conversion of deposition components. Deposition of alkaline marine salts and continental rock dust will have a neutralising effect. The critical load of actual acidity indicates the total load of acidity input the system can tolerate, regardless of its source.

Criteria used

Examples of the chemical criteria used for setting critical loads of acid deposition are shown in Table 1. The criterion Ca/Al≥1.0 was used for calculating critical loads for forest soils. For surface waters, it was required that streamwater ANC≥50 µeq l^{-1}. For groundwater, it was required that ANC≥150 µeq l^{-1} at 2 m depth in the soil.

Table 1. Examples of the chemical criteria suggested for use in setting critical loads of acid deposition

Criteria	Forest soils	Groundwater	Lakes and streams
Location of application	Root zone 0-50 cm	At groundwater level	Lake volume weighted Stream transect
pH	E layer : ≥pH 4.0 B layer : ≥pH 4.4	≥pH 6.0	≥pH 6.0
Alkalinity	≥-300 µeq l^{-1}	≥100 µeq l	≥ 50 µeq l^{-1}
Total aluminium	≤4.0 mg l^{-1}	≤0.1 mg l^{-1}	≤0.08 mg l^{-1}
Labile aluminium	≤2.0 mg l^{-1}		≤0.03 mg l^{-1}
BC/Al molar ratio	≥1.0		≥5

The PROFILE model

Critical loads for forest soils in Sweden were calculated using the steady-state mass balance model, implemented as the Apple Macintosh version of the PROFILE model. Units used are keq ha^{-1} yr^{-1}, if not otherwise specified. The soil profile is divided into four layers, using input data for the thickness of each soil layer (O, A/E, B, C). The criteria were applied in the 0-50 cm layer of the soil, assumed to be the tree rooting depth in Sweden. In the PROFILE model, all soil processes communicate via the soil solution, and the model incorporates chemical weathering, nitrification, nitrogen and base cation uptake to vegetation, hydrological transport of dissolved substances, solution equilibrium for all species, and aluminium solid-liquid equilibrium. In the dynamic model SAFE (Soil Acidification in Forested Ecosystems), kinetic ion exchange is included. Temperature dependence is considered for all individual processes, equilibria and reactions involved. The PROFILE model has been set in an automated framework, reading input data from a file containing the regional data base. This data base is based on the Swedish Forest Survey Programme. The capability of the PROFILE model for calculating weathering rates has been tested on 29 independent determinations of field weathering rates (Sverdrup *et al.* 1992; Jönsson, Warfvinge & Sverdrup 1993). The model calculates weathering rates with an accuracy of ±15%.

The PROFILE model may also be used to calculate the steady-state chemistry in response to a certain deposition input. The model is simply fed with the different deposition scenarios corresponding to different reduction scenarios, and the corresponding steady-state chemistry in terms of BC/Al ratio or alkalinity is calculated. For the steady-state calculation, the prehistoric deposition was used as input to create a frame of reference. To translate the steady-state soil chemistry to ecological effects, the damage model for spruce as shown in Figure 1 was used:

$$f = \frac{BC}{BC + k \, . \, Al} \qquad (1)$$

where $k = 0.40$ for Norway spruce (*Picea abies*). The field data are based on growth *versus* the BC/Al ratio in the B layer, not considering potential higher values in other soil layers. In PROFILE, growth reduction is calculated for each soil horizon, and the plant is allowed to reallocate uptake from one layer to another.

INPUT DATA

All the data in this work are based on the framework of the Swedish national survey of forest data (Rikskogstaxeringen/Standortskarteringen). It consists of a network of stations evenly spread over the complete forest area of Sweden (267 000 km^2). Soil samples were collected down to the C layer at approximately 60 cm depth, and their basic soil parameters were analysed. The list below represents the input data required for the PROFILE model for each site in the calculations: total deposition of sulphate, nitrate, ammonium, base cations, precipitation, runoff and mean annual temperature, soil mineralogy of the C layer, soil texture class for the C layer, base cation uptake and nitrogen uptake. For the catchment weathering rate, depth to bedrock or effective hydrologically active depth must also be specified. Several other parameters, such as CO_2 pressure, soil solution dissolved organic carbon, distribution of uptake and evapotranspiration, and gibbsite coefficients have less influence and are entered as standard values taken from the literature; they generally remain constant between runs and sites. Annual average air temperature, precipitation and runoff were taken from the official statistics of the Swedish Meteorological Institute (SMHI), for each NILU grid (50 km x 50 km). Other input data were derived strictly in accordance with Sverdrup *et al.* (1990).

Uptake

Basic vegetation data have been measured at 27 000 stations, and soil parameters such as Ca/Al ratio, soil solution pH (H_2O), C/N ratio, base saturation, etc, at 1804 sites. For each site, the net long-term uptake is specified, based on the measurements in the survey of base cation and nitrogen contents of stem and branch, combined with site-specific estimates of net long-term forest growth. Each tree species is considered separately, and the total uptake weighted together for each calculation point. In the model, base cation uptake will occur from each layer, but uptake is stopped if the base cation soil solution concentration falls below 15 μeq l^{-1}. Residual uptake is then moved to the next layer.

Mineralogy

The soil mineralogy was derived for 124 sites from measurements by the Swedish Geological Survey (SGU) at Uppsala. Total elemental content of the soil after complete dissolution was determined for all 1804 sites, and analysed using generic wet chemistry methods for Ca, Mg, Na, K, Al, Si, Fe, Ti and trace metals.

The mineralogy was determined by separating the sample into two density classes and two particle size classes, as illustrated in Table 2. Each subsample was analysed for total elemental composition using wet chemistry methods, and subsequently analysed for elements using XRF, and for minerals using XRD and known standards of mixtures of pure mineral phases. The total analysis implies that the minerals in a soil sample are totally dissolved in hot hydrofluoric acid or a lithium borate-alkali melt, diluted in water and analysed using standard methods. The fast-weatherable minerals will tend to concentrate in the heavier fraction, and the analysis can then be carried out on the enriched fraction, even though they may be present in very small amounts in the whole soil sample. Separation of the mineral sample in a liquid with density in the range 2.680 g cm^{-2} will generally concentrate easily weathered minerals 30-70 times. The mineralogy determination is carried out on the silt and clay fraction of the soil, as all particles larger than 50 µm dissolve too slowly to be significant for the weathering rate. These data are used to estimate the mineralogy of the whole soil sample.

Table 2. The mineralogy analysis scheme required each soil sample to be divided into four parts, based on density and particle size

	Light fraction ≤2700 kg m^{-3}	Heavy fraction ≥2700 kg m^{-3}
Sand-silt	I	II
≤2 µm	Felspars Quartz Muscovite Biotite	Pyroxene Amphiboles Epidotes Biotite Nesosilicates
Clay	III	IV
≤2 µm	Vermiculite Montmorillonite Quartz	Chlorite

The UPPSALA model is a back-calculation model for reconstructing the mineralogy from the total digestion analysis in order to provide input to models like PROFILE from simple survey data. After an initial survey of Sweden's soil mineralogy in what was defined as different geological provinces (areas with differing mineralogy but still believed to have originated from similar bedrock types); it was realised that mineralogy for these geological provinces would be correlated to the total analysis chemistry. Total analysis includes Ca, Mg, Na, K, Al, Si, Fe in a totally dissolved sample of soil. The mineralogy model is based on an *a priori* knowledge or assumption on the stoichiometric composition of the minerals of the particular soil. The minerals have been grouped into assemblies with similar composition and dissolution rate. Accordingly, for soils of granitic origin or soils derived from rock of secondary granitic origin (sandstones) as well as originating from different types of schists and shales, it is assumed that the following mineralogical groupings are valid. Muscovite is assumed to comprise muscovite, secondary di-octahedral illite, di-octahedral chlorite and vermiculite of secondary weathered type. Chlorite comprises tri-octahedral chlorite, primary illite and tri-octahedral vermiculite of primary type, as well as biotite, phogopite and glauconite. Hornblende implies all amphiboles, such as hornblende, riebeckite, arfvedsonite, glauconite and tremolite. Epidote comprises all epidotes, zoisites and pyroxenes. Plagioclase has for Sweden been assumed to be oligoclase with 80% albite felspar component. K-felspar is assumed to be 10% albite felspar component. All phosphorus has been attributed to apatite. The equations used in the UPPSALA model for calculating the content of the individual minerals are:

K-felspar = $5.88\ k_20 - 0.588\ Na_2O$
Plagioclase = $11.1\ Na_2O - 0.22\ KF$
Apatite = $2.24\ P_2O_5$
Hornblende = $6.67\ CaO - 3.67\ AP - 0.2\ OL$
Muscovite = $2.08\ K_2O - 0.208\ Na_2O$
Chlorite = $3.85\ MgO - 0.39\ HO - 0.39\ MU$
Epidote = $0.10\ HO + 0.03\ OL - 0.3$
Quartz = $SiO_2 - 0.63\ OL - 0.45\ HO - 0.33\ CL - 0.38\ MU - 0.42\ EP - 0.68\ KF$

where KF = K-felspar; OL = oligoclase; AP = apatite; MU = muscovite; CL = chlorite; QU = quartz; VE = vermiculite; and EP = epidote. The equation is checked by calculating the amount of quartz, and only such sites which lie within the range 95-105% are accepted. Using this criterion reduced the 1913 sites to 1804, and of these approximately 50 were estimated to be calcareous soils to which the mineralogy model does not apply in any case. The weak point in the UPPSALA model is the expression for epidote. Epidote occurs only in small amounts, in most soils of granitic origin. The expressions do not correspond to normative calculations based on absolute stoichiometries for pure minerals, but rather represent a set of empirical field stoichiometries.

Texture

Texture was measured by granulometry and BET/adsorption analysis on the 124 mineralogy analysis samples, and correlated against field texture classification. The texture for all 1804 sites was read from the correlation using the field classification. The simplest way to calculate the soil texture is from a particle size distribution. The conversion from a standard sieve curve to surface area was carried out using:

$$A_{tot} = (x_{clay} \cdot 8.0 + x_{silt} \cdot 2.2 + x_{sand} \cdot 0.3) \cdot \frac{\rho_{soil}}{1000} \qquad (2)$$

where A has the dimension $10^6\ m^2\ m^{-3}$ soil, and where we have the condition:

$$x_{clay} + x_{silt} + x_{sand} + x_{coarse} = 1 \qquad (3)$$

and x_{clay} = the fraction of soil particles less than 2 µm; x_{silt} = the fraction of 2-60 µm; x_{sand} = the fraction of 60-200 µm; and x_{coarse} = the fraction of soil matrix coarser than 200 µm. r_{soil} is the bulk density of the soil in kg m^{-3}. The granulometric approach is used because of difficulties of interference from secondary precipitates and organic matter with the BET measurements on whole soil samples. BET measurement is good for soil samples unaffected by these factors. The granulometric relation was determined by measuring BET surface on a series of soil samples divided up into sand, silt and clay fractions. Untreated fractions and those where the precipitates and organic matter had been removed were used. A relation between the field classification of soil texture and laboratory measurements of exposed surface was found:

$$A_{tot} = 0.093 \cdot e^{(0.51\,TX)} \qquad (4)$$

where TX = the texture class; and A_{tot} = the total surface in 10^6 m^2 m^{-3} soil. The relation is based on determinations of surface area and texture class on 100 soil samples. The texture class is determined in the field by placing a small soil sample on the palm of the hand, spitting on it and rolling the smallest possible string that will still keep together. The texture class is judged from the thickness of the string.

Deposition

Deposition data were prepared by the Institute of Water and Air Research (IVL) at Göteborg, from the Swedish deposition monitoring network and European Monitoring and Evaluation Programme data, modified with filtering factors for different vegetation types. The total deposition is calculated for each calculation point using the filtering factors and the vegetation mixture for that point.

RESULTS

Critical loads of acidity

Critical loads and areas where they have been exceeded for forest ecosystems are shown in Figure 4, and for streams on moraine soils in forested landscapes in Figure 5. The mass balance approach shows more streams and lakes to be sensitive to acidic deposition than the Henriksen model. Both the PROFILE and the Henriksen models include large uncertainties when applied to streams. For the PROFILE model calculations, only average soil depth in each 50 km x 50 km grid was available, which makes the distribution of critical loads within each grid narrower. In the Henriksen model, large uncertainties are connected to the F factor, a substitute variable for weathering rate. Critical loads and areas where they have been exceeded for groundwaters at 2 m depth in moraine are shown in Figure 6.

Chemical effects on soils

Steady-state chemical states were calculated for forest soils, streams and groundwater, in order to be

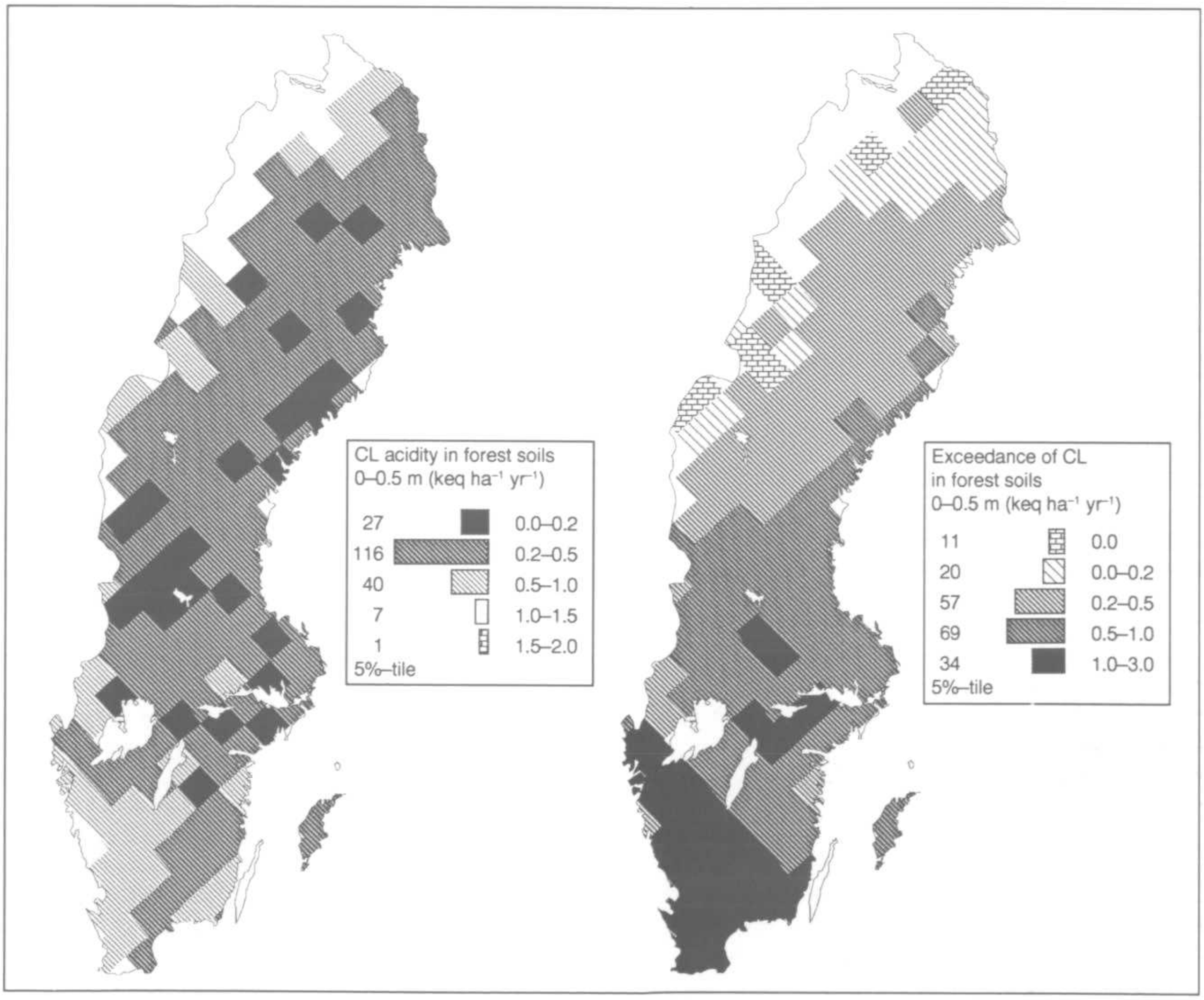

Figure 4. The critical loads and areas where they have been exceeded for Swedish soils; 82% of Sweden's forest area receives more acidic deposition than the critical load

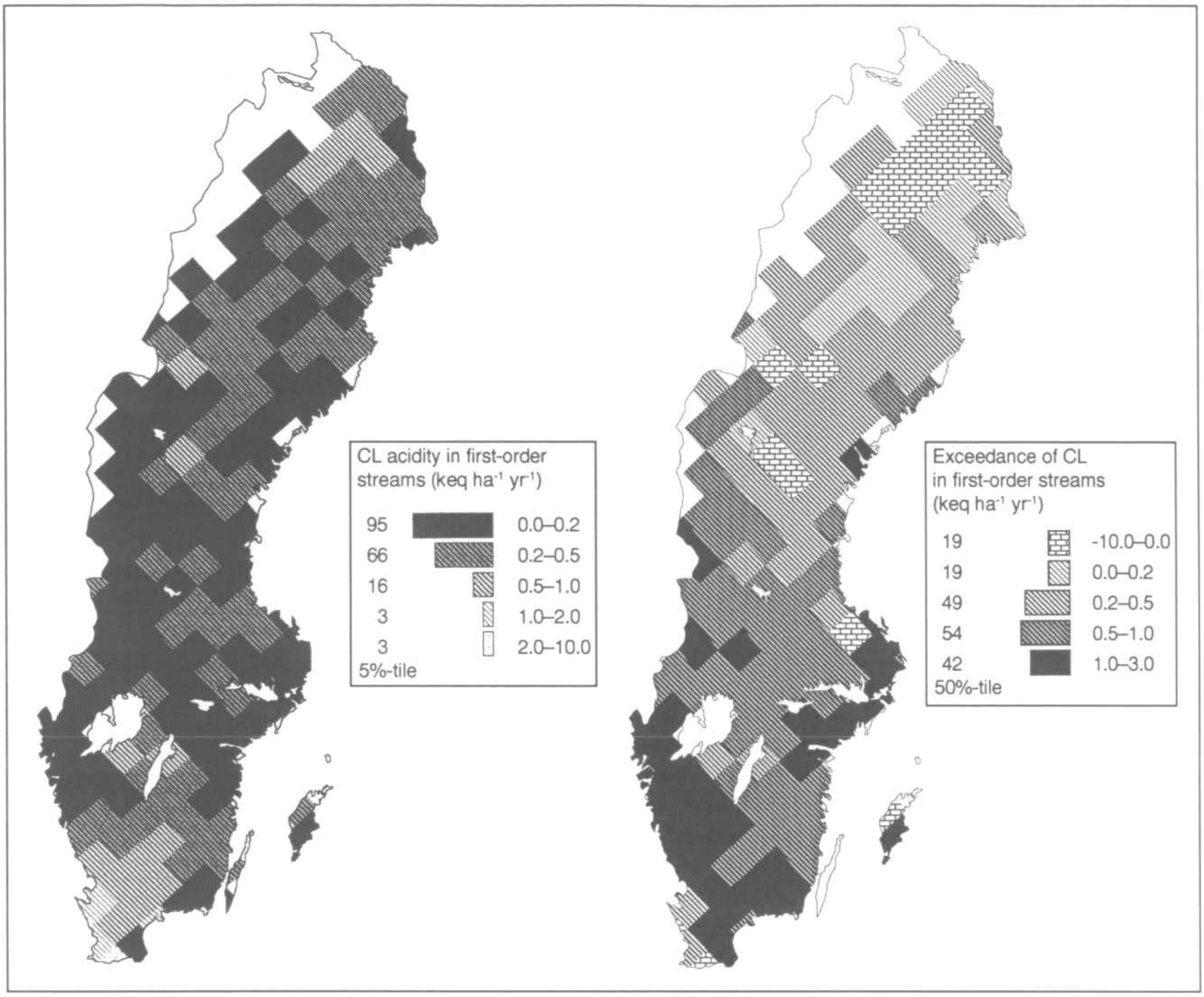

Figure 5. Critical loads and areas where they have been exceeded for streams on moraine soils in forested landscapes

able to judge the effects of different levels of exceedance over critical loads. For forest soils, BC/Al was calculated; for streams, ANC of the water was calculated.

A pre-industrial deposition level (1840) was chosen in order to illustrate what the situation once looked like before the advent of acidic deposition. The different deposition scenarios considered have been listed in Table 3. Future steady-state chemistry corresponding to different deposition levels is illustrated in Figures 7-9. The 60% S/30% N reduction level corresponds roughly to planned emission reductions in Europe.

Table 3. The different deposition scenarios considered in the steady-state calculations

Scenario	Sulphur	NO_3-N	NH_4-N
1990 level	±0%	-	-
Future 1	-30%	-	-
Future 2	-60%	-	-
Future 3	-60%	-30%	-30%
Future 4	-85%	-30%	-30%
1840 level	-95%	-95%	-85%

In southern Sweden, the (Ca+Mg)/Al ratio fell to a lower level at an earlier date on comparable soils. The effect of this change has been illustrated in Figure 10 where the calculated development of the (Ca+Mg)/Al ratio using SAFE has been combined with a growth model (Sverdrup *et al.* 1992), responding to (Ca+Mg)/Al ratio and base cation concentration. From 1920 to 1980, the nitrogen deposition has steadily increased over the stand. The model predicts that the (Ca+Mg+)/Al ratio will fall below 1 around the year 2000.

Table 4 presents suggested regional target loads for areas of Sweden.

Chemical effects on streams and groundwater

Steady-state stream alkalinity was calculated for streams. The numbers shown in Figure 8 are annual

Table 4. Suggested target loads using regional critical loads for Sweden, based on the present calculations

Target load	Skåne	Götaland	Svealand Norrland
kg S ha^{-1} yr^{-1}	6.0	4.0	3.0
kg N ha^{-1} yr^{-1}	6.0	3.0	1.0

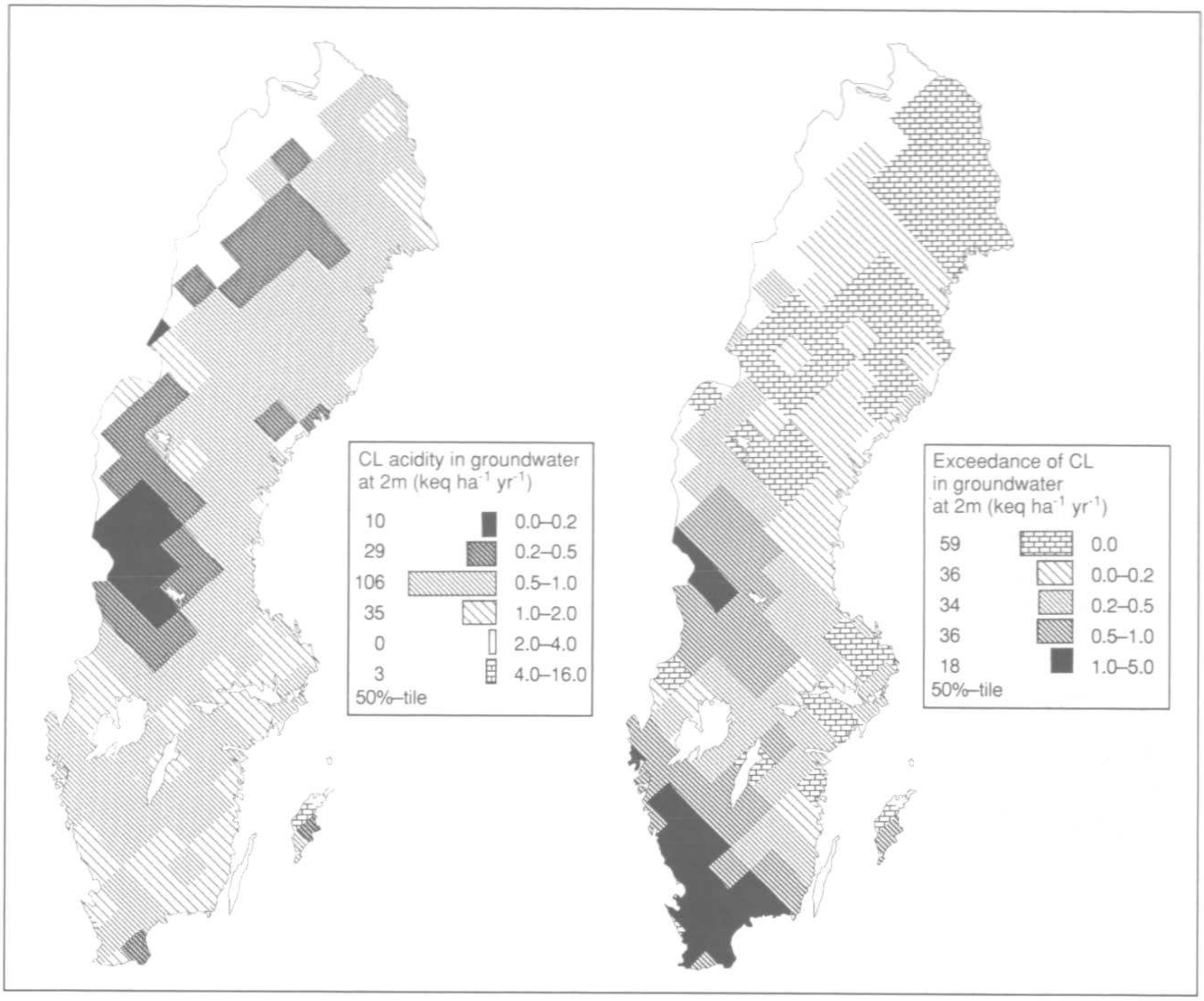

Figure 6. Critical loads and areas where they have been exceeded for groundwater at 2 m depth

water volume averaged values only. Under present deposition levels, the effect on Swedish streams will be very large in terms of adversity for fish. Deposition reductions in the same order of magnitude as required for forests are required to protect 90% of the lakes. For groundwater, alkalinity has been calculated for percolating water to the 2 m depth level (Figure 9). Water may be more alkaline further down in the aquifer, but these values indicate the alkalinity of water replenishing the aquifer.

Ecological effects on forests

Calculated BC/Al was used to calculate future growth reductions, using the response functions shown in Figures 1 and 2. The results for forest growth in Sweden are shown in Figure 10 for current deposition levels and for some reduction scenarios, and are summarised in Table 5. The calculations indicate that acid rain can significantly reduce growth in Swedish forests to a degree where it will be visible in the field. The growth reductions will affect both commercially exploited forests as well as forests not cropped at the present time. Sensitive tree species will be at a disadvantage, and forest tree species composition may change significantly over the long term. Figure 11 shows that the Ca/Al ratio in soil water can deteriorate very rapidly under certain deposition scenarios, especially in high-yield sites.

Table 5. Forest growth loss in Sweden due to soil acidification using known cause relationships in economic terms (10 SEK = 1 US$)

	(Ca+Mg)/Al-ratio						
Region	≥1.0	1.0	0.3	≤0.1	Growth lost	Growth reduction	National loss of profit
	% of forest growth				($10^6 m^3 yr^{-1}$)	(%)	(10^9SEK yr^{-1})
1990 level	19	34	46	1	17	20	4.3-8.7
-30% S	44	34	22	1	14	16	3.5-7.0
-60% S	54	28	18	-	10	12	2.5-5.0
-60% S / -30% N	83	14	3	-	7	9	1.8-3.5
-85% S / -30% N	96	3	1	-	6	7	1.5-3.0
1840 level	100	-	-	-	5	6	1.3-2.5

Economic effects on forests

In Sweden there is 267 000 km^2 of forest, 235 000 km^2 of which is considered productive. The annual net growth is 85 million m^3, approximately 70 million m^3

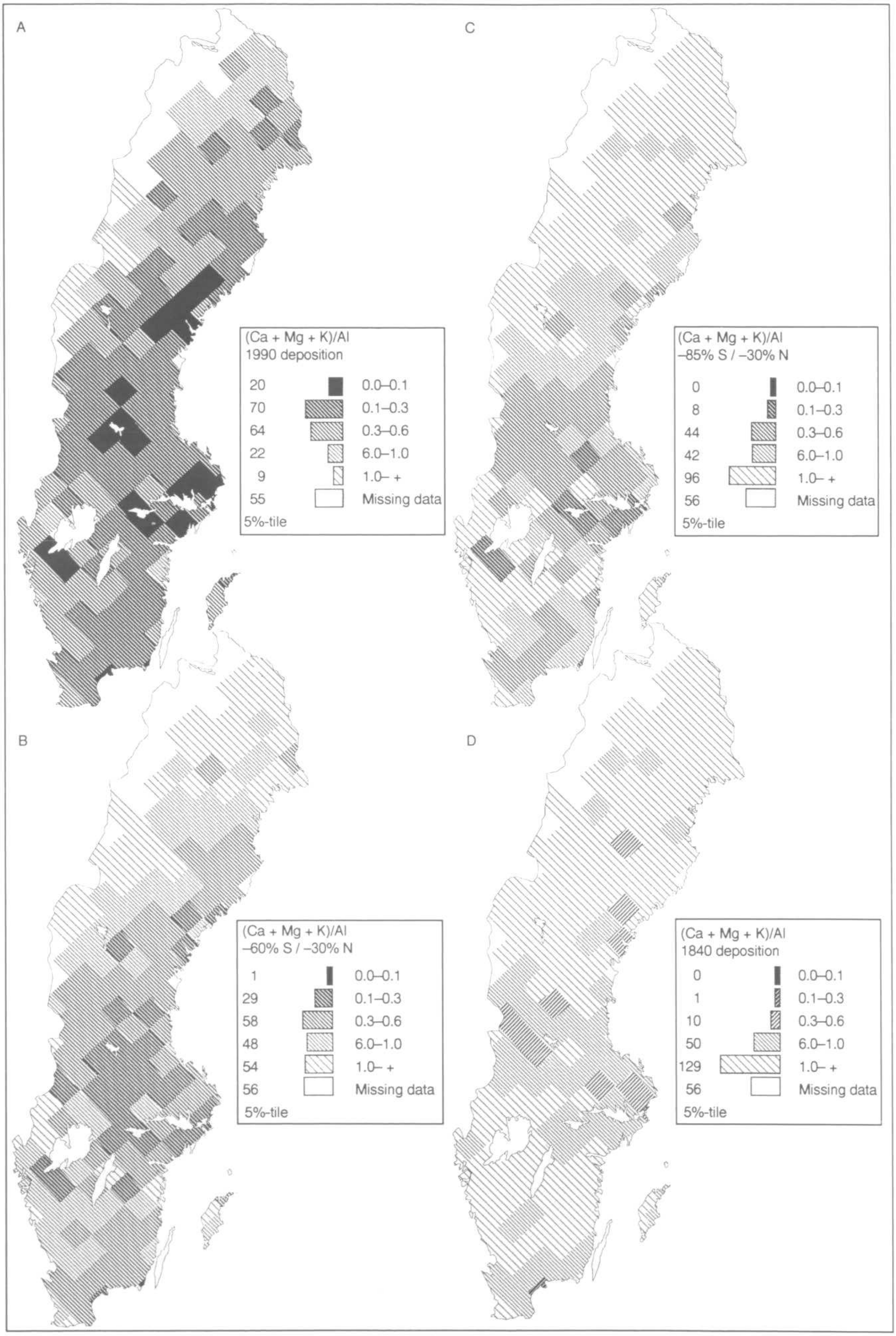

Figure 7. Steady-state (Ca+Mg+K)/Al in the B layer of Swedish soils under A, 1990 deposition level; B, -60% S and -30% N; C, -85% S and -30% N; and D, under 1840 deposition level

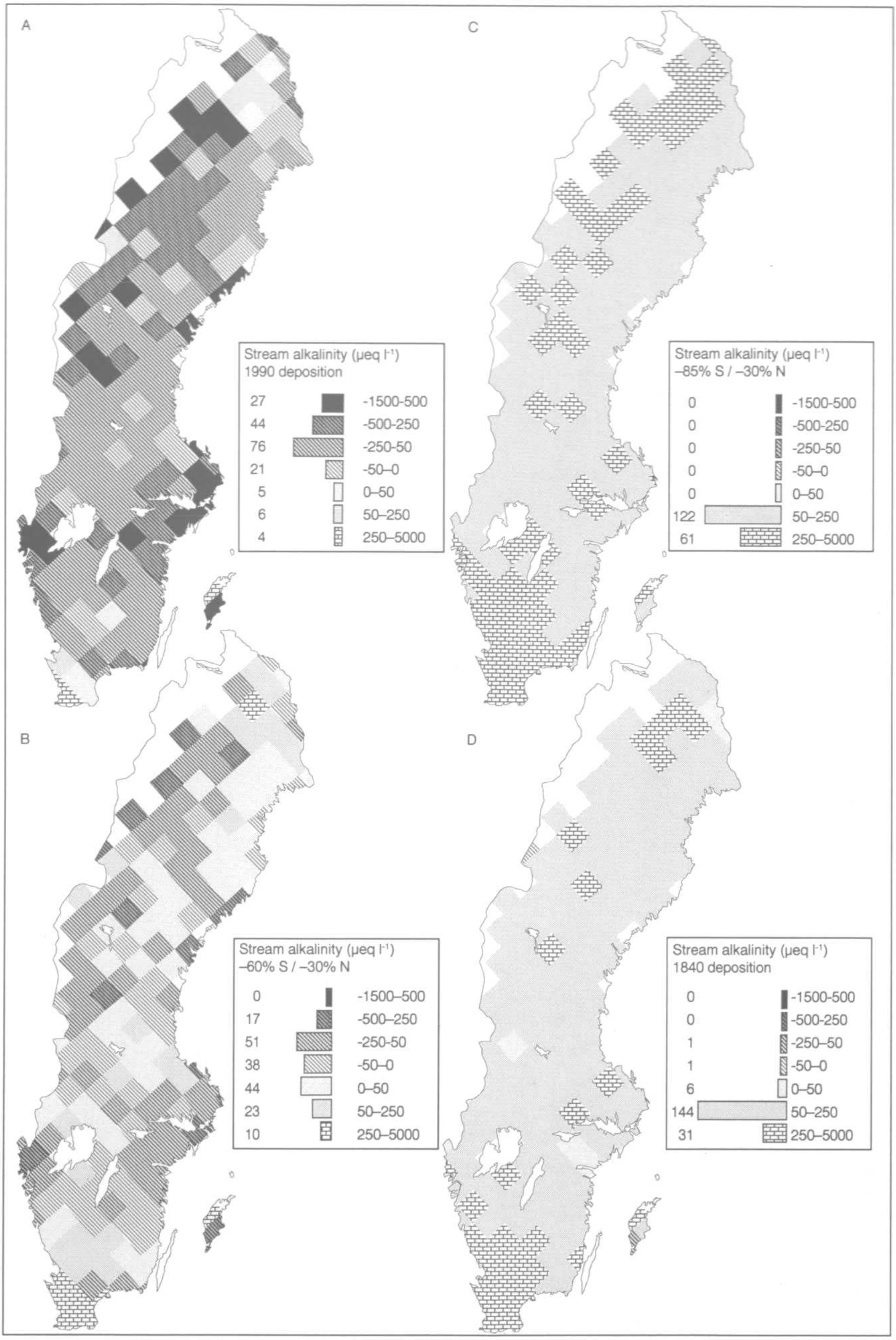

Figure 8. Steady-state alkalinity in Swedish first-order streams and lakes under A, 1990 deposition level; B, -60% S and -30% N; C, -85% S and -30% N; and D, under 1840 deposition level

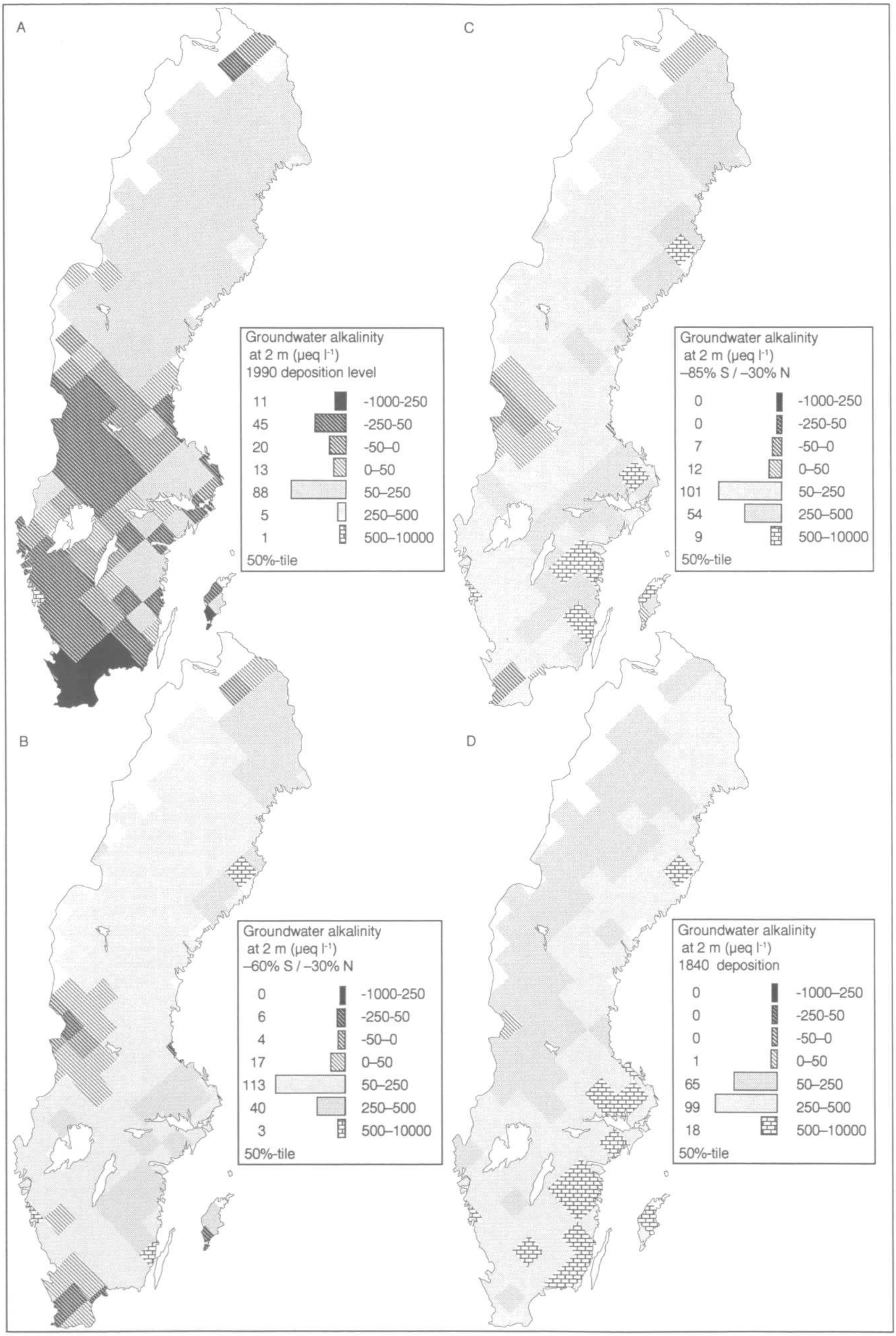

Figure 9. Steady-state alkalinity in groundwater at 2 m depth in moraine soils under A, 1990 deposition level; B, -60% S and -30% N; C, -85% S and -30% N; and D, under 1840 deposition level

Figure 10. Growth reductions in Swedish forests due to the effect of the BC/Al ratio under A, present deposition level; B, -30% S; C, -60% S; and D, -60% S and -30% N. The maps show forest growth as a proportion of the pre-acidification value

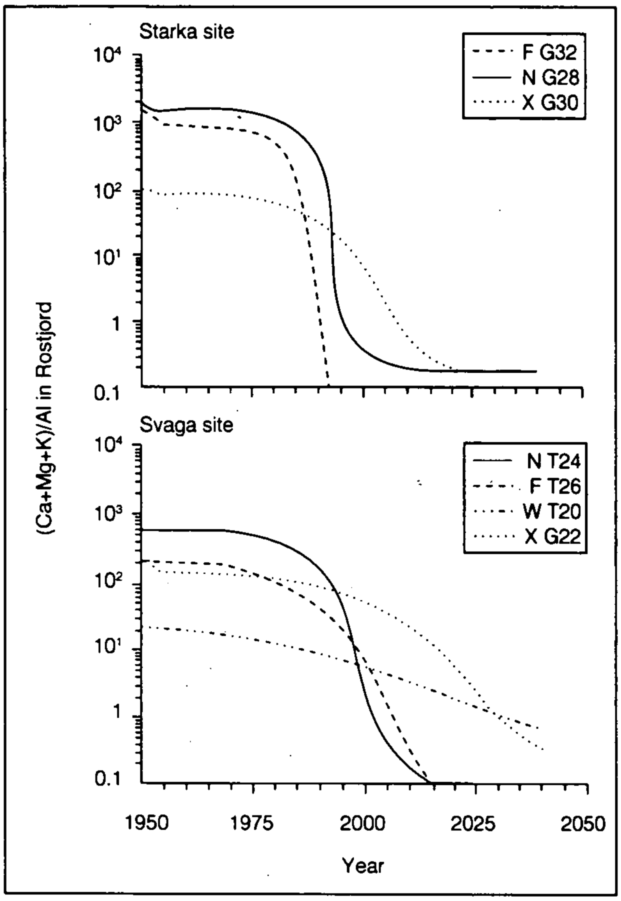

Figure 11. Development of the BC/Al ratio over time in high-yield sites (Starka) and low-yield sites (Svaga), as calculated by the SAFE soil chemistry model. The calculations indicate that we are progressing towards growth collapse in Swedish forest, if the laboratory relationship between the BC/Al ratio and growth holds. Calculations for different Swedish sites indicate that forest collapse on a larger scale will occur during 1990-95 in southern Sweden

being cropped every year. This figure represents a net national profit of approximately 18,000-35,000 million Swedish kronor (SEK) (£3,500 million sterling yr^{-1}). The standing forest is approximately 2500 million m^3. (Net national profit varied in the range 250-500 SEK m^{-3} during 1981-89, 1 ECU=7 SEK, 1 US$=6 SEK.)

We have used the growth impacts listed above to estimate the revenue streams at risk in the Nordic countries. The results are listed in Table 5 for Sweden under different deposition scenarios. Nutrient limitation is estimated to reduce present forest growth by approximately 5 million m^3 yr^{-1}.

The cost impact is increased by the fact that the most productive forest is located in southern Swedish areas calculated to attain very low (Ca+Mg)/Al ratios. The utilisation of increased forest growth and harvest on high-weathering soils may increase the annual production by 1-2 million m^3, even under the 1990 deposition level scenario, which, however, is still rather marginal in comparison with the incurred growth reductions.

DISCUSSION

General

Including uncertainties in the economic calculations will change the absolute numbers of the cost impact, but not the order of magnitude. The best case still implies a net production loss of 15%, compared to the likely case 20% and the worst case 30% of current commercially exploitable forest growth. In a best-case scenario, the annual costs will run into several thousand million SEK, dealing a heavy blow to the Swedish national economy. The cost impact is also increased by the fact that forest production is at present greatest in southern Sweden, the area that will also experience the highest deposition loads and the lowest soil (Ca+Mg)/Al ratios. The forest harvest will be affected only in the long term. The present forest stands will be available for harvesting, but the major economic effect will show up in the next rotation period, when the next harvest is to be taken. If the weathering rate is not based on geochemical properties or mineralogy and texture, all accuracy and confidence in the derived critical load values are lost.

In due time after the soils have been thoroughly acidified, most of Sweden's streams and lakes may become acidified, unless there are drastic emission reductions on the European continent and in the British Isles.

The acidification question can be phrased as: who has the right to utilise the weathering rate of Swedish soils? Does it belong to the individual landowner in Sweden, or to the Swedish nation (under the jurisdiction of the Swedish parliament), or the industries of Europe that have actually taken it for neutralising the acid combustion waste they are at present dumping in Sweden? We think that there is no national sovereignty to defend the trans-boundary unsolicited dumping of industrial waste in neighbouring countries.

Sensitivity analysis

A sensitivity analysis of the PROFILE model shows that the model can calculate the weathering rate for a given soil within a maximal error range of ±40%, the ANC leaching within ±25%, and the BC/Al ratio within ±30% of the best estimate. The analysis was performed by Monte Carlo simulation, with an assumed error range in each parameter of ±10-100%, depending on parameter type. The error ranges are for the 10th and 90th percentiles, ie 80% of the values fall in this range. The most important factors for weathering rate calculations are soil physical parameters, ie soil density, soil specific area and soil moisture content. The second most important parameters are soil solution equilibrium constants, especially soil P_{CO2}, while climatic parameters, deposition and uptake parameters, and soil stratification are of less importance. For calculating the BC/Al ratio, deposition and uptake parameters are important, as well as soil physical constants and soil solution equilibrium constants. For the calculation of ANC leaching, the most important factors are percolation, deposition and uptake, and mineralogy.

The uncertainties in the model calculations will naturally induce some uncertainty in the calculation of critical loads. This uncertainty may alter the exact numbers presented here, but not the magnitude of the numbers. When calculating critical loads and taking all parameter uncertainties into consideration according to the sensitivity analysis, the result varies by a maximum of ±40%. For an examination of the critical load calculation sensitivity, 20 field data points were used within a 50 km x 50 km area in southern Sweden. The cumulative frequency distributions of the calculated critical load for each of the 20 points are shown in Figure 12. The Figure shows that variations between different sites in the same geographical area are more important than variations caused by uncertainties in the model calculations. Furthermore, when comparing the 10/90 and 25/75 percentiles to the medians of the critical load distributions for the 20 points, the uncertainty interval for different predictions can be studied.

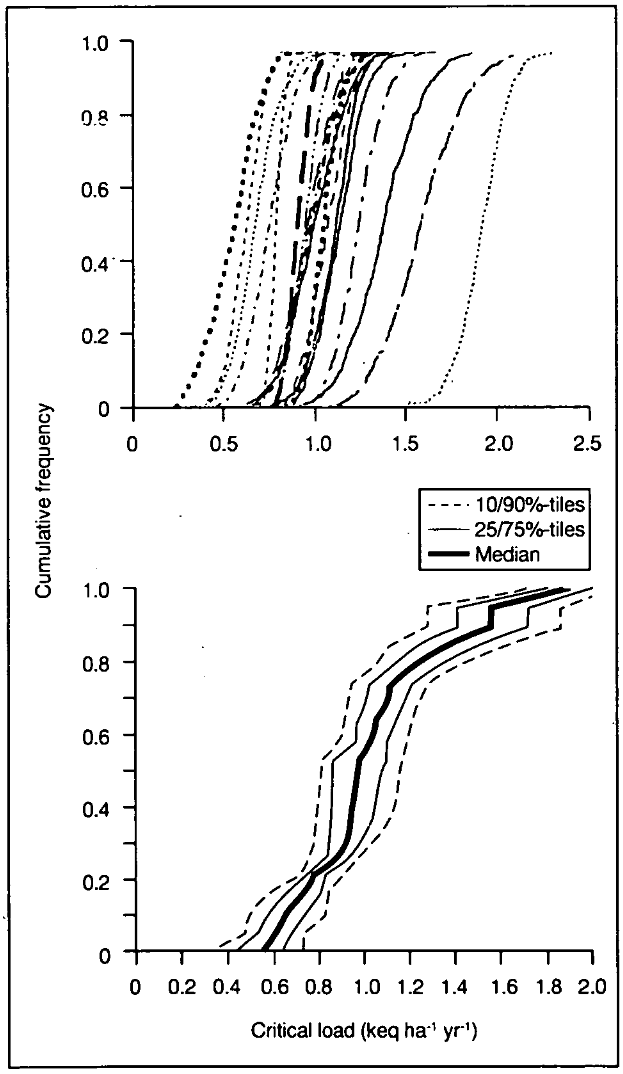

Figure 12. (Top) Cumulative frequency distributions of critical load values at each of the 20 sites within the NILU grid 5861. Variation between sites is larger than the uncertainty for the individual site (Bottom) Distribution of critical load values within NILU grid 5861, showing the cumulative distribution of the median values, the upper and lower 25/75 percentile and the 10/90 percentile

CONCLUSIONS

We can conclude that a minimum deposition reduction over Sweden of 80% for sulphur and 30% for nitrogen is required in order to protect 75% of the groundwater resources, 95% of the forest resource, and 90% of the surface water ecosystems.

Sweden is receiving acid deposition in excess of the critical load for more than 80% of its lake, stream and forest resources. Calculations indicate that the present level of acidic deposition is starting to cause significant economic damage, and will evolve to become a factor that could significantly affect the national economy to the degree where the standard of living is affected. Calculations show that considerably more than a 60% reduction in S deposition and 30% reduction in N deposition are required in order to leave only 11 000 km^2 of forest unprotected and avoid significant economic damage to Swedish forest landowners and the Swedish forest industry.

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Testing the PROFILE model on long-term data

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INTRODUCTION

The PROFILE model for calculating field weathering rates is being used to calculate and map critical loads for acid deposition (Sverdrup 1990; Sverdrup & Warfvinge 1988; Sverdrup, De Vries & Henriksen 1990). PROFILE predicts the steady-state conditions resulting from a constant atmospheric input to a soil. Although the model does not predict the time taken to reach the steady state, this is likely to be decades if not hundreds of years. Testing the model, therefore, requires long-term data. Whilst PROFILE has been tested against some short-term data (Sverdrup *et al.* 1990), few long-term data sets exist that enable a true test; one such set is that from the Classical Experiments at Rothamsted Experimental Station, the main site of the Agricultural and Food Research Council's Institute of Arable Crops Research.

The Classical Experiments were begun by the founder of Rothamsted, J B Lawes, in 1843. Those of particular interest to research into acid deposition and soil acidification are the Park Grass, Broadbalk Wilderness and Geescroft Wilderness Experiments. Long-term data regarding acid deposition to the sites were described by Goulding *et al.* (1986), and the gradual acidification of the soils at the sites was described by Johnston, Goulding and Poulton (1986). The site best suited to testing PROFILE is Geescroft Wilderness. Geescroft field was growing arable crops until the 1880s but, unlike most of the Rothamsted estate, it had not been limed to overcome the (then undefined) acidity that had developed from continued arable cropping. In 1886 an area of 1.2 ha at the eastern end of the field was fenced off and left. Over the ensuing 105 years when the site has remained unmanaged, scrubland and eventually deciduous woodland have developed. The site is now a mature oak (*Quercus*)-dominated woodland. Much of the history of its development, details of tree species, and some soil properties were described by Jenkinson (1971) and Goulding and Johnston (1989).

Throughout the development of the woodland, soil samples were taken periodically, as at all the other Classical Experiments. These samples represent an invaluable resource for assessing real long-term changes in ecosystems and relating them to climate, especially acid deposition. Johnston *et al.* (1986) attempted this for several Rothamsted sites, including Geescroft Wilderness. During the last three years, a new and much more thorough investigation of Park Grass and the Wildernesses has begun. Both historical and current soil samples (to 92 cm) have been analysed for pH, carbon and nitrogen contents, and water-extractable, exchangeable and total cation, anion and toxic metal contents. These measurements are being related to the developing acidity and to the composition of trees and plants growing on the sites. Some typical results are given in Figure 1; and show the greatly increased uptake of toxic metals in oak saplings growing on acid Geescroft Wilderness compared with neutral Broadbalk Wilderness.

Precipitation was first collected and analysed at Rothamsted in 1853. These long-term, but unfortunately not continuous, data allow considerable insight into changes in acid deposition, as shown by Brimblecombe and Pitman (1980) and Goulding *et al.*

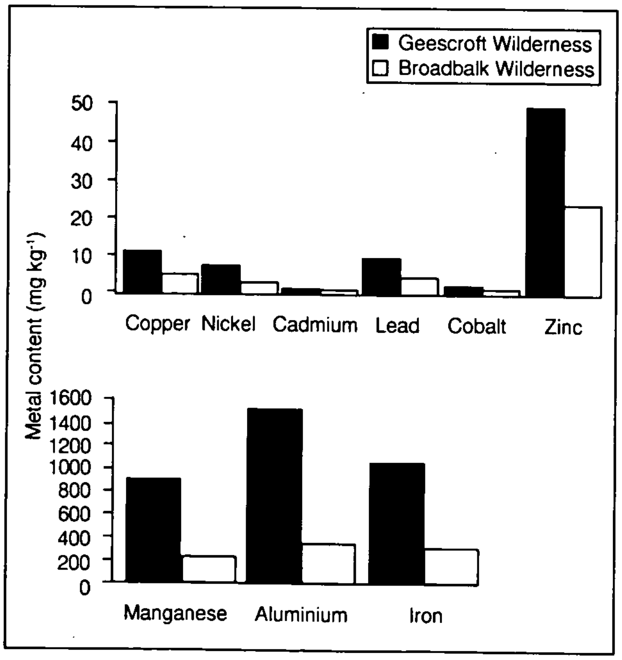

Figure 1. The metal contents (mg kg^{-1}) of oak saplings growing on Geescroft Wilderness (acid) and Broadbalk Wilderness (neutral) in 1991

(1986). Whilst the soils at Rothamsted are not the thin, acid-prone soils on which acidity is a serious current problem, the analyses of current and historical soil samples and of acid deposition offer a unique test of models such as PROFILE. This paper reports a preliminary test using data from the Geescroft Wilderness Experiment.

THE PROFILE MODEL

The model has been described fully by Sverdrup *et al.* (1990). For each soil horizon, the model requires data on soil mineralogy, surface area, moisture content, bulk density, CO_2 pressure, leaching, nutrient uptake, dissolved organic carbon, log K gibbsite (used to define the relationship between pH and Al concentration), and pK values for H/Al and H/Ca exchange. Then, from further data on precipitation, leaching, atmospheric deposition, nitrification, nutrient uptake and soil temperature, it will calculate the steady-state conditions of each soil layer and any streamwater issuing from the soil; results include soil pH, Ca, Mg and K (ie base cation) content, Al, NO_3^- and NH_4^+ content, and weathering rates for each soil horizon. The model includes no timescales, and so predictions are of equilibrium conditions at some future date resulting from constant atmospheric inputs, soil conditions and plant growth. The dynamic form of PROFILE, SAFE (Soil Acidification in Forested Ecosystems), includes timescales and varying atmospheric inputs (Wright *et al.* 1991).

ACID DEPOSITION

Early Rothamsted data were for precipitation composition only. The gauge, built in 1853, was a large lead-lined funnel, of area 4.05 m^2 (exactly 0.001 acres) at ground level. Samples are now collected, and analysed, as part of the UK Secondary Precipitation Composition Network, in a standard 'Warren Spring Laboratory' pattern bulk collection gauge (Devenish 1986; Hall 1986) at 2 m above ground level, as well as from the old large gauge. Figure 2 shows the deposition of SO_4^{2-}-S, NO_3^--N and NH_4^+-N in precipitation since 1853.

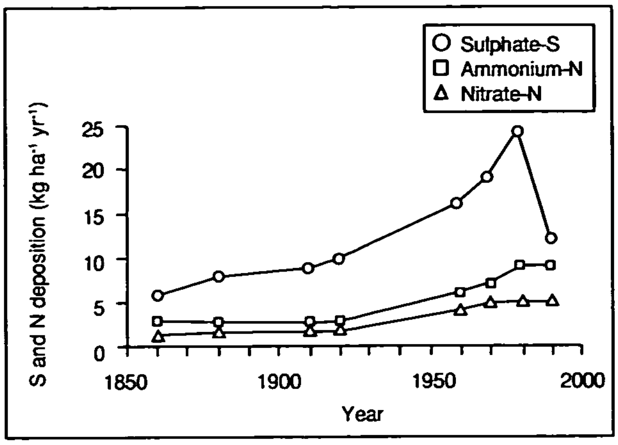

Figure 2. Bulk deposition (kg ha^{-1} yr^{-1}) of sulphate-sulphur and ammonium- and nitrate-nitrogen at Rothamsted Experimental Station from 1853 to the present

PROFILE requires data for the deposition of wet and dry *excess* (anthropogenic) sulphur, called SO_4 in the model, and wet and dry oxidised and reduced nitrogen, called NO_3^- and NH_4^+, respectively, in the model. These are now measured at Rothamsted, but no data exist for dry deposition of nitrogen until the 1970s or of sulphur until the 1960s. In order to calculate dry deposition before this time, we have assumed, in the absence of contradictory evidence, that the ratio of wet/dry S deposition at Rothamsted was the same then as it is now, ie 1. For the various forms of nitrogen, the pattern seems to be much more complex. Present measured ratios at Rothamsted are: wet/dry 'NO_3^-' (NO_2, HNO_3, HONO, etc), 1:4; wet/dry 'NH_4^+' (NH_3 and NH), 1:1.2. The numbers of road vehicles (an important source of NO_2) have increased greatly in recent years and, of course, none existed at the beginning of the Wilderness Experiments. With few data on which to base our estimates, we have assumed that the ratio of wet/dry NO_3^- has increased from 1:1 in 1880 to 1:4 today. Whilst NH_3 emissions have increased recently (Ap Simon, Kruse & Bell 1987), there are few data to show how the ratio of wet/dry NH_3 deposition might have changed over the century. We have, therefore, assumed a constant ratio of wet/dry NH_3 of 1:1.2. The calculated depositions of excess wet and dry SO_4, and of wet and dry NO_3^- and NH_4 are given in Table 1.

Table 1. Acidic deposition to Geescroft Wilderness

	Deposition (keq ha^{-1} yr^{-1})								
		Excess SO_4^{2-}			NO_3^-				NH_4^+
Year	Wet	Dry	Total	Wet	Dry	Total	Wet	Dry	Total
1880	0.38	0.38	0.76	0.11	0.11	0.22	0.18	0.22	0.40
1920	0.55	0.55	1.10	0.14	0.28	0.42	0.27	0.33	0.60
1980	1.07	1.07	2.14	0.45	1.43	1.88	0.64	0.77	1.41
1990	0.53	0.53	1.06	0.45	1.43	1.88	0.64	0.77	1.41

NB Wet deposition measured (as bulk deposition) for NO_3^- and NH_4^+ Wet deposition of excess SO_4^{2-} calculated from bulk deposition of SO_4^{2-} and Na content. Dry deposition of NO_3^- and NH_4^+ measured in 1990, and of SO_4^{2-} in 1980 and 1990; calculated in other years from the ratio of wet/dry in 1990

SOIL ANALYSES

Soil samples were collected from Geescroft Wilderness in 1883, 1904, 1965 and 1991. Collection and preparation were described by Jenkinson (1971). For the purpose of testing PROFILE, some data are required to run the model and others to test its output.

Data required to run PROFILE

PROFILE normally accepts the usual soil horizons: L, O, A_h, etc. The first samples collected from Geescroft, in 1883, were separated into equal 9" (23 cm) depths: 0-23 cm, 24-46 cm, 47-69 cm, 70-92 cm. Subsequent samples have been collected in the same way to ensure continuity, and so the model is fitted to these layers. For this first, preliminary test, data for bulk density, moisture content, surface area and mineralogy were taken from previous analyses of the Batcombe Series soil (Jenkinson 1971; Weir, Catt & Ormerod 1969). The current 30-year average for precipitation was used, and leaching ('runoff rate' in the model) was calculated as the average amount leached from block lysimeters (undisturbed blocks of soil isolated, surrounded by brickwork, and underdrained) of the Batcombe Series soil. Plant uptake was obtained from Jenkinson's (1971) calculations, and analyses of samples were taken in 1991. Nitrification rate was assumed to be the maximum rate of the four suggested by Sverdrup *et al.* (1990) in their model description (0.04, 0.01, 0.005 and 0 kmol m^{-3} yr^{-1}) because no measurements were available. Mean annual soil temperature used, 8.5°C, is the current 30-year average. Values of pK H/Al and H/Ca were calculated, using the Gapon equation, from exchangeable base cation and Al analyses (see below). The CO_2 pressure, dissolved organic carbon (DOC) and log K gibbsite were not known or easily measurable, so appropriate values were selected from the PROFILE manual: CO_2 pressure 100, 20, 20, 20 x atm; DOC 15, 10, 5, 0 mg l^{-1}; log K gibbsite 8.5, 9.5, 9.5, 9.5 for 0-23, 24-46, 47-69 and 70-92 cm layers, respectively.

Data required to test PROFILE

The data required are, for each layer, pH, base cation and Al concentration and saturation, and NO_3^- and NH_4^+ concentration. Weathering rates and other non-measurable parameters are also calculated by the model, but the veracity of these was not assessed. The soil chemistry data were obtained from 1:1 soil/water extracts of <2 mm air-dried soil. Base cation and Al concentrations were measured in M ammonium acetate and KCl extracts, respectively, by inductively coupled plasma emission spectrometry, NO_3^- and NH_4^+ by ion chromatography, and pH using an Orion Ionalyzer EA 920 meter and a Ross Sureflow combination electrode. Soil pH changes are shown in Figure 3.

In the soil samplings of 1883, 1904 and 1965, individual samples were bulked; no standard errors showing soil variability are, therefore, available. Samples taken in 1991 from four replicate holes were analysed separately to show current variability; error bars are shown in Figure 3. It is likely that present heterogeneity in the woodland soil is much greater than that in 1883 when the soil was still growing arable crops, or that in 1904. The error bars, therefore, probably represent the maximum error arising from soil variability. Subsampling and analytical errors were assessed from four replicate subsamples of soil. These errors were very small, generally of the same order as the width of the lines in Figure 3.

TESTING THE MODEL

The model predicts steady-state conditions resulting from constant atmospheric inputs. No idea of the time needed to reach steady state is given. Figures 2 and 3 show that atmospheric inputs have not been constant and (therefore) the soil has not reached a steady state, except perhaps for the surface (0-23 cm) layer. A true test of PROFILE is not, therefore, possible. The most sensible test was taken to be a comparison of model predictions of the effect of 1880 and 1920 atmospheric inputs on '1883' soil with soil analyses made in 1965 and 1991. The effects of 1980 and 1990 atmospheric inputs were also modelled because these inputs are much more acidic than those of 1880 or 1920, but there was a marked decrease in sulphur deposition between 1980 and 1990 (Figure 2; McGrath & Goulding 1990).

Soil pH

Model predictions of soil pH in Table 2 are compared with the measured values in Figure 3. The predicted values are sensible, and those resulting from 1880 and 1920 atmospheric inputs are in reasonable agreement with the measured values for 1965 and 1991, respectively, particularly for the 0-23 cm horizon which had reached an equilibrium pH of 4.2-4.3 by the time the soil was sampled in 1965, and has remained at this value for 25 years. The model predicts that this surface soil will remain within the Al buffer system and not change to the Fe/Al buffer system (pH 2.8-3.8), even under 1980 atmospheric inputs in which potential acidity is almost 5 kmol ha^{-1}

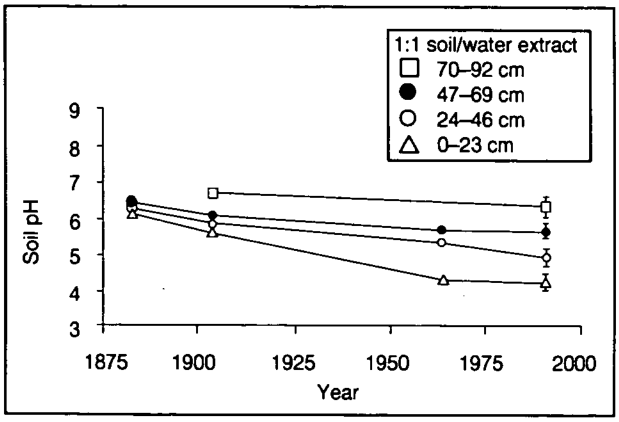

Figure 3. The pH of soil samples taken from Geescroft Wilderness in 1883, 1904, 1965 and 1991

yr^{-1}. It is interesting to compare this prediction with that for the top 3 cm of mineral soil receiving similar acid inputs. PROFILE predicts a steady-state pH of 3.6, compared with the measured value in 1991 of 3.8.

Table 2. PROFILE predictions of steady-state soil pH values resulting from the atmospheric inputs of 1880, 1920, 1980 and 1990 falling on '1883' soil

Horizon (cm)	Soil pH in a 1:1 soil/water extract for the atmospheric inputs in 1880	1920	1980	1990
0-23	4.5	4.3	4.0	4.1
24-46	5.9	4.7	4.3	4.4
47-69	6.6	6.2	4.3	4.4
70-92	6.8	6.6	4.4	4.6

Middle horizons in 1965 were more acid than predicted by PROFILE from 1880 atmospheric inputs. In 1991 the 47-69 cm horizon was still more acid than predicted from the 1920 atmospheric inputs; for other horizons, predicted and measured values agreed quite well.

Generally, the measured pH values are decreasing towards the predicted values arising from the acid inputs of the present day. A simple, linear extrapolation suggests that the 24-46 cm horizon will reach its predicted pH (*ca* 4.4) in the year 2050, the 47-69 cm horizon its predicted pH (*ca* 4.4) in 2200, and the 70-92 cm horizon its predicted pH (*ca* 4.6 at 1990 inputs) in about 2300. These very acid pH values are unlikely to support deciduous woodland (see Figure 1). Likely climax vegetation and the need to reduce inputs are discussed below.

Aluminium, base cation and nitrogen content

The predicted Al, Ca + Mg + K, NO_3^- and NH_4^+ contents of the soils at steady state resulting from 1880 and 1920 inputs are compared with measured values for 1965 and 1991 in Tables 3 and 4, respectively. The model predictions again appear to be sensible, but agreement with measured values is poorer than for pH.

Table 3. Predicted effects of 1880 atmospheric inputs on 1883 soil, as expressed by the steady-state concentrations (μeq l^{-1}) of Al, base cations, NO_3^- and NH_4^+, compared with 1965 soil data

Parameter		Soil horizon (cm) 0-23	24-46	47-69	70-92
Al	Model	67.9	6.9	1.3	0.8
	Measured	397	9.0	4.4	1.9
Ca + Mg + K	Model	180	470	948	1358
	Measured	2012	1665	1866	1750
NO_3^-	Model	54.5	27.8	7.9	7.9
	Measured	606	136	294	ND
NH_4^+	Model	1.1	0	0	0
	Measured	344	11	0	ND

ND = Not determined

Table 4. Predicted effects of 1920 atmospheric inputs on 1883 soil, as expressed by the steady-state concentrations (μeq l^{-1}) of Al, base cations, NO_3^- and NH_4^+, compared with 1991 soil data

Parameter		Soil horizon (cm) 0-23	24-46	47-69	70-92
Al	Model	67.9	6.9	1.3	0.8
	Measured	397	9.0	4.4	1.9
Ca + Mg + K	Model	180	470	948	1358
	Measured	2012	1665	1866	1750
NO_3^-	Model	54.5	27.8	7.9	7.9
	Measured	606	136	294	ND
NH_4^+	Model	1.1	0	0	0
	Measured	344	11	0	ND

ND = Not determined

Aluminium

Predicted Al concentrations resulting from 1880 atmospheric inputs agreed quite well with measured values for the 1965 sampling for all except the surface horizon, where there was six times more Al measured than modelled. This result is surprising as the modelled and measured pH values for the surface layer agreed well. For the 1920 inputs, predicted Al concentrations agreed well with 1991 measured values, except for the 24-46 cm horizon; here, the modelled value was nearly 100 times the measured, whereas measured and modelled pH agreed well.

Base cations

The model predicted much smaller amounts of base cations than those measured, except in the deepest horizon. The most likely explanation is that Ca, Mg and K are cycling through the upper horizon, tree roots, leaves and litterfall, preventing leaching and depletion, a process not taken into account by the model.

Nitrogen

Generally, the model predicted the presence of much less N than was measured; in some cases, the differences were very large - two orders of magnitude in the case of NH_4^+ in the surface soil in 1964. Again, nutrient cycling may be causing this difference.

Sensitivity analysis

The sensitivity of the model to its various inputs was tested. Generally, changing parameters had the expected effects both in terms of direction and magnitude. For example, warmer soils were predicted to weather faster and therefore to be less acid, while wetter weather and more leaching result in a more acid soil; increasing surface area increased weathering rate and decreased acidification. Acidification is predicted to increase with increasing base cation uptake and to decrease with increasing N uptake. The most significant effects arose from atmospheric deposition, nutrient uptake and log K gibbsite. Increasing deposition of S or N increased acidity, and increasing deposition of NO_3^- increased nitrate leaching: atmospheric N is proving to be a very important source of the nitrate leached in all ecosystems (Goulding 1990; Goulding & Poulton 1992).

Log K gibbsite had a very significant effect. Increasing it decreased acidification markedly. The model would only give sensible answers when using the log K gibbsite values of between 6.5 and 9.5 suggested in the manual (Sverdrup *et al.* 1990). Calculated values from measured soil data in any of the years sampled did not give a sensible result, but predicted no acidification at all. It seems that the authors of the model now accept that log K gibbsite is not a good way to model the relationship between pH and Al concentration, and are modifying this part of the model (H Sverdrup, pers. comm.).

Model parameters which do not always produce the results expected are pK values and temperature. Changing values of pK seemed to have no effect on soil chemistry, and warmer soils did not nitrify faster and produce more NO_3^-. Presumably, nitrification rate is defined solely by the value given to the model (0.04 kmol m^{-3} yr^{-1} in our case).

CONCLUSIONS

The PROFILE model gives sensible predictions of soil properties, many of which compare quite well with measured values from the long-term (100 years) Geescroft Wilderness Experiment. PROFILE has a user-friendly menu-driven format in both Apple Macintosh and PC versions, and it permits the user to assess rapidly the effects on soil chemistry of various soil parameters and atmospheric inputs; it, therefore, permits easy estimations of the effects of acid deposition and of critical loads. It also enables the likely effects of climate change to be predicted.

On the other hand, PROFILE requires a lot of data to run it and to test it, including some detailed analyses of soil parameters, which may not be easy to obtain. The model does not always respond to its input parameters in the way in which one might expect; the inclusion of a new submodel to predict pH/Al relationships is an important step forward.

Predictions of Al and base cation concentrations need to be improved if the model is to be truly valuable. The most common problems arising from soil acidity are the toxicity of Al to roots and too large Al/Ca ratios, especially in streamwaters that issue from acid soils. Some work is needed to examine whether nutrient and pollutant cycling in surface soil layers is the only or prime cause of the poor predictions of Al, base cation and nitrogen content.

Assuming that the model predictions are reliable, its application to Geescroft Wilderness gives some alarming results. If present atmospheric inputs are not reduced, then the soil under the woodland will eventually become very acid, pH 4.1 at the surface and pH 4.6 at about 1 m. Such an acid soil is unlikely to support healthy deciduous woodland, and more likely to change gradually to acid heathland or grassland. However, the modelling exercise also offers some hope for the future. The reduction of S deposition between 1980 and 1990 is predicted to result in significantly less acidification at depth in the soil, despite the continuing large N deposition.

To maintain the soil at present pH values or allow some recovery to less acid conditions, especially at the surface, requires deposition to be restricted by returning to pre-industrial emissions of <5 kg ha^{-1} yr^{-1} each of S and N. A more realistic aim might be to limit emissions to those of the early 19th century and accept some acidification of the top 50 cm of soils, whilst protecting subsoils. Surface soil acidity could then be corrected relatively easily with lime, and the very difficult problem of reducing acidity in subsoils avoided.

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Critical loads mapping at the UK Critical Loads Mapping Centre – data requirements and presentation

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INTRODUCTION

The UK Critical Loads Mapping Centre (known in European terms as the National Focal Centre) at ITE Monks Wood is responsible for producing critical and target load maps for Great Britain, and maps showing the exceedance of critical load by acid deposition. Critical and target load maps for soils have already been forwarded to the UN-ECE Co-ordination Centre for Effects (CCE) at Bilthoven in The Netherlands, along with data and maps at the European scale, based on the European Monitoring and Evaluation Programme (EMEP) grid.

In generating critical load maps, the resolution, together with the statistical method used for calculating the critical load, has been taken into consideration. The choice of these factors can influence the resulting maps and may affect subsequent interpretation and use of the data. For this paper, the Laser-Scan Horizon geographical information system (GIS) has been used to generate a series of maps to demonstrate these effects.

DATA FOR MAPPING

The various subgroups of the Critical Loads Advisory Group (CLAG) of the UK Department of the Environment (DoE) provide critical loads data to the Mapping Centre in a variety of formats from available data sources.

The soils subgroup derived critical loads for the dominant soil type for each 1 km square of Great Britain, based on the 1:250 000 maps and data bases of the Soil Survey and Land Research Centre (SSLRC) and the Macaulay Land Use Research Institute (MLURI). The Skokloster classification (Nilsson & Grennfelt 1988) is aimed at forest soils but, at present, all mineral soils are treated in a similar way. For peat areas, the critical load is defined in terms of an acceptable pH shift. To allow for agricultural liming in some areas, a land use modification was applied to the soil critical loads data at the Mapping Centre, using the ITE land use classification data for 1 km squares; the critical load was decreased by one class (to become less sensitive) for each square where either arable or improved grassland was the dominant land use.

The freshwaters subgroup of CLAG is carrying out a sampling programme of lakes throughout Great Britain. One site in each 10 km square is sampled, by selecting a lake (or first-order stream where no lake exists) in the most (geologically) sensitive part of each square. The critical loads data are calculated from chemical analysis of water samples for these sampling points, and the results are then forwarded to the Mapping Centre.

Incoming data to the Mapping Centre are converted to standard ASCII format and loaded on to an ORACLE data base where information can easily be manipulated, eg to convert grid references from letters and numbers to values in kilometres. The data are then passed through various formatting programs to generate vector or raster maps, as appropriate, on the GIS.

Revisions to both data and maps are made following discussion with the appropriate CLAG subgroups. At intervals, agreed versions of data and/or maps are made available to CLAG, the DoE or the UN-ECE CCE in Bilthoven, as required.

PRESENTATION OF MAPS

Traditionally, critical load maps have been presented as coloured grid squares, so the size of the square (the mapped unit) is an important consideration in presenting the data as maps. The maps of soil critical loads are initially presented as 1 km squares of the Ordnance Survey's National Grid for Great Britain, which are coloured according to their critical load value. Subsequently, critical load values for each 10 km and each 20 km square of the National Grid are calculated from the distribution of the 1 km data set and mapped. Figure 1 shows the different maps obtained from plotting the critical load which indicates the modal value (see below) of a typical data set at four different resolutions. The maps for soils are being used as the official national critical load maps and the data are forwarded to the UN-ECE. For freshwaters, the data are mapped with each 10 km square of the National Grid being coloured according to the critical load value of the water sample collected from it.

The maps provide a good indication of the area of

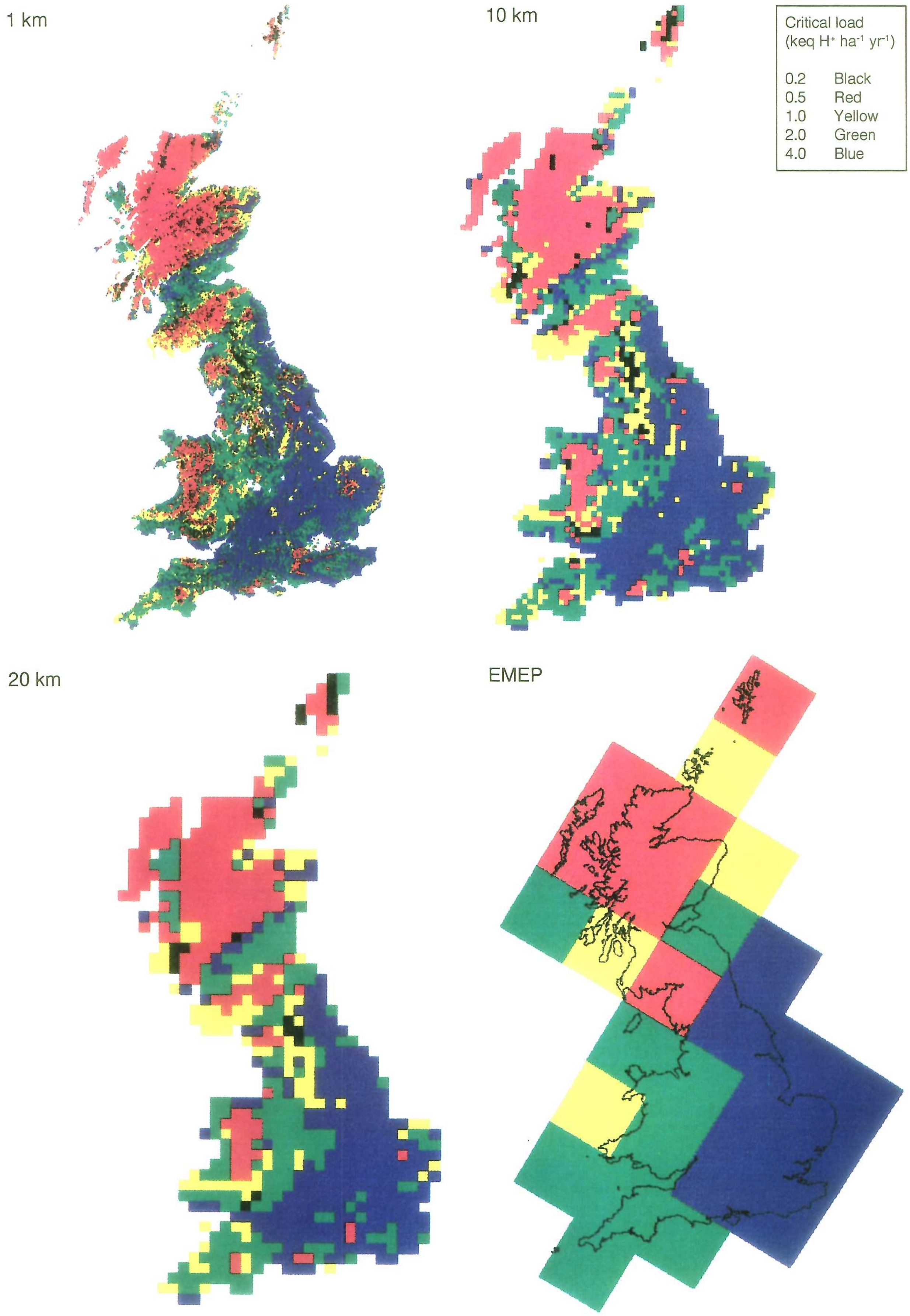

Figure 1. Effects of resolution: plotting the modal soil critical loads (March 1992 data) on 1 km, 10 km, 20 km and EMEP grids

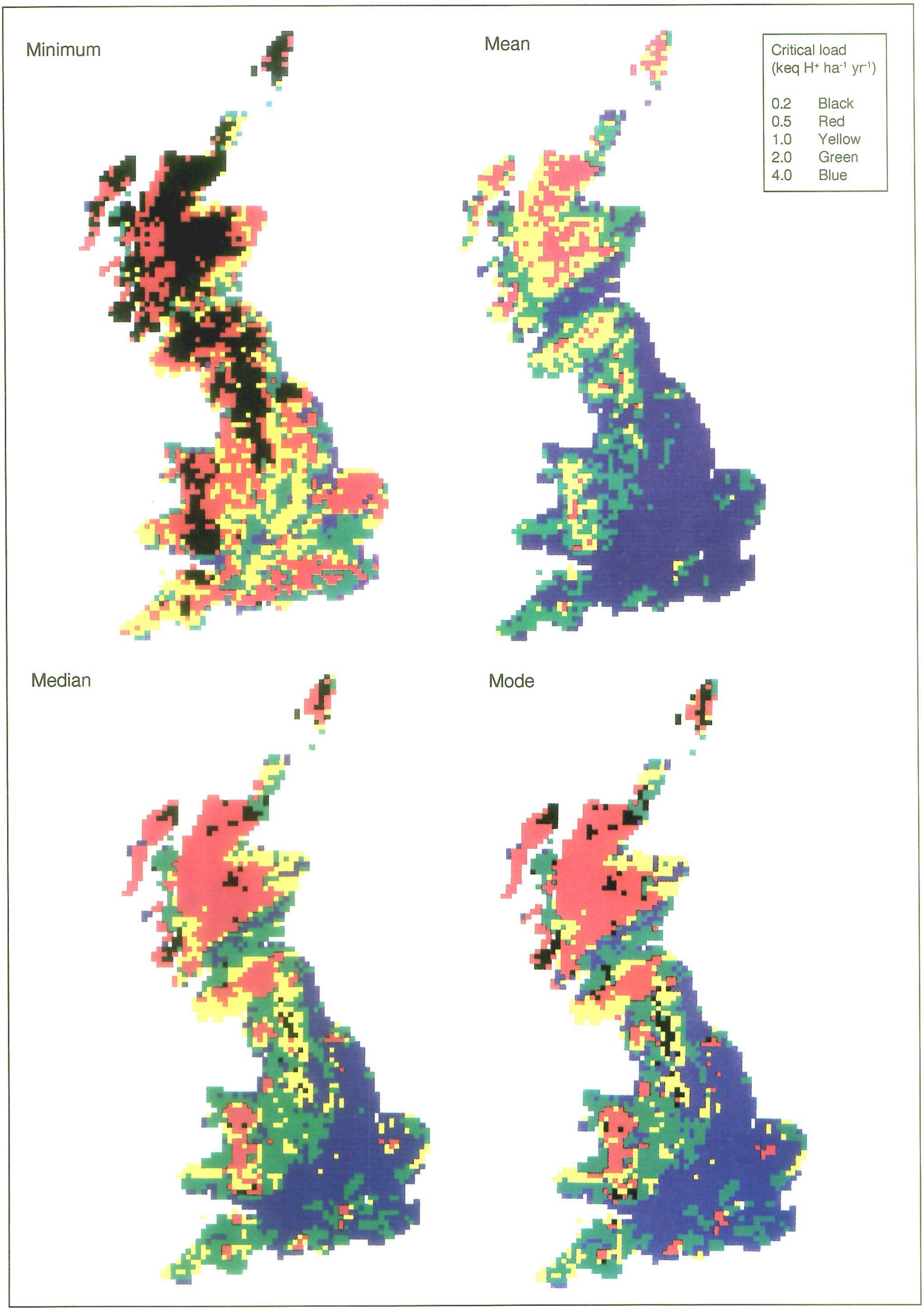

Figure 2. Effects of using different statistics to present soil critical loads data (January 1991) on a 10 km grid

Great Britain, so a coast outline is not usually included. However, a coastline or grid may be added as an aid to the location of grid squares.

Pollutant deposition data are generated by ITE and the Warren Spring Laboratory for 20 km grid squares. These are used to calculate areas where the deposition exceeds the critical load (called critical load exceedances). Such calculations may be made using the 1, 10 or 20 km critical load values, with the same deposition value assigned to each of the 1 or 10 km squares within the 20 km square. Maps of these exceedances are usually produced on a 10 km square grid. In addition, maps have been generated using modelled deposition values which show areas of the country where the critical load will still be exceeded in the year 2005, ie 'not protected' areas. These target load maps are usually presented on a 20 km square grid. Target loads for freshwaters are derived from the lowest critical load value of the four 10 km squares within a 20 km grid. This method is consistent with the CLAG freshwaters group sampling the most sensitive areas.

The statistic selected for mapping the aggregated data will determine the final appearance of the map; different statistics may give very different pictures. The usual maps of soil data (at 10 and 20 km) show a single statistic of the underlying data - the mode of the 1 km data set. However, the 1 km data may also be used to generate different statistics. Figure 2 shows the effect of plotting the minimum critical load, which gives a 'more sensitive' map; the mean critical load, apparently less sensitive; and the median and modal critical loads, which give similar results of intermediate sensitivity. The modal value has been chosen for most of the soil maps as this is consistent with the use of the dominant soil for defining the critical load of each 1 km square. Modal maps have also been found to give similar pictures at different resolutions because the area proportions of the different critical load classes are similar.

To provide information at the European scale to the UN-ECE, the co-ordinates of each 1 km square of the soil critical load map is transformed into the EMEP projection, thus providing information for the 150 km x 150 km squares of the EMEP grid covering the UK (Figure 1). A statistic has to be selected for mapping these data. Traditionally, within the UN-ECE, percentiles of the cumulative distribution of critical load values have been favoured when presenting data at the EMEP scale. UN-ECE maps have used the 1, 5 and 50 percentiles. Each percentile may be calculated from the distribution of critical load values for each EMEP square. Comparison of percentile maps with deposition values enables 'damage' to be assessed. Non-exceedance of a 5 percentile value indicates that at least 95% of the ecosystem area mapped is 'protected'. Low percentiles are usually preferred for European maps as they give comparable results for different countries which may map different receptors using different methods. Low percentiles focus upon the most sensitive elements of the grid square. For the UK, the 25 percentile has been chosen for EMEP-scale target loads data for 2005. The areal distribution of critical loads at the EMEP scale is similar to that for the modal map at 20 km.

When calculating the statistic to be mapped for any of the above maps, a decision has to be made whether to include areas of sea in coastal squares. In general, the Mapping Centre has excluded the sea when calculating critical loads for grid squares. While it makes little difference to the higher-resolution maps, eg 1, 10 and 20 km, it has a significant effect for data plotted on the EMEP grid. Single 150 km squares could be designated as highly sensitive when only a few kilometres of sensitive land are surrounded by sea in the square, while a similar area of high sensitivity on the mainland may have a lower calculated sensitivity for the same percentile. Therefore, for EMEP grids, areas of sea are included in the calculations.

THE GIS APPROACH TO MAPPING CRITICAL LOADS

Critical load maps may be generated using a variety of different computer graphics software programs; however, there are a number of advantages in using full GIS software.

By 'zooming-in' to the screen graphics, the high-resolution 1 km maps can be examined in detail, eg to match the English/Scottish border of the soil maps. In addition, using GIS, different mapped data sets can be overlaid on one another so they can be examined together. For example, the 'not protected' squares from target load maps, catchment areas or other areas of interest can be superimposed on to the various critical load maps.

The Horizon GIS works in conjunction with the ORACLE data base, enabling communication in both directions between the GIS and data base. It allows interrogation of the data base from features (eg 10 km squares) displayed on the screen. In this way, information can be extracted from the data base and displayed for any individual feature or user-specified selection of features, eg all sites with a pH value <4.5 and a critical load value <0.5 keq ha^{-1} yr^{-1}. In addition, there is the facility to classify features on the map according to the data held in the data base, and thereby to generate new maps. Comparisons of soil and freshwater critical load values have been mapped in this way.

CONCLUSIONS

In its role of co-ordinating the mapping of critical loads, the Mapping Centre at ITE Monks Wood has examined the effects of producing maps at different resolutions, and of using different statistics on the data. Discussions within CLAG attempt to ensure that the most appropriate maps and data are presented, both for the UK and Europe. Maintaining a data base of the high-resolution data provides the facility for examining ways of presenting different maps, eg by generating a range of percentile maps for the UN-ECE. The GIS allows flexibility in displaying maps of different resolution (either separately or together), 'zooming-in' for detailed examination of areas of interest on high-resolution maps, and direct access to information held on any area by the ORACLE link.

ACKNOWLEDGEMENTS

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Critical loads of nitrogen for soils

Current methods of calculating critical loads for nitrogen: application to the Excess Nitrogen Experiment with Norway spruce

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INTRODUCTION

The critical loads approach is now widely accepted as a tool for devising pollutant abatement strategies. It aims to maximise environmental benefits by targetting emission controls, so that deposition is reduced in sensitive areas rather than uniformly.

The critical loads concept was first applied to the acidifying effects of sulphur deposition. More recently, attempts have been made to quantify a critical load for nitrogen. This exercise has proved complex because of the numerous biological reactions of nitrogen in the environment and the need to consider both acidification and eutrophication effects. At the time of the Conference in Grange-over-Sands, the most advanced approach for defining a critical load for N for forests was that published in the Coordination Centre for Effects (CCE) mapping manual (Hettelingh, Downing & de Smet 1991). The results of applying this equation to data from the Excess Nitrogen Experiment on Norway spruce *(Picea abies)* were presented by the author at the Conference. Since the Conference, however, the CCE equation has been superseded. This paper describes the application of the new equation to the same data.

In April 1992, a UN-ECE workshop was held in Lokeberg, Sweden, to formulate critical loads specifically for N. The aim was to produce an equation which modelled the reactions of N deposition in the ecosystem and evaluated the thresholds at which adverse effects occurred. It is difficult to test whether this objective has been achieved, because (i) the necessary data are rarely available for a range of ecosystems, and (ii) damage symptoms in the field can seldom be attributed unequivocally to N deposition. Controlled experiments are the next best option to field data for testing critical load equations.

This paper discusses the results and implications of applying the Lokeberg critical load equation to data from the Excess Nitrogen Experiment with Norway spruce. This pot experiment investigated the effects of different chemical forms and loads of N deposition on soil acidification and the health of Norway spruce (Wilson 1990, 1991). Although unrepresentative of the field, it provides most of the data required for the critical load equation. The sensitivity of the critical load to the magnitude and chemical form of N deposition can be examined using these data. Most importantly, critical loads can be compared with the N load at which adverse effects were observed.

THE EXCESS NITROGEN EXPERIMENT WITH NORWAY SPRUCE

The Excess Nitrogen Experiment was set up in response to concerns that atmospheric N deposition was contributing to soil acidification and deteriorating forest health. Potted Norway spruce clones were treated with ammonium-N ($(NH_4)_2SO_4$) or nitrate-N ($HNO_3/NaNO_3$) in simulated rain for over 2.5 years. N was applied to either the foliage or the soil by an automated spray or drip system, respectively. The trees were grown in an acid brown earth soil of the Moretonhampstead series. This experiment investigated:

- the response of Norway spruce to different chemical forms of N;
- the relative importance of foliar uptake of N and soil-mediated processes;
- the N load at which adverse effects occurred.

Nitrogen was applied at 0, 30, 60 and 120 kg N ha^{-1} yr^{-1} above ambient rainfall, referred to as the 0, 30, 60 and 120 kg N treatments. Foliage was analysed bi-annually for total N, nutrient elements, nitrate reductase activity, and amino-acids. Every three weeks, soil leachate was collected and analysed for pH, cations and anions. The growth of the trees was measured at the end of each growing season. The nitrogen budgets calculated for this spruce 'ecosystem' have been used for determining critical loads. Figure 1 illustrates treatment application and leachate collection.

COMBINING EUTROPHICATION AND ACIDIFICATION EFFECTS: THE CCE MAPPING MANUAL 1991

The first critical load maps for N produced by the CCE (Hettelingh *et al.* 1991) aimed to reflect the sensitivity of forests to both nitrate leaching and soil

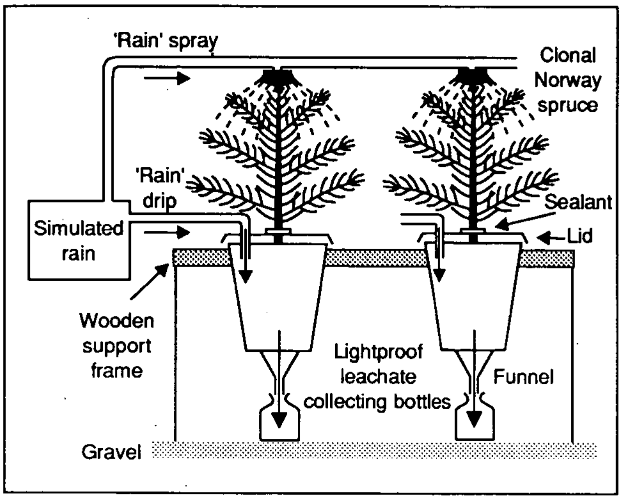

Figure 1. Treatment application and leachate collection in the Excess Nitrogen Experiment with Norway spruce

acidification. If N is immobilised by forests, the potential for nitrate leaching (and consequently acidification) is reduced. On this premise, the critical load was defined as the sum of N uptake by forests and the critical load of acidity for N (equation 1) (Hettelingh *et al.* 1991). The latter was calculated as 1 - S_f, where S_f (the 'sulphur fraction') is the contribution of sulphur to the critical load for acidity (equation 2).

$$CL_N = N_u + (1 - S_f) \, . \, CL_{acidity} \qquad (1)$$

$$CL_N = N_u + \left(1 - \frac{SO_x}{SO_x + NO_x + NH_x - N_u - N_{i,crit}}\right) . \, CL_{acidity} \qquad (2)$$

where N_u = N uptake by forests ('eutrophication term'); $N_{i,crit}$ = critical N immobilisation; and SO_x, NO_x and NH_x are present loads of S and N.

This approach attempts to protect the ecosystem from both the eutrophication and acidifying effects of excess N deposition. N uptake by managed forests is taken as the maximum allowable eutrophication limit. N removed in this way will not contribute to either acidification as a result of nitrification/base cation leaching, or the eutrophication of groundwater, and can be included in the critical load. However, the nitrogen uptake term (N_u) appears twice in the equation, with opposing effects on the critical load. In the sulphur fraction term, the critical $load_{(acidity)}$ apportioned to a given sulphur load will be greater in forests where N uptake is high. Consequently, a high rate of N uptake by forests gives a lower critical $load_{(acidity)}$ for N. In the eutrophication term, however, a higher rate of N uptake results in a larger critical load for N. There are difficulties with including both eutrophication and acidification effects in the same equation, because reactions which limit the potential of N to acidify (N uptake) also increase the risk of eutrophication of vegetation.

In equation 2, N uptake by biomass is treated as an infinite sink, ignoring the potentially adverse effects of high rates of N assimilation. On poor soils, the 'dilution effect' (Lundegardh 1945; Miller & Miller 1988) or competition between NH_4^+ and base cations at root uptake sites (eg Baule & Fricker 1970; Kirkby 1969) may result in nutrient deficiencies or unfavourable N/base cation ratios.

N uptake by biomass is viewed as limiting acidification - which it does insofar as it prevents nitrification of deposited ammonium, and the leaching of base cations by nitrate. However, the acidifying effects of NH_4^+-N uptake by vegetation and microbes are not addressed; 1 mole of H^+ is released for each mole of NH_4^+ assimilated. If N is deposited as nitrate, uptake by biomass will have an alkalinising effect; 1 mole of OH^- is extruded for each mole of NO_3^- assimilated (Reuss & Johnson 1986). Whether N uptake is acidifying or not will depend on the chemical form of N available, and hence the relative contributions of NH_4^+-N and NO_3^--N to total N deposition.

The acidifying effects of N deposition are quantified by partitioning the critical load for total acidity between S and N according to current deposition. The N critical load is dependent on S deposition, and will alter with changes in emissions. The total acidity concept is a useful one because an ecosystem's sensitivity to the acidifying effects of one pollutant will depend on the deposition of the other. However, an acceptable method of partitioning the critical load for acidity between S and N is still required.

SEPARATING EUTROPHICATION AND ACIDIFICATION EFFECTS: THE LOKEBERG WORKSHOP, 1992

The critical load equation for N formulated at the Lokeberg workshop overcomes many of the limitations of its predecessor. The most significant advance has been to calculate separate critical N loads for forests, based on acidification effects and eutrophication effects. This approach gives a critical load for N which is independent of S deposition. The eutrophication equation is concentrated on here, but many of the issues raised apply equally to the acidification equation.

Using the steady-state mass balance method, the critical load of N to protect forests from eutrophication effects is defined as:

$$CL_N = N_{leach} + N_{humus} + N_{denit} + N_{biomass} - N_{fix} \qquad (3)$$

where N_{leach} = acceptable N leaching; N_{humus} = immobilisation in organic matter; N_{denit} = loss by denitrification; $N_{biomass}$ = net accumulation in biomass; and N_{fix} = N fixation.

The eutrophication approach aims to protect the forest ecosystem from 'N saturation', as defined by Aber *et al.* (1989), ie when the availability of inorganic N is in excess of the total combined plant and microbial nutritional demand. Nitrate leaching above an 'acceptable' amount is used as an indicator of N saturation. Some nitrate leaching occurs even in unpolluted forests, usually in winter when demand for N by biomass is low. This natural leaching is accommodated in the N_{leach} term. This term also has to protect groundwater from eutrophication. Sinks for N - immobilisation, denitrification and uptake into biomass - prevent N inputs from being leached and can be included in the critical load. An upper limit is placed on N_{humus} in order to prevent 'destabilisation' of the ecosystem.

A significant improvement on the approach in the CCE mapping manual is that biomass is no longer considered an infinite sink. N uptake is limited to prevent nutrient imbalance. The amount of N that a tree can assimilate without adverse effects on nutrient/N ratios will depend on the extent to which it can be balanced by a long-term supply of base cations. This figure can be calculated using the Nutrient Limitation Model (Warfvinge, Sverdrup & Rosen 1992):

$$N_{biomass} = D_x + W_x - L_x / x{:}N \qquad (4)$$

where D_x = deposition of nutrient x; W_x = supply of x by weathering; L_x = x unavailable for uptake by biomass; and x:N = nutrient/N ratio in biomass.

Nutrient availability will influence the susceptibility of a forest to excess N deposition. 'Type 2' decline in pine (*Pinus* spp.) forests in The Netherlands has been attributed to high ratios of NH_4^+ to base cations in the soil solution which inhibit nutrient uptake, resulting in characteristic deficiencies (Roelofs *et al.* 1985). The most severe dieback occurs on sandy, nutrient-poor, podzolic soils. Field and experimental data show that nutrient deficiencies may limit N uptake - and consequently increase N leaching. In the Excess Nitrogen Experiment, uptake of applied N by Norway spruce was limited by P and K deficiency. When the deficiency was corrected by a PK fertilizer, needle N status increased significantly compared with unfertilized trees (Wilson 1991). Application of a PK fertilizer to mature Sitka spruce *(Picea sitchensis)* plantations in north Wales significantly increased the uptake of N by the trees and reduced the concentrations of nitrate in soil water (Stevens *et al.* 1993).

The Nutrient Limitation Model performs two functions. First, it safeguards against the adverse effects of high rates of N assimilation by limiting N_u so that acceptable nutrient/N ratios in biomass are maintained. Second, it makes N uptake sensitive to nutrient supply.

APPLYING THE LOKEBERG EUTROPHICATION EQUATION TO THE RESULTS OF THE EXCESS NITROGEN EXPERIMENT WITH NORWAY SPRUCE

One of the major advantages of using controlled experiments to test the critical load equation is the availability of data. Few countries attempting to apply the critical load concept to the 'real world' will have the required data available to them. The summary document of the Lokeberg workshop provides guideline values, usually as a range, for countries which have insufficient data. These values are shown in Table 1.

Table 1. Guideline values recommended in the Lokeberg workshop summary document for calculating critical loads (N) based on eutrophication effects

	Guideline values (kg N ha^{-1} yr^{-1})		
Parameter	Lower limit	Upper limit	Other
N_{leach}	2	4	Values from 'pristine' sites
N_{humus}	0	3	Accumulation since glaciation divided by years since glaciation. Values based on C/N ratios
N_{denit}	-	-	Relationship based on data from Sverdrup and Ineson
$N_{biomass}$	-	-	Use Nutrient Limitation Model with critical nutrient ratios, or N uptake in harvested biomass
N_{fix}	0	3	

Using these recommended values simplifies the determination of critical loads, but has certain disadvantages. As guideline values replace site-specific data, critical loads will become less sensitive to the ecosystems they aim to protect. The result may be rather uniform maps of critical loads. There is also the problem of how comparable the maps will be between countries which use guideline values and those which use real data. Furthermore, selecting the lower, upper or intermediate guideline values within the recommended range may make a considerable difference to the critical load.

These issues have been explored by applying the eutrophication equations 3 and 4 to the experimental Norway spruce 'ecosystem'. This exercise looks at:

- the effect of using guideline values and experimental data for nitrogen uptake terms (N_{humus} and $N_{biomass}$);
- the different methods recommended in the Lokeberg summary document for estimating N_{leach};
- the choice of guideline values within the recommended range.

The relationship between the calculated critical loads for Norway spruce and the N inputs at which harmful effects were observed is examined.

The data from the Excess Nitrogen Experiment which have been used in the critical load calculations are shown in Tables 2 and 3. Additional data required to calculate the critical uptake of N by biomass using the Nutrient Limitation Model are given in Table 3.

Table 2. Data from the Excess Nitrogen Experiment with Norway spruce, used to calculate critical loads based on eutrophication effects

Treatment (kg N ha^{-1} yr^{-1})	Water flux, Q (m^3 ha^{-1} yr^{-1})	Nitrate leaching in (control treatments)* (kg N ha^{-1} yr^{-1})	N_{leach}^{a} (kg N ha^{-1} yr^{-1})	N_{leach}^{b} (kg N ha^{-1} yr^{-1})	N uptake ($N_{biomass} + N_{humus}$) (kg N ha^{-1} yr^{-1})	Denitrifcation^c (kg N ha^{-1} yr^{-1})
Ammonium						
0	4959	0.34	27.8	55.5	2.72	1.75
30	4297	0.34	24.1	48.1	27.0	5.74
60	4297	0.34	24.1	48.2	55.0	8.54
120	4904	0.34	27.5	54.9	78.7	10.5
Nitrate						
0	4408	1.99	24.7	49.4	4.02	1.82
30	5014	1.99	28.1	56.3	21.4	5.74
60	4738	1.99	26.5	53.1	39.2	8.54
120	4959	1.99	27.8	55.5	68.9	10.5

*Taken as a 'pristine' ecosystem

[a] N_{leach} calculated using a critical concentration of 5.6 mg N l^{-1} to protect ground flora

[b] N_{leach} calculated using a critical concentration of 11.3 mg N l^{-1} to protect drinking water

[c] N_{denit} calculated using the Sverdrup - Ineson relationship

Table 3. Data from the Excess Nitrogen Experiment with Norway spruce, used to calculate $N_{biomass}$ using the Nutrient Limitation Model

Treatment (kg N ha^{-1} yr^{-1})	K deposition (keq ha^{-1} yr^{-1})	Mg deposition (keq ha^{-1} yr^{-1})	Ca deposition (keq ha^{-1} yr^{-1})	K/N molar ratio in biomass*	Mg/N molar ratio in biomass*	Ca/N molar ratio in biomass*	Weathering rate, K (keq ha^{-1} yr^{-1})	Weathering rate, Mg (keq ha^{-1} yr^{-1})	Weathering rate, Ca (keq ha^{-1} yr^{-1})	Critical N uptake (biomass) (kg N ha^{-1} yr^{-1})
Ammonium										
0	0.08	0.13	0.36	0.37	0.35	1.17	0.03	0.49	0.01	4.05
30	0.08	0.13	0.36	0.52	0.22	1.32	0.03	0.49	0.01	2.93
60	0.08	0.13	0.36	0.39	0.19	1.37	0.03	0.49	0.01	3.50
120	0.08	0.13	0.36	0.42	0.19	1.41	0.03	0.49	0.01	3.37
Nitrate										
0	0.08	0.13	0.36	0.65	0.29	1.60	0.03	0.49	0.01	2.35
30	0.08	0.13	.036	0.62	1.53	1.53	0.03	0.49	0.01	2.47
60	0.08	0.13	0.36	0.41	0.99	0.99	0.03	0.49	0.01	3.77
120	0.08	0.13	0.36	0.39	1.22	1.22	0.03	0.49	0.01	3.91

*Above-ground biomass

Weathering rates (W_x) have been calculated by the PROFILE model for a similar geology at Plynlimon, Wales (W H Robertson, pers. comm.). The limiting nutrient concentrations (L_x) are those suggested by Warfvinge *et al.* (1992), ie 0, 5 and 5 meq m^{-3} for K, Mg and Ca, respectively.

The use of experimental data and guidelines values for N_{humus} and $N_{biomass}$

The effect on the critical load of using experimental data from the Excess Nitrogen Experiment and guideline values for N uptake is illustrated in Figures 2 and 3. In Figure 2, critical loads based on experimental data are calculated using nitrate leaching in the control treatments (taken as a 'pristine' ecosystem) for N_{leach} and N uptake from N budgets for $N_{humus} + N_{biomass}$ (Table 2). Denitrification

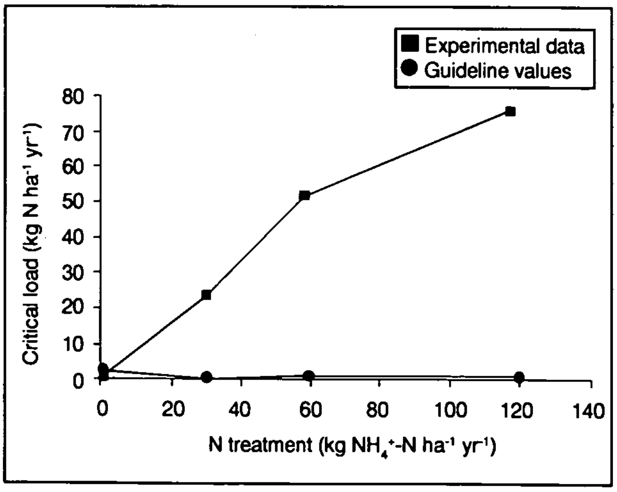

Figure 2. Critical loads (N) for an experimental Norway spruce ecosystem (ammonium treatments) using eutrophication criteria. Critical loads are calculated using minimum guideline values and experimental data for $N_{biomass}$ and N_{humus}, and minimum guideline values for other parameters

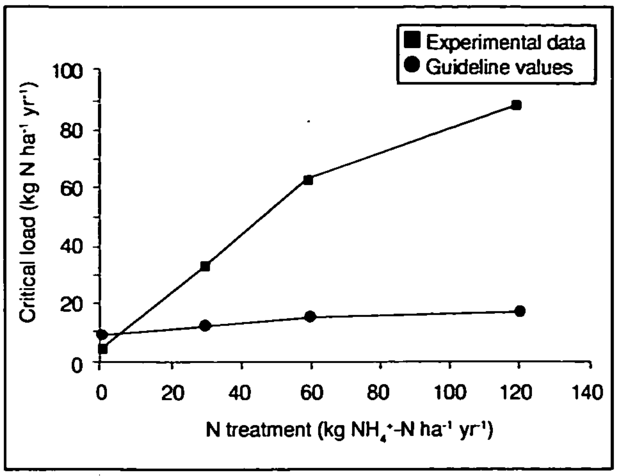

Figure 3. Critical loads (N) for an experimental Norway spruce ecosystem (ammonium treatments) using eutrophication criteria. Critical loads are calculated using maximum guideline values and experimental data for $N_{biomass}$ and N_{humus}, and maximum guideline values for other parameters

and N fixation were not measured in the experiment, so guideline values which give the lowest critical loads have been used. Critical loads based on guideline values are calculated using recommended values for N_{leach}, N_{humus}, N_{denit} and N_{fix} which give the lowest critical load. $N_{biomass}$ is calculated using the Nutrient Limitation Model. The same comparison is made in Figure 3, but using guideline values which give maximum critical loads. N_{denit} is based on the relationship given by Sverdrup and Ineson (Table 2) in Warfvinge *et al.* (1992).

Using real data for N uptake significantly increases the critical load for Norway spruce, compared with using guideline values (Figures 2 & 3), and reflects the high rate of N immobilisation by soil microbes. N immobilisation acts as an almost infinite sink, with 'saturation' or 'break-through' not occurring until N is applied at 120 kg N ha^{-1} yr^{-1} (Figure 4). N assimilation by the trees is limited by P and K deficiency and does not exceed 0.3 kg N ha^{-1} yr^{-1}. This rate of N uptake is acceptable in terms of nutrient/N ratios which remain above values critical for forest health (Huettl 1990). If critical loads are calculated using guideline values, an upper limit is placed on N_{humus}, which ignores a potentially large sink for N, and may considerably underestimate the critical load.

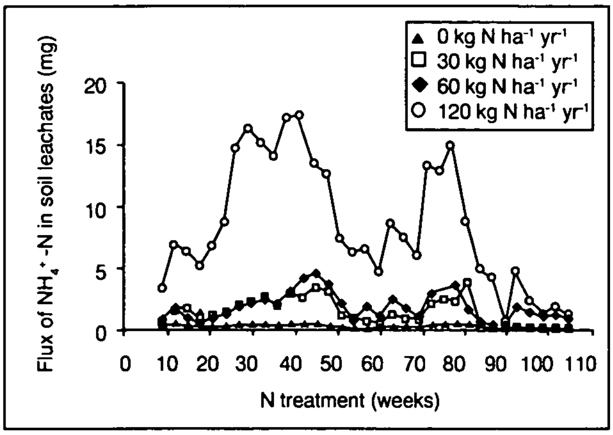

Figure 4. The effect of N load on the flux of NH_4^+-N in soil leachate of ammonium treatments

The aim of limiting N_{humus} is to prevent 'destabilisation' of the ecosystem in the long term. However, in the short term, it contradicts the premise of 'N saturation' - fundamental to the critical load concept - by leaving a sink for N 'unfilled'. As yet there is no empirical evidence to suggest that N accumulation will have adverse effects on forest health. This question will not be answered by short-term studies, and there may be a case for exercising the 'precautionary principle' by setting an upper limit for N_{humus}.

Deposition dependency

Calculating critical loads using experimental data for N_{humus} and $N_{biomass}$ shows that the critical load increases with the load of N applied (Figures 2 & 3). This result reflects the increase in N immobilisation at higher rates of application. The critical load of any forest ecosystem is unlikely to be independent of N deposition because N uptake by biomass increases with N availability, as has been demonstrated both practically (eg Miller 1986) and theoretically (Agren 1985). Three functions in equation 3 may be deposition-dependent: $N_{biomass}$, N_{humus} and N_{denit}. The Nutrient Limitation Model sets a fixed value for acceptable N uptake which is sensitive to nutrient availability, but independent of N deposition. In reality, N assimilation by trees will be approximately proportional to supply up to the point when the critical uptake rate is reached. The Lokeberg summary document recommends that countries which do not have the data required to calculate $N_{biomass}$ according to the Nutrient Limitation Model should use the mean N accumulation over a rotation. This method gives the critical load at present rates of deposition, and does not allow for an increase in $N_{biomass}$ with increasing N load. The critical load will also be deposition-dependent in waterlogged sites, according to the relationship of Sverdrup and Ineson, where denitrification increases with deposition (Warfvinge *et al.* 1992). If critical loads are based on real data, they are likely to be deposition-dependent,and hence reflect existing deposition patterns to some degree.

The chemical form of N deposition

Using guideline values for N_{humus} and $N_{biomass}$ will mask any differential response to the two chemical forms of N. A number of experiments show that most conifers exert a preference for either ammonium-N or nitrate-N (eg Bigg & Daniel 1978; Krajina, Madoc-Jones & Mellor 1973; Van den Driessche 1971). If real data are used, the magnitude of N_{humus} and $N_{biomass}$ will depend on the chemical form of N available. Figure 5 shows critical loads for Norway spruce in ammonium and nitrate treatments, calculated using experimental data for N_{humus}, $N_{biomass}$ and N_{leach}. In the 60 and 120 kg N treatments, the critical load for ammonium exceeds that for nitrate by 14 and 8 kg N ha^{-1} yr^{-1}, respectively.

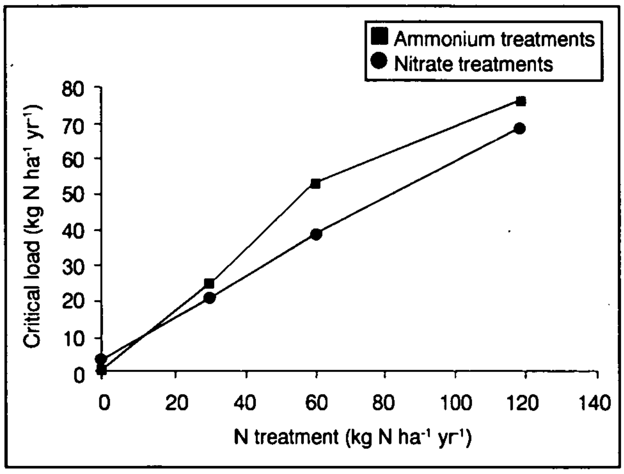

Figure 5. Critical loads (N) for ammonium and nitrate treatments of an experimental Norway spruce ecosystem based on eutrophication criteria. Critical loads are calculated using experimental data. N_{leach} is the NO_3^- flux in control treatments

Uptake of NH_4^+-N by both vegetation and soil microbes is greater than uptake of NO_3^--N. Foliar analyses for amino-acids showed that more NH_4^+-N was assimilated by Norway spruce than NO_3^--N (Wilson 1991). Differential retention of NO_3^--N and NH_4^+-N by soil microbes results in different losses of N from the system in ammonium and nitrate treatments for the same load applied. In ammonium treatments the amount of N retained by the system increases with the amount applied, until saturation in the 120 kg N treatment (Figure 4). In nitrate treatments, the flux of N increases linearly with load (Table 4).

Table 4. Nitrogen flux and retention in ammonium and nitrate treatments of the Excess Nitrogen Experiment with Norway spruce

	Total N flux in leachate (kg N ha^{-1} yr^{-1})		Nitrogen retained as % of that applied	
N treatment (kg N ha^{-1} yr^{-1})	Ammonium treatments	Nitrate treatments	Ammonium treatments	Nitrate treatments
0	2.20	1.98	55.0	67.1
30	7.99	14.6	77.2	59.4
60	9.93	26.8	84.7	59.5
120	46.3	57.1	62.9	54.7

Table 5. Concentrations of NH_4^+-N and NO_3^--N in soil leachate in ammonium and nitrate treatments of the Excess Nitrogen Experiment with Norway spruce

	NO_3^--N concentration in leachate (mg N l^{-1})		NH_4^+-N concentration in leachate (mg N l^{-1})	
N treatment (kg N ha^{-1} yr^{-1})	Ammonium treatments	Nitrate treatments	Ammonium treatments	Nitrate treatments
0	0.07	0.45	0.39	0.08
30	0.03	2.97	1.80	0.07
60	0.05	5.64	2.30	0.09
120	0.04	11.4	9.40	0.09

The chemical form of N deposition and the preferences of species for NH_4^+-N or NO_3^--N can evidently affect the critical load of an ecosystem. It is important that these factors are considered when setting guideline values and determining critical loads.

Estimating N_{leach}

The Lokeberg summary document recommends three ways of estimating N_{leach}:

- critical concentration multiplied by the runoff, Q;
- an estimated N flux, typical of forests which are not N-saturated (2-4 kg N ha^{-1} yr^{-1});
- nitrate leaching measured in pristine ecosystems.

The first method has the advantage over a guideline flux value of using site-specific data for Q. Currently, there is no consensus on the 'critical nitrate concentration', although values for first-order streams in areas of low deposition are being collated (M Hornung, pers. comm.). Alternative approaches use critical nitrate concentrations which (i) prevent changes in ground flora, or (ii) maintain groundwater quality within CEC limits for drinking water. Critical nitrate concentrations are 25 mg l^{-1} (5.6 mg N l^{-1}) (De Vries & Kros 1991) and 50 mg l^{-1} (11.3 mg N l^{-1}) for (i) and (ii) respectively.

Figures 6 (ammonium treatments) and 7 (nitrate treatments) compare critical loads using experimental data and upper and lower limit guideline fluxes for N_{leach}, or N_{leach} calculated by critical nitrate concentrations multiplied by Q.

Critical loads for Norway spruce in both ammonium and nitrate treatments are considerably higher when N_{leach} is calculated using critical NO_3^- concentrations, compared with guideline fluxes for N_{leach}. Critical concentrations of 5.6 and 11.3 mg N l^{-1} increase the critical load by approximately 25 and 50 kg N ha^{-1},

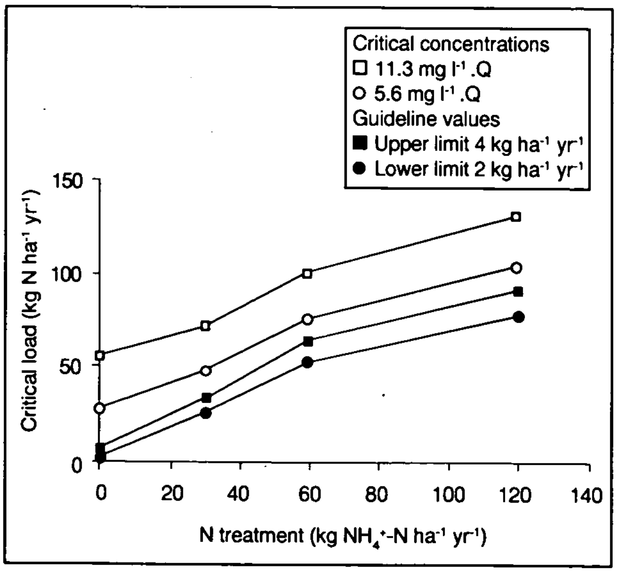

Figure 6. Critical loads (N) for an experimental Norway spruce ecosystem (ammonium treatments) using different values for N_{leach}

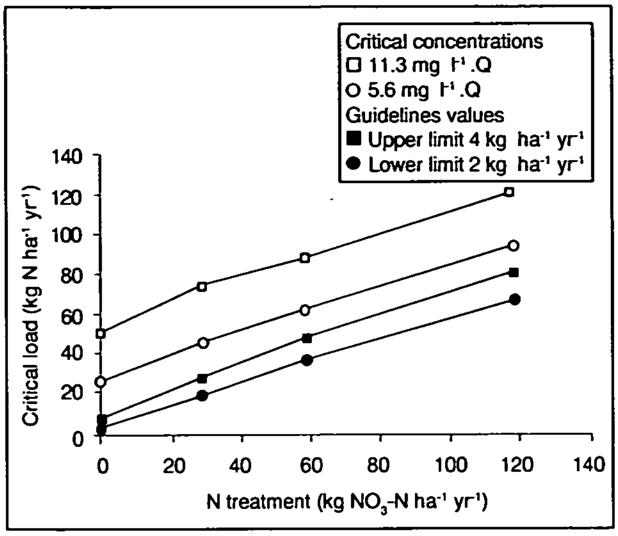

Figure 7. Critical loads (N) for an experimental Norway spruce ecosystem (nitrate treatments) using different values for N_{leach}

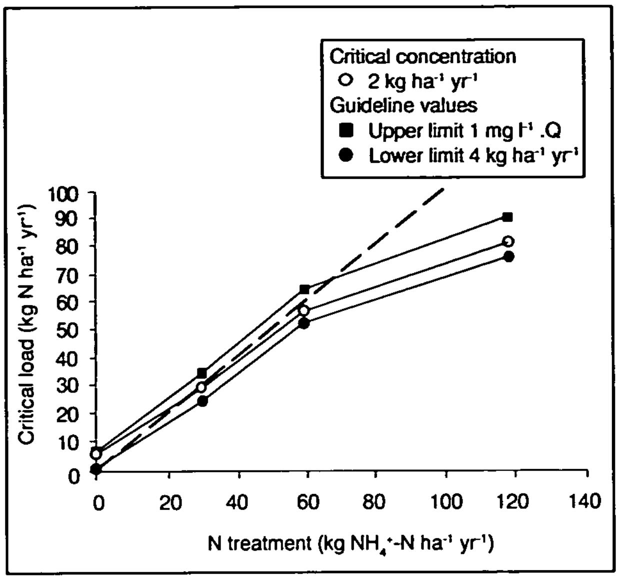

Figure 8. Critical loads (N) for an experimental Norway spruce ecosystem (ammonium treatments) using experimental data and different values for N_{leach}. - - - plots the experimental N input equal to the critical load *(x=y)*

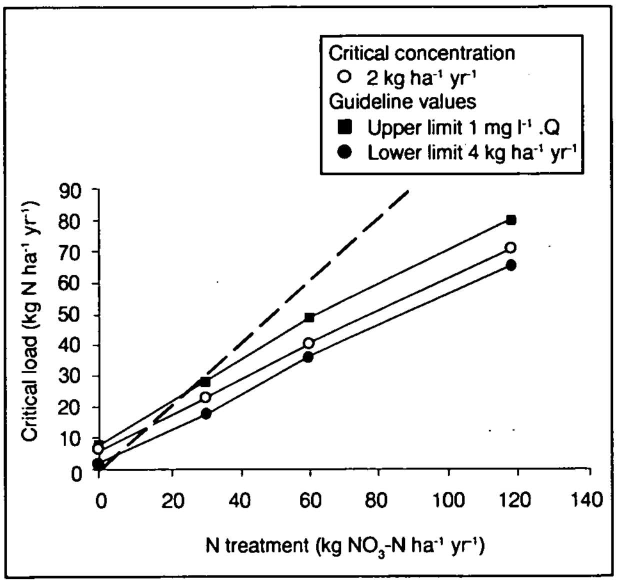

Figure 9. Critical loads (N) for an experimental Norway spruce ecosystem (nitrate treatments) using experimental data and different values for N_{leach}. - - - plots experimental N input equal to the critical load *(x=y)*

and N_{leach} becomes the dominant term in equation 3. The flux in the Excess Nitrogen Experiment is not atypical of runoff from British catchments which ranges from 1500 to 15 000 m^3 ha^{-1} yr^{-1} (Sverdrup, De Vries & Henriksen 1990). Because Q is a large number, N_{leach} will be highly sensitive to the critical nitrate concentration. Figures 6 and 7 show that the critical load differs significantly according to whether the critical concentration aims to protect ground flora or drinking water. Even the lower concentration of 5.6 mg N l^{-1} is likely to be far too high to protect forests from N saturation. Using this value gives an acceptable NO_3^- leaching of approximately 25 kg N ha^{-1} yr^{-1}, which is considerably greater than in forests considered to be N-saturated. In the Beddgelert catchment in north Wales - which is N-saturated to the extent that N outputs exceed inputs - NO_3^- leaching is only 11 kg N ha^{-1} yr^{-1} (Stevens, Hornung & Hughes1989). This suggests that a critical concentration of 5.6 mg N l^{-1} may be more than 2.5 times too high to safeguard against N saturation.

A more appropriate critical concentration might be estimated from NO_3^- concentrations in first-order streams of unpolluted catchments. In the Glendye and Peatfold catchments (north-east Scotland), where mean annual inputs of N in rainfall are 14.7 and 8.3 kg N ha^{-1} yr^{-1}, respectively, the concentration of NO_3^--N in output rivers does not exceed 0.3 mg l^{-1} (Edwards, Creasey & Cresser 1985). In the N-saturated forest at Beddgelert, concentrations of NO_3^- range from 0.17 to 2.4 mg N l^{-1} in streams and from 0.01 to 9.9 mg N l^{-1} in the C horizon of soil (Stevens *et al.* 1989). Although an extensive literature review is required, 1 mg N l^{-1} may be a suitable critical nitrate concentration to protect forests from N saturation. Figures 8 and 9 compare critical loads for the Norway spruce ecosystem calculated using experimental data and either the upper or lower limit guideline fluxes for N_{leach}, or a critical NO_3^--N concentration of 1 mg l^{-1}. Critical loads calculated using the critical concentration of 1 mg N l^{-1} are intermediate between those calculated with the upper and lower guideline fluxes for N_{leach}, indicating that the recommended fluxes are appropriate for safeguarding against N saturation.

Calculating N_{leach} using a critical concentration makes the term more site-specific than using a guideline flux value, but introduces dependency on the runoff term, Q. Critical loads calculated in this way may reflect annual precipitation at a site, rather than the sensitivity of the ecosystem to N inputs. In Britain, where runoff varies from 1500 m^3 ha^{-1} yr^{-1} in the south-east to 15 000 m^3 ha^{-1} yr^{-1} in west Scotland and Wales (Sverdrup *et al.* 1990), the N_{leach} term could range from 1.5 to 15 kg N ha^{-1} yr^{-1} (based on a critical concentration of 1 mg N l^{-1}) for similar ecosystems.

If N_{leach} is calculated using a critical concentration of 1 mg N l^{-1}, it will prevent the eutrophication of groundwater, but may leave forests vulnerable to N saturation where Q is high. In Welsh forests, the acceptable nitrate leaching based on a critical concentration of 1 mg N l^{-1} could be as high as 15 kg N ha^{-1} yr^{-1}, a flux which would normally be considered indicative of N saturation. Forests with a high runoff will generally tolerate higher inputs as N is less likely to accumulate. However, where a high Q gives a large critical load, there is no guarantee that N inputs equivalent to N_{leach} will be lost from the system by leaching rather than assimilated by biomass. Inputs of NH_4^+-N, which are less mobile than NO_3^--N, are more likely to accumulate. The critical load equation 3 assumes that N deposition will be allocated to sinks in the environment according to their fraction of the critical load, but, in reality, this may not be the case.

Choice of guideline values within recommended range

Countries which rely on guideline values for calculating critical loads will have to choose appropriate values from within the recommended ranges. Figure 10 shows the critical N load for the spruce ecosystem calculated using values from the guideline ranges to give minimum, intermediate and maximum scenarios in ammonium treatments. The minimum and intermediate scenarios differ only for N_{leach}. The minimum scenario uses the recommended 'leaching from pristine systems' by taking the annual flux of NO_3^- in the control treatments of the experiment. The intermediate and maximum scenarios use the lower and upper values of the guideline ranges (2 and 4 kg N ha^{-1} yr^{-1}) for N_{leach}. The maximum values for denitrification are from the Sverdrup-Ineson relationship (Table 2). $N_{biomass}$ is calculated by the Nutrient Limitation Model (Warfvinge *et al.* 1992) using experimental data for base cation deposition and cation/N ratios measured in the Norway spruce seedlings used in the experiment (Table 3).

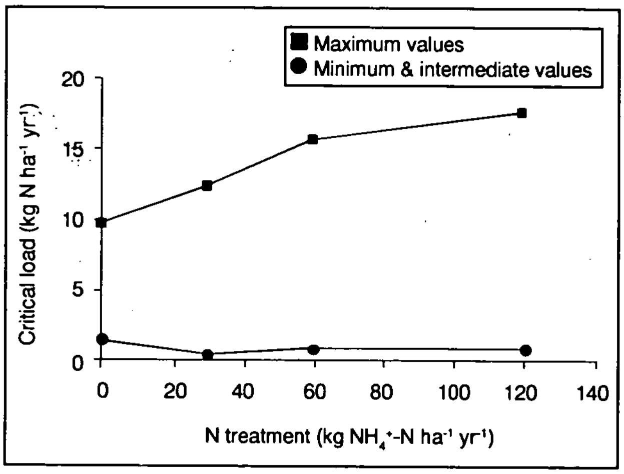

Figure 10. Critical loads (N) for an experimental Norway spruce ecosystem (ammonium treatments) using eutrophication criteria. Critical loads are calculated using minimum, intermediate and maximum guideline values (see text)

In ammonium treatments, there is very little difference between the critical load in the minimum and intermediate scenarios, suggesting that the recommended lower guideline value for nitrate leaching in temperate coniferous forests (2 kg N ha^{-1} yr^{-1}) is consistent with 'natural' leaching at low rates of N deposition for the Norway spruce in this experiment. There is, however, a significant difference between the minimum/intermediate and the maximum scenarios. Using the upper values of the recommended ranges gives a mean critical load of 14 kg N ha^{-1} yr^{-1} - 14 times the mean critical load calculated by the minimum scenario. While this comparison adopts the two extremes, it illustrates the need to choose the 'correct' guideline value. This choice would be aided by a data base of the parameters in equations 3 and 4 for a large number of ecosystems with different characteristics, eg geology, soil type, climate, species, etc. It may be that the guideline ranges would need to be extended, but countries would be given the opportunity to select values which reflect their own types of ecosystems.

Critical load calculations and observed harmful effects

The ultimate test of the validity of the critical load equation is whether calculated values relate to the N deposition at which harmful effects actually occur. The symptoms of European 'Type 1' decline were not reproduced in Norway spruce in the Excess Nitrogen Experiment, but a number of effects considered harmful to ecosystem function were observed.

The critical load for the experimental spruce ecosystem can be assessed by a number of criteria:

- N saturation - leaching of N above 'natural' rates (>2-4 kg N ha^{-1} yr^{-1});
- critical N concentrations in leachate - these should be <5.6 kg N ha^{-1} yr^{-1} to protect ground flora, <11.3 kg N ha^{-1} yr^{-1} to protect drinking water, and approximately 1 mg N l^{-1} to safeguard against N saturation;
- adverse effects on tree nutrition.

These criteria have been used to compare the N load which induced harmful effects in the spruce 'ecosystem' with critical loads calculated by the Lokeberg eutrophication equation.

Empirical critical loads in ammonium treatments

N saturation of the spruce ecosystem can be assessed in terms of an acceptable N flux, a critical NO_3^- concentration, or the N input at which the demand for N is exceeded. Applying the criterion of nitrate leaching to Norway spruce treated with NH_4^+-N gives a critical load in excess of 120 kg N ha^{-1} yr^{-1}, as the flux of NO_3^- was <2 kg N ha^{-1} yr^{-1} in all treatments. Nitrate concentrations in leachate (Table

5) did not approach the lower limit of 1 mg N l^{-1}, which may be an appropriate critical concentration to prevent N saturation. If, however, saturation is defined as the point at which N inputs exceed demands, the critical NH_4^+-N load for this ecosystem could be judged to be between 60 and 120 kg N ha^{-1} yr^{-1}. When 120 kg NH_4^+-N was applied to the soil, a significant 'break-through' or leaching of N occurred, compared with the 0, 30 and 60 kg N treatments (Figure 4). If the aim is to prevent changes in ground flora by maintaining N concentrations in leachate below 5.6 mg N l^{-1}, the critical load would also be between 60 and 120 kg N ha^{-1} yr^{-1} (Table 5). The critical load would be the same if effects on tree nutrition were taken as the criterion for damage. The 120 kg N treatment significantly acidified the soil compared with lower doses, and increased the loss of Mg, Ca and K by leaching. The result was a significant reduction in foliar Mg concentrations in the 120 kg N ha^{-1} yr^{-1} treatments, although not to deficiency levels (Wilson 1991). The critical NH_4^+-N load for Norway spruce in this experiment - invoking all three criteria - can be taken as between 60 and 120 kg N ha^{-1} yr^{-1}.

Empirical critical loads in nitrate treatments

Unlike ammonium treatments, inputs of NO_3^--N to Norway spruce did not acidify the soil or have adverse effects on nutrient status. In nitrate treatments, the critical load should be assessed by N saturation criteria. Nitrate leaching exceeded 4 kg N ha^{-1} yr^{-1} - the upper limit considered acceptable for non-saturated forests - in the 30 kg N treatment (Table 4). The estimated critical N concentration to protect against N saturation (1 mg N l^{-1}) was also exceeded in this treatment (Table 5). Critical N concentrations aimed at protecting ground flora (5.6 mg N l^{-1}) and drinking water (11.3 mg N l^{-1}) were exceeded by nitrogen inputs of between 30 and 60 kg N ha^{-1} yr^{-1} (Table 5). From the point of view of N saturation, the critical NO_3^--N load for the spruce ecosystem would appear to be approximately 30 kg N ha^{-1} yr^{-1}.

The relationship between calculated critical loads and observed harmful effects

Figures 8 and 9 show the critical loads for Norway spruce calculated using the Lokeberg eutrophication equation, for ammonium and nitrate treatments respectively. Values are based on experimental data and either the lower or upper limit of guideline fluxes for N_{leach}, or N_{leach} calculated using a critical concentration of 1 mg N l^{-1}. The dashed line shows the experimental N input equal to the critical load (*x*=*y*). Where the calculated critical load falls below this line, N inputs exceed the critical load. At these points, we would expect to observe 'harmful effects'.

In ammonium treatments (Figure 8), the critical load (calculated using a critical concentration of 1 mg N l^{-1}) is approximately equal to N load in the 30 kg N treatment. The critical load begins to be exceeded at inputs of 60 kg N ha^{-1} yr^{-1}. The critical load calculated with all three methods for N_{leach} is exceeded at inputs of approximately 70 kg N ha^{-1} yr^{-1}. Application of the 120 kg N ha^{-1} yr^{-1} treatment exceeds the calculated critical load by some 40 kg N ha^{-1} yr^{-1}, suggesting that harmful effects should occur above approximately 70 kg N ha^{-1} yr^{-1}, and be observable in the 120 kg N treatment. These predictions correspond well with the N inputs at which adverse effects were actually observed.

In nitrate treatments, critical loads calculated using all three values for N_{leach} are just exceeded in the 30 kg N treatment, indicating that harmful effects should start to occur when N is applied above this rate. As in ammonium treatments, the calculated critical loads correspond well to the empirically derived critical loads.

In the case of this experimental Norway spruce ecosystem, the Lokeberg equation based on eutrophication effects gives a reasonable estimate of the critical load if real data are used. Using guideline values for N uptake, however, considerably underestimates the critical load (Figures 2 & 3) by 13-55 kg N ha^{-1} yr^{-1}. The current approach for determining critical loads reflects the sensitivity of Norway spruce to the chemical form of N deposition, if real data are used. Lower critical loads were obtained for NO_3^--N deposition than for NH_4^+-N deposition, consistent with the occurrence of harmful effects at lower inputs of N in nitrate treatments (Figures 8 & 9). The critical load for NO_3^--N was lower because nitrate was less readily assimilated by biomass than ammonium. This fact was only revealed because experimental data were used for N uptake.

DISCUSSION

A critical load for nitrogen is a relatively new concept, and there has been little opportunity to test the current approach against real data. Using data from the Excess Nitrogen Experiment with Norway spruce indicates that the Lokeberg equation based on eutrophication effects is a reasonable model. Calculated critical loads accurately predicted the N load at which adverse effects occurred. The sensitivity of Norway spruce to nitrate and ammonium forms of N and the magnitude of N inputs were also successfully modelled by the equation.

This exercise raised a number of questions about the use of guideline values generally, and their use for the N_{humus}, $N_{biomass}$ and N_{leach} terms specifically. For Norway spruce in this experiment, critical loads calculated using guideline values were a very poor estimate of the N input which had harmful effects. As critical load calculations become more dependent on guideline values, they will become less sensitive to the ecosystem they aim to protect. Maps of critical loads may begin to look quite uniform across Europe. However, the current approach provides limited opportunities to input site-specific data into the Lokeberg eutrophication equation. If critical loads are calculated using the Nutrient Limitation Model, the supply of the limiting nutrient by weathering and deposition (W_x and D_x) are the only terms which are site-specific for non-waterlogged sites. If the critical N/nutrient ratios are made species-specific, this parameter will also be tailored to a particular ecosystem.

Inevitably, some countries will use real data and others guideline values to calculate critical loads. This practice is likely to result in very different values for similar ecosystems. Countries using a standard data base will have to make decisions on whether to use upper, lower or intermediate guideline values - which could also significantly affect the critical load. If the input data are not comparable between different countries, the validity of the critical load maps for influencing international emission strategies at an international level is questionable.

Applying the Lokèberg equation to data from the Excess Nitrogen Experiment highlighted the need to use real data for N_{humus}. Using guideline values underestimated the critical load by placing an upper limit on N_{humus}, and masked the sensitivity of Norway spruce to both the magnitude and chemical form of N deposition. Placing an upper limit on the amount of N which an ecosystem is permitted to immobilise contradicts the basic aim of a critical load to 'prevent N saturation', because this sink may be larger than the recommended value. It may, however, protect the ecosystem against destabilisation in the long term. This raises the issue of the timescale for which we expect our present decisions to protect forest ecosystems.

Both the currently recommended methods for calculating N_{leach} have limitations. N_{leach} calculated using a critical concentration multiplied by Q will protect groundwater, but may give an 'acceptable' N flux which is indicative of saturation (if Q is high). Conversely, in a dry area, the criterion of acceptable N flux could be met, while the N concentration exceeds that required to maintain groundwater quality.

Realistically, few countries will have the data required to calculate critical loads, and it will be necessary to supply guideline values. An extensive data matrix encompassing a wide range of ecosystem characteristics is required so that guideline values can be approximated to site data as far as possible.

It is only by applying the critical load equations to real data that theoretical limitations and practical problems will be discovered. Much would be gained by applying the equation to forest ecosystems already damaged by atmospheric N deposition. It would be useful to test the robustness of the equation to different chemical forms of nitrogen and different harmful effects, using data from declining forests in The Netherlands where high rates of NH_4^+-N inputs have caused nutrient deficiencies, and N-saturated forests such as Beddgelert in Wales which are leaching NO_3^- at high rates. If the critical load concept is to play a role in emission reduction strategies, it is essential to demonstrate a clear link between calculated critical loads and N inputs having deleterious effects in the field.

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Critical loads of nitrogen for upland forests in Wales – effects of plantation age

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For ten years or more, staff at ITE Bangor have studied the effects of upland land use change on the biogeochemical cycling of nutrients. Using the small catchment approach, a primary objective has been to assess the effects of afforestation with exotic conifers, mainly Sitka spruce (*Picea sitchensis*) on soils, soil waters and streamwaters. Analysis of streamwater from a small number of agriculturally unimproved moorland and young forest stands at Llyn Brianne and Plynlimon reveals low concentrations of nitrate (Figure 1). These observations are, therefore, in line with the traditional view that these systems are nitrogen-limited and are unlikely to lose nitrogen through the process of nitrate leaching. Figure 1 also shows that the mature, 50-year-old spruce plantation at Beddgelert has a considerably higher streamwater nitrate concentration. In fact, the inorganic nitrogen budget for the Beddgelert catchment reveals a state approaching nitrogen saturation. Streamwater inorganic nitrogen outputs (entirely as nitrate) have averaged 14.6 kg N ha^{-1} yr^{-1} since 1982, whereas wet deposition inputs, which admittedly underestimate total inputs, have averaged 10.3 kg N ha^{-1} yr^{-1} (Stevens *et al.* 1993). It is clear that the trees, forest floor litter or soil microbial biomass of the Beddgelert system are unable to immobilise the available nitrogen, unlike the moorland and younger tree crops.

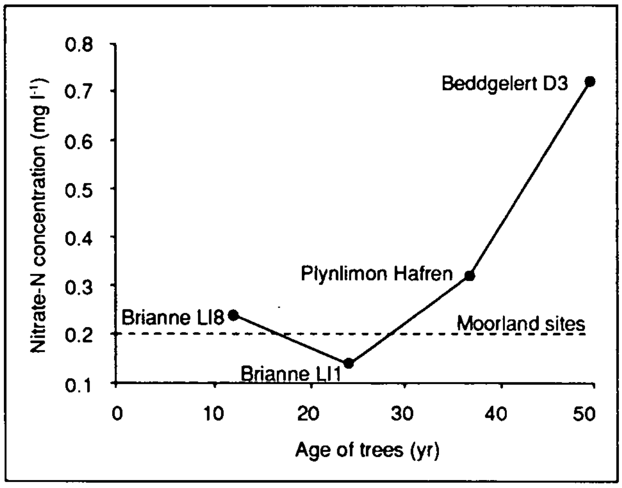

Figure 1. Streamwater nitrate concentrations at ITE forest and moorland sites in Wales

Because of the comparatively large nitrate leaching losses (for an upland ecosystem) at Beddgelert, there is concern that acidification of soils will be occurring at a faster rate than that attributable to acid deposition and biomass uptake of base cations. This concern is based upon the acidifying effect of nitrification. Nitrification is the conversion to nitrate-N of ammonium-N derived from mineralisation of soil organic matter or entering the system as atmospheric deposition. For each nitrate ion generated by nitrification, two hydrogen ions are released. If the nitrate is taken up by plants, the hydrogen ions are re-immobilised in the vegetation and there is no overall increase or decrease in hydrogen ion concentration in the system (Reuss & Johnson 1986). If the nitrate is in excess of plant or soil microbial demand, it may be leached. In this case, the hydrogen ions may:

- leach from the soil, resulting in stream acidification;
- exchange for Al or base cations on the soil exchange complex, leading to enhanced Al concentrations in streams or a reduction in soil base saturation;
- lead to enhanced weathering rates and the generation of Al minerals, which in turn will increase the extent of Al saturation on the soil exchange complex.

Acidification of soils by atmospheric deposition of nitrogen compounds will be mainly through nitrification of ammonium-N inputs, coupled with leaching of the nitrate-N generated. Acidification resulting from wet deposition of nitrate-N and dry deposition of NO_x will depend upon the relative proportions of acidic and basic cations accompanying the incoming and outgoing nitrate.

Acidification due to the nitrogen transformations in soil may be estimated using the method proposed by van Breemen, Driscoll and Mulder (1984) and applied by Berdén *et al.* (1987):

$$\text{Acidification} = (NH_4{}^+{}_{in} - NH_4{}^+{}_{out}) + (NO_3{}^-{}_{out} - NO_3{}^-{}_{in})$$

At Beddgelert, the average rate of acidification from this process for the period 1982-90 is 1.00 keq H^+ ha^{-1}

yr^{-1}. This figure compares with acid deposition (wet only) inputs of 0.94 keq H^+ ha^{-1} yr^{-1} and a critical load for acidity of 0.2 keq H^+ ha^{-1} yr^{-1} for the soil at Beddgelert (K R Bull, pers. comm.), although a degree of 'double accounting' of the nitrogen inputs is included in these calculations. In Britain, the maps of critical load exceedance of soils have been prepared using acid deposition inputs as the only acidifying influence. Nitrogen transformations have so far been excluded from these exercises, but it is clear that, in sites such as Beddgelert, this source of acidification may have a substantial impact.

It would be easy to dismiss the Beddgelert site as 'unusual'. However, there are a number of plausible and undoubtedly interlinked explanations why this and other mature spruce plantations may be unable to utilise available nitrogen, resulting in it being lost as nitrate.

i. Inputs of inorganic-N, primarily as dry deposition and cloud water deposition, may be greater to taller, older crops.
ii. Rates of mineralisation and nitrification of forest floor and pre-existing soil organic matter may be greater in older crops, either as a result of improved microclimatic conditions or increased N concentrations in throughfall (see i).
iii. Mature crops have a reduced demand for nitrogen, which results in greater rates of leakage of the N made available in throughfall and mineralisation. Reduced demand for N may be the result of smaller growth increments (because of the physiological age of the trees or deficiencies of other nutrients such as P and K), or increased internal retranslocation of N. Inputs of nitrogen in wet deposition over most of Wales are significantly greater than at pristine sites, and these inputs may be greater than the trees' requirement in the later stages of the crop cycle.

In order to test the hypothesis that there is a relationship between nitrate leaching rates and spruce crop age, the Forest Nitrogen Survey was established. In 25 upland catchments in mid- and north Wales, wet deposition inputs, throughfall, soil water and stream solute concentrations were measured every four weeks for one year until November 1991. Five of these catchments were agriculturally unimproved moorland or grassland, and there were five catchments in each of four forest age categories; 0-15, 16-30, 31-45, and greater than 45 years. Data from the first six months of this study indicate a very clear relationship between streamwater nitrate concentration and forest age (Figure 2), and a similar trend to that in Figure 1. Only in forests older than 30 years are stream nitrate concentrations higher than 0.2 mg N l^{-1}, but in the oldest sites nitrate concentrations reach 1.0 mg N l^{-1}. Initial data analysis indicates that throughfall inorganic-N fluxes increase linearly with age, but the increase is insufficient to explain the trend in streamwater nitrate concentrations. Also, rates of mineralisation and nitrification in the soil increase with crop age, and particularly in crops older than 30 years. A combination of (i) and (ii) above, and probably (iii) are, therefore, the likely causes of the observed leaching losses.

It may be concluded that soils of crops older than 30 years will be acidifying through the influence of nitrogen transformations, especially as more than half of the inorganic-N entering the system in rain or throughfall is in the ammonium form. In much of Wales, where the critical load for acidity of soils is exceeded by wet deposition inputs alone, total exceedance of the critical load for acidity will be substantially more than currently recognised.

The critical load for nitrogen *as a nutrient* is clearly exceeded in crops older than 30 years. In much of upland Wales, wet deposition inputs of inorganic-N are around 10 kg N ha^{-1} yr^{-1}, and evidence from Beddgelert indicates that total inputs are nearer 20 kg N ha^{-1} yr^{-1} if throughfall fluxes are used as a surrogate (Stevens *et al.* 1990). For there to be no leaching losses of nitrogen, immobilisation within the tree biomass or soil must occur. However, immobilisation within the soil microbial population or soil organic matter only delays the losses until a phase of increased mineralisation occurs, such as following clearfelling. It may therefore be argued that, to avoid acidification, rates of atmospheric N inputs should be balanced, in the long term, by the removal of nutrients in timber at harvest. The 50-year-old trees at Beddgelert contain 128 kg N ha^{-1} in harvestable timber and 428 kg N ha^{-1} in all above-ground biomass (Stevens *et al.* 1988), equivalent to rates of accumulation of 2.5 and 8.5 kg N ha^{-1} yr^{-1} respectively. Allowing for an acceptable (and arbitrarily defined) rate of acidification of, for example, 2.5 kg N ha^{-1} yr^{-1}, conventional (stem only) and whole-tree harvesting would require the target load for nitrogen to be set at 5 and 11 kg N ha^{-1} yr^{-1} respectively. Whole-tree harvesting is probably not a long-term solution, however, as losses of base

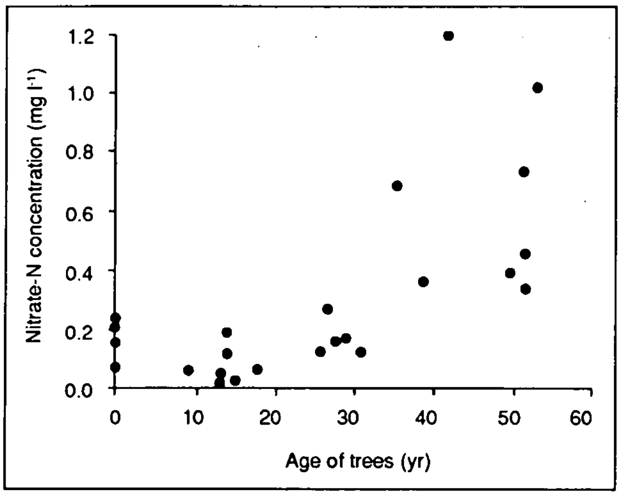

Figure 2. Streamwater nitrate concentrations at the 25 Forest Nitrogen Survey catchments. Plotted points are means of the first six months of the study. Moorland sites are plotted at age 0

cations in harvested material would also result in significant soil acidification. For example, at Beddgelert, 279 kg Ca ha^{-1} would be removed by whole-tree harvest at a site where the soil pool of extractable Ca is only 112 kg Ca ha^{-1} to the base of the rooting zone, and the total calcium content of the soil is 1390 kg ha^{-1} (Stevens *et al.* 1988). Unless this rate of removal is balanced by rain or dust inputs of Ca, significant soil base cation depletion will result.

Unless whole-tree harvesting is practised, in which case fertilization with base cations and P will be necessary, inputs of N from the atmosphere will need to be reduced substantially from around 20 kg N ha^{-1} yr^{-1} (total inputs) to 5 kg N ha^{-1} yr^{-1}. Ideally, there should be a greater reduction of ammonium-N inputs than nitrate-N. Ammonium-N is derived mainly from agricultural sources, and each mole of ammonium contributes two moles of hydrogen ion to the soil if nitrified and leached, even if this process occurs indirectly via the soil organic matter. At forest sites studied in the Forest Nitrogen Survey, results to date indicate that ammonium-N occurs in larger concentrations than nitrate-N in both incident rainfall and in throughfall, and it is therefore the more significant acidifying influence. Emissions of ammonia from intensive agriculture and the subsequent deposition in nearby woodland have been recognised as the major acidifying influence in woodlands in The Netherlands (van Breemen *et al.* 1982). It is something of a surprise, then, that the substantially less intensive animal production in upland Wales can also be the source of significant acidification of forests.

In the Forest Nitrogen Survey, it has been concluded that a substantial reduction in nitrogen inputs to forests is necessary to reduce acidification rates. However, emissions of nitrogen pollutants from industry, transport and agriculture are stable or increasing (Skeffington & Wilson 1988). We have shown that forest systems younger than 30 years immobilise incoming nitrogen, but the implications of increasing nitrogen deposition raise a number of questions regarding the future ability of these systems to continue acting in this way.

- What is the role of nitrogen on ecosystem acidification and what are the effects on other nutrients?
- What is the maximum, or critical, load for nitrogen which can be immobilised by a young Sitka spruce stand?
- What are the comparative effects of inputs of nitrate- or ammonium-N on soil and plant processes and pools?

These questions will hopefully be answered by an experiment in the hills near Aber, some 9 km east of Bangor, in which ammonium-N and/or nitrate-N is being added in a water spray to the forest floor of a 30-year-old Sitka spruce plantation at present leaching 4.2 kg N ha^{-1} yr^{-1} as nitrate and receiving inputs of 17 kg N ha^{-1} yr^{-1}. It is a randomised block experiment (Figure 3) containing three replicate

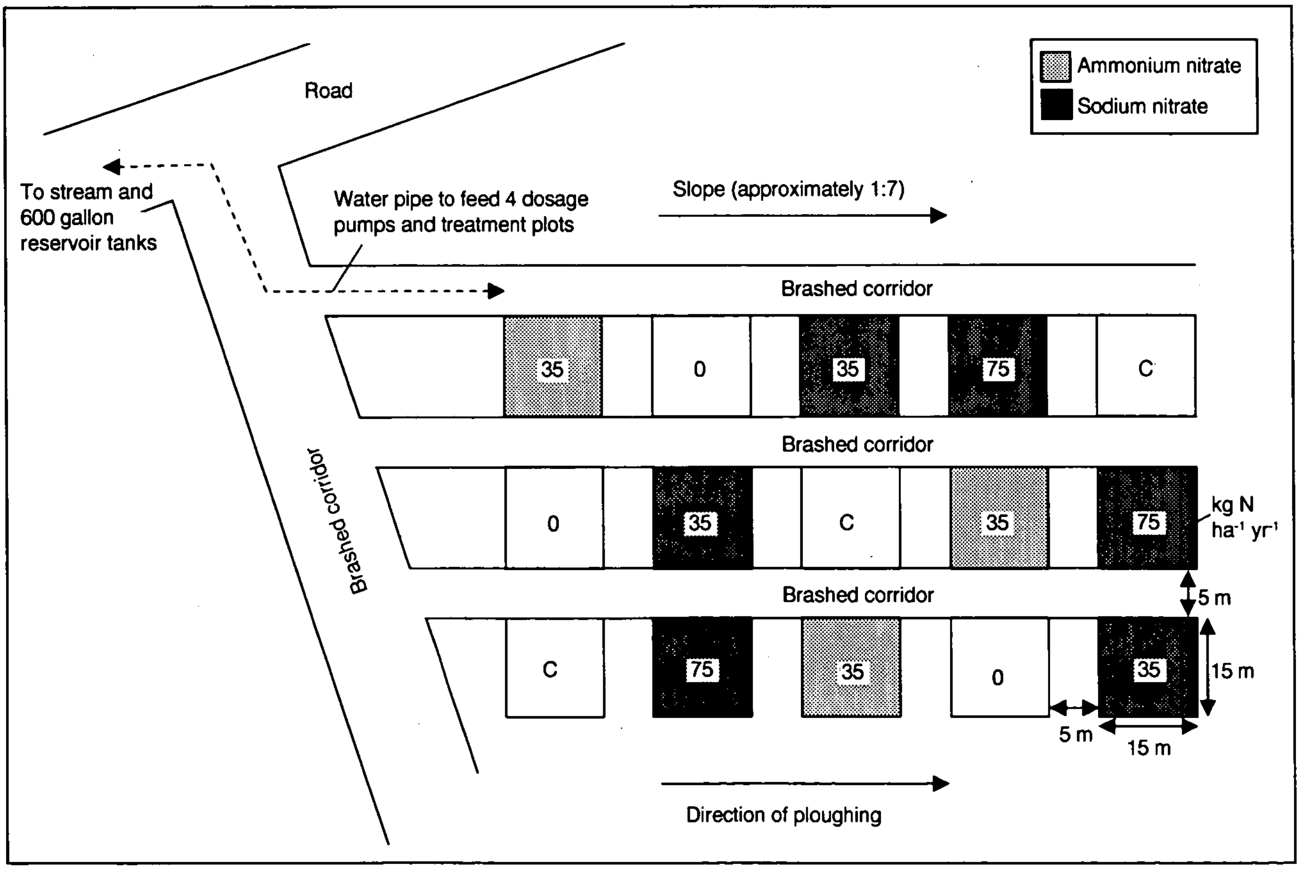

Figure 3. Layout of the Aber nitrogen critical load experiment. Each plot contains seven rows of spray tubing running parallel to plough furrows

plots of each of five treatments:

- control, no additional N, unsprayed
- zero N, sprayed with water only
- 35 kg N ha^{-1} yr^{-1} as sodium nitrate
- 35 kg N ha^{-1} yr^{-1} as ammonium nitrate
- 75 kg N ha^{-1} yr^{-1} as sodium nitrate

The additions represent extra nitrogen applied to the forest floor, and the total inputs, including throughfall and stemflow, are around 50 kg N ha^{-1} yr^{-1} in the 35 kg N spray plots. Spraying takes place approximately weekly, except in extended periods of dry or freezing weather. As a result, the additional nitrogen is applied at moderate concentration and avoids shock effects.

In each of the 15 plots, throughfall and soil water are collected and analysed every two weeks. Application of these treatments is intended to take place for 3-4 years. The first year was completed in October 1991, and ^{15}N is now being added to the N in the spray as an aid to identifying the sinks for the additional N. Results to date indicate that there is a significant sink for N in this forest; 100% of the ammonium nitrate and >75% of the nitrate in the sodium nitrate has been retained after one year of spraying. Sampling of soils, forest floor, tree biomass and litterfall, together with estimation of denitrification and soil water leaching losses, mineralisation and nitrification rates, will provide the basis for modification and development of models describing the effects of enhanced rates of nitrogen inputs to spruce plantations, and for the determination of a critical load.

Results from the Aber experiment will be integrated into an international network of Nitrogen Saturation Experiments (NITREX), comprising six European countries and funded by the CEC Science and Technology for Environmental Protection (STEP) programme. The network consists of either N addition or removal experiments across a nitrogen input pollution gradient from 5 kg N ha^{-1} yr^{-1} in Norway to 50 kg N ha^{-1} yr^{-1} in The Netherlands. The overall objectives of NITREX are to determine:

- the threshold for nitrogen saturation in coniferous forest ecosystems;
- when and how recovery occurs following removal or reduction of N inputs (forest roof experiments);
- the critical load for N in coniferous forests in Europe;
- the role of N in ecosystem acidification and forest decline.

ACKNOWLEDGEMENTS

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Critical loads for waters

Critical acidity loads for UK freshwaters – introduction, sampling strategy and use of maps:

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INTRODUCTION

The clear cause-effect relationship established between acid deposition and surface water acidification over the last decade (eg Battarbee *et al.* 1990; Last & Watling 1991) suggests that the most obvious method for controlling and reversing acidification is to eliminate or reduce acid deposition, especially over regions sensitive to acidification.

Even before the scientific basis for this relationship was fully determined and accepted by all countries, Norway and Sweden had already persuaded the United Nations Economic Commission for Europe (UN-ECE) to set up a Convention on Long Range Trans-boundary Air Pollution and to promote protocols for reducing the emissions of both sulphur and nitrogen oxides.

The sulphur protocol which encouraged Member States to reduce national emissions of SO_2 by a minimum of 30% by 1993, based on a start year of 1980, was adopted in 1985. Membership of the '30% Club' expanded rapidly, but a number of net sulphur-exporting countries, most notably the United Kingdom, did not join. Nevertheless, the UK accepted the importance of SO_2 reductions in 1986 and began the planned introduction of flue gas desulphurisation (FGD) to a small number of large British power stations, such as Drax B in Yorkshire. Subsequently, the UK has signed the European Community Directive for large combustion plants, which requires a 60% decline in emissions from these sources by 2003, based on a 1980 datum.

Despite the political success of the 30% Club, this approach to emission reduction has inadequacies, mainly because reductions need to be targetted more specifically at regions and countries with large emission sources; it is also sensible to optimise reductions to gain the most benefit to damaged regions (Bull 1991). For example, it might be more important to remove almost all emissions from power stations directly upwind of acidified regions and have lower reductions for power stations whose emissions fall largely in the sea or on land with high neutralising capacity.

For this reason, the critical loads concept has been devised and embraced by European countries, and the UN-ECE intend to replace the S protocol (30% Club) in 1993 by a critical loads approach to emission control.

The most commonly used general definition of critical loads is that of Nilsson and Grennfelt (1988):

'the highest deposition of acidifying compounds that

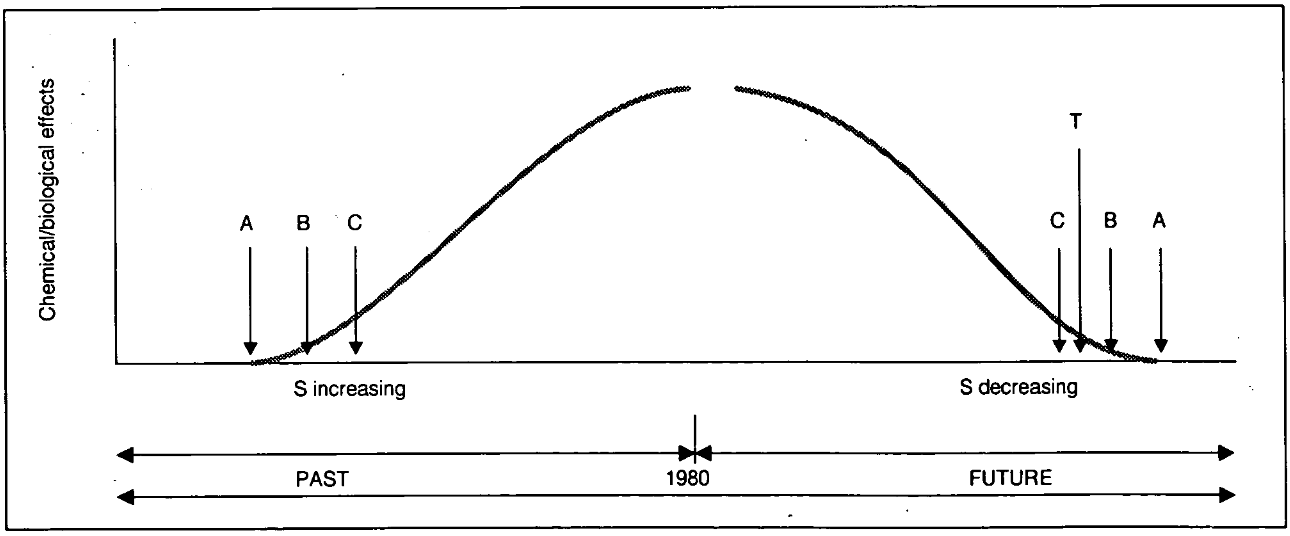

Figure 1. Critical and target loads concept graph (source: Bull 1991). The critical load for a site is exceeded at point A, points B and C represent critical loads for progressively less sensitive organisms. As sulphur deposition decreases in the future, a target load (T) can be chosen to protect selected species, or ideally to enable full recovery (point A)

will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function'.

This idea can be most simply illustrated as in Figure 1, where a chemical or biological effect is related to an increased acid loading over time. If the critical load is less than the actual measured S load at any site, then the critical load is exceeded. Maps of exceedance within and between countries can be drawn up and used for policy-making and the fixing of target loads.

The Nilsson and Grennfelt definition is quite ambiguous, mainly because of the subjectivity required to judge a long-term harmful effect and the multiplicity of effects of acid deposition on ecosystems at all structural and functional levels. Consequently, the definition can be improved if the term harmful is omitted and if critical loads are related to specific aspects of the system. Then, with reference to Figure 1, the critical load for the system as a whole (ie an individual site) is exceeded when the first change occurs in the aquatic ecosystem that can be related to acid deposition.

Additional critical values can, in theory, be defined as loads increase and as progressively less acid-sensitive organisms in the ecosystem decline or disappear. Target loads may then be set according to the need to protect any selected species or groups of species. A knowledge of species ecology in relation to water acidity/alkalinity is a prerequisite of the critical loads approach (Ormerod, pp 115-118).

In the UK freshwaters programme, three different but complementary approaches have been used to set critical loads for individual waterbodies. These are the diatom model (Battarbee *et al.* 99-102), the steady-state water chemistry model (Harriman & Christie, pp 103-108), and the MAGIC model (Jenkins & Shaw, pp 109-114). The first two of these have been used to map critical loads on a 10 km x 10 km grid square basis throughout the UK, and critical load exceedances on a 10 km x 10 km grid square basis using modelled sulphur deposition data mapped on a 20 km x 20 km basis. The critical load maps have also been used along with future sulphur deposition projections to map regions where critical loads are still exceeded by the year 2005. The MAGIC model has been used more selectively to explore the effect of timelags, land use and other factors in achieving target loads at different sites. The output of all methods is being compared, especially at Acid Waters Monitoring Network sites (Patrick *et al.* 1991).

MAPPING WATER CHEMISTRY

Strategy

Despite the abundance of detailed water chemistry data for many sites in key acidified regions of the UK, no data base of water chemistry for the whole of the UK suitable for critical load mapping was available before the start of the critical loads project in the UK. Indeed, it was thought that much existing information was inadequate to satisfy the specific requirements of

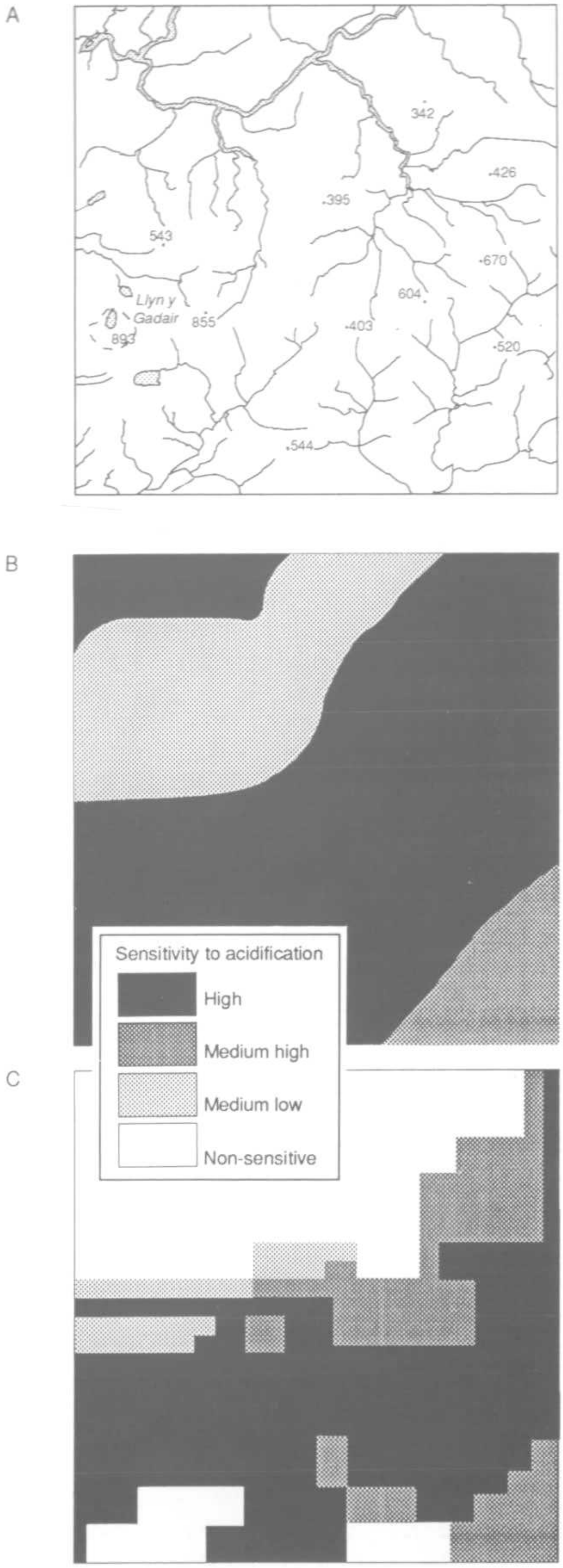

Figure 2. An example of site selection within a 10 x 10 km square (SH 71). The distribution of surface waters within the square is shown in A. The sensitivity of solid geology (B) (Edmunds & Kinniburgh 1986) is modified using soil and land use data to produce maps of freshwater sensitivity on a 1 km grid square basis (C) (ITE Monks Wood). Within the area of highest sensitivity, the highest-altitude standing waterbody is selected, in this case Llyn y Gadair (A)

critical load mapping. For this reason and to preserve uniformity of sample collection and analysis, a major programme of water chemistry sampling for the whole UK was organised. A compromise between spatial resolution and resource availability dictated a sampling grid of 10 km x 10 km in regions of high or medium sensitivity to acidification, and a grid of 20 km x 20 km in low- or no-sensitivity areas, requiring the collection and analysis of *ca* 1500 water samples. In addition, as only one sample per site could be analysed, standing waters were targetted in preference to running waters (because of the inherently more variable water chemistry of the latter). Where no suitable standing waters were available, low-order streams were used.

Site selection

An example of site selection for a single 10 km x 10 km grid square is shown in Figure 2. The criteria used for selecting individual waterbodies were as follows.

1. Catchment: high-sensitivity areas were defined using the geological sensitivity map of Edmunds and Kinniburgh (1986), the national (1:625 000) map of solid geology and, where available, detailed soil and land use maps
2. Altitude: the waterbody at the highest altitude was selected within the area of greatest catchment sensitivity
3. Size: a minimum size of 0.5 ha was specified and large waterbodies extending between grid squares were excluded.

Water sampling programme

The status of the water sampling programme as of June 1992 is shown in Figure 3. All sites predicted to be sensitive to acidification in mainland Scotland, northern England, Wales and the west country, and at least half the sensitive sites in Northern Ireland have been sampled. In addition, sites in non-sensitive areas within these regions have also been sampled. The type of site selected for sampling (including some sites which have been chosen but not yet sampled) is shown in Figure 4. The majority of sites are natural lakes, although regional exceptions are the Pennine hills of northern England, where artificial reservoirs predominate, and eastern Scotland, where streams have been sampled owing to the lack of standing water.

At each site two water samples were taken, one for major ion chemistry and one for total phosphorus. In addition, notes were taken on lake and catchment features and epilithic samples were collected for later diatom analysis. Some first-choice sites, although suitable on the map, proved inaccessible (eg permission for access was refused). In these cases, an alternative second-choice site was sampled.

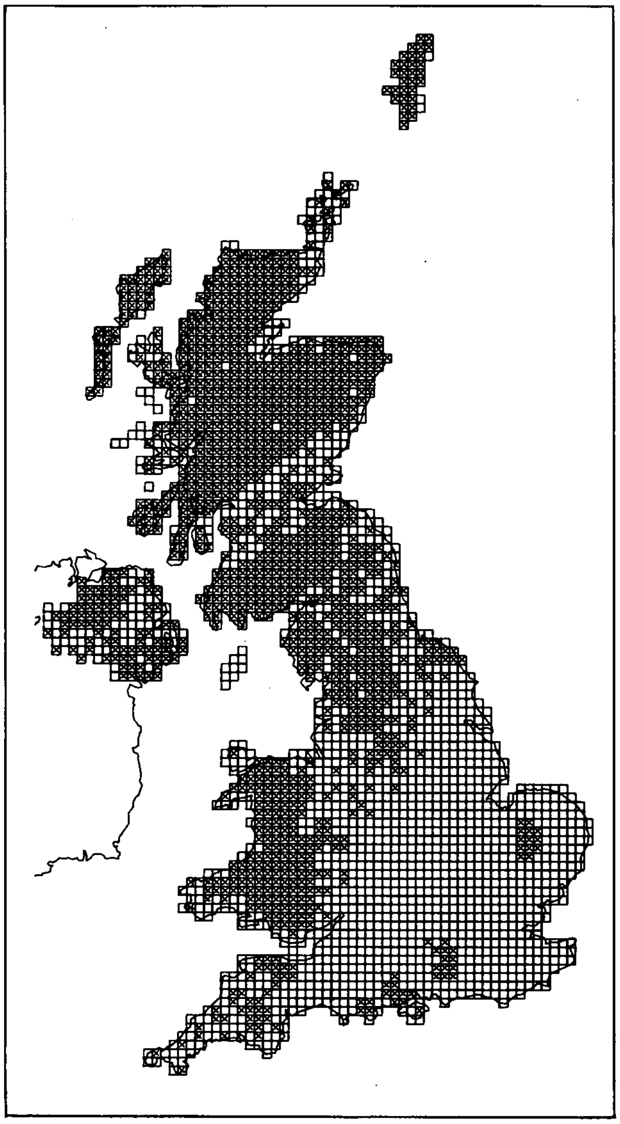

Figure 3. Sites sampled within 10 km x 10 km grid squares (as of 30.6.92)

Figure 5 shows the distribution of sites by choice. A full list of sites is given in Battarbee *et al.* (1992).

Sampling started in Scotland in May/June and September/October 1990. In 1991 sampling was carried out in May and September for northern England and Wales. This strategy to sample systematically region by region was altered in September 1991 when a new deadline of May 1992 was set for completion of the work by the Department of the Environment. Consequently, the remaining sensitive sites in England, Northern Ireland, Orkney and the Shetlands were given priority, and these were sampled in October/November 1991 and May/June 1992. Sites still to be sampled are in non-sensitive areas, and will be completed in September/October 1992.

Chemical analysis

Chemical analysis was carried out at the Freshwater Fisheries Laboratory of the Scottish Office Agriculture and Fisheries Department. On arrival, a 100 ml fraction was filtered through a 0.45 µm Durapore membrane filter for major anion and cation

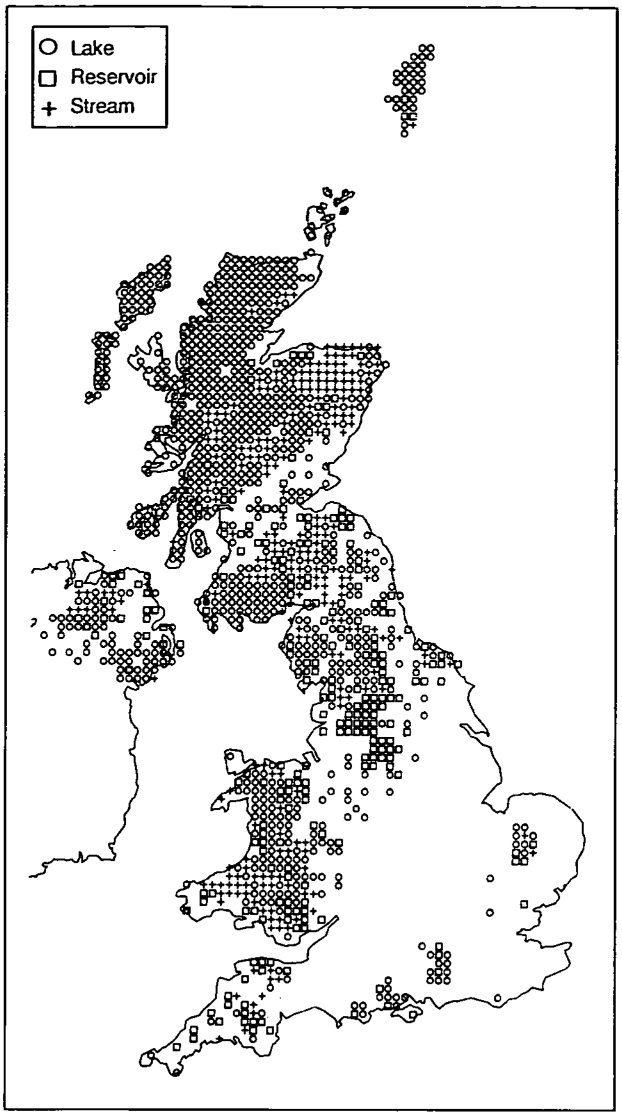

Figure 4. Site selection according to type (lake, reservoir, or headwater stream)

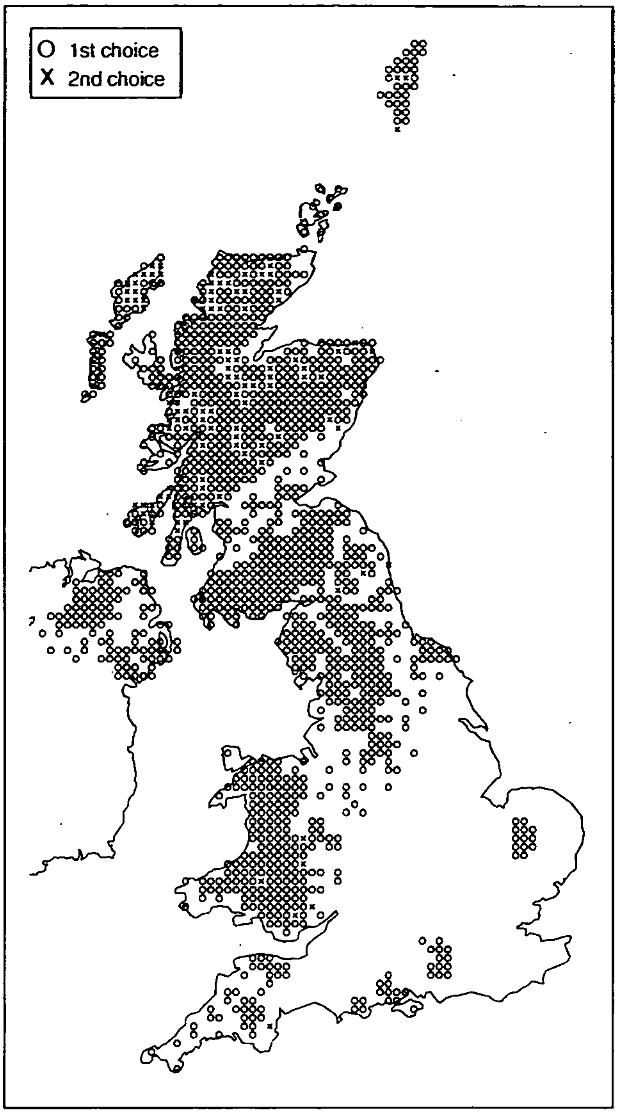

Figure 5. Sites sampled according to choice

analysis. Alkalinity and pH were determined as soon as possible on an unfiltered fraction. The methodology has been described previously (Harriman, Gillespie & Morrison 1990) and is the same as that used for the Acid Waters Monitoring Network (Patrick *et al.* 1991).

Part of the analytical procedure requires twice-yearly involvement in analytical quality control (AQC) checks to maintain control of analytical errors. Analytical results were entered on a relational data base (PARADOX) and transferred to University College London on disc. Critical loads for each 10 km x 10 km grid square were calculated on either LOTUS or QUATTRO PRO spreadsheets (Henriksen model) or using PARADOX (diatom model), and were produced as critical load maps by ITE Monks Wood.

DISCUSSION

The results of the national water quality mapping are presented elsewhere in this volume (Harriman & Christie, pp 103-108; Battarbee *et al.*, pp 99-102). Because of time and resource constraints, the data have not yet been fully evaluated, but in the next three years both the critical loads and critical load exceedance data will be validated by considering:

- problems with models associated with high sea-salt influences;
- within-square problems of spatial and temporal variability in water chemistry;
- differences in sensitivity between standing waters and headwater streams;
- problems of estimating sulphur deposition for individual sites with regard to altitude, land use and aspect.

Because of these problems and issues the present maps should be regarded as being provisional, especially for parts of the country where clearly anomalous values have been produced (eg the north-west of Scotland).

In addition, the maps should be used cautiously. Because they are based on the chemistry of only a single sample from one site in each grid square, and because each site was chosen to be the most sensitive site in the square, the maps do not show the critical load for the whole square, neither do they indicate how much of any square is exceeded, or the mean exceedance of the square. They indicate, in theory, the lowest critical load and the highest exceedance, respectively, reached by at least one waterbody in that square.

Consequently, because of these sampling and scale factors, the present maps are not directly useful for the management of individual waterbodies and their catchments (except, of course, for the 1500 or so catchments used in the study so far). Their function is to indicate regions where acidification has occurred for use by the UK Department of the Environment in the forthcoming renegotiations of the sulphur protocol, where they will be scaled up to the level of the European Monitoring and Evaluation Programme grid square (150 km x 150 km).

Although critical load values, where they have been accurately determined, should stay relatively constant through time, exceedance levels in the future will change because of other factors. Increases in nitrogen deposition, afforestation, and changing climate may all offset the beneficial effect of reduced sulphur deposition. A parallel commitment to improved modelling and to detailed monitoring at key sites until at least the year 2005 is needed to evaluate fully the relative importance of these factors.

ACKNOWLEDGEMENTS

The critical loads programme described here is funded by the UK Department of the Environment under contract PECD 7/12/48. Grateful thanks are due to all members of the Environmental Change Research Centre, UCL, and staff at the Freshwater Fisheries Laboratory, SOAFD, for assisting with field sampling, and to members of the Critical Loads Advisory Group (freshwaters subgroup) for many hours of useful discussion. We are also grateful to landowners for giving permission to sample.

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Setting critical loads for UK surface waters: the diatom model

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PRINCIPLES

In the UK, the emphasis placed on palaeolimnological research into lake acidification has allowed an approach to critical loads to be devised using diatom-based pH reconstruction. The stratigraphic analysis of diatom assemblages of dated lake sediment cores from low alkalinity lakes in areas of high sulphur deposition invariably shows clear evidence of recent acidification (Figure 1).

Because diatoms are amongst the most sensitive organisms to acid stress, it can be logically argued that this point of change in a lake sediment record represents the time at which the 'critical load' for that lake was exceeded. Detailed examination of dated cores from 53 lakes with varying sensitivity to acidification throughout the UK uplands (eg Anderson *et al.* 1986; Flower, Battarbee & Appleby 1987; Battarbee *et al.* 1988; Atkinson & Haworth 1990; Jones *et al.* 1993) shows variations in the degree of acidification ('critical load exceedance') at any site according to the base cation concentration of the water and the amount of S deposition received by the site. The relationship is so strong that a dose-response model can be proposed (Battarbee 1990) to predict the acidification status of UK freshwaters with confidence, and thereby to calculate critical loads.

Although the S deposition received at a site at the time of the initial acidification is not known (because it usually occurred in the 19th century), it can be derived empirically from the dose-response model. Comparison of contemporary water calcium and S deposition data for the 53 sites (Battarbee *et al.* 1992) shows that almost all acidified sites have Ca/S ratios of <70:1 (where Ca is in μeq l^{-1} and S in keq ha^{-1} yr^{-1}). The ratio 70:1 is then the 'critical ratio'; sites with lower values exceed the critical load, and the critical load for any site can be calculated from this ratio and a knowledge of pre-acidification calcium values (Ca_o) determined using the Henriksen F factor (Harriman & Christie, pp 103-108). This ratio requires statistical validation, and should be regarded as preliminary.

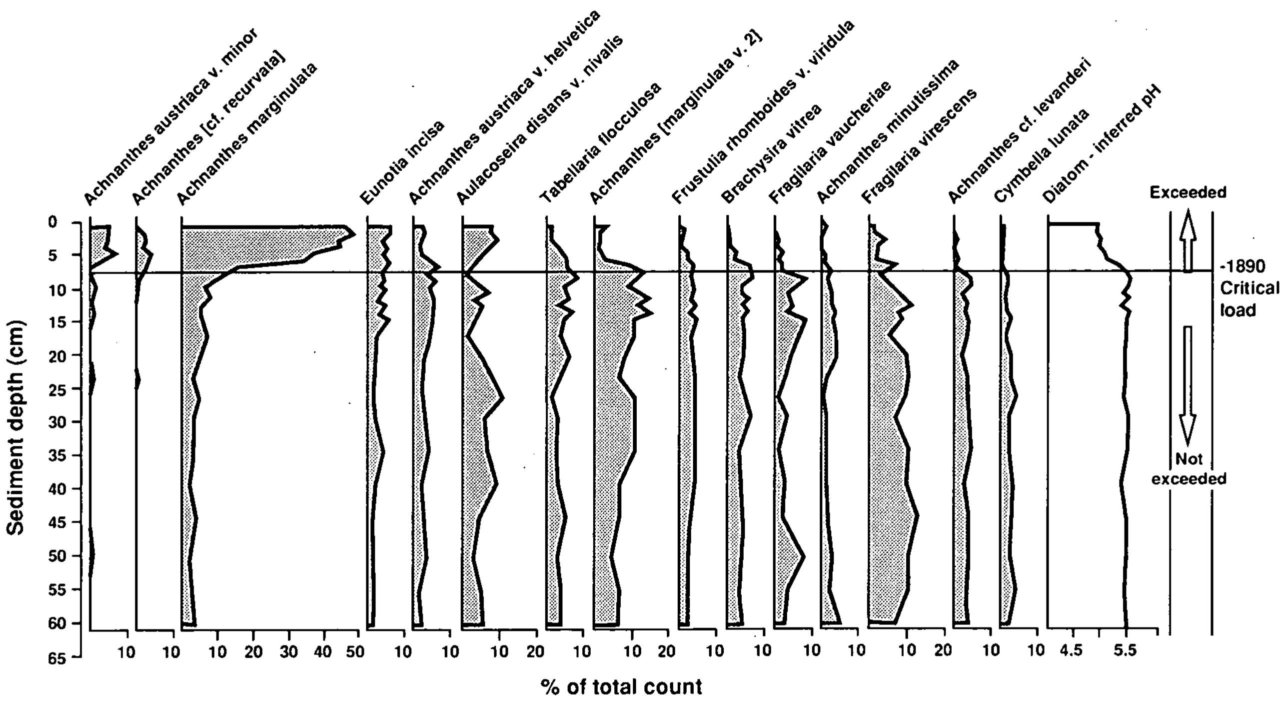

Figure 1. Diatom diagram from Lochnagar, Scotland, showing point of critical load exceedance in the late 19th century (source: Jones *et al.* 1993)

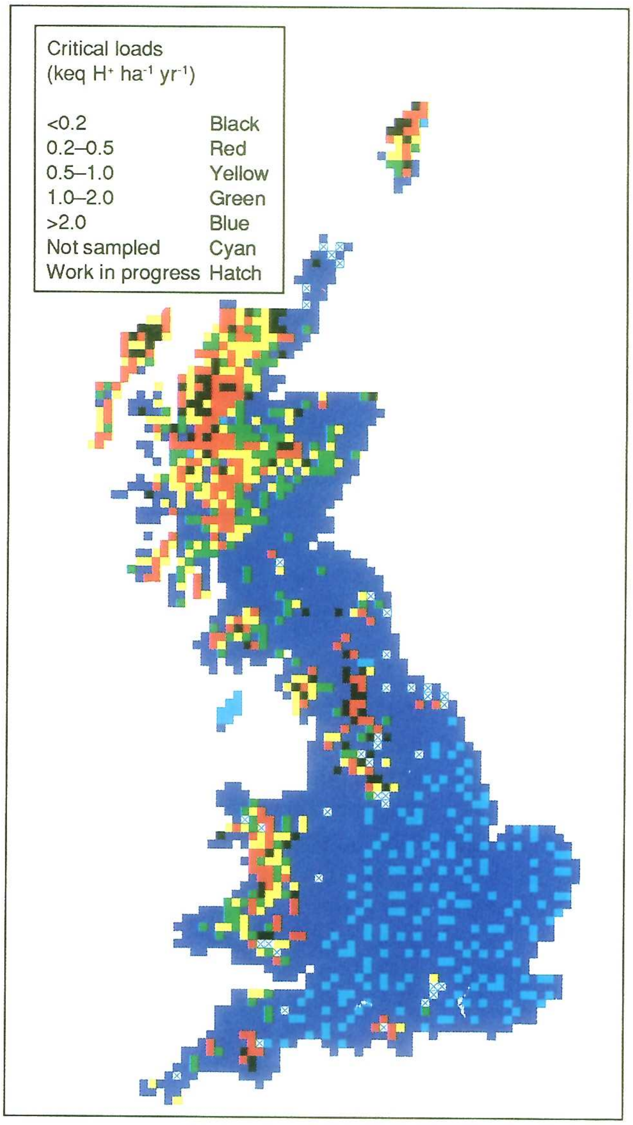

Figure 2. Map of critical loads for sulphur, expressed as acidity, for the UK, on a 10 km x 10 km grid (diatom model)

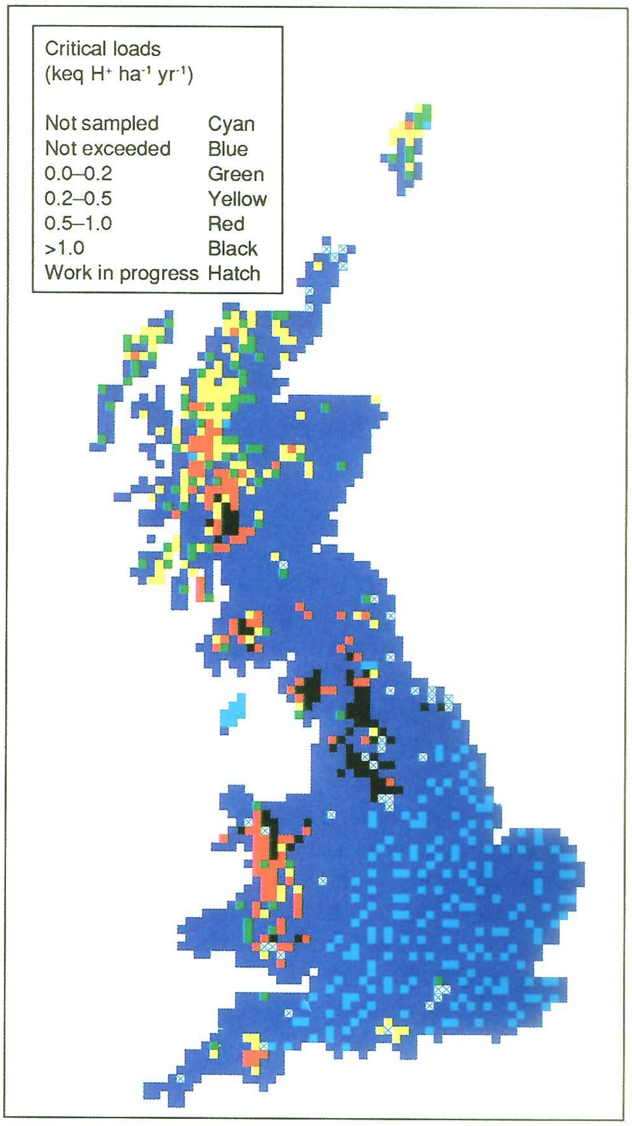

Figure 3. Map of critical load exceedance for sulphur, expressed as acidity, for the UK, on a 10 km x 10 km grid (diatom model)

METHODS

To map critical loads in the UK using the diatom model, water samples were obtained from selected sites in each 10 km x 10 km grid square (Kreiser, Patrick & Battarbee, pp 94-98). Chemical analysis of all ions was carried out at the Freshwater Fisheries Laboratory of the Scottish Office Agriculture and Fisheries Department, and the data, including calculated values for Ca_o, were transferred on disc to University College London. Subsequent data analysis was carried out using the data base PARADOX, and initial maps were made using MAPICS software (MAPICS 1985). Final critical load maps were produced by ITE Monks Wood, where critical load exceedance maps for the present day and for the year 2005 were printed using modelled S deposition based on data for 1986-88 and projected S deposition data for 2005, respectively.

RESULTS

Critical load and critical load exceedance maps for the UK using this approach are shown in Figures 2 and 3 respectively. The main areas of exceedance at present occur in expected areas, where acidification problems are already well known. These include the Cairngorms, the south-west Highlands, Galloway, Cumbria, the Pennines and Wales. However, exceedance is indicated in many other regions that are either not expected (eg north-west Scotland) or not previously studied (eg Dartmoor, New Forest). At this stage, there has been no detailed site-by-site assessment, and further work is required to assess the extent and seriousness of acidification problems in such areas.

In some cases the exceedance may be more apparent than real, related to uncertainties and problems with primary data. For the north-west of Scotland, for example, erroneous results may be caused by a combination of underestimation of the calcium values and an overestimation of S deposition. Calcium values in this windswept coastal region are unreliable because of the uncertainty of the sea-salt contribution to the base cation concentrations, and S deposition values may be unreliable because of errors in extrapolation from widely spaced monitoring stations and because of uncertainties in

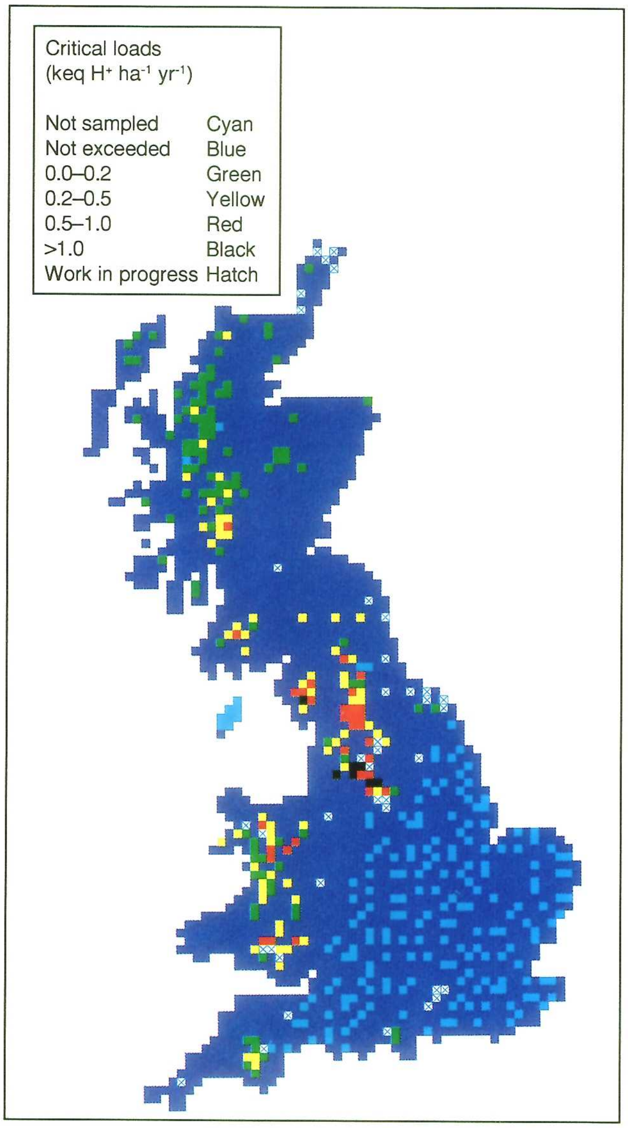

Figure 4. Map of projected critical load exceedance for sulphur, expressed as acidity, for the UK in the year 2005, on a 10 km x 10 km grid (diatom model)

adjusting for the effect of altitude. These problems affect all critical load models, and new research to evaluate these problems is urgently required.

Despite these concerns, it is clear that planned reductions in S emissions by the year 2005 will not be enough to decrease S deposition below the critical values for many sites. According to projections of future S deposition, areas where significant critical loads exceedances will remain include the Trossachs, Galloway, Cumbria, the Pennines and Wales (Figure 4). However, the number of exceedances is reduced and those that remain are generally of a lower class (cf Figure 3).

DISCUSSION

Critical loads calculated using the diatom model are complementary to the standard Henriksen steady-state water chemistry model adopted in the UN-ECE mapping programme across Europe and used also for the UK (Harriman & Christie, pp 103-108). For mapping purposes, the Henriksen model is constrained by the use of a preselected ANC value (often ANC = 0). This is inappropriate for setting the critical load for a site because the actual ANC at which the critical load is exceeded, according to the diatom model, varies from site to site. Setting ANC = 0 excludes exceedance sites with positive alkalinity, and setting a higher ANC could erroneously include naturally acid waters as exceedance sites. The diatom model is not linked to ANC in this way; it indicates the first point of change for any site, whatever ANC threshold is crossed. As such, the diatom model, in theory, generates the base critical load for the site.

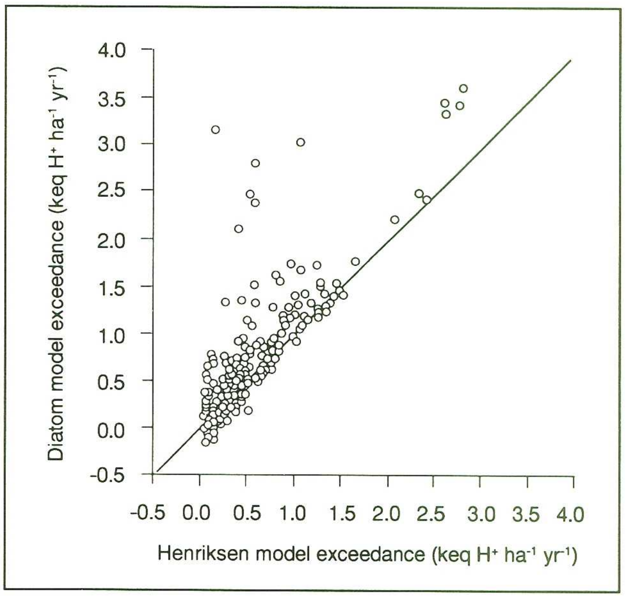

Figure 5. Comparison of critical load exceedances from the Henriksen and diatom models

Consequently, the diatom model is likely to show larger numbers of sites with low critical loads and with high exceedances than the Henriksen model when the ANC = 0 value is used. A comparison of the exceedance maps using the two models shows this likelihood to be true. In almost all cases, the diatom model generates exceedances either in the same or a higher class (Figure 5).

The strength of the Henriksen model is its ability to generate a critical load for any given ANC value and to relate this to *species* distribution, allowing maps to be produced for any selected species. For example, ANC = 0 is used to protect brown trout *(Salmo trutta)* populations. Protection for other species may require higher or lower ANC values. In contrast, the diatom model cannot specify critical loads for individual taxa in this way, although by indicating the base critical load for the *site* it can be used to prevent ANC values being set too high for any individual waterbody.

ACKNOWLEDGEMENTS

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Evaluation of the steady-state water chemistry method for surface waters

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CONCEPTS AND DEFINITIONS

At the Skokloster workshop (Nilsson & Grennfelt 1988), a suitable definition of critical acid deposition load to a specific ecosystem was agreed to be:

'the highest deposition of acidifying compounds that will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function'.

This generalised definition was later modified to describe important components of the ecosystem. Thus, for surface waters, the critical load is defined as:

'the highest deposition of acidifying compounds that will not lead to chemical changes which result in long-term harmful effect on aquatic biota'.

Using this definition, a link is made between acidic inputs, chemical changes in surface waters, and biological response. As will be seen later, these links are incorporated into the formulation used to calculate critical loads and exceedance values. Because of the time constraints imposed by the tight schedule set by the Task Force on Mapping of the United Nations Economic Commission for Europe (UN-ECE) and because of the logistics of the sampling programme (see Kreiser, Patrick & Battarbee, pp 94-98), only one sample was taken from each 10 km x 10 km square in the UK, and in non-sensitive regions sampling was limited to one from each 20 km x 20 km square. While these maps satisfied the requirements of UN-ECE for determining sulphur protocols for SO_2 emission reductions, their use for other purposes (eg as a guide for forest planting) may be inappropriate.

The steady-state method discussed in this paper is commonly referred to as the Henriksen Empirical Model (HEM), the development of which has previously been described (eg Henriksen 1984; Henriksen & Brakke 1988).

METHODOLOGY

Critical load calculation

Unlike dynamic models such as MAGIC, the underlying assumption of the steady-state water chemistry method is that inputs of atmospheric pollutants are in steady state with outputs. The present-day water quality is therefore used to calculate the pre-acidification base cation weathering rate. Any other factors (ie afforestation) which affect base cation concentrations are also incorporated into the present-day water quality data. No time component is specified in this method and, consequently, the rate of recovery of acidified waters cannot be calculated unless repeated sampling is employed until stable exceedance values are obtained. An important consequence of using this method is that the calculated pre-acidification weathering rate should remain relatively constant, irrespective of future changes in water quality resulting from changes in inputs or afforestation.

The basic formulation of critical loads of surface waters using HEM is:

$$CL_{acid} = (BC_o^* - ANC_{limit}) \,.\, Q - BC_d^* \,.\, R \qquad (1)$$

where CL_{acid} = critical load of acidity; where * indicates the non-marine component; BC_o = excess base cation concentration prior to acidification; ANC_{limit} = critical level appropriate to a target organism; Q = runoff; BC_d^* = non-marine base cation concentration in precipitation; and R = rainfall.

No data are available for BC_o, so it is calculated from the equation:

$$BC_o^* = BC_t^* - F\,[SO_{4t}^* = SO_{4o}^*] \qquad (2)$$

where BC_t^* = present-day excess base cation concentration; SO_{4t}^* = present-day excess sulphate concentration; and SO_{4o}^* = pre-acidification excess sulphate concentration.

The F value in this formula determines the proportion of extra base cations leached by increased sulphate inputs. This F factor can be calculated on the basis of an empirical equation:

$$F = \text{Sine}\,[\,90\,.\,\frac{(BC_t^*)}{S}\,] \qquad (3)$$

where S is the base cation concentration at which it is assumed that F = 1.0. S can vary between 200 and 400 $\mu eq\ l^{-1}$, depending on the location.

Thus, most systems with base cation levels >200 µeq

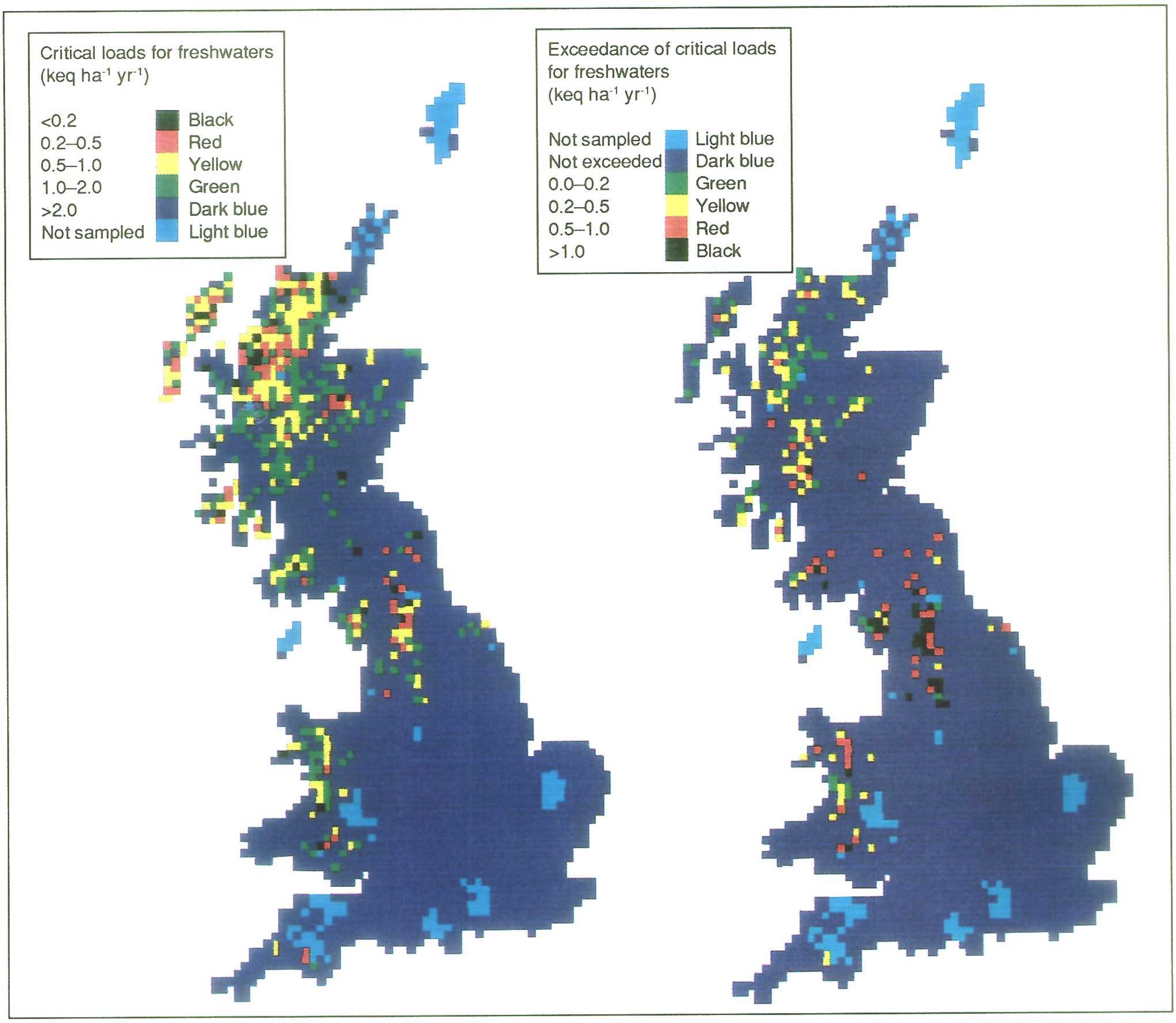

Figure 1. Critical load and exceedance maps for the UK, based on the steady-state water chemistry (Henriksen) model

l^{-1} would have an F factor close to unity. The only remaining unknown factor is the pre-acidification sulphate level (SO_{4o}^*). Data from the most sensitive pristine sites in Scandinavia indicate SO_4^* levels around 20 µeq l^{-1}, and these values increase with decreasing sensitivity. A suggested equation is:

$$SO_{4o}^* = 15 + 0.16 . BC_t^* \qquad (4)$$

However, in some areas of western USA, excess SO_4 levels are close to 10 µeq l^{-1}; therefore, the intercept value may vary within a 5-15 µeq l^{-1} range, depending on location.

A further step is required to complete the calculation of critical loads (and exceedance estimates). An appropriate transfer function must be established to link the critical ANC limit in the equation to an appropriate biological indicator. This requires a certain knowledge of factors which influence the survival of selected biota, but in many cases such information is available (eg for salmonid fish).

In calculating critical loads from the current UK data set, the following criteria were utilised (see Sverdrup, De Vries & Henriksen 1990).

- A critical chemical value of the acid neutralising capacity was equal to zero.
- A runoff value was calculated as rainfall/1.15.
- An excess base cation value in precipitation was 8 µeq l^{-1}.
- Any water with a base cation value in precipitation of 8 µeq l^{-1} was given a Henriksen F value of 1.0 (ie S = 400).
- SO_{4o} was always >15, even for negligible base cation levels.
- Modified (eg for elevation, vegetation) S inputs were based on 20 km^2 grids; therefore the same input value was allocated to the four equivalent 10 km square blocks.
- In areas with high sea-salt inputs, the use of Cl (or Na) to calculate excess SO_4 (and excess Ca, Mg, K and Na) can occasionally produce zero or small negative values. If the Cl calculation gives negative values, then the Na calculation is used.

If the Na calculation is negative, then a zero value is recorded.

It should be noted that SO_{4t} may be lower than SO_{4o} in high sea-salt areas. When these two values are subtracted and multiplied by F, a negative value is obtained, thus increasing the BC_o value rather than reducing it.

Exceedance of critical loads

The critical load values calculated by the HEM method are for total acidity. If exceedance values are required for specific components of the total acidifying inputs (ie SO_4), then the calculation simplifies to:

$$SO_{4Ex} = (BC_o^* - ANC_{limit}) \,.\, Q - BC_d^* \,.\, R - SO_{4\ input}^* \qquad (5)$$

where $SO_{4\ input}^*$ is an estimate of the sulphate load, including both wet and dry deposition.

Negative values derived from this equation indicate that the critical load is exceeded by sulphur inputs alone. The current critical load and exceedance maps (1992) based on HEM and modified input data are shown in Figure 1. These can be compared with those produced from the diatom model which show a greater number of exceeded sites because a variable ANC_{limit} value was used (Battarbee *et al.*, pp 99-102).

EVALUATION OF HEM METHOD

Uncertainties with critical load estimates

During the early phase of the critical load mapping exercise, it was recognised that the following key questions should be addressed before the selection criteria and calculated critical loads could be validated.

- Which components of the Henriksen equation are likely to introduce the greatest uncertainties in the critical load calculation?
- Will critical loads vary from class to class on a temporal basis?
- How certain are we that the selected site will adequately represent the most sensitive square in the catchment?

In a preliminary study of these questions, the temporal data from the Acid Waters Monitoring Network (AWMN) were utilised, along with selected data from the critical loads survey.

The first question was addressed by systematically checking the responsiveness of critical loads to each component of the steady-state equation, in particular the estimate of the pre-acidification weathering BC_o^*. The first uncertainty arises with the calculation of the present-day non-marine base cation level BC_t^*. For sites with chloride levels >500 $\mu eq\ l^{-1}$, to which the standard sea-salt corrections were applied, the excess sodium and magnesium values were occasionally negative, resulting in small or negative critical loads. When the negative values were corrected to zero, a more realistic range of critical loads was produced. A detailed appraisal of the effect of high sea-salt deposition on critical load calculations is under way to examine the efficacy of sea-salt correction factors.

The second component which influences the BC_o^* estimate is the F factor, which determines the increase in base cation leaching caused by increased pollutant inputs. It is related to the measured base cation value BC_t^* by the equation:

$$F = \text{Sine}\ [\ 90\ .\ \frac{(BC_t^*)}{S}\ ]$$

where S is the perceived base cation value associated with an F factor of 1. The preferred value of S in the critical loads equation is 400 $\mu eq\ l^{-1}$, but values of 200 and 100 $\mu eq\ l^{-1}$ have been suggested for certain regions.

While some uncertainty may exist over the true value of the F factor, the effect of F on the BC_o^* calculation is relatively small for the more sensitive sites with low base cation levels. For the sensitive AWMN sites, there was no significant movement from one critical load class to another as a result of varying the S value between 100 and 400 $\mu eq\ l^{-1}$. While other uncertainties exist with the other components of the equation (eg SO_{4o}^*), the above factors appear to exert the greatest influence on the critical load values.

The second question addresses the temporal component of water quality variables and, again, this can be investigated on a preliminary basis using the monthly data from the AWMN sites. Intuitively, we would expect that critical loads would be highest for water samples at the highest pH, while the best relationships between biological status and critical loads would be for water samples at the lowest pH. For most small upland lakes the seasonal pH variation is relatively small, and consequently the seasonal variation in critical loads is also small. The variation in critical loads calculated from individual samples is likely to be greater for upland streams than upland lakes, and to be more strongly correlated with pH (or ANC). An example of pH/critical load relationships for a small upland lake and stream in the monitoring network is shown in Figure 2, indicating that the choice of upland lakes to reflect steady-state chemistry is probably correct. Clearly, it would be unreasonable to expect a stable relationship between mean critical loads and those calculated from the chemistry of samples at the maximum and minimum pH; the difference between mean and minimum critical loads for most of the network sites is quite small compared with the differences between mean and maximum critical load values (Figure 3).

The third question about the representativeness of the sampling site has yet to be investigated in detail as it requires intensive sampling in a random selection of 10 km grid squares. For squares with

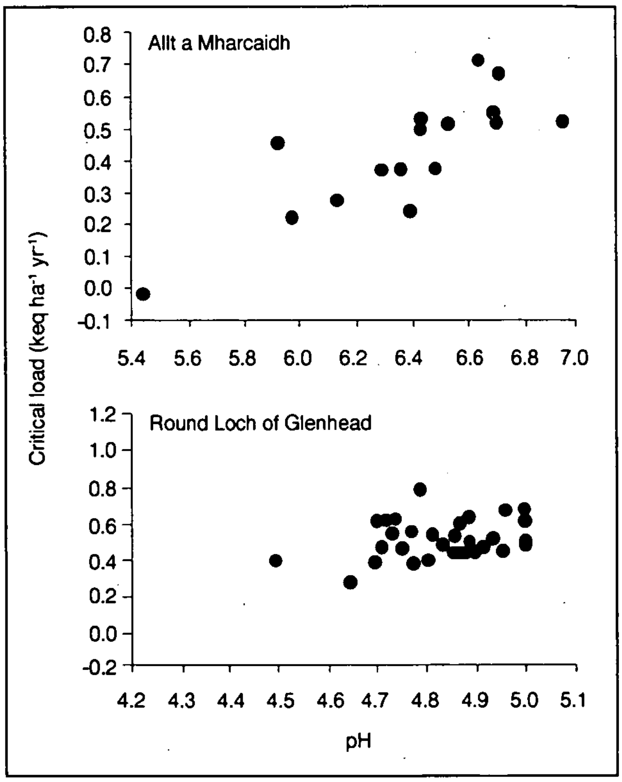

Figure 2. Variation in the critical load/pH relationship for one upland lake and stream selected from the Acid Waters Monitoring Network

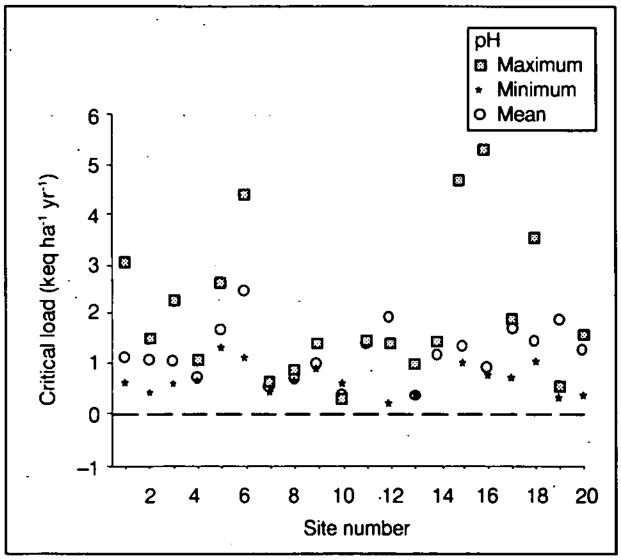

Figure 3. Critical loads for the Acid Waters Monitoring Network sites calculated from the chemistry at maximum, minimum and mean pH

available data, the selected sites for the mapping exercise appear to represent the most sensitive waters in the 10 km square, but a more representative set of squares should be sampled before the uncertainties can be quantified.

Uncertainties with exceedance estimates

The major uncertainty with exceedance calculations is that the input data, although corrected for elevation and vegetation characteristics in the square, are available only for 20 km grid squares. Thus, the same input data are allocated to the four 10 km grid squares associated with each 20 km grid square. This may result in underestimates of inputs to upland areas and overestimates for lowland areas. Validation of exceedance squares is not a straightforward matter because of the uncertainties associated with critical loads and input estimates, but, if the present-day water quality exhibits a negative ANC and is also exceeded using HEM, then the estimates can be treated with a greater degree of confidence.

ANC_{limit}/BIOLOGICAL INDICATORS

In the HEM formulation, the critical base cation leaching for a given indicator organism is given by:

$$BC_{leach,crit} = (BC_o^{*} - ANC_{limit}) \cdot Q \qquad (6)$$

where ANC_{limit} represents a tolerance function for any selected indicator organism. In the present calculations, a value of zero is used where:

$$ANC = HCO_3 + A^- - H^+ - \Sigma Al^{n+} \qquad (7)$$

or

$H^+ + \Sigma Al^{n+} = HCO_3^- + A^-$ when ANC = 0

where A^- = the concentration of organic anions and ΣAl^{n+} = the sum of a positively charged aluminium species.

From the above relationship, it can be seen that the ANC_{limit} can be calculated if toxic levels of H^+ and Al^{n+} are known for the selected organism. For many salmonid fish, a pH around 5.5 and Al^{n+} level of about 30 µg l^{-1} are known to reduce survival (Harriman, Gillespie & Morrison 1990). When these values are incorporated into equation 7, an ANC value close to zero is obtained for clearwater lakes. As A^- increases, so does the ANC value for the same pH and Al^{n+} criteria. Therefore, if the pH and Al^{n+} criteria are similar over a range of A^- levels, then the ANC_{limit} may be a negative value (higher critical load) at high A^- levels.

Any organisms responding to higher pH levels (5.5-6.0), such as certain invertebrate and diatom species, will have a correspondingly higher ANC_{limit} and consequently the critical load of the stream or lake will be lower. Survival of aquatic organisms may not depend on mean pH and Al levels but may be determined by the frequency of acid episodes during the most sensitive development phase. This may not be a problem in lakes, where chemical changes are attenuated, but in streams a pH shift of 2 units is not uncommon. Using pH/flow data for any selected stream, it is possible to determine the association between mean pH and frequency of episodes below any selected critical pH level. Thus, a functional relationship between ANC_{limit}, mean pH and critical pH and Al levels can be determined.

CRITICAL LOADS AND NITROGEN INPUTS

The present exceedance maps (Figure 1) are derived from sulphate inputs only, while the calculated critical load values reflect the ability to withstand *all* forms of acidifying inputs. Therefore, following the agreement on sulphur emissions, the UN-ECE will next consider a protocol for nitrogen - the second major acidifying input.

Unlike sulphur, any uptake and release of nitrogen in terrestrial and aquatic ecosystems is predominantly biologically mediated, and many of the reactions can either consume or produce acidity. At a recent workshop in Lokeberg, Sweden, provisional recommendations were made to incorporate nitrogen processes in critical load calculations. In the context of surface waters, it was accepted that increased nitrate leaching was the key signal of net acidification in any catchment. In pristine regions, nitrate concentrations are invariably <5 µeq l^{-1} and often at the limit of detection (<1 µeq l^{-1}), while impacted regions exhibit much higher nitrate concentrations in runoff. Comparisons of input-output nitrogen budgets show considerable variability in nitrogen ($NO_3 + NH_4$) retention in catchments, ranging from virtually 100% retention to a two-fold greater export than input (Dillon & Molot 1989; Henriksen 1988). Catchments exhibiting high nitrate leaching are frequently located in areas receiving high inputs of nitrogen and sulphur. Catchments exhibiting high nitrogen retention values are located in both high and low N deposition areas.

On the basis of the nitrate leaching criteria, the critical load exceedance equation 5 can now be modified to:

$$Ex_{acid} = (BC_o^* - ANC_{limit}) \,.\, Q - SO_{4input} - NO_{3leach} \qquad (8)$$

The major problem with this formulation is that it only expresses the *present-day* acidifying potential of nitrogen inputs. It must, therefore, be considered as a steady-state model with an implicit assumption that uptake and removal of nitrogen remain constant during the vegetation cycle.

In practice, many nitrogen removal processes are rate-limited (eg denitrification and in-lake retention), being proportional to nitrogen inputs. Consequently, the nitrate leaching term in equation 8 can be substituted by an input and removal component, ie:

$$NO_{3leach} = N_{dep} - N_{upt} + N_{tr} \qquad (9)$$

where N_{upt} = N uptake in forests which are subsequently harvested; N_{tr} = total N retained in catchment; and

$$N_{tr} = N_{den} + H_{hum} + N_{lr} \qquad (10)$$

The two dynamic components in equation 10 are denitrification (N_{den}) and in-lake retention (N_{lr}), while the greatest uncertainty rests with the estimate of long-term changes in the accumulation rate of nitrogen in soil humus (N_{hum}). Even small changes in the mineralisation of this large nitrogen pool may dramatically affect nitrate leaching rates (see Kamari *et al.* 1992).

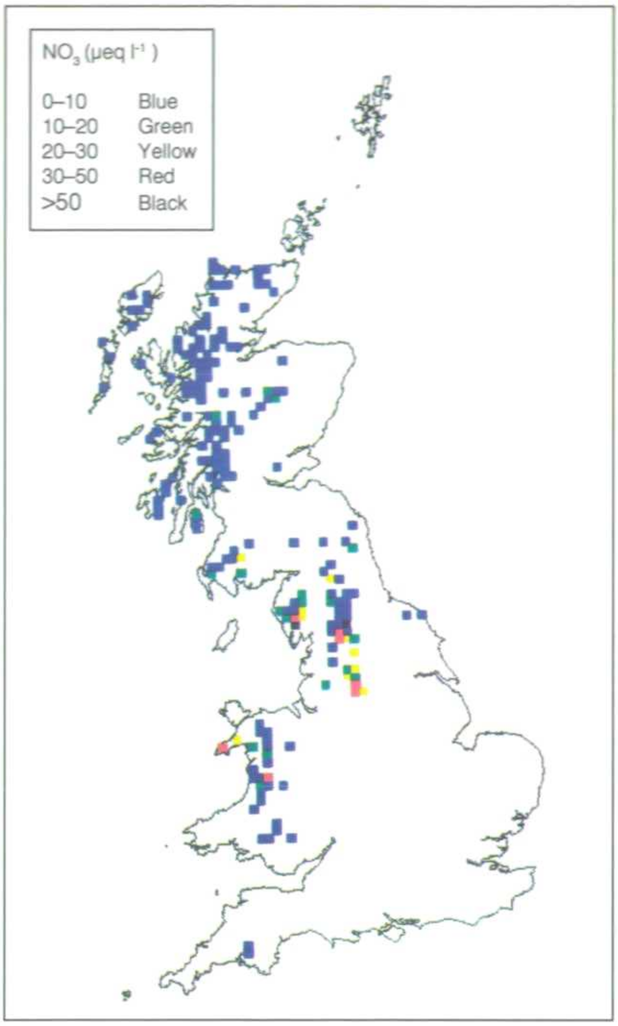

Figure 4. Nitrate concentrations at UK sites where critical load values are exceeded

Future development of critical load models for nitrogen will not only require a more dynamic approach, so that the future acidifying potential of an increase in N inputs can be calculated, but must also incorporate short-term changes in H_{hum} and NO_{3leach}. While changes in N_{hum} and NO_{3leach} may be small during natural forest cycles, they may be significant in managed forests, particularly at harvesting. Ironically, the NO_3 leaching term could also change as an indirect effect of sulphur inputs, eg where forest dieback results in a reduction in nitrogen uptake.

At the present time, it is possible to use the modified Henriksen steady-state model (equation 8) to calculate the present-day exceedance of total acidity. This calculation would increase the present number of exceeded 10 km squares (197) by a small but significant amount. The areas where nitrate leaching would contribute most to exceedance values are found in Cumbria, the Pennines, and parts of Wales. In Scotland, nitrate levels are generally low in the most sensitive areas and contribute little to exceedance values (Figure 4).

CONCLUSIONS

The steady-state water chemistry (Henriksen) model provides a simple and practical method of estimating critical loads for freshwaters. It can be applied to both running and standing waters over a full range of hydrological conditions. Although not required for this particular exercise, it could be used to assess the effects of land use changes where base cation leaching or pollutant inputs are likely to change (ie afforestation).

The advantage of this technique compared with soil critical loads is that the values generated reflect the conditions prior to the onset of acidic deposition. Consequently, any emission reductions are calculated on the basis of a return to pristine conditions rather than the present status, as in the case of soils.

Perhaps the greatest advantage of this method is the incorporation of a chemical limit term which links critical loads to a biological response. Providing tolerance criteria are known, any biological indicator can be used depending on local or national requirements.

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Comparison of empirical and dynamic model approaches for surface waters

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INTRODUCTION

The purpose of determining critical loads is to set goals for a future deposition rate of acidifying compounds such that the environment is protected. The definition of the critical load for freshwater is: 'the highest load that will not lead in the long term (within 50 years) to harmful effects on biological systems' (Nilsson & Grennfelt 1988). Critical loads for water are defined, therefore, with the purpose of protecting a chosen sensitive receptor; for example, water acidification can lead to the decline or disappearance of natural fish populations. Commonly used criteria to protect aquatic organisms are that the runoff water should have positive alkalinity and a concentration of labile inorganic aluminium less than 50 $\mu g\ l^{-1}$ (Nilsson & Grennfelt 1988; Henriksen & Brakke 1988).

Once the protective criteria have been set and the timescale for these criteria to be met has been decided, the concept of the critical load for a catchment can then be regarded as a fixed value, altering only with major changes in chemical processes occurring within a catchment, eg brought on by afforestation. It represents a threshold specific to that catchment, below which no harmful effects should occur. Critical loads will differ for a given area as well as from site to site, depending upon the inherent sensitivity of the natural environment. Two methods have been advanced for calculating critical loads for freshwaters: steady-state water chemistry and dynamic modelling (Sverdrup, De Vries & Henriksen 1990). Dynamic models ('level 2' approach to critical load determination), however, are difficult to parameterise and calibrate for a site, requiring substantial data input, but perform an essential role within the critical loads exercise. The empirical ('level 1') approach assumes a steady state with respect to inputs to a catchment from the atmosphere, and take no account of timelags in response or of changes in land use; it therefore does not describe the time dependence of critical loads or assess the influence of land use. Only dynamic modelling can account for changes over time, such as the depletion of element pools in soils (Henriksen, Lien & Traaen 1990). This paper compares the two approaches, explores the impact of afforestation on critical load calculation, and assesses the inter-relationships between sulphur and nitrogen deposition in estimating critical loads.

THE HENRIKSEN EMPIRICAL MODEL

This static model for surface waters is based upon the empirical link between present-day water and precipitation chemistry identified by Henriksen (1980), based on a regional study of lakes in Norway. The model is static in that it specifies the water chemistry resulting from a given change in deposition without specifying the time at which this new water chemistry will exist. Information as to the length of time required to achieve steady state following a change in acid deposition can be gained, but only through further experimentation. Conversely, the model cannot calculate a required deposition loading to achieve a specified water chemistry if a time limit is set. The empirical model takes no account of any future changes in land use and assumes that soils will de-acidify at the same rate at which they acidified. This soil buffering ability is represented in the model by an empirical factor relating the ratio of Ca and Mg to SO_4^{2-} ions in solution.

THE DYNAMIC MAGIC MODEL

MAGIC (Model for Acidification of Groundwater In Catchments) is an intermediate-complexity process-oriented model for constructing acidification history and predicting future acidification over time periods of decades to centuries (Cosby *et al.* 1985a, b). MAGIC makes use of lumped parameters on a catchment scale and focuses on chemical changes in the soil caused by atmospheric deposition, vegetation, and leaching to runoff. The processes in MAGIC include atmospheric deposition, sulphate adsorption, cation exchange, CO_2 dissolution, precipitation and dissolution of aluminum, chemical weathering, uptake and release of cations by vegetation, and export in runoff.

MAGIC has been used extensively in a variety of applications at sites in both North America and Europe. Application of MAGIC to the whole-catchment experimental manipulations of the RAIN project shows that this intermediate-complexity lumped model predicts the response of water and

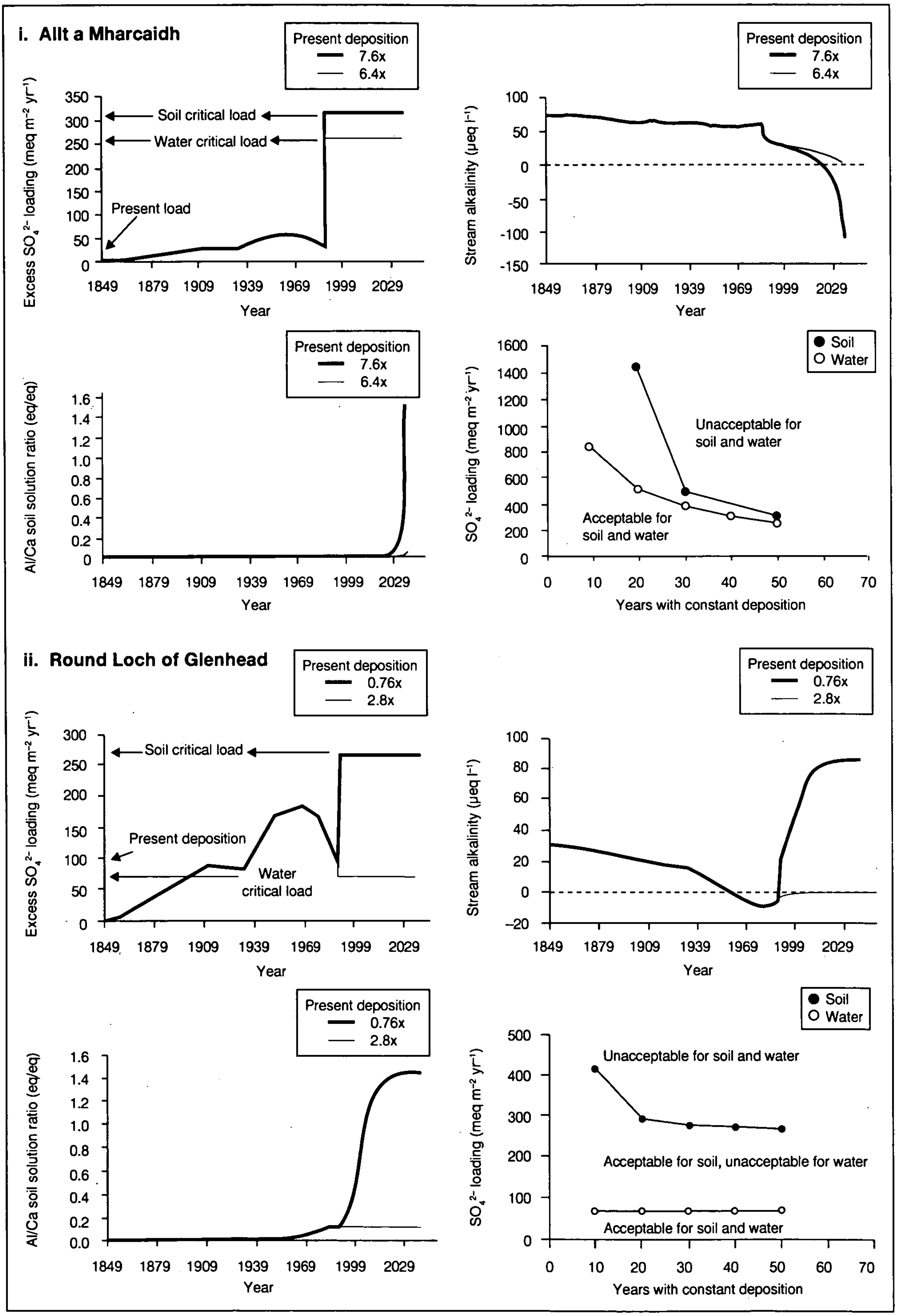

Figure 1. The historical sulphate deposition, stream alkalinity and soil water Al/Ca response at (i) Allt a Mharcaidh and (ii) Round Loch of Glenhead. Predictions of future response assume deposition held constant at the relevant sulphur loading into the future. A lower sulphur load is necessary if the critical chemistry is to be achieved over a shorter timescale

soil acidification to large and rapid changes in acid deposition (Wright *et al.* 1990). These results reinforce other evaluations of MAGIC, such as comparison with palaeolimnological reconstructions of lake acidification (Jenkins *et al.* 1990). The major processes by which forests influence surface water and soil acidification are also represented in MAGIC (Jenkins *et al.* 1990). In addition, several of the assumptions in MAGIC have been tested experimentally (Grieve 1989). Together, these applications indicate that MAGIC provides a robust tool for predicting future soil and water acidification following changes in acid deposition. It is one of several dynamic models included in the handbook on mapping critical loads, produced by the United Nations Economic Commission for Europe (Henriksen *et al.* 1989).

To calculate critical loads, the model is calibrated to a catchment using available soil and water chemistry and land use data. The model is then used in predictive mode to determine the sulphur deposition required to achieve some predefined critical chemistry in soil or surface water at some predefined time in the future. This critical chemistry is conventionally taken as a surface water alkalinity of zero (to protect brown trout *(Salmo trutta)*) and soil solution Al/Ca ratio (eq/eq) of 1.5 to protect forests, although, in practice, a critical load can be determined on any chemical parameter in the soil/ water system. The timescale is taken as 50 years as this is the most appropriate timescale over which deposition reductions and critical chemistry should be achievable. The level of deposition required is assumed to be reached immediately and held constant at that level for 50 years. MAGIC is then run repeatedly with different levels of deposition until the critical chemistry is achieved. This deposition is assumed to be the critical load for sulphur, assuming that the loading and retention of nitrogen compounds are not changed from present-day conditions.

These concepts are illustrated in Figure 1. In Figure 1(i) (Allt a Mharcaidh), current and past sulphur deposition is not sufficient to depress stream alkalinity below zero or the Al/Ca ratio above 1.5, because the critical load is not currently exceeded at this site, and so the sulphur deposition is increased in the model to achieve alkalinity zero and thereby quantify the critical load. Figure 1(ii) (Round Loch of Glenhead) shows a site where present deposition exceeds the critical load for water and so must be reduced to achieve zero alkalinity, although the soil is not as sensitive.

COMPARISON OF TECHNIQUES

The empirical and MAGIC techniques for calculating critical loads generally compare favourably (Figure 2), and in most cases produce the same critical load category. There are, however, differences between the two methods caused by differences in the dynamic and equilibrium assumptions utilised (Figure 2). Across a wide range of critical loads, from sensitive to insensitive, differences mainly occur at the least sensitive sites (ie high critical loads), where the MAGIC critical load is generally higher than the Henriksen empirical critical load, whilst at the most sensitive end of the range (ie low critical loads) the opposite is true. The less sensitive sites are characterised by well-buffered chemistry with high pH and alkalinity, influenced by soils which have retained a high acid buffering capacity. To deplete this soil store of base cations will either take many years of acid deposition at a relatively low sulphur loading, ie as described by the equilibrium concept assumed in the empirical critical load, or 50 years of a higher sulphur loading, as described by the dynamic MAGIC model. Sensitive sites are characterised by low pH and low, or in many cases negative, alkalinity and have soils with low base saturation and a large adsorbed sulphate pool. At these sites, the MAGIC critical load is lower than the empirical critical load, because the empirical approach assumes equilibrium conditions over an indefinite timescale whereas the dynamic modelling approach assumes a finite timescale. The MAGIC critical load is lower because these systems require longer than 50 years to recover, and the base cation store in the catchment soils which have been depleted by many years of acidic deposition and forest growth must be replenished by weathering inputs before recovery can occur.

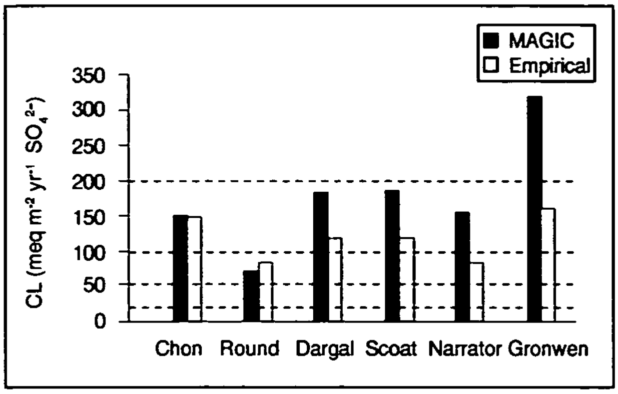

Figure 2. MAGIC and empirically calculated critical loads are in general agreement as regards critical load class

The question of uncertainty in critical load calculation is difficult to address as much of the variance in surface waters will be due to the changing flow regime, and in particular the chemical changes associated with high flows. It must be remembered that the MAGIC model simulates only mean annual chemistry and takes no account of these flow/ chemistry relationships. Uncertainty can be introduced into the model simulations, however, by incorporating known measurement errors and spatial variability in the parameters within the catchment at the calibration stage (Jenkins & Cosby 1989). In this way, uncertainty bands for the model simulations can be presented as maximum and minimum values for output variables, including target loads (Figure 3). A range of critical loads can also be calculated

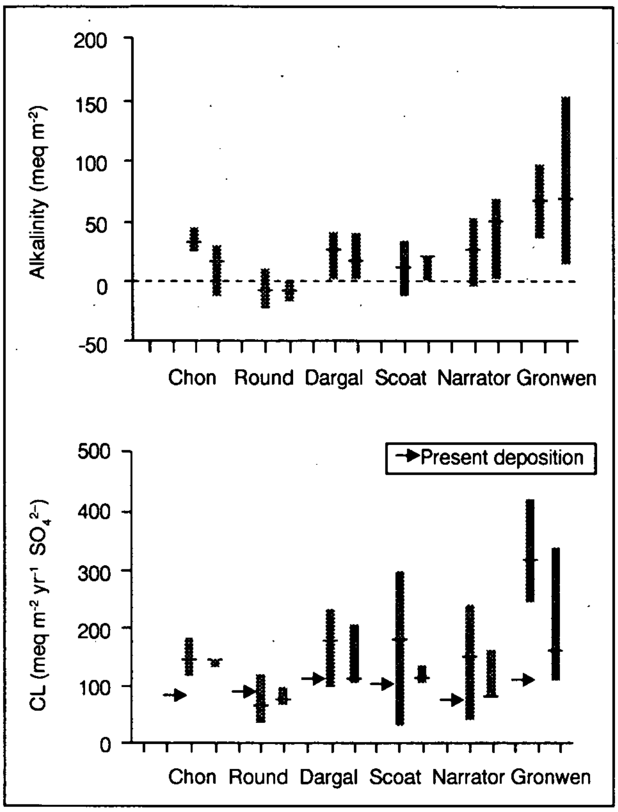

Figure 3. Comparison of the uncertainty in model simulations of stream or lake alkalinity (top) or critical loads (bottom). In each case, the vertical bars represent the range of observations, or of model simulations derived by varying the input parameters across a likely range of error, and the horizontal bar is the mean. For each site, two values are shown: MAGIC simulations on the left in each case; observed (top) or empirical model simulations (bottom)

empirically for samples collected across a wide range of flows. The ranges calculated from the two techniques demonstrate a good match (Figure 3), although the representations of uncertainty have little statistical significance, and further work is necessary, in particular to assess the importance of episodes to biological response and to relate the mean chemistry to extreme events.

The empirical approach to critical load calculation, based on the long-term weathering rate at a site, is time-independent, ie steady-state conditions are assumed, so that the timescale over which the new equilibrium at the critical chemistry will be achieved is irrelevant in the calculation. It is also assumed, perhaps more crucially, that the site is capable of recovering to the target chemistry level and that, prior to the onset of acidification, the water chemistry was equivalent to, or less acidic than, the target chemistry. Furthermore, the influence of the catchment soils, in particular their ability to adsorb and desorb ions through time, is only implicitly included in the empirical model, which assumes that the soil acidification process is completely reversible in the very long, or equilibrium, timescale. Soil chemical processes may mitigate or delay acidification through sulphate adsorption and base cation exchange, or extend the acidification effect after acidic deposition is reduced through soil recovery (adsorbing base cations and desorbing sulphate). This time-dependent effect can be identified using MAGIC, and Figure 4 (also illustrated in Figure 1) indicates that the target year by which the critical chemistry is to be reached produces variation in the calculated critical load.

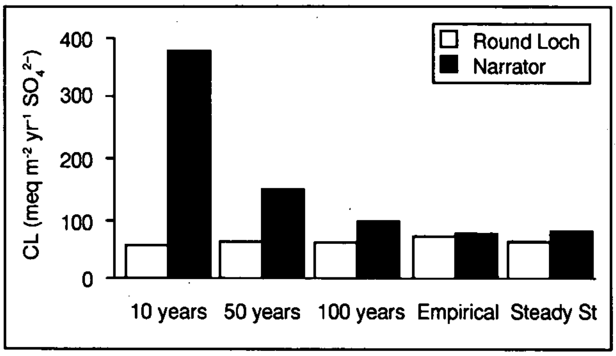

Figure 4. The time dependency of the critical load calculation using MAGIC. The longer the timescale over which the critical chemistry is to be achieved, the lower the critical load required to protect sites currently in exceedance, such as Round Loch. Where a site has to acidify in order to reach the critical load, as at Narrator, the opposite is true

THE INFLUENCE OF AFFORESTATION

The role of trees in the critical loads concept requires careful consideration. As well as being sensitive receptors for which critical loads need to be determined, trees play a crucial role in the soil and water acidification process by the uptake of base cations for growth, changing hydrological behaviour, and by scavenging pollutants from the atmosphere thereby increasing the deposition. Figure 5 shows critical loads calculated for the forested Loch Chon catchment in central Scotland using the MAGIC model (see Jenkins *et al.* 1990 for details). Three future afforestation scenarios are utilised in the critical load calculations:

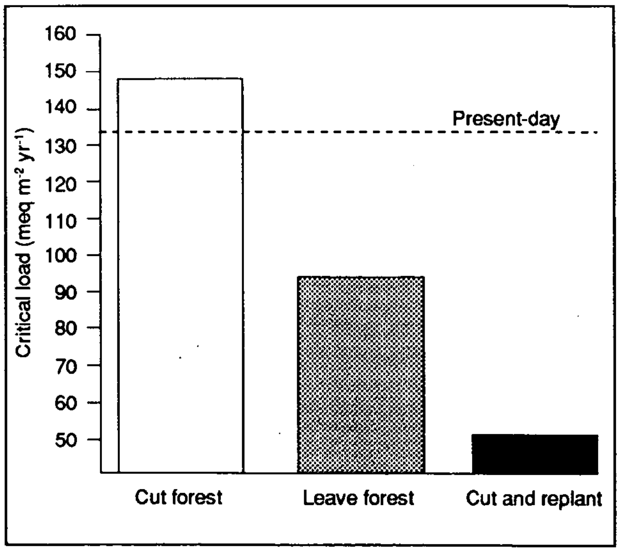

Figure 5. Critical loads calculated using MAGIC under three future afforestation scenarios

- leave the forest in place for a further 50 years;
- cut the forest immediately and leave as a moorland site for 50 years;
- cut the forest immediately and replant the entire area.

The results (Figure 5) show that the acidification stress occurring within the catchment, due to the second-rotation forest, is clearly reflected in the calculation of a low critical load, whilst the relative recovery following deforestation with no subsequent replanting changes the catchment processes to afford a slightly higher critical load than for the 'constant' forest scenario. Clearly, the calculation of critical loads for soil and freshwaters in areas where plantation forestry is a major land use requires that these impacts be considered.

THE INFLUENCE OF NITROGEN DYNAMICS

Although nitrogen is dealt with in an extremely simplified manner in MAGIC, uptake being modelled as a first-order function, the model can be used to illustrate the influence of nitrogen dynamics in calculating critical loads for sulphur. An increase in nitrate leaching from a catchment in surface waters may occur as a result of increased nitrate deposition, decreased plant uptake of nitrate (perhaps due to sulphur-induced stress on the organisms), or through a change in climatic conditions leading to increased mineralisation of nitrogen in the soil. Describing this increased nitrate leakage simply as a decrease in catchment uptake, we can evaluate the relative effect on sulphur target load. If nitrate increases in surface water, the target load of sulphur decreases (Figure 6).

CONCLUSIONS

In addition to the factors considered here, long-term climate change will potentially have an important effect on calculated critical loads for soils and water. This effect can only be realistically quantified through the application of dynamic models. Such models will, however, need to incorporate explicitly the processes and driving variables leading to increased nitrate leaching from catchment soils. The role of land use and climate change and the interaction between sulphur and nitrogen dynamics make the use of dynamic models mandatory within the critical loads framework. It is also clear that the models represent a powerful tool in the determination of target load strategies and for assessing the consequences of not achieving critical loads.

Critical loads estimated using MAGIC represent the change in deposition flux which must be made immediately to achieve zero alkalinity, and so must be regarded as a 'best case', as sulphur deposition is more likely to be reduced gradually over a long time period. On the other hand, if an equilibrium timescale is considered such that the recovery time of the system is of no great importance, then the empirical calculations are adequate. Under the empirical critical load deposition scenario, however, it is possible that a number of surface waters in the UK will not be restored to positive alkalinity within the lifespan of the existing population.

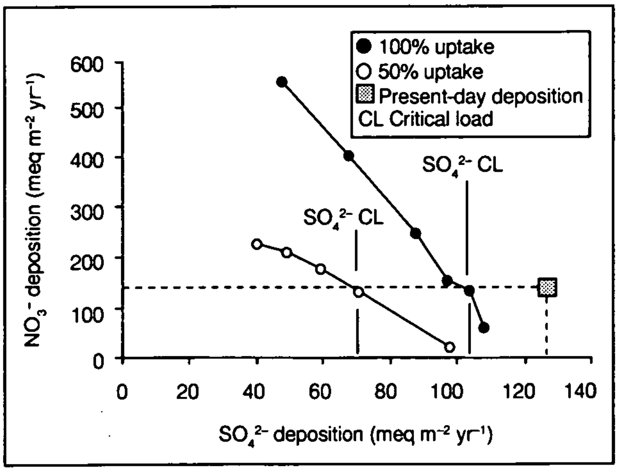

Figure 6. The interaction between sulphur and nitrogen critical loads. The graph shows MAGIC calculations of critical loads for sulphate at a given N deposition, assuming 100% or only 50% N uptake. Sulphate critical loadings for these two assumptions at the present rate of N deposition are shown on the graph

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Assessing critical levels of acid neutralising capacity (ANC) for stream organisms using empirical data

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INTRODUCTION

Central to the concept of critical loads or levels is the notion that biological systems have limits of tolerance to environmental factors - usually loads of pollutants of anthropogenic origin - beyond which damage or dysfunction may occur (Nilsson 1986). Indeed, biological systems provide the key element in the determination of critical loads, providing the definitive targets upon which change through physical, chemical or energetic processes is judged to be adverse (Nilsson 1986). As an example, we might suggest that fish populations decline to unacceptable levels when the acid neutralising capacity (ANC) in surface waters falls below 0 μeq l^{-1} (see Harriman & Christie, pp 103-108).

How might this simple concept - of defining biological end-points beyond which there is damage - be applied in practice? The biological systems at risk from the exceedance of critical loads are essentially those that make up the science of ecology: they comprise whole ecological units, such as ecosystems, or individual components at any organisational level between the subcellular and the community. Moreover, ecosystem components for which we might wish to assess critical loads involve not only structural aspects, such as community composition, but also functional features, such as biogeochemical cycling, energy transfer and production (and these are only examples). The literature is replete with examples showing how all these structural components of ecosystems, along with their functional linkages, have been damaged by the addition of pollutants. As a further complication, damage may arise because the level of a given pollutant is exceeded (eg a concentration in air or water), or because a load is exceeded (a volume or mass of pollutant deposited over a given time).

The barriers to understanding the effects of pollutants under such a complex array of possible permutations are many, and arise not least through the dearth of appropriate data. We might choose to generate such data experimentally, in which case a further choice arises over whether we should generate them in the field or in the laboratory. Both approaches have disadvantages, laboratory data, for example, often being unrealistic or of limited scale. In the field, however, manipulations on units of an appropriate size are difficult, often unreplicated, and subject to unintended intrusions on to the given experimental design (Hurlbert 1984). In either case, laboratory or field, the generation of data sufficient enough to allow a comprehensive understanding of critical load exceedance for all the potential targets would be a vast undertaking.

DETERMINING CRITICAL LOADS EMPIRICALLY

An alternative approach might be to generate the required data to set critical loads empirically. Through this method, biological attributes would be measured under field conditions at different loads or levels of pollution. Relationships between biological status and pollution would then be derived statistically in ways analogous to a dose-response curve in ecotoxicology; the curve is represented by logistic or linear regression, or by relationships derived in more than one dimension using discriminant analysis (eg Ormerod *et al.* 1988). Here, the major assumption is that the observed biological conditions have arisen as a result of the pollution climate. In few instances can we show this to be true by assessing real ecosystem change (see the work by Battarbee *et al.*, pp 99-102); instead, we rely on the inference that present-day conditions are the result of change that has already occurred (and, in fact, are in equilibrium). A further assumption is that the dose-response curves derived in this empirical way are robust enough to be repeatable and predictable in other instances, eg under other ecological circumstances. This assumption might not be a problem, provided that the effect of pollution subsumes other ecological factors which influence the distribution, abundance and biology of organisms, and the functioning of ecosystems. In freshwaters, some of these assumptions about empirically modelling relationships between pollution status and biological conditions have been tested to some extent (see Ormerod & Edwards 1991).

THE EMPIRICAL DETERMINATION OF CRITICAL LOADS IN FRESHWATERS

Partly because of the many difficulties in deriving critical loads or levels experimentally, much work in freshwater systems has been aimed at deriving critical conditions through the empirical approach (with notable exceptions for individual organisms, such as fish). Here, empirical relationships have been derived between biological conditions and those features of water quality which either mediate the effects of acid deposition, or change as a result of it: pH, aluminium concentration, total hardness, ANC (see Ormerod *et al.* 1988). In this way, these parameters might be regarded as critical levels of water quality which are affected by differing loads of deposition. In other words, relationships between water quality and the biological status of freshwaters represent transfer functions through which the impact of different deposition loads on water catchments can be understood. Thus, the determination of a critical deposition load for freshwaters depends on the load of pollutants which cause a given critical threshold (a critical level) of a chemical parameter to be crossed. This critical threshold, in turn, is determined by the level at which a given biological or ecological change occurs.

For which organisms or ecosystem components in freshwaters should we assess critical loads or levels in this way? In addition to fish, there are many organisms in or around freshwaters which have a recognised value to conservation (eg otters *(Lutra lutra)*, amphibia, riparian birds). At the same time, however, the current emphasis on biodiversity means that we should not ignore all the other organisms in freshwaters which also contribute to species diversity, and to genetic diversity within species. Impacts on species richness through changing acid base status are well recognised. We might also recognise that there are important ecological processes in freshwaters, such as primary and secondary production, though so far there are few clear data on how such processes are critically affected by changing acid base status.

The need to assess critical loads and levels for all ecosystem components might be circumvented if we could assess critical conditions for a small range of indicators whose status reflected conditions in the wider ecosystem. In freshwaters, for example, macroinvertebrates can provide important information in this way. In the case of acidification, their role in indicating the status of fish, stream macroflora and even some riverine birds has been well demonstrated.

TWO CONTRASTING USES OF THE EMPIRICAL DETERMINATION OF CRITICAL CHEMICAL CONDITIONS FOR FRESHWATER ORGANISMS

There are two general methods through which stream pH, aluminium concentration, or ANCs might be related to biological characteristics:

1. a *'top-down' approach*, where given chemical values are stipulated, and biological characteristics relative to the stipulated conditions are then assessed. For example, we might decide an ANC of 0 μeq l^{-1} is important, and then determine the biological characters of surface waters with ANC above and below this level. By this approach, we could judge which biological features may or may not be protected when we set a critical level at this value.
2. a *'bottom-up' approach*, through which important biological characteristics are first recognised, and then the conditions required to maintain them are determined. The biological characteristic might be the presence of a species important to economics or conservation. For example, we might wish to determine the ANC at which brown trout *(Salmo trutta)* populations are no longer sustained.

Both these approaches will have value under different circumstances. For example, the top-down approach will be particularly useful in instances where temporal trends in chemical data have been determined or are predicted; biological losses or gains might then be simulated. This approach will be valuable in predicting the general biological character of surface waters on the basis of critical loads derived from particular groups of organisms. For example, diatoms have been used to show the chemical conditions at which the first, critical, point of change has occurred (see the work by Battarbee *et al.*, pp 99-102).

By contrast, the bottom-up approach will be useful where we wish to determine critical loads or levels for any of the large array of organisms present in surface waters.

A demonstration

Both these approaches, the bottom-up and top-down, are demonstrated here, using data collected from upland streams throughout Wales during 1984. Macrofloral assemblages, macroinvertebrate assemblages and trout densities at 87-99 sites are related to ANCs determined from mean chemistry (as the sum of base cations minus acid anions). Similar methods might be applied to data on pH, aluminium concentration, or base cation concentration. The data collection and analyses are described more fully elsewhere (Wade, Ormerod & Gee 1987; Ormerod, Wade & Gee 1987; Ormerod *et al.* 1988). For macroinvertebrate and macroflora, the data were used to identify assemblages by a multivariate technique for classification, TWINSPAN. The same data sets were

Table 1. The taxon richness of invertebrates, density of brown trout and modal invertebrate/floral assemblage type at sites in upland Wales characterised by different mean ANC. The values are means (with SD), the values for trout being back-transformed from the log distribution

ANC category (µeq l^{-1})	n	Mayflies	Stoneflies	Caddis	All taxa	Modal invertebrate (M) or floral (F) class		n	Trout density (nos 100 m^{-2})
-200 to -50	6	0 (0)	6.2 (1.2)	3.3 (2.1)	16.2 (4.4)	M4	F3	4	0.73 (0.5)
-50 to 0	26	0.5 (1.0)	7.3 (1.9)	4.1 (3.2)	21.6 (9.8)	M4	F3	20	11.0 (0.8)
0 to 50	29	1.3 (1.4)	6.9 (1.4)	4.3 (2.4)	27.2 (8.3)	M3	F2/3	29	24.3 (0.6)
50 to 450	26	3.4 (2.2)	7.7 (2.4)	7.1 (5.3)	33.6 (15.3)	M1	F2	25	39.2 (0.7)
F		18.2***	NS	4.1**	6.9***			5.4**	

also used to determine relationships between species richness and chemistry at each of the sites.

The top-down approach

At the array of sites included in the survey, the median ANC was close to zero, and ranged from -250 to 450 µeq l^{-1}. Thus, four arbitrary ANC classes were determined within this range, and the biological character of sites in each class was assessed (Table 1). The species richness of mayflies (Ephemeroptera), caddis (Trichoptera) larvae and total invertebrates all varied significantly between the classes, as did the density of brown trout. In nearly all cases, there was a progressive gradation in biological character between classes, rather than a step change at any given ANC (eg ANC 0). For some groups, such as stonefly (Plecoptera) nymphs, there was no reduction in species richnesses with decreasing ANC; this pattern is expected from previous knowledge about their distribution in relation to acidity (see Wade *et al.* 1987).

Bottom-up approach

For this approach, biological features such as taxon richness and trout density were related to ANC by regression. Additionally, with macroinvertebrate and macrofloral assemblages classified using the TWINSPAN procedure, the significance of differences in ANC between the resulting site groups were assessed by ANOVAR.

The taxon richness of mayflies, caddis and all invertebrates increased significantly in each case with ANC (Table 2). Thus, for any given ANC, the average number of taxa might be predicted. Rearranging these regressions (Table 3), we can assess the average ANCs required for given numbers of invertebrate species. Similarly, trout densities increased significantly with ANC so that streams of ANC 0 would, on average, have 14 fish per 100 m^2, and at ANC 50 they would have 24 fish per 100 m^2 (Table 2). Again, we can rearrange these regressions to determine the mean ANC required for given fish densities.

Use of the TWINSPAN approach enables us to see further how the ANC values required for certain biological characters can be assessed. In the macroinvertebrate classification, four typical assemblages (Groups M1-M4) were found at the sites, diversity generally increasing with pH or ANC. Species present in the more diverse groups included a range of mayflies and caddis not present at lower pH. In the floral classification, three typical assemblages were apparent (Groups F1-F3); species diversity again increased with pH, with the most diverse group characterised by *Lemanea* (Ormerod *et al.* 1987).

The mean ANCs at sites occupied by each of the groups are shown in Table 4. In each case, values differ significantly between groups, and show also a progressive reduction from group M1to M4, and from F1 to F3. Determination of a critical ANC level is thus possible for each group, and choice depends on which is selected as the target for protection. For example, to prevent the formation of an impoverished invertebrate fauna (M4), we might suggest ANC should not fall below 13 µeq l^{-1} (the mean + 1 SD). If we wished to prevent the formation of an impoverished macrofloral assemblage (F3), we might suggest ANC should not fall below 40 µeq l^{-1}. This classification approach has also been examined

Table 2. Regression relationships (y = a + b ANC) between biological characteristics and ANC for upland Welsh streams

Parameter	a	b	F	r^2
Nos of mayfly species	1.12	0.017	51.5***	0.38
Nos of caddis species	4.49	0.016	8.5**	0.08
Nos of invertebrate taxa	24.5	0.074	19.3***	0.18
Trout density (log (nos 100 m^{-2} + 1))	1.18	0.004	13.5***	0.15

Table 3. Regression relationships (ANC = a+ b * biological parameter) for predicting the ANC required for different taxon richness of invertebrates, and different densities of brown trout

Parameter	a	b
Nos of mayfly species	-6.35	22.0
Nos of caddis species	2.0	5.4
Nos of invertebrate taxa	-37.4	2.5
Trout density (log (nos 100 m^{-2} + 1))	-12.1	35.7

with respect to other important resources. For example, an ANC of around 35 $\mu eq\ l^{-1}$ is a threshold at which the occurrence of dippers *(Cinclus cinclus)* along Welsh streams becomes increasingly unlikely, and at which their breeding performance is impaired (Ormerod *et al.* 1986, 1991).

Table 4. Differences in mean ANC (with SD, n) between Welsh streams occupied by different macrofloral and invertebrate assemblages

Invertebrate assemblage	ANC	Floral assemblage	ANC
M1	105.9 (85.8, 13)	F1	88.8 (93.0, 10)
M2	39.8 (51.1, 18)	F2	50.7 (68.9, 38)
M3	31.1 (60.2, 34)	F3	-6.3 (46.2, 37)
M4	-27.5 (40.1, 22)		
F	14.5***		12.4***

DISCUSSION

From this work, it can be shown that ANC (or some other parameter of acid base status) can be related to the biological status of freshwaters. The relationships are strong, highly significant statistically, repeatable between regions, and robust when applied to other locations than those at which the relationships were originally developed (eg Rutt, Weatherley & Ormerod 1990; Ormerod & Edwards 1991). Moreover, there is evidence from short-term manipulative studies which change water chemistry that biological systems behave in ways at least partly consistent with the predictions from the relationships in Tables 2 and 3 (Ormerod *et al.* 1990; Weatherley & Ormerod 1990). In addition, the organisms whose status is related to ANC can be shown to have a wider indicator value in freshwater ecosystems.

At the same time, however, several difficulties remain with accurately assessing a given level of ANC at which damage through critical load exceedance can be said to have occurred. In summary, these difficulties include:

1. a graded response in biological status across a wide range of ANC (or acid base status), which makes the definition of a single end-point difficult to define;
2. the role of natural variation in chemistry as responsible for at least part of the graded response;
3. mathematical uncertainty in relationships between ANC in surface waters and given loads of pollutants, particularly when nitrogen is added to the anion load (Hauhs *et al.* 1986).

Among these difficulties, the third is dealt with elsewhere in this volume by Harriman and Christie (pp 103-108), whilst the first and second are to some extent overcome by the approach of Battarbee and his co-workers. Since their system, of using diatoms, specifies a point of *change*, a fixed end-point representing damage can be identified. Water quality conditions responsible for crossing such end-points can also be specified. The importance of work described here is that, using the top-down approach, the significance of such end-points can be placed into the context of wider ecosystem components.

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Estimating the relative contribution of SO_x, NO_y and NH_x inputs to effects of atmospheric deposition

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INTRODUCTION

Atmospheric pollutant inputs to ecosystems include sulphur dioxide (SO_2), nitrogen oxides (NO_x), ammonia (NH_3) and their reaction products. Emissions of these gases derive from very different sources. Most of the SO_2 and NO_x derives from combustion sources, such as in electricity generation and from vehicles. By contrast, NH_3 is primarily a consequence of livestock agriculture, with over 80% estimated to derive from the volatilisation of animal wastes (Buijsman, Mass & Asman 1987). Emission controls of these pollutants, therefore, require very different strategies, so that it is of interest to identify the relative contribution of each component to atmospheric deposition. This information is useful in identifying the most efficient way of reducing deposition to meet critical and target loads. Effects of atmospheric deposition loads may be divided into three issues: eutrophication effects of total nitrogen deposition, effects of total acidifying deposition, and effects of ammonia deposition on base cation uptake.

Total nitrogen deposition is important where ecosystem effects are a consequence of eutrophication from fixed atmospheric N (eg Bobbink *et al.* 1992). The relative contribution of NO_x and NH_3 emissions may be found by summing the deposition of total reactive oxidised nitrogen (NO_y) and reduced nitrogen (NH_x), where these groups include the precursor gases and their products following atmospheric transformation (eg NO_2, HNO_3, NO_3^-, NH_3 and NH_4^+). The dry deposition inputs must be quantified for each of the gases and aerosols, as well as inputs in precipitation (wet deposition).

Total acidifying deposition derives from each of NO_y, NH_x and oxidised sulphur inputs (SO_x), which include SO_2 and sulphates (SO_4^{2-}) formed by atmospheric transformation. Assessing the relative contribution to acidification from SO_x, NO_y and NH_x is a more uncertain task because acidification from deposited N depends on its fate within the receiving ecosystem.

Ecological effects of NH_x deposition *per se* have also been recognised, such as nutrient imbalances with base cations (K^+, Mg^{2+}, Ca^{2+}), but have received less attention in their own right, being usually included with N eutrophication effects (eg Bobbink *et al.* 1992). They are considered separately here because they are a particular consequence of NH_3 emissions. In this case, it is of interest to examine the relative contribution of the different forms of NH_x deposition.

In the following sections, the different components of deposition are considered, with dry deposition of NH_3 taken as an example to show how annual inputs may be estimated. This and other N inputs are summed for example sites in the UK to assess their relative contribution to N deposition. Ecosystem processes affecting the acidification deriving from N compounds are then considered, and the same UK examples used to provide ranges for the relative contribution of NO_y and NH_x to acidification. The results are discussed in relation to the implementation of critical loads and requirements for emission controls.

QUANTIFYING THE COMPONENTS OF DEPOSITION

The different components of atmospheric deposition include dry deposition of gases (eg SO_2, NO_2, HNO_3, NH_3) and aerosols (eg SO_4^{2-}, NO_3^-, NH_4^+), as well as wet deposition of the ionic species in precipitation. A further input is the direct impaction of cloud droplets on to vegetation (cloudwater deposition). Wet deposition is the best quantified of these terms, with extensive results from precipitation monitoring networks allowing regional deposition fields to be mapped (eg UK Review Group on Acid Rain (UKRGAR) 1990). Cloud droplets exist in the size range 5-30 µm diameter, which permits efficient impaction on to vegetation surfaces. Coupled with the large concentrations in such droplets, high-altitude sites that are frequently enshrouded by cloud may receive significant pollutant inputs by this mechanism (Fowler, Cape & Unsworth 1989). Atmospheric aerosols provide the condensation nuclei for the formation of cloud droplets, but because these particles exist in much smaller size ranges, typically 0.1-1 µm, impaction is very inefficient. Other mechanisms for deposition are also inefficient in this size range, so that such aerosols deposit very slowly and are generally only a small component of the total deposition.

Dry deposition of the precursor gases represents a major contribution to total deposition, and provides the most uncertain term in its calculation, particularly for NH_3 dry deposition, which has only recently been studied using methods allowing regional extrapolation for a range of ecosystem types (see Sutton, Pitcairn & Fowler 1993). From micrometeorological measurements, the affinity of vegetation for dry deposition may be described using a 'canopy resistance' (r_c). By coupling estimates of r_c, derived from short-term flux measurements, with resistances for the turbulent atmosphere (r_a) and the quasi-laminar boundary layer at leaf surfaces (r_b), a total resistance (r_t) can be found. The reciprocal of r_t is referred to as the deposition velocity (V_d) and may be coupled with monitored air concentrations (c) to estimate long-term (eg annual) deposition fluxes (F_g):

$$r_t = r_a + r_b + r_c = V_d^{-1} \qquad (1)$$

$$F_g = -V_d \chi \qquad (2)$$

Figure 1 shows an example of measured NH_3 V_d and r_c for a moorland in southern Scotland (Sutton, Moncrieff & Fowler 1992). The large V_d and small r_c indicate that the vegetation is an efficient sink for NH_3, with deposition limited by atmospheric resistance.

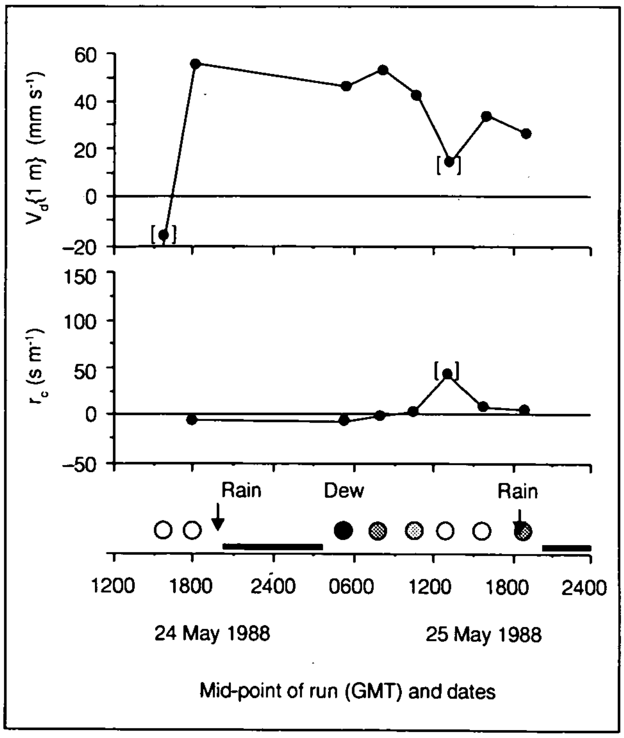

Figure 1. Deposition velocity (V_d) and canopy resistance (r_c) measurements of NH_3 at Fala Moor, southern Scotland. Canopy wetness is shown at the foot of the graph, from dry conditions (open circle) to fully wet (black circle). A horizontal bar denotes night-time. Runs with a large measurement uncertainty are shown in brackets

The mean r_c for these measurements was 5 s m^{-1}. Ammonia exchange is, however, highly dependent on land use, and both NH_3 emission and deposition occur over other vegetation types, such as grazed grassland and fertilized croplands, which are less efficient for NH_3. On a regional scale, ammonia exchange reduces the overall input by NH_3 dry deposition. However, larger air concentrations are maintained, allowing an increased deposition to semi-natural ecosystems.

An example of the use of such measurements is shown in Table 1, where monitored air concentrations are combined with calculated average V_d to provide estimates of annual NH_3 dry deposition fluxes. Four examples of unfertilized sites in Britain are shown to reflect a range of input fluxes and land uses. Air concentrations are larger at the eastern England sites, allowing increased fluxes. In addition, because of the small size of r_c and increased turbulence over rougher forest vegetation, which provides smaller r_a and r_b, dry deposition is much larger over forests compared with short vegetation. Rural background air concentrations are used in these calculations, so that dry deposition may be much larger near local NH_3 sources.

Table 1. Estimated annual NH_3 dry deposition to example unfertilized sites in the UK (further details provided by Sutton *et al.* 1993)

Site	χ NH_3 (μg m^{-3})	V_d^* (mm s^{-1})	NH_3 deposition (kg N ha^{-1} yr^{-1})
Southern Scotland			
Upland forest, eg Glentress	0.58	38.1	5.7
Upland moor, eg Fala Moor	0.55	18.5	2.6
Eastern England			
Lowland forest, eg Thetford	3.2	50.2	41.1
Lowland heath, eg Breckland	2.6	13.8	9.3

*Calculated using r_c = 5 s m^{-1}

RELATIVE CONTRIBUTIONS TO TOTAL NITROGEN AND NH_x DEPOSITION

The different components of nitrogen deposition may be summed to estimate the relative contribution of NO_y and NH_x, as shown in Table 2 for the same example sites considered above. Dry deposition fluxes of NH_3 from Table 1 are combined with NH_4^+ inputs in wet deposition, and with aerosol and cloud-water deposition. A similar approach is used to calculate total NO_y deposition, accounting for dry deposition of NO_2 and HNO_3 as well as wet, cloud and aerosol deposition of NO_3^-.

Table 2 shows that NH_x typically accounts for 60-80% of the total N input for these examples of unfertilized and semi-natural ecosystems. The fraction is largest

Table 2. Relative contribution of NO_y and NH_x to total N deposition for example unfertilized sites in the UK, calculated using NH_3 dry deposition from Table 1. Details of other wet and dry deposited inputs provided by Sutton *et al.* (1993). Inputs in kg N ha^{-1} yr^{-1}

Site	NO_y deposition	NH_x deposition	Total N deposition	% deposition from NH_x
Southern Scotland				
Upland forest, eg Glentress	7.1	11.8	18.9	62
Upland moor, eg Fala Moor	5.5	7.9	13.4	59
Eastern England				
Lowland forest, eg Thetford	12.4	48.4	60.8	80
Lowland heath, eg Breckland	9.9	16.6	26.5	63

for the lowland forest, where NH_x contributes about 80% of the nitrogen input and NO_y only 20%. In addition to showing the importance of NH_x deposition to total N inputs, the Tables also show the relative contribution of NH_3 dry deposition. For these examples, NH_3 contributes 33-85% of the NH_x input. At specific near-source locations (ie adjacent to livestock farms), NH_3 dry deposition is expected to dominate both NH_x and total nitrogen deposition. In general, because NH_3 has a shorter atmospheric residence time than NO_2, sites in NH_3-polluted regions will be most dominated by the NH_x inputs, while remote sites will have the largest NO_y/NH_x deposition ratio.

While these values are only examples, they may be contrasted with estimated emissions of NO_x and NH_3. The UK emission of NO_x has been estimated to be 750 Gg N yr^{-1} (UKRGAR 1990), whereas recent NH_3 emission estimates range between 170 and 390 Gg N yr^{-1} (see Sutton, Pitcairn & Fowler 1993). On the basis of these emissions, the large contribution of NH_x to total N deposition might seem surprising. However, as noted above, NH_3 dry deposition depends on land use, with fertilized agricultural vegetation generally showing net emission or a balance between emission and deposition over the year. Hence, net dry deposition is limited to unfertilized sites which consequently receive larger inputs than might be expected.

CONTRIBUTION OF NO_y AND NH_x TO TOTAL ACIDIFYING DEPOSITION

Biological interactions of deposited nitrogen

Estimating the contribution of N compounds to ecosystem acidification is complicated because of interactions of the deposited ions within the ecosystem. Because N is a major nutrient, much of the deposited input may be taken up by plants or immobilised in soil organic matter, affecting the extent of soil acidification (Binkley & Richter 1987). The problem is not so significant with deposition of SO_x, as much smaller amounts of S are accumulated within biomass and soil organic matter. As a consequence, in most ecosystems, outputs of SO_4^{2-} by leaching equate closely with atmospheric SO_x inputs over a few years.

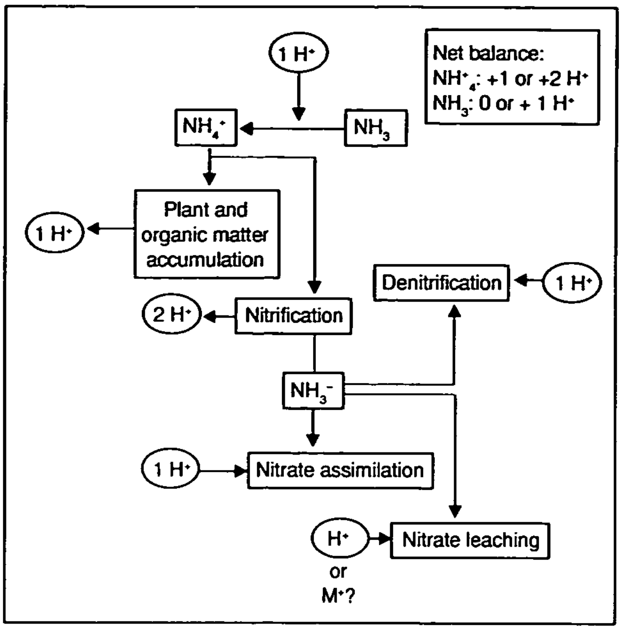

Figure 2. Effects of atmospheric NH_3 or NH_4^+ input on soil acidity. M^+ represents a metal ion that might be leached from the soil

The effects of N transformations on acidification are demonstrated for NH_x inputs to soils in Figure 2. Deposition of NH_4^+ followed by accumulation in biomass and soil organic matter is acidifying, because the N is stored as organic $R\text{-}NH_2$ forms. Uptake results in an equivalent net release of H^+ ions back to the soil to maintain electro-neutrality. By contrast, mineralisation of soil organic matter is de-acidifying, so that the actual acidification by this mechanism depends on net accumulation in biomass and organic matter. In the long term, this requires harvesting of biomass or litter removal.

If NH_4^+ is left in the soil and nitrified to NO_3^-, the effect is again acidifying:

$$NH_4^+ + 2O_2 \rightarrow NO_3^- + H_2O + 2H^+ \qquad (3)$$

Accumulation of NO_3^- in biomass may neutralise this acidity, as the NO_3^- is reconverted to $R\text{-}NH_2$ forms. In addition, any NO_3^- that is denitrified results in an equivalent consumption of H^+, neutralising soil acidity. Alternatively, NO_3^- may be leached from the soil accompanied by base cations or aluminium, resulting in net acidification.

Nitrification of NH_4^+ in the soil does not always occur, however, and, where NH_x deposition is very large or there are shortages of other nutrients (Gundersen & Rasmussen 1988), NH_4^+ may accumulate allowing NH_4^+ leaching. Though not shown in Figure 2, potentially the effect would be de-acidifying, especially where the leached NH_4^+ is derived from mineralisation of organic N originally deposited as NO_y (M S Cresser, pers. comm.). However, nitrification may be expected in subsequent ground- and surface waters, so this effect is temporary.

Figure 2 also shows that deposition of NH_3 or NH_4^+ has differing effects. Conversion of dry-deposited NH_3 to NH_4^+ consumes one H^+ ion, so that NH_3 has no net acidifying effect if accumulated in biomass or denitrified. Equally, nitrification from deposited NH_3 produces one H^+, rather than two as for NH_4^+. Ammonium is, therefore, more acidifying than NH_3, and in many cases represents a major acidity input compared with free H^+, SO_2 and NO_2 deposition. However, it should be noted that ammonia emission occurs as NH_3, and that NH_4^+ is a product of H_2SO_4 and HNO_3 neutralisation, where these acids derive from SO_2 and NO_x emissions. Hence, while much of the acidifying deposition arrives as neutral NH_4^+ salts, the acidity originates from SO_2 and NO_x.

Because of the effect of atmospheric transformation, it is useful to consider acidifying inputs in two distinct ways. One is to consider inputs of the *actual deposited species.* In this way, the acidification from each of the ions and gases deposited (eg SO_2, SO_4^{2-}, NH_3, NH_4^+, H^+, etc) is summed, and account is taken of the different acidifying effects of each species. This approach is relevant for detailed assessments of the form in which acidity is input to ecosystems. Alternatively, acidifying inputs may be treated in terms of the *equivalent emitted pollutants.* In this approach, the acidifying effect of each of the deposited species is treated as if it were the precursor gas; thus removing the effect of atmospheric transformation; the method is therefore useful for assessing the relative contribution of different emissions (SO_2, NO_x, NH_3) to acidifying deposition.

Critical loads treatment of deposited N interactions in ecosystems

The estimation of critical loads for acidity has largely treated the acidifying deposition in terms of the equivalent emitted pollutants. Hence, all inputs of SO_x, NO_y and NH_x are assigned the acidifying effect of SO_2, NO_2 and NH_3. In the mass balance approach (eg Schulze *et al.* 1989), the nitrogen transformations discussed above are associated with the critical load calculation, rather than with the deposition. All deposited N, apart from that accumulated in vegetation and organic matter, is assumed to be acidifying, and all NH_x left in the soil is assumed to be nitrified. This approach, as outlined by Schulze *et al.*, states that the critical load for acidity is met if (expressed as equivalents):

$$F_{SOx} + F_N < W_{bc} + A_N + I_{Nnet} \qquad (4)$$

where F_{SOx} = total deposition of SO_x; F_N = total deposition of NO_y and NH_x; W_{bc} = base cation weathering; A_N = biomass accumulation of N; and I_{Nnet} = net N immobilisation. Further modifications of this approach have been developed (eg Grennfelt & Thörnelöf 1992), such as accounting for acceptable nitrate leaching, base cation deposition and denitrification.

The approach, therefore, takes some account of the nitrogen transformations. However, it fails to separate the different possible effects of NO_y and NH_x deposition, which becomes important if, for example, N accumulation is different for NO_3^- or NH_4^+. The approach also entails some circularity, as N accumulation is expected to be a function of F_N. As a consequence of these effects, it becomes harder to examine the relative contribution of SO_2, NO_x and NH_3 emissions to the critical load exceedance.

Relative contributions to ecosystem acidification

A complementary approach to that outlined above is to associate N transformations with the atmospheric deposition side of the equation. This approach is adopted here, the aim being to examine, in a simple manner, the relative magnitude of the different components of acidifying deposition. By examining deposition in this way, it is possible to consider inputs both in terms of the actual deposited species and as the equivalent emitted pollutants.

To demonstrate the different relative contributions of the deposited species, total acidifying inputs are calculated for the same example sites considered in Tables 1 and 2. This calculation is shown first in Table 3 for the suite of *actual deposited species.* The range of possible acidification deriving from NH_3 and NH_4^+ inputs may be found according to Figure 2, and a similar approach is used for NO_y inputs. Nitric acid deposition is acidifying if left in the soil and ultimately leached, but neutral if accumulated in vegetation. Conversely, NO_3^- inputs are neutral if left in the soil, but de-acidifying if accumulated in vegetation. Deposition of SO_4^{2-} is not shown in this Table as, in itself, it has no net acidifying effect, the reason being, as with NO_3^-, that the acidity derives from the associated input of H^+ ions.

Table 3 shows the fluxes of the deposited species in kg (H^+, S or N) ha^{-1} yr^{-1}, and, underneath in italics, ranges of the possible acidification (mmol H^+ m^{-2} yr^{-1}) for each species. The Table is revealing in that it highlights the importance of each of the actual deposited species to ecosystem acidification. If plant uptake and denitrification are assumed to be negligible and nitrification complete, the upper figures are appropriate for the N species. In this case, the largest contribution to acidification at each of the sites is from NH_3 in dry deposition and from NH_4^+ in wet deposition. The contribution to acidification that is deposited as NH_x is shown in the final column. Even assuming complete biomass accumulation and immobilisation of all deposited N, the Table shows that about 40% of the acidity is input as non-acidic NH_x. If accumulation and immobilisation are unimportant, then 60-75% of the acidity is deposited as NH_x.

Table 3. Ranges of possible acidifying deposition to example unfertilized sites in the UK. Inputs expressed as the actual deposited species.. For each site, the first figure is the deposition (kg ha^{-1} yr^{-1}) as H, N, or S. The second figure (in italics) is the equivalent H^+ input (mmol m^{-2} yr^{-1}). Ranges of possible acidification for N components depend on the fate of the deposited species (see Figure 2). Dry deposition of free H^+ in aerosols is assumed to be small and is not calculated

	Wet deposition [a]				Dry deposition					Cloudwater deposition [d]			H^+ input	
Chemical species	H^+	NH_4^+	NO_3^-	SO_2 [b]	NO_2 [c]	HNO_3 [c]	NH_3 [c]	NH_4^+ [c]	NO_3^- [c]	H^+	NH_4^+	NO_3^-	Total H^+ (mmol m^{-2} yr^{-1})	% of positive inputs from NH_x
H^+ produced (mol/mol)	+1	+1 or +2	-1 or 0	+2	0 or +1	0 or +1	0 or +1	+1 or +2	-1 or 0	+1	+1 or +2	-1 or 0	-	-
Southern Scotland														
Upland forest,	0.35	4.5	3.5	3	1	1.5	5.7	0.6	0.2	0.11	1.0	0.9	*76-211*	40-61
eg Glentress	*35*	*32-64*	*-25-0*	*19*	*0-7*	*0-11*	*0-41*	*4-9*	*-1-0*	*11*	*7-14*	*-6-0*		
Upland moor,	0.35	4.5	3.5	3	1	0.6	2.6	0.6	0.2	0.02	0.2	0.2	*66-162*	39-59
eg Fala Moor	*35*	*32-64*	*-25-0*	*19*	*0-7*	*0-4*	*0-19*	*4-9*	*-1-0*	*2*	*1-3*	*-1-0*		
Eastern England														
Lowland forest	0.25	6.5	4.5	10	4.3	3.2	41.1	0.8	0.4				*105-540*	37-74
eg Thetford	*25*	*46-93*	*-32-0*	*63*	*0-31*	*0-23*	*0-294*	*6-11*	*-3-0*	-	-	-		
Lowland heath	0.25	6.5	4.5	10	4.3	0.7	9.3	0.8	0.4					
eg Breckland	*25*	*46-93*	*-32-0*	*63*	*0-31*	*0-5*	*0-66*	*6-11*	*-3-0*	-	-	-	*105-294*	37-58

a, from Warren Spring Laboratory (1988)
b, from UKRGAR (1990)
c, from Table 1 and Sutton, Pitcairn and Fowler (1993)
d, from Fowler, Cape and Unsworth (1989)

Alternatively, these deposition data may be viewed as the *equivalent emitted pollutants*. Because all inputs are treated as either SO_2, NO_x or NH_3, the calculation of possible acidification is simplified, and accounting for the range of possible effects of N deposition may be expressed (in moles) as:

$$H^+ = 2F_{SOx} + (0\text{-}1)F_{NOy} + (0\text{-}1)F_{NHx} \qquad (5)$$

The results of applying this equation to the same sites as in the previous Tables are shown in Table 4. In this case, the input of free H^+ in rain and cloudwater deposition is not treated separately. It is included implicitly because it is a result of atmospheric reaction of emitted SO_2 and NO_x. Ranges of total acidifying inputs to the soil are given for each site, and agree well with those calculated in Table 3, particularly as the components of deposition are given different weighting in the two approaches to account for atmospheric transformation. Apart from the approximations in estimating deposition, the only difference between the Tables is the small contribution of free H^+ in dry deposited unneutralised SO_4^{2-} aerosols, which is not easily measured and excluded from Table 3. It is expected to contribute less than dry deposition of NH_4^+ aerosols and to have a minor effect on the calculated figures.

Table 4. Ranges of possible acidifying deposition to example unfertilized sites in the UK. Inputs expressed as the equivalent emitted pollutants

	Deposited species (kg S, N ha^{-1} yr^{-1}) (mmol H^+ m^{-2} yr^{-1})				% acidifying deposition attributable to equivalent emitted pollutants		
Site	SO_x [a]	NO_y [b]	NH_x [b]	Total H^+ input	SO_2	NO_x	NH_3
Southern Scotland							
Upland forest,	12	7.1	11.8				
eg Glentress	*75*	*0-51*	*0-84*	*75-210*	36-100	0-24	0-40
Upland moorland,	12	5.5	7.9				
eg Fala Moor	*75*	*0-39*	*0-56*	*75-170*	44-100	0-23	0-33
Eastern England							
Lowland forest,	20	12.4	48.4				
eg Thetford	*125*	*0-89*	*0-346*	*125-560*	22-100	0-16	0-62
Lowland heath,	20	9.9	16.6				
eg Breckland	*125*	*0-71*	*0-119*	*125-315*	40-100	0-22	0-38

a, from UKRGAR (1990)
b, from Table 2

The results in Table 4 demonstrate the relative contributions of different emissions to the acidifying input. If all the deposited N is accumulated and immobilised, then, by definition, all acidification derives from SO_2 emissions. By contrast, where these effects are not important, N deposition would contribute approximately 50-80% of the acidifying deposition for these examples. Ammonia emissions are also seen to be the main component of acidifying N deposition, contributing up to 30-60% of the acidifying deposition for the sites shown.

Tables 3 and 4 show in a simple manner the ranges of possible acidification deriving from deposited inputs. Accordingly, no particular values for N transformations are given. These depend on biomass production and, therefore, on ecosystem type (forest, moorland) and management (harvesting). They are also expected to depend on the magnitude of deposition, as well as on interactions with other nutrients. For example, short-term increased growth of trees deriving from increased N supply would result in increased base cation uptake by the trees, depleting soil base saturation and increasing acidification, which would limit the mitigating effect of biomass accumulation. Biomass accumulation will, however, have little effect in unharvested ecosystems (eg unharvested forest nature reserves). In this case, N uptake is balanced by mineralisation, so that where nitrification occurs the upper figures for the N contribution to acidification are expected to apply. Equally, in moorland areas where net accumulation of biomass is limited (removal by burning), the N contribution will approach the upper figures.

DISCUSSION

The Tables show the contribution of the different deposited pollutants to each of N eutrophication, acidification and NH_x effects. By definition, the latter depend specifically on NH_3 emissions, though the figures show that NH_x also contributes substantially to the other effects. In particular, the deposition of N as NH_x is larger than for NO_y for each of the example sites by a factor of 1.4-4. Although NH_3 emissions in the UK are less than for NO_x, the deposition of NH_x to the example sites is greater because the semi-natural ecosystems of interest for critical loads receive large rates of NH_3 dry deposition, whereas agricultural land is often a net source of NH_3.

Much of the scientific focus has been on the acidifying effects of deposition. In this case, N transformations may mitigate the effect of NO_y and NH_x, so that deposition of SO_x is most important. However, where N removal is limited, NH_3 emissions may make a major contribution (up to 30-60% for the examples given). The magnitude of this contribution has important implications for emission control policy. At present, control is focused on SO_2 and NO_x emissions (eg Commission of the European Communities 1989). From the example figures shown above, it is clear that such measures will have limited effectiveness for both acidification and N eutrophication in the absence of reductions in NH_3 emissions. Additionally, such measures would have no benefit in the case of NH_x-specific ecological effects. Though these NH_x effects have received less attention in their own right, they may be particularly important because of local deposition near agricultural sources. In addition, Bobbink *et al.* (1992) point out that the critical load for NH_x nutrient imbalances can be as small as 11 kg N ha^{-1} yr^{-1} for non-nitrifying soils.

The importance of each of the three pollutant response groups (N eutrophication, acidification and NH_3 effects) also highlights the need for improved integration of different critical loads. Alongside the development of critical loads for acidity, there has been some development of critical loads for N eutrophication (eg Bobbink *et al.* 1992), though there is also a need to further distinguish critical loads for NH_x effects (eg De Vries 1992). This distinction would be useful, because they represent a specific response to NH_3 emissions. A potential constraint is that some deposited NO_y may be assimilated by vegetation and subsequently mineralised to form NH_4^+. In this case, NH_4^+-nutrient imbalances might be defined in response to total N deposition. However, such imbalances would not be completely expected in situations of excess N, particularly for woody plants, which show a preference for uptake of NH_4^+ (Schulze *et al.* 1989), leaving NO_3^- to be leached from the soil. Hence, NH_4^+ effects are expected to be most closely coupled with NH_x deposition.

Separating these three pollutant response groups would allow each critical load to be mapped and compared with the relevant deposition to show exceedances in each case. It would be attractive to overlay these three maps of critical load exceedance (E) and apply the most sensitive effect (E_{max}) for each area in a combined exceedance map, ie:

$$E_{max} = \text{largest of } (E_{acidity} \text{ or } E_{totalN} \text{ or } E_{NHx}) \qquad (6)$$

However, this procedure is complicated because each of the different exceedances and underlying critical loads relate to deposition of different quantities: SO_x+NO_y+NH_x for acidity, NO_y + NH_x for nitrogen eutrophication and NH_x for NH_x/base cation ratios, respectively. Reducing deposition on the basis of E_{max} at a particular date would not necessarily reduce the other exceedances. For example, if $E_{max} = E_{acidity}$ and emission controls reduced S deposition only, then the critical load for acidity might be met, but E_{totalN} and E_{NHx} would remain unchanged. As a consequence, protection of an area requires each of the three critical loads to be met. Nevertheless, these issues could be integrated qualitatively in a combined map, showing where each of the three critical loads were exceeded. This map would show up regional patterns in the three effects and the degree to which they overlapped.

One benefit of integrating the critical loads for N eutrophication and NH_x alongside that for acidity is that such integration would allow for ecosystem responses that are currently treated as mitigating effects in the critical load for acidity. For example, where a soil is non-nitrifying, this factor might be seen protecting it from acidification due to NH_x. However, it is in such conditions that the critical load for NH_x, in the context of imbalances in nutrient cation uptake, is most likely to be exceeded. Hence, integrating the three critical load exceedances would mean that the effect was still recognised.

A further example of such an effect is denitrification from forest soils, which is sometimes added to the critical load as a removal mechanism alongside biomass N accumulation (Grennfelt & Thörnelöf 1992). Because denitrification (producing both N_2 and N_2O) emissions increase in response to N deposition, ecosystem acidification from N inputs would tend to be limited (eg Aber *et al.* 1989).

Conversely, enhanced N_2O emission might in itself be considered a detrimental environmental effect and an indicator of having exceeded an N eutrophication critical load. Again, integration of the three exceedances would show which effect was the most sensitive.

There is also some evidence that enhanced N deposition can result in feedbacks which limit net NH_3 dry deposition to semi-natural ecosystems (Sutton *et al.* 1993). The suggestion is that the elevated N supply increases plant nitrogen status, allowing extended periods of NH_3 emission, which may also be associated with increased plant senescence. This process would mitigate against each of the three pollutant responses, as the deposition term used to compare with critical loads would be reduced. Such a response may be visualised as diluting the effects of deposited N by exporting the NH_3 from the most contaminated areas. Clearly, there is a problem for the critical loads approach, as the reduced deposition is itself a response to having exceeded the critical load. A possible solution would be to estimate deposition to damaged sites as if for healthy ecosystems, where NH_3 deposition was more rapid. At present, there is a requirement for further measurements to examine how widespread this phenomenon is, and also to ensure that NH_3 inputs are quantified on the basis of deposition to healthy sites.

CONCLUSIONS

Atmospheric pollutant inputs to ecosystems can be quantified by combining estimates of wet and cloud-water deposition, together with inputs by dry deposition of gases and particles. Dry deposition of gases (SO_2, NO_x and NH_3) is one of the most uncertain terms, especially for NH_3. Rates of NH_3 dry deposition depend on the vegetation type, though rapid deposition occurs for the semi-natural ecosystems of most interest for critical loads. Ammonia deposition is particularly sensitive to the canopy resistance, which for these sites is estimated at 5 s m^{-1}. For the four example sites in the UK, NH_3 dry deposition ranges from 3 to 41 kg N ha^{-1} yr^{-1}, which represents 33-85% of the total NH_x input at these sites.

Such dry deposition inputs may be combined with the other components of deposition to estimate the relative contributions to total nitrogen deposition (NO_y+NH_x) and acidifying deposition (SO_x+NO_y+NH_x). For the same example unfertilized sites, NH_x is estimated to contribute 60-80% of the total N deposition, with the greatest percentage over lowland forest where NH_3 dry deposition is largest. Total N deposition for these examples is estimated to be 10-60 kg N ha^{-1} yr^{-1}, though in practice some UK sites probably receive up to 100 kg N ha^{-1} yr^{-1} at woodlands close to large NH_3 sources.

Estimating the relative contribution to acidification is less simple because interactions within the ecosystem mitigate the acidification from the deposited N species. Nevertheless, ranges of the possible acidifying contributions may be defined. The acidifying inputs may be considered in two ways, as the *actual deposited species* or as the *equivalent emitted pollutants.* The former assesses the contribution of individual gases and ions deposited (eg SO_2, SO_4^{2-}, NH_3, NH_4^+, H^+), while in the latter the acidifying effects of SO_x, NO_y and NH_x are treated as for the precursor gases (SO_2, NO_x, NH_3), removing the effect of atmospheric transformation. Expressed as the equivalent emitted pollutants, NH_x contributes up to 30-60% of the acidifying input, demonstrating the importance of NH_3 emissions. The contribution is even larger when expressed as the actual deposited species (up to 60-75%), showing that most of the acidity is actually deposited as NH_x. The magnitude of each of these figures demonstrates the need to include NH_3 in emission control policy.

Although most attention has been given to acidifying deposition, effects of both total N deposition and NH_x *per se* have also been recognised. It is useful to distinguish these issues because they define three pollutant response groups for SO_x+NO_y+NH_x, NO_y+NH_x, and NH_x deposition, respectively. Separating these three groups defines effects in relation to the appropriate emissions, and also allows combined exceedance maps to be constructed showing areas not protected for each effect. Integrating critical loads for these three groups would also account for responses that tend to mitigate ecosystem acidification, such as non-nitrifying soils which are sensitive to NH_x-nutrient imbalances. Nevertheless, other ecosystem responses may remain untreated by this approach. An example is a possible feedback which limits NH_3 dry deposition to severely polluted semi-natural ecosystems, and which may itself be a response to having exceeded the critical load for N deposition. As a result, some caution is needed when comparing critical loads with deposition estimates that are based on inputs to N-polluted sites.

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Workshop reports

A number of workshop sessions were held during the Conference to discuss questions relating to the critical load concept and its application. The following section presents the questions discussed and the main points from the discussions.

Question 1 *Can we identify biological targets for use in determining critical loads of acidity for non-forest soil/plant systems? Can critical chemical limits be set for these targets?*

The group considered that it was important to establish the aims of the 'protection' afforded by a given critical load. Thus, the aim may be to protect, for example, a given rare species, a given ecosystem or the maximum species diversity. If it was to protect a given rare species, then that species becomes the indicator; if to protect given ecosystems, then a characteristic species of the system could be used as an indicator.

An alternative approach would be to identify those ecosystems most sensitive to acidification: lowland valley mires, raised mires, lowland heaths, upland mountain heaths, epiphytes, acid-neutral grasslands. However, the group was doubtful that sufficient data were available to enable chemical limits to be set for individual species or the sensitive communities. A decline in pH was known to be more important at certain critical points; eg at pH <6.5, phosphorus availability increased, and below pH 4.5, calcicoles tended to be absent from communities; other existing relationships between soil chemical parameters and species occurrence were probably too imprecise for the setting of critical loads. Manipulative experiments should be carried out on some of the sensitive plant assemblages.

Question 2 *Should land use effects be incorporated into calculations of critical loads? If so, which effects and how should they be incorporated?*

The group concluded that land use effects should be incorporated as they were, potentially, major modifiers of the critical load.

It was agreed that the principal land uses which should be considered were forestry and intensive agriculture.

The effects which should be incorporated for forestry were considered to be scavenging effects, base cation uptake, fertilizer additions, and the accumulation of acid litter and humus.

Agricultural effects to be incorporated were base cation uptake/removal, nitrification and fertilizer addition. Some of these various effects would effectively increase the critical load, while others would lead to a decline. The group also stressed that it would be important to incorporate temporal changes in land use and to consider the scale at which land use was incorporated into critical load calculations.

The effects of land use could be incorporated into calculations of critical loads through the use of dynamic models or by incorporating the relevant values into the mass balance equation. It was also felt that critical load maps should be presented both with and without the incorporation of land use factors. Geographical information systems offer a methodology for exploring the impact of the various land use effects on critical loads.

Question 3 *Can we use the same biological targets in determining critical loads of acidity for all types of forest? If so, what are the most suitable targets - roots, mycorrhizae, soil flora, soil fauna, or some other? Can we set chemical limits for a range of forest types, whether using consistent or different biological targets?*

The group concluded that no single, overall, biological target could be used for all forests/woodlands as different criteria would apply if a forest was maintained primarily for production or conservation purposes.

In a production forest/woodland, any change in production would be the main biological indicator of adverse effects. Tree crown condition was not felt to be a useful indicator because of the many confounding factors which might influence crown condition.

In 'conservation' forests/woodlands, ground flora species might provide useful indicators.

The group also pointed out that changes in tree growth and health provided 'targets' related to changes in ecosystem function, while changes in ground flora indicated changes in ecosystem structure.

It was considered that current knowledge of the relationships between variations in mycorrhizal communities, soil fauna or flora, and soil chemistry was not precise enough to form the basis for the setting of chemical limits for use in calculating critical loads. However, it was felt that further analysis of available data bases should be carried out.

Concern was expressed about the lack of dose-response data for tree fine roots, although these are currently the most widely used biological indicator.

Question 4 *Should we use fish invertebrates, diatoms or other organisms as biological indicators when calculating critical loads for surface waters? Would they have the same chemical limits?*

The group concluded that different groups of organisms could provide the best indicators for different aims, eg protection of rare species, maintenance of fish stocks, maintenance of biodiversity, economic *versus* conservation considerations.

Recent work had suggested that diatoms form valuable indicators; they were sensitive to change and provided a historical record. They were, however, best suited to setting critical loads for lakes rather than streams. It was also felt that the public and decision-makers related better to a critical load set to maintain fish stocks rather than to prevent changes in diatom populations.

It was concluded that information was needed on the effect of exceedance of a critical load, set using an organism, or a group of organisms, on other organisms.

The use of ANC = 0 as a critical chemical limit, as in the UK, was discussed. It was felt that, because of the graded nature of biological response, indicators could be chosen to match any arbitrary value of ANC.

The group concluded that at present diatoms offered the best biological indicators for a range of aims, ie conservation, maintenance of fish stocks, but that the existing diatom model needed validating. However, the use of other groups of aquatic organisms as biological indicators should be explored further.

Question 5 *Should we use chemical or biological criteria in setting critical loads of nitrogen for soil/plant systems? Define the most suitable criteria.*

The group initially considered what criteria could be used. Discussion was based around:

- loss of species diversity
- tree health
- visible *versus* invisible effects
- increased nitrate concentrations in the soil
- use of mycorrhizae

It was agreed that the aim should be to use biological criteria. The group then considered what data were available for each of the above criteria, and on which a dose-response relationship could be based. Problems were identified in the case of all the criteria, either because of a lack of data or because given responses could result from causes other than increased nitrogen deposition, eg frost damage and impacts of pathogens on tree health. Impacts of sulphur deposition were also a confounding factor when considering some effects, eg M_g deficiency.

Nitrogen-sensitive ecosystems could be identified, eg actively growing *Sphagnum* mires, montane moss heaths, spring-fed fens, unimproved lowland grassland, lowland heaths and chalk grasslands. Precise limits could not, however, be set for critical loads for these systems as yet, but they were likely to be low, *ca* 8-10 kg N ha^{-1} yr^{-1}.

In the case of forest systems, a different critical load might be used if the forest was maintained for timber production or for conservation purposes. The N saturation approach might be useful in the case of production forests, but changes in ground flora would be more appropriate criteria in forests primarily maintained for conservation.

Question 6 *Are the formulations of weathering rate in the PROFILE model well supported? What research is needed to overcome any weaknesses in the model?*

The group organised its discussions around a number of questions, as reflected in the following report.

1. *Are the reaction rate coefficients used in the model valid?*

The main concerns were the application of laboratory-derived coefficients to the field situation, the application of coefficients derived in single mineral experiments to poly-mineral soils, and the fact that the model predictions of the main mineralogical controls on weathering seemed to contradict perceived wisdom on mineral stabilities.

The group felt these issues remained unresolved and that further research was needed on reaction rates of certain minerals (epidote, chlorite and hornblende, for example), plus further testing of the model by application to a wider range of soils/mineralogies for which detailed mineralogy and soil solution data were available. Further sensitivity analysis of the model was also required.

2. *Is the role of organic acids adequately dealt with in the model?*

It was agreed that organic acids were not adequately considered, but it was also felt that there was a lack of good experimental data and rate coefficients for organic acids. The question of organics must be dealt with, as they were such an important factor in many soils, but the required research presented considerable experimental difficulties.

3. *Should the model incorporate the effects of mineral coatings on mineral weathering?*

It was concluded that available evidence suggested that ion diffusion through coatings was rapid compared with weathering rates. Therefore, coatings would not have a significant effect and need not be incorporated into the model.

4. *Should the gibbsite coefficient be used as the control on aluminium release?*

It was agreed that this was not an appropriate control (Dr Sverdrup pointed out that this part of the model was soon to be changed). The group was in favour of controls related to mineral weathering, ion exchange, organic complexation, and precipitation reactions.

5. *Are surface area effects dealt with realistically in the model?*

The increase in mineral reactivity with increasing surface area (decreasing particle size) needed further evaluation using field and laboratory data.

Question 7 *How is it possible to validate critical loads - can a programme be devised?*

The group addressed the questions in relation to specific target systems: soils, vegetation and surface waters. However, two main approaches were available: field monitoring and manipulative experiments. It was stressed that monitoring-based approaches necessitated reliable data on pollutant inputs, and on weathering rates for each site monitored.

Soils Responses could be tested using increased or reduced pollutant loadings, but these required detailed information on current soil properties, and there were problems in defining the 'controls'. Another major problem was the slow rate of change of many soil properties in response to acid input - soil solution chemistry responded faster to changes in inputs and might form a better basis for evaluating critical loads.

Vegetation The response of vegetation to pollutant loadings (as opposed to atmospheric concentrations) was generally indirect and soil-mediated. Studies of vegetation change-response, therefore, needed parallel soil data and studies. As with soils, the timescale of change-response was long, and long-term studies were therefore required. The criteria of damage also needed definition: change in performance parameters or loss of species.

Waters Validation was probably easier than with soils or vegetation; comparison of sites from areas in which the critical load was exceeded with those from sites where the critical load was not exceeded would be useful (a similar approach could be used with vegetation). It was agreed that biologically important criteria could be defined for validation; the link between water chemistry and performance of aquatic species was clearer than similar relationships between vegetation performance and soil chemistry; water chemistry interpreted changes in acid pollutant loading to catchments, while parallel changes in the catchment soils might be imperceptible.

Conclusions

The validity of critical loads could be tested experimentally, but the timescale of response to changed loadings was a problem both with soils and vegetation.

A survey and monitoring approach to determine responses to variations in loading would be valuable in the context of vegetation and surface waters.

Damage (adverse effects) required clearer definition in the context of critical loads for all the main receptors currently being considered.

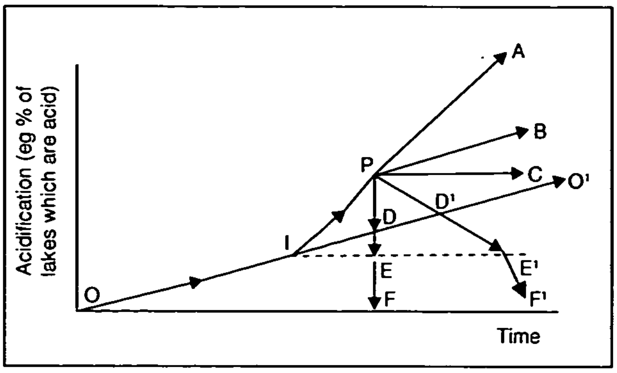

Figure 1. Conceptual representation of acidification and recovery of soils and surface water. $O - O^1$ = natural acidification; I = Industrial Revolution; P = present; P - A = no action; P - B = reduce to natural rate of acidification; P - C = no change in present status; P - D = reduce to natural state; P - E = restore to pre-industrial state; $P - D^1$ and $P = E^1$ = and, if so, how fast?

Question 8 *What should be the aim of the critical loads approach, given that most of Europe is not in a pristine state?*

The group first discussed the slope $0 - 0^1$ (Figure 1) in the context of the natural acidification of waters and soils. It was agreed that, based on diatom records over the past 200-300 years, and comparisons of the properties of waters and soils in pristine areas, the slope 0 - 1 for waters would be close to zero but for soils it would show a more positive increase. The slope I - P reflected the significant increase caused by post-industrial acidification in soils and water.

The group then discussed what the final acidification status of soils and waters would be, given the current approach for calculating critical loads. It was suggested that waters and soils be considered separately.

Waters – standing

Critical load calculations using the Henriksen, MAGIC or diatom model did not maintain the *status quo*, but restored waters to pristine levels along the route $P - E^1$. The rate of recovery was difficult to predict but recoveries, due to current reductions of SO_2 emission, had been measured in Galloway lochs over the timescale of a decade. The diatom method, which could be modified to give target loads based on the point of first change of diatom flora, would provide site-based information using a variable ANC_{crit} at each site.

Alternatively, the fixed ANC_{crit} used in the Henriksen method, and set at a value of zero, was felt to be appropriate as it did not produce any negative critical loads, whereas suggested values of 20 or 50 would produce a significant number of negative values.

Waters – running

It was recognised that critical loads may be lower in sensitive upland streams draining into standing waters. However, the current methods would allow for critical load calculations even during acid episodes, although there was no method validation for the diatom method.

The advantages of the freshwater methods were that:

- chemical changes were easier to measure;
- the slope P - E^1 was likely to be much steeper than soils, giving a quicker recovery;
- they incorporated land use change into the calculations.

Soils

The first point to be made was that the soil methods were generally conservative and would only maintain the *status quo* along line P - C. It was nevertheless felt that the peat maps developed in the UK did allow for some degree of recovery along P - E^1, thus explaining their greater sensitivity. Suggested timescales of recovery were long (100 years). The group discussed in detail the possibility of using a fossil indicator to identify the point of change (1) in soils, but no suitable organism was identified.

Changes in trace metal levels were also suggested as a possibility. The group was convinced that the land use aspect should be included in determining soil critical loads. Maps with and without land use influence should be available, with an indication of how each type of land use influenced the soil classification.

Finally, the group discussed the possibility of using models to estimate pre-industrial base cation weathering rates '0', thus identifying point 1. Validation would be extremely difficult.

Conclusion

The group concluded that the aim of the critical loads approach should be, whenever possible, to return waters and soils to their pre-industrial status.

The extent of any recovery should be carefully monitored and compared with both modelled changes and the current biological and chemical status of pristine ecosystems on the fringe of western Europe.

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The concept of 'critical loads' is of great ecological, economic and political importance. Although a simple concept – the threshold at which a pollutant load causes harm to the environment – turning this basic idea into scientifically acceptable criteria is an ambitious undertaking. Nevertheless, governments are pressing ahead with European legislation on air pollution based on acceptable critical loads. However, there has been little opportunity for the ecological community as a whole to participate in these vital discussions on European policy, or for those who have been involved to present the results of their deliberations to their scientific peers. To help remedy this situation, the Institute of Terrestrial Ecology organised a Conference in 1992 for all interested parties. It generated new ideas, re-assessed existing views, and sparked some controversies. This book documents the proceedings of the Conference and will be invaluable in the further development of the critical loads concept.

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