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MODELLING STUDIES ON SOIL-MEDIATED RESPONSE TO ACID DEPOSITION AND CLIMATE VARIABILITY

Maria Holmberg

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Title: Modelling studies on soil-mediated response to acid deposition and climate variability

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Abstract: The impact of acidifying atmospheric precipitation and climate variability on forest soil was studied using three approaches: (i) dynamic process-oriented modelling, (ii) static vulnerability assessment, and (iii) non-linear response pattern identification. The dynamic soil acidification model MIDAS is presented, with applications at the plot and catchment scale in Norway, Sweden and Finland, which show that growing vegetation contributes to soil acidification and which illustrate that the recovery phase is not symmetrical to the acidification phase. I report the results of an analysis using the SMART acidification model and the DEPUPT nutrient uptake model for selected deposition and forest growth scenarios at the Integrated Monitoring site of Hietajärvi, eastern Finland. The results show the importance of how the present day deposition is estimated. A combinatory matrix approach is described that uses regional data together with expert judgement to provide an assessment of groundwater sensitivity to acidification in Europe, without the need for detailed mathematical process formulations. I also demonstrate the variability that is introduced in the Finnish critical loads of sulphur for forest soils by using alternative criteria and by extending the critical loads model to include organic complexation of aluminium and the leaching of organic anions. The impact of climate variability on runoff water quality is illustrated with an empirical stream water model that builds on artificial neural networks for reproducing patterns in the observations of TOC, N_{tot} and P_{tot} at Hietajärvi, and also at Valkea-Kotinen, an Integrated Monitoring site in southern Finland. The stream water model is used to predict changes in element fluxes from these forested catchments in response to climate change.

Keywords: dynamic soil model, groundwater sensitivity matrix, artificial neural network, empirical stream water model, acidification, climate change

Academic dissertation

Systems Analysis Laboratory
Helsinki University of Technology

Modelling studies on soil-mediated response to acid deposition and climate variability

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Publications

The dissertation consists of the present summary article and the following papers:

[I] Holmberg, M., Hari, P. and Nissinen, A., 1989. Model of Ion Dynamics and Acidification of Soil: Application to Historical Soil Chemistry Data from Sweden. In: Kämäri, J., Brakke, D.F., Jenkins, A., Norton, S.A. and R.F. Wright (Eds.), Regional Acidification Models. Geographic Extent and Time Development, pp. 229-241. Springer-Verlag, Berlin.

[II] Holmberg, M., 1990. Model of Ion Dynamics and Acidification of Soil: Simulating Recovery of Base Saturation. In: Fenhann, J., H. Larsen, G.A. Mackenzie & B. Rasmussen (Eds.), Environmental Models: Emissions and Consequences, pp. 359-368. Elsevier Science Publishers B.V., Amsterdam..

[III] Warfvinge, P., Holmberg, M., Posch, M. and Wright, R.F., 1992. The Use of Dynamic Models to Set Target Loads. Ambio 21:369-376.

[IV] Holmberg, M., Rankinen, K., Johansson, M., Forsius, M., Kleemola, S., Ahonen, J. and Syri, S., 2000. Sensitivity of soil acidification model to deposition and forest growth. Ecological Modelling 135:311-325.

[V] Holmberg, M., Johnston, J. and Maxe, L., 1990. Mapping Groundwater Sensitivity to Acidification in Europe. In: Kämäri, J. (Ed.), Impact Models to Assess Regional Acidification, pp. 51-64. Kluwer Academic Publishers, Dordrecht.

[VI] Holmberg, M., Mulder, J., Posch, M., Starr, M., Forsius, M., Johansson, M., Bak, J., Ilvesniemi, H., and Sverdrup, H., 2001. Critical loads of acidity for forest soils: Tentative modifications. Water, Air, and Soil Pollution: Focus 1: 91-101

[VII] Holmberg, M.., Forsius, M., Starr, M. and Huttunen, M. An application of artificial neural networks to carbon, nitrogen and phosphorus concentrations in three boreal streams and impacts of climate change. To appear in Ecological Modelling.

Contributions of the author

Holmberg developed the MIDAS model in I, II and III, applied the DEPUPT model in IV, and designed the artificial neural networks in VII. In I and IV she carried the main responsibility for outlining the work, analysing the results, drawing the conclusions and writing the article. She participated in writing III. Holmberg initiated the matrix approach to groundwater sensitivity in V and the article was written jointly by the authors. She implemented the modifications to the Finnish critical loads calculations in VI and co-ordinated the documentation of the work. In VII, Holmberg had the main responsibility for structuring the work and analysing the results, while all authors joined in drawing the conclusions and writing of the article.

Preface

The wild cat stalking its quarry inhibits the desire to spring prematurely, and controls to a deliberate end its eagerness for the instant gratification of a natural appetite.

F.M. Alexander (Man's Supreme Inheritance, Mouritz, 1996, London, p. 23).

I wrote this summary while employed by the EU-CNter project (QLK5-2001-00596) at the Research Programme on Global Change at the Finnish Environment Institute. Dr. Martin Forsius supported and inspired my work, both as Programme Manager and as enthusiastic and talented colleague, generously sharing his knowledge and visions. I much appreciate his suggestion we should look into neural networks. Martin contributed structure and clarity to the articles he co-authored and I appreciate also his comments to this summary.

Director General Lea Kauppi, Research Director Juha Kämäri and Programme Director Mikael Hildén contribute to the stimulating atmosphere at the Finnish Environment Institue (SYKE). I thank Lea and Juha in particular for they engaged me to work, first with acid sulphate soils, at SYKE's predecessor the National Board of Waters and the Environment, and later gave me the opportunity to continue on the series of articles that constitute this thesis.

I thank warmly all my colleagues at SYKE for kindly sharing their understanding and friendship, particularly Pirkko Kortelainen, Sirpa Kleemola, Tim Carter, Matti Johansson, Pekka Vanhala and Jussi Vuorenmaa. My warmest thanks are extended to Sirkka Vuoristo for skilful graphical work and to Ritva Koskinen for problem-free layout work. I am grateful also to Roger Munn, who checked the language of an early version of the summary. All remaining bugs have crept in after he saw the text.

My sincere thanks are due to Professor Pekka Kauppi and Research Professor Ilkka Savolainen for their constructive criticism in the preliminary examination.

The collaboration with my co-authors was very enjoyable and I thank them all warmly for generously giving their expertise and contributing time and effort to the articles: Pertti Hari, Ari Nissinen, Per Warfvinge, Maximilian Posch, Richard. F. Wright, Katri Rankinen, Matti Johansson, Martin Forsius, Sirpa Kleemola, Johanna Ahonen, Sanna Syri, John Johnston, Lena Maxe, Jan Mulder, Michael Starr, Jesper Bak, Hannu Ilvesniemi, Harald Sverdrup and Markus Huttunen.

Professor Pertti Hari, University of Helsinki, twenty years ago, gave me the standards of realism, focus and honesty in science. I am grateful for Pepe's teaching and encouragement, and no, it's not only because of his high standards that this took me so long. I appreciate also his helpful comments to this summary. I thank all my former colleagues at the University of Helsinki, especially Hannu Arovaara, Hannu Ilvesniemi and Ari Nissinen. In particular Annikki Mäkelä's knowledgeable presence and artless friendship was, and continues to be, enjoyable.

If these pages reflect some understanding of the behaviour of boreal forest soils, the origin lies in 18 years of instructive discussions with Michael Starr. The misinterpretations are, naturally, my own. I thank Mike also for meticulous review, not only of the two papers he co-authored, but also of article IV. Co-operation with Max Posch is always a gift and I thank him for the professional perseverance he brings into it, mixed with large doses of cultural and linguistic humour. To Dick Wright I am grateful both for professional advice and kind friendship. I thank Johanna Ahonen, Katri Rankinen and Sanna Syri for insightful discussions concerning modelling and Markus Huttunen for contributing advice on neural networks and hydrological model results to article VII.

Articles I and II were written while I was a research assistant of the Academy of Finland at the Department of Silviculture, University of Helsinki. Funding by the Ministry of the Environment and the Ministry of Agriculture and Forestry through the Finnish Acidification Research Programme HAPRO is also gratefully acknowledged. I appreciate warmly Per Warfvinge's efforts as lead author of article III, to which the Air Group of the Nordic Council of Ministers contributed funds.

The EU Financial Instrument for the Environment (LIFE) financed the work on article IV within the project LIFE95/FIN/a11/EPT/387. A summer in the YSSP programme and a spring on a Peccei scholarship at the International Institute for Applied Systems Analysis (IIASA), provided the background for article V. I thank IIASA, and especially Professor Leen Hordijk who was the inspiring leader of the Acid Rain Project. The Air group of the Nordic Council of Ministers gave financial support also to article VI. Work on article VII has been financed by the Finnish Global Change Research Programme FIGARE, Academy of Finland, Ministry of Agriculture and Forestry) and the project CNter (QLK5-2001-00596, EU 5th framework).

Professor Raimo P. Hämäläinen provided an engaging studying atmosphere, for which I am most grateful. My warmest thanks are due also to Professor Esa Saarinen, whose lectures last spring helped me rediscover the joy in writing. I am sincerely grateful to my Alexander teacher Richard H. Gilbert, who showed me how to look for what happens between stimulus and response and gave me the means whereby I could write this thesis. I feel gratitude towards all fellow Alexander teachers who generously have shared their insights on conscious constructive control, especially Soile Lahdenperä and Reinaldo Renzo, and I thank particularly Matti Harilo, Auli Hohenthal, Christina Huldén and Stiina Kaikkonen, for ample opportunities to practice inhibition.

I am deeply grateful to my parents for the innumerable aspects of life and caring, and to Birgitta Buttenhoff and Tor-Erik Holmberg, with families, for generously sharing their lives with me. I thank my sister especially for her encouraging attitude of never giving up and my brother for warmly cherished child-hood memories of the sound of waves meeting the bow of a sun-warmed dinghy.

With love and gratitude to my most demanding critics and warmest supporters: Christoffer, Lars and Leni Sundman.

Helsinki, October 2003 Maria Holmberg

Contents

1 Introduction

1.1 Context

The purpose of this thesis is to discuss the strengths and limitations of model-based reasoning used to study the environmental effects of human activities. My arguments are based on experiences gained while developing and applying three types of methods to assess the soil-mediated response to acid deposition and climate change:

(i) dynamic modelling of soil acidification;
(ii) static assessment of regional acidification risk; and
(iii) empirical modelling of stream water quality with artificial neural networks.

Articles I to VII give the details of these modelling studies. The spatial scales of the models range from individual soil plots to a Europe-wide grid (Fig. 1) and they are formulated as (i) dynamic models (I,II,III,IV); (ii) static assessment methods (V,VI) and (iii) artificial neural networks (VII).

The work reported in the articles was carried out at the Department of Silviculture of the University of Helsinki, the International Institute of Applied Systems Analysis (IIASA) and the Finnish Environment Institute. The work was part of the Finnish Acidification Research Programme HAPRO

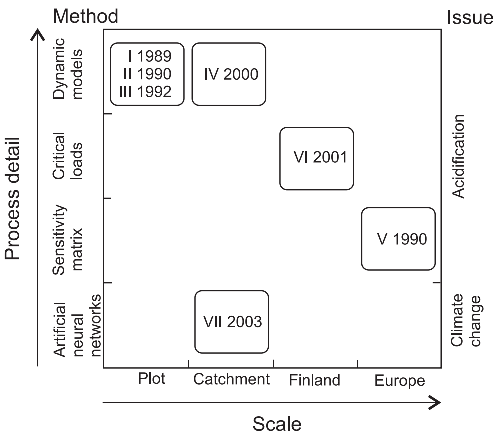

Figure 1. Schematic view of the distribution of process detail and scale of the studies discussed in this summary.

(Kauppi *et al.* 1990) and the Finnish Global Change Research Programme FIGARE (Käyhkö and Talve 2002), and contributed to two research projects by the Nordic Council of Ministers Air Group (Wright *et al.* 1991, Holmberg 2000), an EU-LIFE project (Forsius *et al.* 1998b), as well as to the EU-CNTER project (Gundersen *et al.* 2002).

In this summary, I present conclusions concerning the balance between simplicity and process detail in model development and argue for transparent and accessible methods in environmental assessment directed at backing up international agreements.

1.2 Background

Modelling

A model is a mapping of one set of reality onto another. The mapping, or function, may be verbal, mechanical, mathematical or algorithmic. Models are developed to provide an alternative view on the subject matter, because the phenomenon is too extensive spatially or temporally or by nature too complex to be grasped by direct observation. Practical implementations include forecasts under different scenarios.

By looking for a formulation of the central processes causing the phenomenon the investigator wants to be able to learn more about what factors determine the response of the system or to say something about how the phenomenon is likely to develop over time or in space. This knowledge may then highlight questions or processes that merit more specific research because too little is known about their cause-effect relationships.

The model may also assist decisions concerning the control - either private or societal, local or regional - of the variables that drive the system's response. Triggered by the concern about how man's activities affect the environment mathematical models are applied to study the relative importance of different driving variables and to analyse the consequences of various future pathways of, for example, energy use.

Facilitated by the exponential increase of computational capacity, numerical solutions of mathematical formulations are used to quantify the environmental effects of energy production and use. Since the 1980s, models are used also to assist decision-making as integral parts of assessment frameworks with the purpose of optimizing societal actions restricted by economic and environmental constraints (Alcamo *et al*. 1987, Alcamo *et al.* 1990, Bull 2001).

Acidification and climate change

Burning fossil fuels releases sulphur into the atmosphere and if combustion takes place at high temperatures nitrogen oxides are emitted, too, while nitrogen in manure from animal farming adds to atmospheric ammonium. The concentrations of sulphur and nitrogen compounds in air are highest near the emission sources but since these substances form aerosols and travel long distances with air masses their concentrations may be raised even in areas having no major point sources.

Vegetation surfaces filter out atmospheric sulphur and nitrogen and rain brings them to the ground, where they engage in the biological and chemical processes that form the quality of soil, groundwater and surface water leading to acidification. The problem of acidification was one of the central concerns in environmental research during the last few decades (Ulrich 1983, van Breemen *et al.* 1984). The emphasis was initially on areas in Europe and North America, followed more recently by an awakening to the global extent of acidification (Bouwman *et al.* 2002).

Sulphur emissions in Europe declined since 1980 (Fig. 2, Vestreng 2003). A decrease in global emissions of acidifying compounds is projected (Nakicenovic and Swart 2000), although not yet seen (Galloway 2001). In Finland, a large-scale restructuring of energy production and industry in combination with investments to comply with national and international requirements that aimed at improving air quality brought the SO_2 emissions down to 73 500 t yr^{-1} in 2000, which is less than fifteen per cent of what was emitted in 1980. Nitrogen emissions continued to increase until 1987 in Finland but then decreased by twenty five per cent to 210 000 t yr^{-1} in 2000, while ammonia emissions in the 1990s went down approximately ten per cent to 33 000 t yr^{-1}.

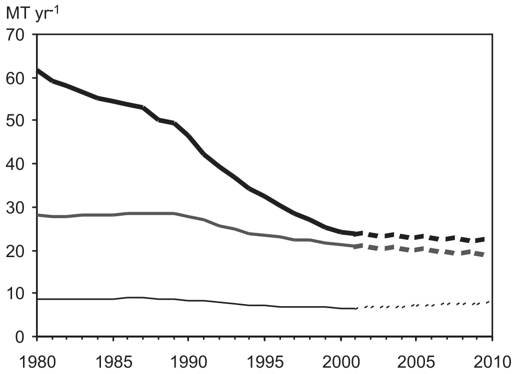

Figure 2. European emissions of SO2 (upper), NOx (as NO2, middle) and NH3 (lower line), with reported values 1980 - 2001 and projected values 2002 - 2010. Drawn from data in Vestreng, V. 2003. Review and Revision. Emission data reported to CLRTAP. MSC-W Status Report 2003. Meteorological Synthesizing Centre-West. Norwegian Meteorological Institute. Oslo. 134 pp.

By 2010, Finland's annual emissions ought to be below 110 000 t sulphur, 170 000 t nitrogen oxides, and 31 000 t ammonia (Ministry of the Environment 2002). The role of nitrogen oxides emissions in the impact of transboundary air pollution will increase in Europe in the future (Johansson *et al.* 2001a). With decreasing sulphur deposition, European and North American lakes show clear signs of recovery from acidification (Stoddard et al. 1999, Mannio 2001, Skjelkvåle *et al.* 2001). In response to sulphur deposition levelling off at around 1999 a slower acidification recovery of Finnish lakes is anticipated in the future (Forsius *et al.* 2003).

Atmospheric concentrations of carbon dioxide, methane, nitrous oxide and halocarbons increased strongly during the last century, giving rise to the greenhouse effect and its associated changes in climate. Although some sources indicate a natural origin for the observed changes in climate (Saarnisto et al. 2002), evidence suggesting major anthropogenic responsibility for perturbations of climate is widespread enough (Houghton et al. 2001) to motivate international efforts to slow down the increase in greenhouse gases in the atmosphere, and to stimulate innovations in the design of industrial and energy production processes low in carbon dioxide emissions (Soimakallio and Savolainen 2002). Whether the variability in temperature and precipitation is driven by natural causes or by human activity, changes in climate will affect many ecosystem processes, including fluxes of elements from forested catchments.

Dynamic soil models

Soil plays a central role in the response of forests and lakes to air pollution and climate change. In remote areas, where the long-range transport of pollutants dominates over point sources the atmospheric SO_2 concentrations are comparatively low and the direct effects on forest canopies and surface waters are small in comparison to effects mediated through changes in soil. Forest soil is a complex aggregate of mineral and organic components, water and air. It can be considered and described as a dynamic system in which its state is determined by the interaction of a number of geochemical, biological and physical processes.

Soil is a system of three phases: solid, aqueous and gaseous. The solid phase consists of extensive mineral and organic components with large reactive surfaces, which are covered with a thin aqueous film even when the soil is dry. The extensive reservoirs of elements in the mineral and organic components, combined with the large reactive surfaces give rise to long response times of the soil system. Because of the long response time, experimenting and monitoring in the field or laboratory give only limited results within a few decades, which is a long time in comparison with most research projects.

The desire to better understand the reactions and processes of plant nutrients in soils drove most of the work in soil chemistry from 1850 to 1970 (Sparks 2001). Important progress in the theory of cation exchange in the 1950's was followed by intense discussion about the role of H and Al in soil acidity in the 1960's (Sparks 2001). This development in soil science laid the foundation for a theoretical framework in which it became possible to study the impact of the deposition of acidifying substances on soils.

Reuss (1980) formulated the basis of later soil acidification models using established principles of soil chemistry. He predicted that changes in soil acidity and base cation status may occur much later and continue far longer than the major acid input, due to the dampening effect of sulphate adsorbing properties of the soil (Reuss 1980). The MAGIC model (Cosby *et al*. 1985a, Cosby *et al*. 1985b) began as an extension of the conceptual approach by Reuss and Johnson (1986). It was later developed to include also reactions involving organic acids (Cosby *et al* 1995). MAGIC is applied to predict acidification and recovery, land-use change and climate change (Ferrier *et al.* 1993, Wright *et al.* 1998, Cosby *et al.* 2001). The PROFILE model (Sverdrup and Warfvinge 1988) simulates silicate weathering in different soil horizons and the SAFE model (Warfvinge *et al.* 1993) uses its results to simulate the dynamics of soil acidification in several layers of soil.

The SMART model (de Vries *et al*. 1989) was developed as an improvement of the first soil submodel of the RAINS framework (Kauppi *et al.* 1985, Alcamo *et al.*1987), which was based on the concept of buffer ranges (Ulrich 1983). SMART is used to study the response of soil and water to acid deposition and forest harvesting in Finland, both in its basic form (Kämäri *et al.* 1995, Forsius *et al.* 1997), and extended to include nutrient cycling (Ahonen *et al.* 1998, Kämäri *et al.* 1998, Ahonen and Rankinen 1999), and geographic information system data (Bilaletdin *et al.* 2001).

Short-term variation in stream water chemistry in response to acid deposition was first modelled by Christophersen *et al.* (1982) using a two-reservoir hydrologic model with a chemical submodel. The MIDAS model in articles I, II and III of this thesis is a simplification of a vertically distributed model, developed by coupling a modified version of the kinetic cation exchange submodel by Oksanen *et al.* (1984) to an adaptation of Pingoud's (1982) infiltration model (Holmberg *et al* 1985). MIDAS is described in detail in Holmberg (1990). Development of the ACIDIC model (Kareinen *et al.*1998) was inspired by experiences with MIDAS.

Gobran and Bosatta (1988) used the approximate kinetics method introduced by Bosatta (1983) to study the time at which the shift to aluminium dominance in soil solution occurs and they conclude that pH is not an appropriate variable to use as an indicator of soil changes. Ågren and Bosatta (1996) developed a model to simulate the decomposition of soil organic matter with applications for climate change.

The effects of emission reduction scenarios at integrated monitoring sites in Europe were studied with the models MAGIC, SAFE and SMART (Forsius *et al.* 1998a, Forsius *et al.* 1998b). SMART was used in article IV to combine the effects of forest growth and acidifying deposition. SMART modelling also inspired procedures for calculating critical loads, both in a static (Posch *et al.* 2001a) and a dynamic framework (Schmiemann *et al.* 2002), and forms the basis of the development of dynamic calculations of regional critical loads (Posch *et al.* 2003).

The temporal development of the state variables (base saturation, soil solution chemistry) of the dynamic soil models is driven by information about historical and future deposition of sulphur and nitrogen. The deposition model DAIQUIRI is used in Finnish studies to produce time series of values for the driving variables that are calculated by modelling the transport and deposition of sulphur (Johansson *et al.* 1990) and nitrogen (Syri *et al.* 1998), while accounting for both dry and wet deposition (EMEP/MSC-W 1998). DAIQUIRI is described in detail by Johansson (1999) and Syri (2001).

Steady-state methods

The sensitivity of an ecological system is a measure of its response to variations in certain external driving functions. Apart from the driving variables, certain physical system characteristics, or sensitivity indicators, determine the amplitude of the response and the response time of the system. If the sensitivity indicators are chosen such that they are independent of time and of the driving functions, it is possible to differentiate between the impact of physical characteristics on one hand and the temporal development of the driving variables on the other.

When combined with the values of the driving functions at one or several points in time, the static sensitivity assessment yields an assessment of the risk of a certain response as a snapshot or a continuous development in time. Article V presents an assessment of the sensitivity and risk of acidification of groundwater in Europe.

In Europe, decisions on large-scale reductions of acidifying emissions are made on the basis of critical loads of sulphur and nitrogen, mostly for forest soils, but in some areas also for lakes (Hettelingh *et al.* 2001, Posch *et al.* 2001a). The critical load is defined as a quantitative estimate of the loading of one or more pollutants below which significant harmful effects on specified elements of the environment are not likely to occur according to present knowledge (Nilsson and Grennfelt 1988).

The introduction of the critical loads concept made it feasible to account for environmental effects in determining reduction targets. The second Sulphur Protocol, signed in 1994, aimed at cost-efficient emission reductions with the environmental targets determined in terms of critical loads. Using the tree as the biological indicator organism, which is assumed to respond with poorer health or reduced growth to increasing aluminium concentrations in soil solution (Sverdrup and Warfvinge 1993), a limit is set to the leaching of acid neutralising capacity in order to derive the maximum allowable acidifying deposition. Critical loads are calculated with the mass balance model SMB (Umweltsbundesamt 1996, Posch *et al.* 1999). The critical loads are fed into the integrated assessment model RAINS (Alcamo *et al.* 1990, Amann *et al.* 1995, Schöpp *et al.* 1999), which gives regional information on the most cost effective way of reducing deposition.

Recent abatement strategies for sulphur and nitrogen emissions in Europe and North America are determined employing critical loads to set emission targets (UNECE 1999, European Commission 1999). The framework for evaluating critical loads for Finland was set up by Johansson (1999), building on earlier modelling studies on acidification (Johansson *et al.* 1990) and critical loads (Forsius *et al.* 1992). The total uncertainty of integrated assessment modelling of acidification is dominated by uncertainty in the critical loads calculations (Syri *et al.* 2000), which implies that the critical loads methodology is among the first modules that should be revised in the integrated assessment framework.

Acidifying deposition over Europe was substantially reduced since its peak in the 1970s primarily as a result of the widespread introduction of technological solutions that allow combustion with lower emissions. The shift towards cleaner energy production occurred in response to international agreements and national legislation triggered by concern about the environmental effects of soil and water acidification. The concept of critical loads is the central tool for assessing the link between effects and deposition, widely used to translate the environmental protection criteria into regional emission reduction requirements.

It may be argued, in the case of acidification, that it is more important to develop and apply a method that allows the emission reductions to be motivated and quantified than to formulate a scientifically objective definition of the cause-effect relation between acidifying emissions and environmental effects. A similar role for science, society and institutions may be seen in the context of sustainable development (Hukkinen 1998). The pragmatic application of critical loads succeeded in reducing acidifying deposition in Europe. Nevertheless, a methodological analysis is relevant in order to increase the understanding and the scientific basis for further actions. Criticism of the critical loads concept (Cronan and Grigal 1995, Løkke *et al.* 1996) triggered a study to explore the consequences of the assumptions in calculating critical loads for forest soils. The results of that study are documented in article VI.

Artificial neural networks

Fluxes of elements that enter soil via precipitation alter the chemical concentrations of runoff in response to complex hydrological, geological, biological and chemical processes in catchment soils. The full range of operation of these responses is difficult to grasp with process-oriented models. Few models encompass all the relevant processes and if they do cover the key phenomena, the parameterisation and application to other sites than those the models were developed for is rarely successful. Instead, information may be extracted from the data by methods directed at exploring the information contained in the data, especially time series analysis (Kantz and Schreiber 1997), spectral analysis (Kirchner *et al.* 2000) or artificial neural networks (Clair and Ehrman 1998, Lischeid 2001a,b, Aitkenhead *et al*. 2003).

Artificial neural networks (ANNs) are global non-linear models (Kantz and Schreiber 1997). They are represented by mathematical algorithms that form a non-linear mapping between a set of input variables to a set of output variables. They developed from simplified models of how neural signals propagate in living organisms (Freeman 1994, Bishop 1995, Kohonen 1995, Cotterill 1998). The key elements of an ANN are the nodes, which mimic neural synapses. In the nodes, algorithms combine a linear weighted sum of the input variables with a non-linear transfer function to yield the output variable(s). The weights are chosen by an iterative algorithm with the aim of minimizing the diffference between the calculated and the desired output.

Artificial neural networks are applied to rainfall-runoff modelling (Shamseldin 1997) and to predict the response of hydrology and geochemical fluxes to changes in climate (Clair and Ehrman 1998). Levine and Kimes (1998) extrapolated soil organic carbon spatial data sets with artificial neural networks. Lischeid used artificial neural networks to study short-term dynamics and long-term trends in hydrochemical time series (Lischeid *et al.* 1998, Lischeid 2001a, Lischeid 2001b). There are also examples of rainfall forecasting (Luk *et al.* 2000) and environmental time series prediction (Aitkenhead *et al.* 2003) with ANNs. Non-linear relationships in ecological variables are better captured with artificial neural networks than with linear multiple regression (Gevrey *et al.* 2003). An application of artificial neural networks to simulate stream water quality is presented in article VII.

Model evaluation

Although it may not be possible to demonstrate the predictive reliability of any model of a complex natural system in advance of its actual use (Oreskes *et al* 1994, Oreskes 1998), it is clear that much information can be gained by a thorough assessment of model quality. Several authors give guidelines for testing model results against observations (Klepper and Hendrix 1994). The suggestions include recommendations for careful use and interpretation of quantitative techniques and qualitative estimation (Janssen and Heuberger 1995), and ideas of how to overcome the uncertainty of experimental data (Monte *et al*. 1996). Also methods for testing hypotheses against experimental data instead of the goodness-of-fit are suggested (Loehle 1997). Tiktak and van Grinsven (1995) recommend that simulation results from simple, lumped parameter models should be compared with those obtained by detailed, mechanistic models that, in turn, can be compared with results from field observations.

Decisions to use models for predictive or management purposes may be based on subjective evaluation, personal preferences or difficulties of implementation, instead of an objective analysis of model performance (Vanclay and Skovsgaard 1997). The responsibility for providing information about model behaviour for as objective choices as possible lies with the modellers. Vanclay and Skovsgaard (1997) list five tasks to examine in order to create structure in the often chaotic terrain of model evaluation: logic and bio-logic; statistical properties; characteristics of errors; residuals; and sensitivity analyses. In acid rain modelling, uncertainty and sensitivity analyses are not standard procedures (Hordijk and Kroeze 1997), not even for integrated assessment models that are developed as tools to assist policy makers in evaluating different abatement options.

Acidification models are often poorly identifiable, because of long time scales and the complex nature of the processes involved. A major restriction to a stringent evaluation of soil acidification models is the discrepancy between the time scales of models and the available observations of soil properties. Because of the nature of the soil processes, models operate in the time period of 1800 or 1900 to 2030 or 2100, whereas there are very few series of soil data that extend in time over ten years or include observations from more than a few points in time. When dynamic soil models first were applied to study acidification (Reuss 1980, Oksanen *et al.* 1984, Cosby *et al.* 1985a), systematically monitored soil data were not widely available. Long term soil monitoring is still not frequent. By now, there are many more predictions of soil acidification or recovery (for example de Vries *et al.* 1989, Warfvinge *et al.* 1993) but I am not aware of any studies revisiting earlier model predictions in the light of recently reported soil data. Revisiting earlier predictions is interesting because the models were, at the time, calibrated totally independently of data that is now available.

1.3 Aims

The aims of this thesis are to develop and apply methods for determining the soil-mediated response to changes in the atmosphere. Modelling studies documented in articles I-VII were directed at estimating the impact of acidifying compounds (sulphur, nitrogen) and climatic variables (temperature, precipitation) on boreal forest soil and stream water. The mathematical representations were (i) differential equations (I,II,III); (ii) algebraic equations (IV,VI); (iii) piecewise linear functions (V) and vector functions (VII). The models were used to (i) study relative impacts of different driving variables (I,II,III,IV); (ii) explore the constraints of an integrated assessment of control options (V,VI); (iii) test a novel method of reproducing empirical observations (VII). These models deal with the impacts of acidifying deposition on forest soil (I,II,III,IV,VI) and on groundwater (V), and with the response of stream water quality to changes in climate (VII).

The advantages and drawbacks of the approaches are discussed after presenting the results of applying the different methods to answering seven questions, using articles I – VII together with some new material presented in this summary. The questions are:

1) what is needed to predict the trajectories of soil acidification and recovery? (I, II, III, IV)
2) how do the old predictions compare with new observations from Skåne, Sweden (I, II)
3) how do the old predictions compare with new observations from Birkenes, Norway (III, summary)
4) what are the relative impacts of forest growth and deposition on soil base saturation? (IV)
5) what is needed to assess sensitivity and risk of groundwater acidification in Europe? (V)
6) how do alternative assumptions change the results of critical loads of Finnish forest soils? (VI)
7) how can stream water concentrations of TOC, N_{tot} and P_{tot} be modelled empirically? (VII)

2 Materials and methods

2.1 Dynamic modelling at the plot and catchment scale

2.1.1 Models

The single-layer dynamic soil model MIDAS (Model of Ion Dynamics and Acidification of Soil) was developed (I,II) to simulate the temporal development of base saturation and ions in soil solution in the uppermost layer of mineral soil. MIDAS falls into a category between the kinetic model of Gobran and Bosatta (1988) and the equilbrium model of Reuss and Johnson (1986) and its followers (Cosby et al. 1985a, de Vries et al. 1989). The fractions of base cations (Ca^{2+},Mg^{2}) and acid cations (H^{+},Al^{3+}) on the cation exchange complex and the soil solution concentrations of these ions are totalled on a charge equivalent basis to represent the state variables of the model (I).

Kinetic equations for cation exchange control the dynamics of the state variables, together with external driving variables that account for deposition, weathering and the net effect of biological turnover on base cations and hydrogen ions in soil solution (I, II). The values of the rate coefficients are chosen so that cation exchange is always faster than the transport of ions through soil. When cation exchange is in equilibrium, the kinetic equations are reduced to the expression presented by Gaines and Thomas (1953). Soil depth and volume wetness are fixed parameters that are used to calculate the residence time of soil solution.

In MIDAS, constant rate weathering replenishes the pool of base cations on the soil exchange complex and consumes an equivalent amount of H^{+} (Fig. 3). Decomposition of organic material releases base cations into solution and consumes H^{+}. Nutrient uptake by plants consumes base cations and releases H^{+}. The apportioning of acid cations into H^{+} and Al^{3+} in soil solution is calculated from an equilibrium with a solid hydroxide phase. The state equations are solved numerically by a fourth-order Runge-Kutta algorithm and the auxiliary equations by Newton-Raphson iteration (I, II). In article I, MIDAS was calibrated by altering the ion exchange equilibrium and changing the rate of nutrient uptake by vegetation until a fit with observed base saturation was achieved.

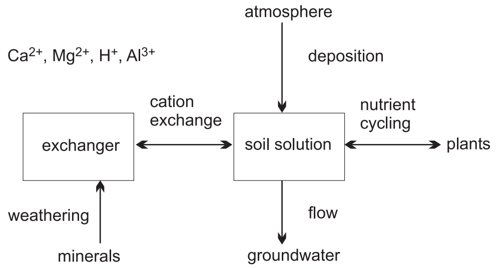

Figure 3. Pools and fluxes included in dynamic soil model MIDAS (From Holmberg, M., 1990. Model of Ion Dynamics and Acidification of Soil: Simulating Recovery of Base Saturation. In: Fenhann, J., H. Larsen, G.A. Mackenzie & B. Rasmussen (Eds.), Environmental Models: Emissions and Consequences, pp. 359-368. Elsevier Science Publishers B.V., Amsterdam).

MIDAS was used in articles I and II to simulate the dynamics of base saturation and pH using observations from soil plots in Skåne, Sweden (Falkengren-Grerup *et al.* 1987) and in article III to simulate base saturation in Birkenes, Norway, Stubbetorp, Sweden and Yli-Knuutila, Finland (Wright *et al.* 1991). In III, only base saturation results by MIDAS are presented because the soil solution concentrations predicted with MIDAS are not directly comparable to the stream water concentrations given by the other models MAGIC (Cosby *et al.* 1985a, 1985b), SMART (de Vries *et al.* 1989), and SAFE (Warfvinge *et al.* 1993).

SMART (de Vries et al. 1989) calculates base saturation and concentrations of major anions and cations in soil solution and catchment runoff. It was used in article IV to simulate the dynamics of base saturation under a set of assumptions concerning deposition and forest growth at Hietajärvi, Finland (Starr et al. 1998). In article IV, deposition scenarios based on domestic and international emission reductions were derived using the long-range transport matrices developed by the Meteorological Synthesising Centre-West (EMEP/MSC-W) of the Co-operative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (Barrett et al. 1995). Estimated historical sulphur (Mylona 1993) and nitrogen depositions (Asman and Drukker 1988, Alveteg et al. 1998) were used. These data were input to DAIQUIRI, a deposition model developed for studying how national and international emission reduction strategies influence deposition of sulphur and nitrogen in Finland (Johansson et al.1990, EMEP/MSC-W 1998, Syri et al. 1998). Historical and future values of annual deposition and forest nutrient uptake values for Hietajärvi were derived with DEPUPT (Johansson et al. 1996, Forsius et al. 1998a,b).

2.1.2 Sites

Skåne

Few data sets are available that provide analysis of soil acidification over the long term. Falkengren-Grerup *et al.* (1987) re-analysed old soil samples from 1949 using the same procedures as for the analysis of samples taken in 1984. The sites are located in Skåne, southernmost Sweden, in a region that received high levels of acid deposition in the latter half of the 20th century. Observations of cation exchange capacity and the amounts of exchangeable H^+, Al^{3+}, Ca^{2+} and Mg^{2+} were used to calibrate the MIDAS model (I,II). For that purpose, base saturation was calculated as the sum of Ca^{2+} and Mg^{2+} divided by the sum of Ca^{2+}, Mg^{2+}, H^+ and Al^{3+}. In this summary I report results pertaining to a beech site (no 3, Falkengren-Grerup *et al.* 1987) and compare them with newer observations from the same region (Falkengren-Grerup and Tyler 1991). The site is located in a beech forest growing on a well developed podzol with a mor layer thicker than 5 cm and an eluvial horizon of ca 15 cm, on a moraine formed by Cambrian sandstone. The spruce site is former arable land, first planted in 1889 and replanted in about 1965. The soil is developed from a sandy moraine and the humus layer is a mull (Falkengren-Grerup *et al.* 1987).

Birkenes

The soils at Birkenes (0.41 km^2), southern Norway, are mainly podzols and brown earths on granitic bedrock with an average depth of 0.4 m, covered by an 80-year old Norway spruce stand (III, Wright *et al.* 1991). Deposition is monitored at Birkenes since the 1970s (SFT 2002a) and base saturation is measured every five years since 1970 (SFT 2002b).

Stubbetorp and Yli-Knuutila

The Stubbetorp catchment (0.9 km^2) has a productive forest of Scots pine and is situated on the eastern coast of Sweden. It is dominated by bedrock of gneissic granites with a generally thin overburden comprised of stony till, with podzolic and brown forest soils of a mean depth of 0.8 m. Yli-Knuutila (0.07 km^2) in southern Finland has podzols and brown earths developed on clays and sands of an average depth of 1.5 m. The one hundred year old spruce and pine forest of Yli-Knuutila was later felled.

Hietajärvi

The forested catchment Hietajärvi (4.6 km^2) in eastern Finland is part of the network of the UN-ECE International Cooperation Programme on Integrated Monitoring (Bergström *et al.* 1995, Starr *et al*. 1998). The soils are Fibric Histosols, Haplic and Ferric Podzols, with a shallow fluctuating water table. The forests are mainly mature or old Scots pine.The estimated present day catchment average base saturation was 47%.

2.1.3 Acidification and recovery (I,II,III,IV)

Scenarios, Skåne

The deposition of SO_4-S for the years 1949 to 1984 at the south Swedish beech site was estimated with the Energy-Emission and Long-Range Transport Module of the RAINS model (Alcamo *et al.* 1987). No filtering was assumed for the beech stand and its sulphur deposition peak in 1980 was 1.6 gm^{-2}yr^{-1}. At the spruce site, however, the sulphur deposition was increased by a filtering factor of 1.6 compared to deposition to open field (Ivens *et al.* 1989). The deposition of calcium and magnesium was kept at a constant level of 0.2 g gm^{-2}yr^{-1} (I), which is comparable to 0.3 g gm^{-2}yr^{-1} bulk deposition of Ca^{2+} and Mg^{2+} in the Gårdsjön area (Grennfelt *et al.* 1985). The base cations leached from the canopy are part of the nutrient cycle and in the MIDAS application they are incorporated in the net production of base cations by biological activity.

The MIDAS model was calibrated to reproduce observed base saturation in 1949 and in 1984 at the Swedish site (I) and the calibrated model was used to simulate the response of base saturation to different levels of reduction of sulphur deposition from its peak in 1980. A ten per cent reduction, fully effective in 1985 was input to the model in (I); and in (II), deposition was linearly decreased by thirty, sixty and eighty per cent from 1980 to 1995. In this summary I compare the predicted rate of change of base saturation to that reported by Falkengren-Grerup and Tyler (1991).

Scenarios, Birkenes

With the soil data from Birkenes, Stubbetorp and Yli-Knuutila, MIDAS was compared to the models MAGIC, SAFE and SMART (III, Wright *et al.* 1991). Predictions of future base saturation were made with three scenarios: (i) a base case scenario with continued deposition at the 1987 level; (ii) minimum reductions: sulphur thirty and nitrogen ten per cent; and (iii) maximum reductions: sulphur fifty five and nitrogen forty per cent. The reductions were implemented by decreasing deposition linearly from 1987 to 2005, after which it was kept constant.

New soil data, Birkenes

Here I discuss the predicted base saturation values in the light of recent observations of deposition and soil properties at Birkenes (SFT 2002a, SFT 2002b). Values of exchangeable calcium and magnesium (meq kg^{-1}) from five layers or horizons in four plots were observed at Birkenes in 2001 (SFT 2002b). For these twenty layer-specific observations, I calculated the relative change from 1984 to 2001 in divalent base saturation, that is (Ca+Mg)/CEC. The maximum and minimum of these values were used to indicate a range of potential values for the catchment average divalent base saturation in 2001, to be compared with model simulations. In article III, target loads, or the level of deposition that satisfies the soil solution chemical criterion by the year 2037, were caculated with MAGIC, SMART and SAFE for Birkenes, Stubbetorp and Yli-Knuutila, and in this summary I will briefly mention the findings for terrestrial ecosystems.

2.1.4 Relative impact of driving variables (IV)

The SMART model was calibrated to data from Hietajärvi for the years 1988 to 1994. The values of 14 parameters were adjusted in order to obtain a visually satisfying fit with observations from six consecutive years of the values of eight components of stream water quality and one observation of soil base saturation (Bleeker *et al.* 1994, Ahonen *et al.* 1998). In article IV, three emission reduction scenarios, with three timing variations and two variations in the level of observed deposition, were combined with five forest growth scenarios to explore the relative impact of deposition and forest growth on the predicted future base saturation values.

Domestic and European emissions of 1990 approximate the average for the period studied and were used in DAIQUIRI, together with average meteorological data for 1985-1994 to calculate present-day deposition values. Emissions of SO_2, NO_x and NH_3 in the year 2010 were taken from different future alternatives investigated by the European Commission to define options for the international negotiations on emission reductions (Amann *et al.* 1996, Amann *et al.* 1997). The modelled historical and future depositions in the Hietajärvi region were derived from the country-level emissions of the EU Acidification strategy by using the long-range transport matrices provided by EMEP/MSC-W (Barrett *et al.* 1995) incorporated in DAIQUIRI. All modelled deposition time series were adjusted to observed present-day deposition, estimated from measurements of bulk, throughfall and open field deposition (Ukonmaanaho *et al.* 1998).

Uncertainty in the estimates of present deposition levels was examined by evaluating three sets of deposition values as representative of present conditions. First, the year of the lowest observed sulphur and nitrogen deposition (1993) was chosen as the calibration point for simulation ‘A’. Second, the year of the most acid deposition (1988) was used to calibrate past and future deposition values (‘E’). For all other simulations, the average observed values (1988-1994) were used for the present conditions. In five different forest growth reconstructions, the form of the growth curves and the present volume were varied. The objective was to represent extreme cases of present fast and slow growth. All forest growth scenarios were combined with the medium deposition scenario.

2.2 Static assessment of regional acidification risk

2.2.1 Groundwater sensitivity to acidification in Europe (V)

Sensitivity matrix

The impact of acid deposition on groundwater is determined by the neutralising properties of the overlying soil and those of the water-bearing body itself. Cation exchange and mineral weathering are the primary processes that contribute to neutralising acid deposition. The composition of the recharge to groundwater is influenced by the residence time of the water in the unsaturated zone, which increases with the depth and decreases with the permeability of the soil, and by the mineral composition and organic matter content of the soil. The residence time of the groundwater in the saturated zone determines the time available for neutralising reactions. Calcareous bedrock is easily weathered, whereas silicate bedrock weathers slowly.

A qualitative measure of the neutralisation capability of the overlying soil and the aquifer, or inversely, the acidification sensitivity, was estimated using a sequence of two-dimensional decision matrices (Fig. 4). The matrices represent best available scientific judgement of the synergistic effects of the processes that contribute to the acidification of groundwater in Europe. The basic geochemical and physical characteristics that influence the chemical behaviour of soil and groundwater were aggregated into ten indicators. The indicators were discrete variables that represent either a classification of the original continuous physical phenomenon (soil depth, texture) or a derived entity (neutralising capability). The total sensitivity was obtained by aggregating the individual indicators, and the risk of European groundwater acidification was estimated by combining the sensitivity of the aquifer with its deposition load.

European data input

Soil depth, texture and base cation content were compiled from the Food and Agriculture Organization (FAO) Soil Map of the World (1974). Aquifer size and mineral composition were taken from the International Hydrological Map of Europe (IAH-UNESCO 1970-1985). The area was subdivided by a grid system with individual cells of 1.0º longitude by 0.5º latitude. The individual grid cells were larger than some essential map features, such as individual soil types and aquifers, and up to seven soil types and six aquifers per grid were included in the input data set.

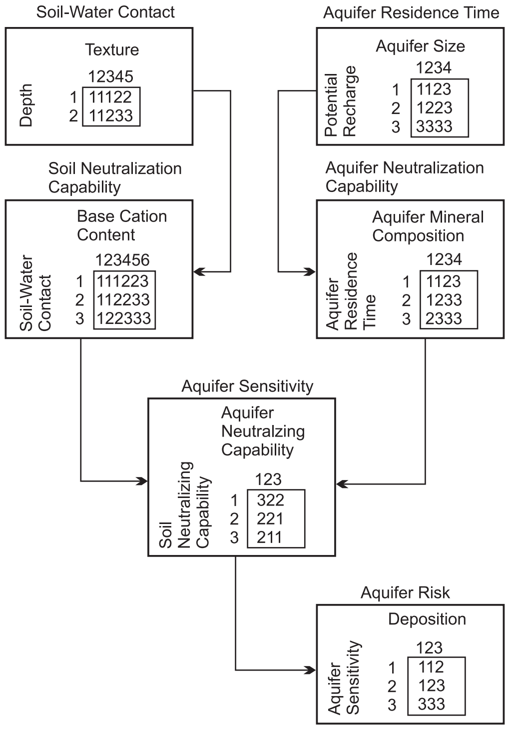

Figure 4. Matrix representation of piecewise linear functions to assess groundwater sensitivity and risk to acidification (From Holmberg, M., Johnston, J. & Maxe, L. 1990. Mapping Groundwater Sensitivity to Acidification in Europe. In: Kämäri, J. (Ed.), Impact Models to Assess Regional Acidification, pp. 51-64. Kluwer Academic Publishers, Dordrecht).

As the data were not related spatially within the grid cells, three approaches were used to choose the soil and aquifer data to represent the grid cells. First, the soil type and the aquifer with the largest area were combined to represent the dominant characteristics of the grid cell. To estimate the best and worst cases all soil types and aquifers covering at least 15% of the grid cell were systematically combined and the lowest and highest sensitivity values were recorded.

Data for each of the 1 844 grid cells were passed through the decision matrices and assigned a sensitivity class. The estimated 1980 sulphur deposition pattern (Alcamo *et al.* 1985) was used to derive the risk of groundwater acidification by combining deposition with sensitivity class for each grid cell.

Evaluation

The responses of the assessment method were evaluated by studying the output variations under different input conditions: concerning the assignment of indicators into classes; the choice of ratings used in the matrices; and the spatial representation of the data.

In article V, the results were not compared to observed groundwater acidification or other regional acidification risk assessments, since such studies were not available at the time. In this summary, I comment upon the usefulness of the method in article V in the light of results from a Europe-wide assessment of critical loads for forest soils (Posch 2002); a Czech study on groundwater quality and vulnerability (Hrkal *et al.* 2002); and Norwegian and Finnish groundwater monitoring observations (Banks *et al.* 1998, Backman *et al.* 1999). I also mention the results of Maxe (1999), who evaluated the performance of the method of article V in relation to a detailed Swedish groundwater study.

2.2.2 Critical loads of sulphur for forest soils in Finland (VI)

Critical loads concept

The critical load of sulphur for forest soil is the maximum sulphur deposition the uppermost 0.5 m of soil can bear in the long run without its beginning to leach more aluminium than base cations, on a molar basis (Umweltsbundesamt 1996, Posch et al. 2001b). If the molar ratio of base cations, especially calcium, to aluminium is persistently low ((Bc/Al) < 1.0), aluminium toxicity to the tree roots may ensue (Sverdrup and Warfvinge 1993), which in turn may lead to a decline in health and growth of the trees.

To derive critical loads of acidity the charge balance of the ions in the soil leachate flux is calculated. The leaching of acid neutralising capacity is defined in terms of the leaching of bicarbonate and organic acids, protons and aluminium. The leaching and uptake of base cations is balanced by the net input through deposition and weathering.

Finnish data input

Finnish data on mineral weathering, base cation deposition and uptake by vegetation were derived by Johansson (1999). Base cation (Ca, Mg, K, Na) and chloride deposition were interpolated from the data from the years 1993-95 of a nationwide network of stations measuring monthly bulk and seasalt correction was made using Na as a tracer. Base cation weathering was estimated from information concerning the effective temperature sum and the total element content of Ca and Mg in the C-horizon (Johansson and Tarvainen 1997).

The long term average net uptake of N, Ca, Mg and K bound in the stem and bark biomass is evaluated from annual average potential forest growth, biomass density and element contents. The denitrification rate is assumed to be proportional to the net incoming nitrogen – which is adjusted with the denitrification factor, depending on the soil type. Forest inventory information on regional distribution of tree species is combined with satellite image assessment of total forest area in each EMEP50 grid cell.

Tentative modifications

In article VI we examined some of the assumptions underlying the current critical loads calculations and tested modifications to methods of deriving critical loads of acidity for forest soils. From a systems analytical point of view, the revision of the method of calculating critical loads of forest soils can be structured into four levels: (i) the choice of indicator; (ii) the definition of a criterion of unwanted effects of the emissions; (iii) the method of calculation used to link emissions to effects; and (iv) the data used in the calculations. Our tests in article VI were focused on levels (ii) and (iii). The tests concerned two alternative criteria, namely critical base-saturation and non-negative acid neutralising capacity. We also explored the consequences of extending the soil model to include organic aluminium complexes and the leaching of organic matter.

The reference case was the standard method used in Finland to calculate critical loads for forest soils (Johansson 1999). An alternative criterion concerning soil base saturation was implemented in article VI by requiring a base saturation above thirty per cent in the organic layer and above fifteen per cent in the mineral soil. The second alternative criterion that we tested was to require that no leaching of acid neutralising capacity take place (Johansson 1999). This eliminates the criterion concerning soil solution concentrations of base cations and aluminium from the calculation and in turn we avoid basing the results on an ambiguous biological response.

Equilbrium with aluminiumhydroxide is a poor predictor of the activity of inorganic aluminium in soil solution, particularly for forest floors (Walker *et al*. 1990), but also for strongly acidified mineral soil horizons (Mulder and Stein 1994) and may lead to a considerable overestimation of the Al^{3+} activity. In article VI, the critical loads model was extended with empirical equations based on laboratory equilibrium experiments conducted with northern spruce forest soils (Mulder 2000, Table I in article VI). Organic acids in soil solution contribute anions to the acid neutralising capacity and reduce the critical load of acidity. The impact of organic anions was included through an empirical equation for the dissociation of organic acids (Oliver *et al.* 1983, Table I in article VI).

In the tests using the base saturation criterion and the organically extended model, the critical load values were calculated separately for the organic layer - the uppermost 5 cm - and the mineral layer – down to 45 cm below the organic layer. The overall results for hypothetical profiles with a depth of 50 cm were obtained by weighting the layer results with the layer thickness.

The modified critical loads were compared to the reference case using information on how much the 1995 sulphur deposition values exceeded the critical loads. First, we summed across all calculation points (3083 in Finland) the amount by which deposition exceeds the critical load, multiplied by the corresponding ecosystem area. This sum of the difference between deposition and critical load, weighted with ecosystem area, was then divided by the total ecosystem area of the Finnish forest soils, giving the Average Accumulated Exceedance (AAE) (Posch *et al.* 1999).

2.3 Empirical stream water model (VII)

2.3.1 Artificial neural networks

An empirical model of stream water concentrations of total organic carbon (TOC), total nitrogen (N_{tot}) and total phosphorus (P_{tot}) was designed using artificial neural networks (VII), with the purpose to study the impacts of climate change scenarios on the leaching of carbon and nitrogen to the catchment runoff (Ilvesniemi *et al.* 2002). The feedforward networks used one hidden layer and were trained by the backpropagation algorithm (Freeman 1994), implemented in *Mathematica®* by Hunka (1997) and modified to be used in article VII. Training and testing was performed separately for carbon, nitrogen and phosphorus. The criterion was to maximise the efficiency of the network to simulate concentration values.

The driving variables were chosen to reflect climate and site characteristics. Daily air temperature, precipitation and runoff observations were used as input variables, together with effective temperature sum, days since peak runoff, and information on the temperature, precipitation and runoff during a period of days before the water quality observation was made. In order to explore the strengths and weaknesses of artificial neural networks in simulating stream water quality the usefulness of the network-based model was tested by comparing its results to an alternative method, namely the flow-weighted average concentration.

2.3.2 Sites and driving variables

The catchments Valkea-Kotinen (30 ha) and Hietajärvi (464 ha) were monitored as part of the United Nations Economic Commission for Europe (UN ECE), International Cooperative Programme on Integrated Monitoring (Bergström *et al.* 1995). Both catchments are small headwater catchments situated in remote, unmanaged forested areas in southern and eastern Finland, respectively. Daily temperature and precipitation observations are from nearby Finnish Meteorological Institute weather stations, Lammi for Valkea-Kotinen and Lieksa for Hietajärvi.

The best-performing concentration network was combined with runoff calculated for present and future conditions using a watershed simulation tool (Huttunen and Vehviläinen 2001). The implications of two future scenarios of climate change were interpreted within the FINSKEN (Carter *et al.* 2002) project for seasonal changes in temperature and precipitation values in Finland over the period 2040-2069. The seasonal changes were used to generate daily temperature and precipitation values for weather stations closest to Valkea-Kotinen (Lammi) and Hietajärvi (Lieksa) with CLIGEN (Carter *et al.* 1995).

3 Results and discussion

3.1 Dynamic modelling of soil acidification
3.1.1 Plot scale (I,II)

Calibration and prediction

When MIDAS was calibrated (I) with a fixed weathering rate of 0.045 eq $m^{-2}yr^{-1}$, it was found that the observed base saturation at the south Swedish beech site could be reproduced when the net biological turnover rate was given the value –0.04 eq $m^{-2}yr^{-1}$. This means that more base cations were bound into growing vegetation than were released from decomposing organic matter and leached from the canopy. According to the results of this calibration, eighty per cent of the vegetation's net nutrient demand is supplied by weathering. In other words, the observed change in base saturation was partly induced by natural acidification caused by growing forest that consumes base cations from weathering mineral particles.

With a ten per cent decrease in deposition (I), base saturation at the beech site decreases from 3.2 per cent in 1949 and stabilises towards 2030 at 1.7 per cent. The molar ratio of Al^{3+} to Ca^{2+}+Mg^{2+} in soil solution drops below 1.0 between 1955 and 1975 but increases after that and approaches 3.5 in 2030 (I). The beech site, with the smallest initial amount of exchangeable bases, shows the smallest maximum rate of change in base saturation and the smallest total leaching of base cations. The maximum rate of change in base saturation is largest at the spruce site, with the largest initial amount of exchangeable base cations and it also receives a larger acid load than the other sites, because of the filtering effect of the spruce canopy. Correspondingly, the spruce site shows the largest total leaching of base cations and requires the longest time to stabilise (I).

Compared with SMART (Posch *et al.* 1989), MIDAS predicts similar changes in pH. As MIDAS is a kinetic parallel to the cation exchange part of SMART, it is expected to perform similarly with respect to those features that involve only cation exchange. The advantage of MIDAS is that observed trends in base saturation can be produced with a minimum of calibration using a simple model that includes only the aggregated cation exchange of H^++Al^{3+} versus Ca^2+Mg^{2+}.

Prediction compared with extrapolation

At the south Swedish beech site, the exchangeable pools of calcium and magnesium, expressed as a percentage of the cation exchange capacity, decreased from 3.2 per cent in 1949 to 2.0 per cent in 1985 (Falkengren-Grerup *et al.* 1987). When extrapolating to 1989 using the results of Falkengren-Grerup and Tyler (1991), the pools decreased to 1.8 per cent by 1989. The model simulations result in a base saturation value of 2.0 per cent or slightly higher in 1989 (I).

In 1989, the rate of change of base saturation, expressed as exchangeable calcium and magnesium, shifts from negative to positive with the eighty per cent reduction scenario (II). With the sixty and thirty per cent reduction scenarios the shift to increasing base saturation is delayed by a few years. The scenario of a sixty per cent reduction in sulphur deposition from 1985 to 1995 brings the deposited amount of sulphur to its 1950-level, a development reported also by Falkengren-Grerup and Tyler (1991). As ammonium and nitrate in deposition continued to increase, the total acidity and acidifying potential of the deposition remained almost unchanged (Falkengren-Grerup and Tyler 1991). The net acidifying effect of the ten or thirty per cent sulphur reduction scenarios is then closer to reality.

With the scenarios which reduce deposition by ten, and thirty, per cent the simulated base saturation stabilises at 2.0 per cent (I, II), meaning that the base cation pool in 2050 would approach sixty per cent of its value in 1949. This prognosis is more optimistic than the linear extrapolation of Falkengren-Grerup and Tyler (1991), which takes the base cation pool of 2050 down to twenty per cent of its value a century earlier.

The non-linear structure of the cation exchange equation, which is the core of dynamic soil acidification models such as MIDAS, will always lead to the rate of change of base saturation approaching zero if the driving variables are constant for a long enough period. The time it takes for the system to stabilise will depend on the size of the pools of exchangeable cations and the rate of deposition, weathering, nutrient uptake and percolation (I). At the simulated south Swedish spruce site the initial base saturation was thirty per cent and this larger base cation pool then decreases almost linearly throughout the simulated 100 years (I).

Recovery

By 2030, the beech site recovers its full base saturation of the 1950s in the eighty per cent reduction scenario, whereas the spruce site continues to acidify (II). The response of soils to a pulse in deposition was studied by plotting transient base saturation simulated with the sixty per cent reduction scenario against the average deposition of SO_4-S of the past five years (Fig. 5, II).

The transition from point 1 to point 2 in Fig. 5 represents the period of increasing deposition, i.e. from 1950 to 1985, and corresponding acidification. Reducing deposition in the period 1985 to 1995 (going from point 2 to 3) stops the acidification at the beech site: base saturation decreases no more and soil solution pH increases. As weathering replenishes the pool of exchangeable base cations in absence of a large flux of mobile anions, the base saturation of the beech site slowly increases towards its initial value.

It takes 75 years to go from point 3 to 4, and by 2070 the 1950-level is still not reached. The long recovery times were already demonstrated by Cosby *et al.* (1985b). The pathway of base saturation (Fig. 5) at the beech site following a sixty per cent cut in deposition shows hysteresis, meaning that the recovery phase is not symmetrical to the acidification phase (II). A similar response pattern was shown by Reuss *et al.* (1987) for pH patways of surface waters.

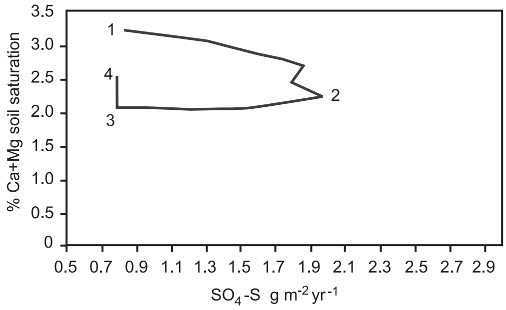

Figure 5. Base saturation (Ca + Mg expressed as per cent of cation exchange capacity) as a function of sulphur deposition at a south Swedish beech site (From Holmberg, M., 1990. Model of Ion Dynamics and Acidification of Soil: Simulating Recovery of Base Saturation. In: Fenhann, J., H. Larsen, G.A. Mackenzie & B. Rasmussen (Eds.), Environmental Models: Emissions and Consequences, pp. 359-368. Elsevier Science Publishers B.V., Amsterdam).

At a certain level of deposition, the reversibility of soil acidification depends on the initial base saturation, the weathering properties of the soil and the base cation demand of the vegetation. At the spruce site the base cation demand of the growing vegetation is so much larger than the amount of base cations supplied by deposition and weathering (I,II) that the endpoint (4) continues to move further away from the starting point (1).

3.1.2 Catchment scale (III,IV)

Model comparison

The models MAGIC, SMART, SAFE and MIDAS all suggest that the soils of the three calibrated sites Birkenes, Stubbetorp and Yli-Knuutila were acidified over the past 140 years. Although the models do not agree in detail over the magnitude and timing of the acidification, they do not contradict each other.

The model results indicate that Birkenes will continue to acidify, unless sulphate deposition is reduced by more than fifty five per cent of its 1987 value. Stubbetorp is close to being acidified by the level of deposition it received in the late 1980s, according to the model predictions, while Yli-Knuutila seems safe from acidification, even if the deposition were to continue for more than fifty years at the 1987 level (III).

For base saturation at Birkenes (Fig. 6, III), simulated without any reductions of deposition, SMART predicts nearly total depletion of Ca and Mg before 2050, while SAFE base saturation stabilises almost at the 1984 level. MAGIC and MIDAS base saturation predictions lie in between these extremes and indicate a continuing decline without stabilisation.

Prediction compared with observation

Deposition of sulphur at Birkenes decreased by thirty per cent from the years 1984-1992 to the period 1992-2001, while nitrogen stayed at a relatively high level (SFT 2002b). This observed development is close to the minimum reduction scenario (ii) in article III. Expected future deposition at Birkenes is closer to the minimum than the maximum reduction scenario used in III (Larssen *et al* 2003).

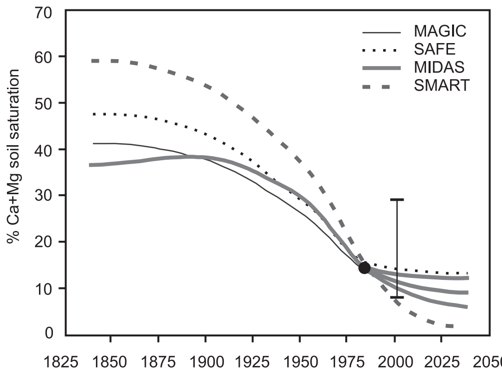

Figure 6. Base saturation (Ca + Mg expressed as per cent of cation exchange capacity) as function of time at Birkenes, Norway (Modified from Warfvinge, P., Holmberg, M., Posch, M. & Wright, R.F. 1992. The Use of Dynamic Models to Set Target Loads. Ambio 21:369-376). The thick, unbroken line shows results of simulations with MIDAS for three deposition scenarios (i) no reduction, lowest line; (ii) -30% S and -10 % N reductions, middle line; and (iii) -55 % S, -40 % N reductions, highest line. Results from simulations with the other models are shown only for the no reductions scenario. The vertical band shows the range of change observed in per cent Ca+Mg saturation (SFT 2002. Årsrapport, jordkemi 10 - 2001. Rapport. Statens forurensningstilsyn, Norwegian Pollution Control Authority, Oslo.).

Although the decrease in simulated base saturation levels off with all deposition scenarios, no recovery is predicted, not even with the maximum reduction scenario (Fig. 6). Base saturation at Birkenes clearly increased in the upper soil layers from 1984 to 2001 (SFT 2002b), due to an increase in exchangeable K, Mg and Ca and lower acidity.

The four plots for collecting soil profiles at Birkenes are very different and Reuss (1990) concludes that is seems virtually impossible to come up with an aggregation that has reasonable and defensible confidence limits. He does, however, present average values per horizon O, E, Bh, Bs and C, but no values pooled for the 0.4 m compartment used in the modelling exercise (Wright *et al.* 1991, III). As it is not clear how the individual horizons are represented in the catchment pooled values used in article III, the predicted base saturation values cannot be directly compared with recently observed values (SFT, 2002b).

In stead, to compare catchment average model simulations to recent observations, I plotted the range of potential change in the catchment average (Ca^{2+}+Mg^{2+})-saturation, translated from the reported individual horizon-specific observations (SFT 2002b). From Figure 6 one sees that the recent soil observations at Birkenes indicate a better potential for recovery than the model simulations.

The usefulness of dynamic model predictions should be evaluated both with respect to observed changes and against the results of other models. Many factors make this evaluation problematic, the most difficult being the lack of long-term consistent soil monitoring data. Large uncertainties are introduced by the process of aggregating individual soil chemical measurements into variables representative of the whole catchment. These uncertainties are even larger in the case of single-layer models.

Target loads

The criterion for no damage to the terrestrial ecosystem is expressed as the molar ratio of Ca^{2+} and Al^{3+} in MAGIC and SMART while SAFE uses the ratio between the sum of Ca^{2+}, Mg^{2+} and K^+ and total inorganic Al. For the one-compartment models MAGIC and SMART, the criterion is applied to the whole soil column, while SAFE takes the lowest value of the horizons in the root zone. To set target loads for sulphur, deposition was adjusted so that the calculated criterion was close to 1.0 by the year 2037. It was assumed that future N deposition would not exceed the demand of the vegetation more than it does today.

For Birkenes, the three models calculated very similar target loads, close to the level of the 1990 deposition. For Stubbetorp and Yli-Knuutila, however, the models give quite different target loads. MAGIC and SMART suggest that the forest soils at Stubbetorp could tolerate twice the S load of 1990, while the results of SAFE indicate that even a total removal of S deposition would not bring the soil condition to acceptable levels.

The differences are caused by the way soil stratification is dealt with in the different models. The single-layer models MAGIC and SMART register only average concentrations in the whole soil compartment, which may overestimate the base cations and underestimate the inorganic aluminium in the rooting zone.

The way aluminium chemistry is modelled may be the largest source of uncertainty and error in the calculations of critical and target loads. In the model versions in III, aluminium concentrations were calculated from the pH of soil solution, assuming that the solubility of aluminium hydroxide phases controls soil solution concentrations. Subsequently, this assumption is challenged by empirical and modelling studies (Nissinen 1999, de Wit 2000).

Nissinen *et al.* (1998) propose that weak acid dissociation and complexation with organic matter control the concentrations of aluminium and protons in soil solution. Also, de Wit (2000) found that organic complexation control of aluminium described well aluminium concentrations in both forest floor and mineral soil and suggested that substitution of the gibbsite control of aluminium solubility with an organic matter complexation control would improve description of cation concentrations in acidification models.

Relative impact of deposition and forest growth

The simulated soil effective base saturation at Hietajärvi decreases throughout the 20th century in response to forest growth and acid deposition (IV). The deposition time series 'A', obtained by assuming the lowest measured annual deposition to be representative of the present level, gave the highest effective base saturation values throughout the simulation period. The future base saturation simulated with the highest deposition time series 'E', with the present levels calibrated to the highest observed deposition values, coincides with the 'D'-results, using average present deposition and highest future emissions. The results of all other scenarios lie between these extremes.

Recovery of base saturation values is seen with the 'C' scenario, which combines low future emissions with high forest growth. With 'B', which uses low forest growth together with an emission scenario aiming at joint optimization of acidfication and ozone, future base saturation stabilises at about the present level.

The assumptions concerning present and future deposition at Hietajärvi introduced the largest source of variability in the base saturation values simulated by SMART (IV). The forest growth scenarios have a smaller impact than the deposition scenarios, but larger than the timing scenarios. The impact of the timing of the reductions was smaller than the impact of the level of deposition reduction or the impact of forest growth.

The year chosen to represent present deposition had a large effect on the results. This is understandable in the light of the large year-to year variation in deposition observations at this site. Such large variation is not uncommon, and emphasises the importance of long-term monitoring (IV). In this case, the observed 6-year average deposition values were closer to the modelled values for 1990, given by the long-range transport matrices provided by EMEP/MSC-W, and incorporated into DAIQUIRI. This is a point in favour of using modelled deposition values rather than observations of only one or two years.

The SMART performance was judged to be useful for the application in IV because the simulated pH history accorded with the diatom-inferred pH history of the lake in the catchment. The year to year variation in the 6 years of observed stream water quality had only a small impact on the calibration over the time period 1900 – 2050. It might therefore be argued, that too few observations were available to evaluate thoroughly the performance of SMART at Hietajärvi, It was decided, however, that the application was useful to analyse the sensitivity of the model to the driving variables deposition and nutrient uptake.

Lumped-parameter models that describe the whole catchment in terms of only one soil profile are sensitive to the way in which the soil variables are derived. The base case deposition runs in IV were calibrated to a catchment-average value of present-day soil effective base saturation at Hietajärvi. The value obtained was forty seven per cent, which compares with an observed minimum value of fifteen per cent for the uppermost mineral soil (0-5cm) and an observed maximum of seventy six per cent for the O-horizon. If peatland values had been included in the aggregated value, a different standard for the calibration would have been obtained, with consequences for the model parameters obtained as a result of calibration and for the simulated future values.

3.1.3 Implications concerning dynamic modelling of soil acidification

A simple dynamic model structure is sufficient to illustrate the combined impact of acid deposition, constant-rate weathering, cation exchange with constant selectivity coefficients, and constant rate removal of nutrients into growing vegetation. Because of the non-linear form of the cation exchange equation, MIDAS predicted less future loss of base cations at the south Swedish beech site than a linear extrapolation of the observed base cation pool. Nevertheless, the 1950-level of base saturation would not be reached at the beech site in 2070 even if simulated with a sixty per cent reduction in the acidifying deposition.

The pathways followed by base saturation and soil solution pH are not symmetrical in the acidification and recovery phases and low acid deposition is required over long periods of time to restore acidified soils and surface waters.

Interpretation of recent soil base saturation observations from Birkenes indicates a better potential for recovery from acidification than what MIDAS and other models predicted.

The most basic and important task in modelling soil processes is defining the boundaries of the system to be studied. This includes also defining the state variables and the processes that are most central for the behaviour of the system to be studied. The choice of system boundaries, although it starts from a theoretical picture of what is to be studied, depends on the data available and it will always influence the results and the consequences that can be drawn. This choice concerns for example the number of soil horizons that are considered and also the method used to aggregate from the plot to the catchment and regional scales.

In their study on cation exchange in podzolic forest soils, Nissinen *et al.* (1999) found large differences between the soil horizons, which implies that single-layer model predictions of cation leaching may be biased (Nissinen 1999). With the recent development of in situ measurement techniques (Sparks 2001), future soil modelling studies may be better informed as regards the true variation of the elements in soil. The interpretation of data on observed soil properties may also benefit from data assimilation techniques such as the Kalman filter (Heuvelink and Webster 2001).

Among the process-formulations used to predict the response of boreal forest soils to acidification, the methods describing aluminium chemistry and complexation with organics may be improved by empirical findings concerning the aluminium reactions in soils high in organic matter (de Wit 2000). This question becomes relevant especially if model predictions concerning soil aluminium concentrations are used as background information for setting emission reduction targets. The value of model studies directed at informing work on air-pollution prevention strategies would be enhanced by reporting on the consequences of the choices made concerning system boundaries, state variables and process-formulation.

3.2 Static assessment of regional acidification risk

3.2.1 Groundwater sensitivity (V)

Highly sensitive areas

The piecewise linear functions (Fig. 4) for assessing groundwater sensitivity to acidification (V) yield a high sensitivity value only if both aquifer and soil neutralising capabilities are rated poor. An area is assumed to have a poor soil neutralising capability if the base cation content of the top 0.5 m is below 400 keq/ha in the FAO Soil Map of the World. The aquifer neutralising capability is rated poor if the potential annual recharge is low and the area does not contain basic silicate or carbonate rocks and the size of the aquifer is not extensive, as interpreted from the productivity classes given by the International Hydrological Map of Europe (IAH-UNESCO 1970)

When the dominant (largest area) soil type and aquifer of each grid cell were passed through the decision matrix tree (V) to evaluate the sensitivity of groundwater acidification, 27% of the grid cells were assigned a high sensitivity value and 29 % low sensitivity. Large areas of Norway, Sweden and Finland were classified as highly sensitive as well as were parts of Scotland, the Alps, Poland, the Czech and Slovak republics, Hungary, northern Portugal and north-western Spain.

Impact of spatial representation and indicator classification

The average sensitivity value for all 1844 grid cells in V was 1.93 and the risk of acidification 2.13, taking into account the sulphur deposition for the year 1980. The results for the best and worst cases encompass the dominant values (Tables 1 and 2), with the majority of the grid cells assigned a low sensitivity to groundwater acidification in the best case and a high sensitivity in the worst case. When the classification of any indicator in V was changed by one unit the probability of a change in the acidification sensitivity was about 0.2. In contrast, the probability of changing the risk assessment by changing the deposition class was about 0.7. In the decision matrix tree (Fig. 4), the ratings of the lowest two tables: sensitivity and risk, were the most influential for the overall output. Changes in the other coefficients of the piecewise linear functions affected the results very little.

It is difficult to find evidence to contradict or support an assessment of how sensitive a certain ecosystem is. A sensitivity estimate should be compared with observed rates of change of one or several variables that describe the state of the system under different trajectories of the driving functions. The assessed risk, on the other hand, can be contradicted or supported by observations of actual acidification, in the light of actual deposition.

Table 1. Distribution of sensitivity values obtained with different approaches to the spatial representation of the data in each grid cell.

Grid cells (% of 1844 cells) with sensitivity class value		
Spatial representation	Low (1)	High (3)
Best case	51%	6 %
Dominant	29 %	27 %
Worst case	7 %	55 %

Table 2. Average sensitivity and risk values (range 1 to 3) for all 1844 grid cells, obtained with different approaches to the spatial representation of the data in each grid cell.

Spatial representation	Sensitivity	Risk
Best case	1.62	1.79
Dominant	1.93	2.13
Worst case	2.23	2.41

Comparison with critical loads of Europe

A map of critical loads is presented by Posch (2002), showing, for about 3800 grid cells (0.5º longitude x 0.5º latitude) the sulphur deposition at which ninety five per cent of the forested ecosystems in the grid cell will be protected from acidification. The map reflects the neutralising capability of the uppermost soil (0.5m), evaluated with the steady-state Simple Mass Balance (SMB) model.

The capability of soil to neutralise acidity, defined in V as a combination of depth, texture and the base cation content of the overlying soil describes the same properties as the critical load estimates. Because the division of the range of sensitivity values and critical loads into classes is arbitrary it was possible to make an association between the divisions used in V and in the map of critical loads of forest soils. For the purpose of comparing V with critical loads the high sensitivity class was associated with critical loads of sulphur of below 200 eq/ha/yr and low sensitivity with loads above 800 eq/ha/yr.

A small number of grid cells in the Czech Republic, western Norway, the Alps, central France, Spain and Portugal come out as highly sensitive to groundwater acidification (V), although according to Posch (2002) these areas may receive high sulphur deposition without violating the acidification criterion for forest soils. Except for these grid cells, the regional distribution of aquifer sensitivity in V corresponds well to the European map of critical loads of sulphur (Posch 2002).

Although a quantitative comparison of these two methods of assessing regional sensitivity to acidification is beyond the scope of this summary, some qualitative remarks can be made. The piecewise linear functions in V rank an aquifer as highly sensitive only if both soil and aquifer neutralising capabilities are poor. If the two methods would be coherent with regard to their interpretation of soil neutralising capability, the aquifer sensitivity map of V would give the ranking as low or medium, instead of high, to the coastal areas in western Norway, Spain and Portugal that have soils that are not very sensitive to acidification in the analysis by Posch (2002).

The incoherence is due both to the coarser spatial resolution in (V) and to the matrix parameters in V (Fig. 4). Good soil neutralising capability comes out of the matrix with the highest base cation content (reflecting good weatherability), or as a combination of good soil water contact (fine textured soils, deeper than 0.5 m) and reasonable base cation content. There are probably enough shallow soils with low base cation content in these coastal grid cells to make the areas come out with low soil neutralizing capability in V.

Comparison with studies from the Czech Republic, Finland, Norway and Sweden

The map of aquifer sensitivity (V) shows areas of high sensitivity in north-eastern, south-western and central parts of the Czech Republic. Hrkal *et al.* (2002) expressed the resistance of individual catchments to surface water acidification as the difference between the pH of runoff and that of precipitation.

Out of fourteen catchments they found four with low resistance on granite and paragneiss in the north-east and north-west; and four with high resistance on gneiss, paragneiss and serpentinite in the south-west and west and in central Czech Republic (Hrkal *et al* 2002, Hrkal and Fottová 1999). Comparing the classification of these individual catchments, five that were classified as having medium or high resistance to surface water acidification by Hrkal *et al.* (2002) are located in areas estimated as less resistant (showing high or medium sensitivity) in the aquifer sensitivity map of V.

Hrkal (2001) mapped the vulnerability of groundwater in the Jizerské Mountains, northern Czech Republic, by combining information on elevation, morphology of the terrain, areal extent, vegetation cover and lithology of the aquifer in a geographical information system (GIS). The most vulnerable areas were situated in the highest altitude belt; with high total rainfall, slopes exposed to the west and completely covered by mature forest, while the least vulnerable areas were in low-altitude areas receiving less precipitation on east-facing slopes with no forest (Hrkal 2001). The scale is much smaller than that of V thus it is not possible to compare the results. The methods differ in that elevation and morphology of the terrain are explicit variables in Hrkal (2001) while these variables implicitly influence soil depth, soil texture and aquifer potential recharge in V. The vegetation cover did not influence the results of V but it could be included through its effect on total deposition, which would influence not the sensitivity but the risk of groundwater acidification.

The greatest change in groundwater quality since the Geological Survey of Finland began monitoring in 1969 was the systematic acidification in 1975-1990 followed by recovery by the mid 1990s (Backman *et al.* 1999). In the map showing the results of the matrix approach in article V, small areas of medium sensitivity are found in the north of Finland, along the western coast, in south-west and south-east, while the rest of the country is ranked highly sensitive.

The Geological Survey found high pH values in south-west, explained by the presence of limestone and long retention time of the large groundwater reserve (Backman *et al.* 1999). The monitoring sites in sand and till in Häme and Uusimaa, at which Backman *et al.* (1999) found rapid response to changes in deposition, are located in highly sensitive grids in the map of article V. In the light of these findings, Finnish groundwater monitoring observations do not contradict the matrix approach in article V.

Almost all of Norway comes out as highly sensitive to groundwater acidification in the results of article V. Even with low deposition, a highly sensitive area is assessed to have a high risk, as a result of the weightings in the last matrix in Figure 4. In contrast, Banks *et al.* (1998) found little evidence of significant acidification of Norwegian groundwaters by acid deposition. This indicates that a better choice of weightings for the last row of the matrix would be (2, 3, 3) or (2, 2, 3).

For groundwater chemistry from 2338 wells in southern Sweden, Maxe (1999) compared the results of the vulnerability matrix in V with an additive assessment method (Jacks and Knutsson 1982) and with calculations by the PROFILE model (Sverdrup and Warfvinge 1988). She found that the vulnerability matrix (V) and the additive assessment gave comparable results: both mis-assigned ten per cent of the wells and gave a good representation in about sixty per cent of the cases (Maxe 1999). In Maxe's comparison (1999) the mechanistic weathering model PROFILE did not improve the regional assessment of groundwater to acidification, although PROFILE did provide a reasonable quantification of the weathering.

3.2.2 Modifications to critical loads calculations (VI)

When we used the extended critical loads model in article VI, which accounted for the complexation of aluminium by organic matter and the leaching of organic anions, and compared the critical loads with deposition for the year 1995, the modified critical loads were exceeded, on average, by only eighty per cent of the degree to which the standard critical loads were exceeded. Table II in article VI gives the details of this comparison, where the average accumulated exceedance (AAE) was 5 eq $ha^{-1}yr^{-1}$ for the standard approach, and 4 eq $ha^{-1}yr^{-1}$ for the calculation with aluminium and soil organic matter complexes and leaching of organic anions.

The modified critical loads became more stringent when the criterion was changed from the standard one, connected to the molar concentrations of base cations and aluminium in soil solution. The critical base saturation limit results in an AAE value of 17 eq $ha^{-1}yr^{-1}$. Allowing no deterioration of the acid neutralising capacity of the soil, implies even lower critical loads and higher AAE (25 eq $ha^{-1}yr^{-1}$) for forest soils. When the leaching of organic anions is accounted for, the leaching of acid neutralising capacity is higher, and the critical load lower, the more organic anions are leached.

The averaging approach disregards the chemically and physically layered structure of the podzolic soils common in Finnish forests. In reality, the precipitation that infiltrates the mineral soil is chemically modified by its contact with the organic layer. It is not clear how much the area exceeded would change at the country level if the podzolic layering were accounted for. For sites where tree roots function predominantly in the organic layer any criterion concerning the effects of aluminium on tree roots may be irrelevant, or very seldom violated.

The tests in VI illustrate the variability of the critical load values of acidity of forest soils that can be introduced by changing the criterion or by varying the calculation method. The results probably do not, however, represent the extreme values of critical loads that could be derived. For instance, assuming a thicker organic layer or separately accounting for forests growing on peatland and forests growing on mineral soils would give lower average accumulated exceedance values.

Other sources of variability related to assessing the influence of organic matter and organic acids are the empirical parameters corresponding to the upper mineral soil that were used for the whole 45 cm of mineral soil. The assumed site density and the concentration of dissolved organic acids in soil solution also influence the resulting critical load values. A constant value of 20 mg C L^{-1} was used for dissolved organic carbon in VI, which clearly is a gross generalisation.

3.2.3 Implications concerning static assessment of regional acidification risk

In contrast to even twenty years ago, in estimating the environmental effects of human actions, we are not restricted by computing resources or the means of graphical display. In the world of technological advances, it is even more important that simple, transparent and accessible methods are used to provide targets for emission reductions as background information for international negotiations.

Preferably, scientists from a broad range of disciplines including earth sciences, medicine, political and economic sciences, should be able to comprehend the methods and evaluate the implications of their inherent assumptions. Ideally, these scientists would also have the resources to inform the lay public about the consequences of different strategies. Efforts that invite us to discuss the methods and strategies are made in the documentation of the work of the Coordination Center for Effects (Posch *et al.* 2003) and in the reports from the AIR-CLIM Project (Alcamo *et al.* 2002).

The combinatory matrix approach in article V is an example of an alternative method to evaluate the sensitivity of soils and aquifers on a regional scale. The process-based assumptions concerning conditions leading to either high or low sensitivity are apparent in the matrices in Figure 4. Because the matrix approach is more qualitative than quantitative in nature, it is possible to use it to represent complex interactions in a transparent form.

To my knowledge, no similar matrix approach is documented since V. However, I think it merits further attention as an accessible method for evaluating the consequences of non-linear reactions in a piecewise linear framework. The technical implementation of the matrix method can be simplified with geographical information systems, and the ratings can be calibrated against the observed responses in different conditions (e.g., Banks *et al.* 1998, Backman *et al.* 1999).

The critical load approach is a gross simplification of a very complex system. For Finland, where forests mainly grow on podzolic soils and peatlands, critical loads may be biased, implying overly sensitive soils, if they are calculated without accounting for organic complexation of aluminium or the leaching of organic anions. The results are especially misleading if the criterion that is used is not relevant (de Wit 2000). In general, it would be preferable to use a criterion concerning the behaviour of the system as a whole, rather than one isolated feature, such as forest health.

The fact that there is an ambiguous link between forest health and changes in soil variables that can be modelled by simple mass balance or dynamic models supports the use of a simpler criterion, relating to a higher level in the system hierarchy. The choice of criterion determines the targets that are obtained with the critical loads calculations. Ideally, the criteria would evolve from a wide discussion involving all sectors of society, not solely chosen on the basis of scientific investigations of a limited scope. The importance of the soil parameters used in modelling was also pointed out by Johansson *et al.* (2001b), who in addition found that using a smaller grid size and accounting for ecosystem-specific dry deposition velocities would increase the area where critical loads are exceeded.

3.3 Empirical stream water model (VII)

3.3.1 Model performance

Using solely the variables of precipitation and temperature related input failed to provide enough information to reproduce observed variations in concentrations of TOC, N_{tot} and P_{tot} (VII). By including variables derived from daily runoff the dynamics of snowmelt were incorporated and consequently all other precipitation related variables could be omitted. Efficient reproductions of stream water concentrations were obtained with networks with at least 13 input variables including sampling month, site specific and temperature and runoff related variables (Fig. 7).

The 13-input networks were chosen for the climate change predictions because they were efficient both when comparing concentrations (R_{eff}=0.7 to 0.8) and fluxes (R_{eff} = 0.9) without including too many input variables. In comparison, also the flow-weighted

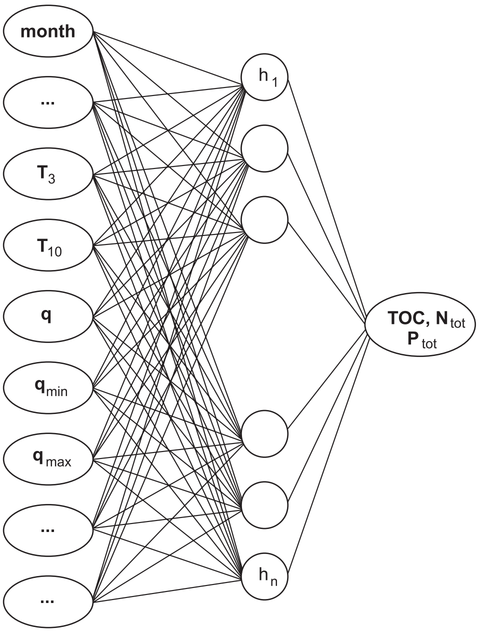

Figure 7. Artificial neural network used to simulate concentrations of TOC, Ntot and Ptot (From Holmberg, M., Forsius, M., Starr, M. & Huttunen, M. 2003. An application of artificial neural networks to carbon, nitrogen and phosporus concentrations in three boreal streams and impacts of climate change. To appear in Ecological Modelling.)

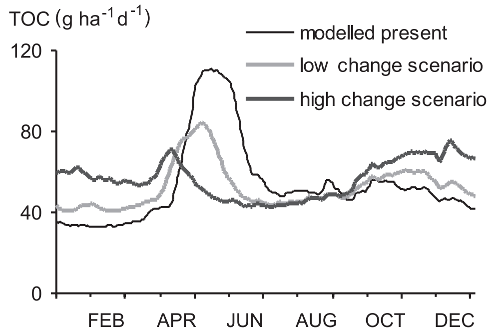

Figure 8. Predicted daily values of fluxes of TOC at Hietapuro for present conditions (1990's) and for the low climate change scenario (2050's) and the high scenario (2050's). (From Holmberg, M., Forsius, M., Starr, M. & Huttunen, M. 2003. An application of artificial neural networks to carbon, nitrogen and phosporus concentrations in three boreal streams and impacts of climate change. To appear in Ecological Modelling.)

average concentration was quite efficient in reproducing fluxes (R_{eff} = 0.8).

Many different sets of network weights were equally efficient in reproducing observed concentrations. A robust selection procedure for choosing network parameters would be needed to ensure low-noise predictions.

3.3.2 Climate change predictions

With the low climate change scenario, the total annual fluxes of TOC, N_{tot} and P_{tot} for a hypothetical year in the 2050s were within the range of –2.0 % to 1.0 % of the fluxes modelled for the present climate conditions (VII). The high change scenario resulted in 26% higher fluxes of TOC, N_{tot} and P_{tot} at Valkea-Kotinen and 4.0% to 6.4% higher fluxes at Hietapuro. The pronounced spring peak in fluxes at present is predicted to be replaced by more subdued but persistent fluxes (Fig. 8). The predicted changes were primarily determined by changes in the amount of runoff.

The response of element concentrations in stream water to climatic variability is a complex interplay of hydrological and biogeochemical processes. High concentrations in the stream may be a result of high biogeochemical activity in the soil matrix and/or a prolonged period of drought followed by an intensive precipitation event.

The approach of VII was to attempt to identify the temperature and precipitation patterns leading to observed high and low element concentrations in the stream. The observed concentrations were, however, strongly dominated by the impact of hydrological conditions, although no simple regression could be found between concentrations and runoff. Next, statistically separating the effects of temperature and runoff on the concentrations would be worth while before further development of artificial neural networks to explore how changes in climate may affect stream water concentrations and leaching fluxes in the future.

3.3.3 Implications concerning artificial neural networks to model stream water chemistry

Although the empirical model based on artificial neural networks did not catch all extreme values, it reproduced most of the dynamics in the observations of TOC, N_{tot} and P_{tot}. The model performed better than the alternative, which was to simulate one set of concentrations based on flow-weighted average concentrations of another set of observations. Good estimates of fluxes were obtained both with concentrations simulated with the neural networks and with flow-weighted averages.

Artificial neural networks proved to be a useful method for empirical modelling of the non-linear responses of stream water concentrations to runoff and temperature. The method can be further improved by placing more emphasis on stringent tests to support the selection of network structures and parameters. For process identification, the neural networks approach has to be guided by theoretical process understanding and statistical methods of separating the impacts of different drivers.

In a changing climate, future predicted increases in stream water TOC, N_{tot} and P_{tot} fluxes at the sites studied, namely Valkea-Kotinen and Hietajärvi, were primarily determined by changes in the amount of runoff and not concentration. Therefore the description of the hydrological response to climatic variables is of decisive importance for realistic estimates of changes in element leaching.

To obtain more informative predictions for changing climatic conditions, it is necessary to identify more clearly the interaction of temperature, precipitation and runoff in driving the dynamics of TOC, N_{tot} and P_t in stream water. The critical processes that influence the chemical and microbiological activity are freezing, thawing and drying of soil and the flushing of elements after a dry spell. It is likely that these processes cannot be fully accounted for by empirical methods and a combination of artificial neural networks with process formulations would be worth exploring in the context of simulating stream water concentrations and leaching fluxes from forested catchments.

4 Conclusions and recommendations

This summary examined three methods for assessing the soil-mediated response to acidification and climate change: dynamic modelling, static risk assessment and empirical modelling with neural networks. The spatial scales of the modelling studies range from individual soil plots to a grid over Europe. The methods and their development were documented in detail in articles I to VII. The articles and this summary contribute to the background work directed at exploring the consequences of different emission reduction strategies and at evaluating the advantages and limitations of impact assessment methods.

Dynamic process-oriented models make it possible to study how long it takes for soils to react to changes in the atmosphere and allow projections to be made of the response behaviour into the future. How useful they are for practical applications is determined by how realistic their process descriptions are – for example the control of aluminium solubility – and how well the model parameters approximate the actual process rates in nature. In podzolic soils in particular, appropriate model parametrisation means, for example, accounting for the layered structure of the profile. Simulations with the MIDAS model indicated, at one site in south Sweden, less drastic future depletion of base cations than estimated in the literature. For the Birkenes catchment, MIDAS simulations predicted slower recovery than recent observations indicate.

The combinatory matrix for static sensitivity assessment provides a transparent and accessible representation of qualitative process knowledge in a stringent form, without requiring detailed quantitative information on process rates. The use of sensitivity matrices to study the risk of groundwater acidification in Europe could not be contradicted by comparison with groundwater quality observations from the Czech Republic, Norway, Sweden and Finland. The sensitivity matrix approach offers a user-friendly alternative method of evaluating the sensitivity of soils and aquifers on a regional scale. The technical implementation of the matrix can be simplified with geographical information systems, and the method's ability to realistic screening of areas of high and low sensitivity may be improved with observations of changes and reaction times.

Artificial neural networks driven by variables related to temperature, runoff and site characteristics provide an efficient means for reproducing non-linear patterns of stream water concentrations of TOC, N_{tot} and P_{tot}. The method can become even more useful for the purpose of future projections of fluxes and concentrations if more stringent tests are developed to inform network design and parameterisation.

In summary, we need dynamic models as tools to improve our understanding of forest soil behaviour by combing theoretical knowledge with empirical observations. In particular, model-based reasoning is necessary if we want to back up international negotiations with informed arguments concerning the effects of our actions on our environment, on a scale that extends in time and space beyond our immediate perception. At the same time, we need to be aware of the limits of the models, that their quantitative – and seemingly objective results are always dependent on the subjective qualitative choices that were embedded in the model design. As it is not possible to avoid the subjective component, it is all the more important to be clear and open about our model's inherent assumptions, so that the disturbance can be minimized. The more transparent the design, the easier it is to decide how and for what purpose the model results may be applied. Our mandate to generalize the results is limited especially by our choice of system boundaries, by the process-formulations used and by the criteria we apply to set targets.

Key findings:

1. A simple dynamic model structure is sufficient to illustrate the combined impact of acid deposition, constant-rate weathering, cation exchange with constant selectivity coefficients, and constant rate removal of nutrients into growing vegetation.
2. In Skåne, south Sweden, the model predictions did not deplete the pools of exchangeable base cations as fully as was reported in the literature on the basis of linear extrapolation of soil observations.
3. Soils in Birkenes may recover better than models predicted, in the light of recent observations of base saturation.
4. Assumptions concerning future forest growth had a smaller impact on simulated future base saturation values than had the choice of the level of present-day deposition.
5. Piecewise linear functions provide a simple and robust method of assessing sensitivity and risk.
6. For Finland, where forests mainly grow on podzolic soils and peatlands, critical loads may be biased, implying overly sensitive soils, if they are calculated without accounting for organic complexation of aluminium or the leaching of organic anions
7. Artificial neural networks are well suited for reproducing the non-linear patterns of stream water concentrations of TOC, N_{tot} and P_{tot} and for exploring the consequences of changes in climatic conditions on leaching fluxes.

Recommendations for future work:

1. The role of soil organic matter should be accounted for in static and dynamic calculations of boreal forest soil response.
2. The potential of the combinatory matrix approach to evaluating non-linear response in a piecewise linear framework merits further attention.
3. The use of empirical stream water quality models based on artificial neural networks may be further improved by stringent tests to guide model parameterisation and by exploring ways to utilise information concerning freezing, thawing, drying and flushing of soils.

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