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MATTI JOHANSSON

Integrated models for the assessment of  
air pollution control requirements

MONOGRAPHS

*of the*

Boreal Environment Research

# BOREAL ENVIRONMENT RESEARCH

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MONOGRAPHS OF THE BOREAL ENVIRONMENT RESEARCH

**13**

Matti Johansson

**Integrated models for the assessment of  
air pollution control requirements**

Yhteenveto: Yhdennetyt mallit ilmansaasteiden päästörajoitus-  
vaatimusten arvioinnissa

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## List of original publications and the author's contribution

This study synthesizes the following original publications, which are referred to by their Roman numerals in the text. The author's contribution is explained separately for each article. In addition, some previously unpublished results and figures are included in this study.

**I** Johansson M., Savolainen I. & Tähtinen M. 1989. The Finnish integrated acidification model. In: Kämäri J., Brakke D., Jenkins A., Norton S.A. & Wright R.F. (eds.), *Regional acidification models. Geographic extent and time development*, Springer-Verlag, Berlin Heidelberg, pp. 203–211.

**II** Johansson M. & Savolainen I. 1990. Regional acidification model for forest soils. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer-Verlag, Berlin Heidelberg, pp. 253–269.

**III** Johansson M., Kämäri J., Pipatti R., Savolainen I., Tuovinen J.-P. & Tähtinen M. 1990. Development of an integrated model for the assessment of acidification in Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer-Verlag, Berlin Heidelberg, pp. 1171–1193.

**IV** Sverdrup H., Warfvinge P., Frogner T., Håøya A.O., Johansson M. & Andersen B. 1992. Critical loads for forest soils in the Nordic countries. *Ambio* 21(5): 348–355.

**V** Johansson M. & Janssen P. 1994. Uncertainty analysis on critical loads for forest soils. In: Grasman J. & Van Straten G. (eds.), *Predictability and nonlinear modelling in natural sciences and economics*, Kluwer Academic Publishers, Dordrecht, The Netherlands, pp. 447–459.

**VI** Johansson M. & Tarvainen T. 1997. Estimation of weathering rates for critical load calculations in Finland. *Environmental Geology* 29(3/4): 158–164.

**VII** Forsius M., Johansson M., Posch M., Holmberg M., Kämäri J., Lepistö A., Roos J., Syri S. & Starr M. 1997. Modelling the effects of climate change, acidic deposition and forest harvesting on the biogeochemistry of a boreal forested catchment in Finland. *Boreal Environment Research* 2(2): 129–143.

**VIII** Syri S., Johansson M. & Kangas L. 1998. Application of nitrogen transfer matrices for integrated assessment. *Atmospheric Environment* 32(3): 409–413.

**I** Author's contribution: Modification and application of transfer matrices, calculation of deposition scenarios, simulations with the soil impact model, integration of results.

**II** Author's contribution: Responsibility for manuscript, derivation of deposition scenarios, development of input data for impact analysis, simulations with the soil model, uncertainty analysis.

**III** Author's contribution: Application of transfer matrices, derivation and integration of deposition scenarios for all impact models, simulations with the soil impact model, analysis of soil impact results.

**IV** Author's contribution: Calculations with Finnish data, selection of output indicators, discussion of results and method differences, presentation of the steady-state mass balance method and weathering rate estimation method for Finland.

**V** Author's contribution: Responsibility for manuscript, derivation of input data, use of UNCSAM software package for the forest soil critical load data in Finland, assessment of uncertainty results.

**VI** Author's contribution: Coordination of the manuscript preparation; analysis and calculation of weathering rates with different methods and using correction factors from fraction size differences. Calculations of critical loads and their exceedances with different weathering rates.

**VII** Author's contribution: Derivation of scenarios for deposition, uptake, forest growth and harvesting, simulations with the SMART model, assessment of results.

**VIII** Author's contribution: Responsibility for task coordination, basis for the integration of different transfer matrices; development of comparison method and assessment of results jointly with other authors.

## Symbol notation and abbreviations in this study

symbols:

<i>Alk</i>	alkalinity (acid neutralizing capacity)
<i>Bc</i>	(physiologically active) base cations: $\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^{+}$
<i>BC</i>	base cations: $Bc + \text{Na}^{+}$
<i>BC2</i>	divalent base cations: $\text{Ca}^{2+} + \text{Mg}^{2+}$
<i>BS</i>	soil base saturation, $BS = BC_{\text{exch}} / CEC$
<i>CD</i>	critical deposition (deposition is at critical load)
<i>CEC</i>	total cation exchange capacity of soil matrix, $CEC = (BC + \text{Al} + \text{H})_{\text{exch}}$ (usually Fe and Mn are ignored)
<i>CL</i>	critical load
<i>eq</i>	equivalent or moles of charge, $\text{mol}_c$ ; if <i>X</i> is an ion with molecular weight <i>M</i> and charge <i>z</i> , then $1 \text{ g } X = M^{-1} \text{ mol } X = z \cdot M^{-1} \text{ eq } X$ ; $1 \text{ gS} \propto 1 \text{ g} \cdot 2 \text{ eq mol}^{-1} \cdot (32 \text{ g mol}^{-1})^{-1} = 0.0625 \text{ eq}$ and analogously $1 \text{ gN} \propto 0.0714 \text{ eq}$
<i>ETS</i>	effective temperature sum
<i>Ex</i>	exceedance (of critical load)
<i>K<sub>gibbs</sub></i>	gibbsite equilibrium constant
<i>N</i>	nitrogen
<i>NH<sub>3</sub></i>	ammonia (emission)
<i>NH<sub>x</sub></i>	ammonium (deposition), as $\text{NH}_3$
<i>NO<sub>x</sub></i>	nitrogen oxides (emission), as $\text{NO}_2$
<i>NO<sub>y</sub></i>	nitrogen oxides (deposition)
<i>O<sub>3</sub></i>	ground-level (tropospheric) ozone
<i>Q</i>	runoff
<i>S</i>	sulfur
<i>SO<sub>2</sub></i>	sulfur dioxide
<i>T</i>	temperature

as superscript:

\* sea-salt corrected (using Na as tracer, thus  $\text{Na}^*_{\text{dep}} = 0$  and  $BC^*_{\text{dep}} = Bc^*_{\text{dep}}$ )

as subscript:

<i>crit</i>	critical (threshold)
<i>den</i>	denitrification
<i>dep</i>	deposition
<i>exch</i>	exchangeable
<i>im</i>	immobilization
<i>le</i>	leaching
<i>max</i>	maximum
<i>min</i>	minimum
<i>nu</i>	net uptake
<i>sust</i>	sustainable
<i>tu</i>	total uptake
<i>we</i>	weathering of soil minerals



## grid systems:

0.25° × 0.125°	the national grid of 52 × 88 cells; each cell size is 0.125° longitude × 0.25° latitude (approximately 14 km × 14 km in southern Finland at 60°N) and the lower left corner of the grid system is at 19°E, 59.5°N
150 km × 150 km	the polar stereographic projection of the EMEP/MSC-W calculation grid of cells approximately the size of 150 km × 150 km; note, that only at 60°N the cells have an area of about 22500 km <sup>2</sup> , which decreases to about 16200 km <sup>2</sup> in the very southern tip of Europe

## abbreviations:

CDF	Cumulative Distribution Function
CLE	Current Legislation: emission scenario of the RAINS model on the effects of national and European Union legislation on reducing emissions
CLIM	Critical Load Integrated Model system
CLRTAP	Convention on Long-range Transboundary Air Pollution
COD	Coefficient Of Determination
CRP	Current Reduction Plans (on air pollutant emissions, officially announced by the countries through UN-ECE)
DAIQUIRI	A model for Deposition, Air Quality and Integrated Regional Information
DEPUPT	A model for estimating the temporal development of site-specific DEPositions of sulfur, nitrogen, chloride and base cations and nutrient UPTakes by forest growth
DIM	Dynamic Integrated Model system
EMEP/MSC-W	Co-operative Program for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe, Meteorological Synthesizing Center – West
EU	European Union
FEI	Finnish Environment Institute (Suomen ympäristökeskus, SYKE)
FMI	Finnish Meteorological Institute (Ilmatieteen laitos, IL)
HAKOMA	The Finnish Integrated Acidification Model (HAppamoitumisen KOkonaisMAlli)
HAPRO	The Finnish Acidification Research Programme 1985–1990 (HAppamoitumisPROjekti)
IAM	Integrated Assessment Model
IIASA	International Institute for Applied Systems Analysis (in Laxenburg, Austria)
MFR	Maximum (technically) Feasible Reductions: emission scenario of the RAINS model of IIASA
NFC	National Focal Center (for Mapping Critical Loads and Thresholds, under CLRTAP)
PUC	Partial Uncertainty Contribution
RAINS	Regional Acidification INformation and Simulation model of IIASA
REF	REFErence emission scenario of the RAINS model of IIASA addressing the smaller emission figure for a country from CRP and CLE scenarios
SILMU	The Finnish Research Programme on Climate Change (Suomalainen ILmakehän suurMUutosten tutkimusohjelma) 1990–1995
SMB	Steady-state Mass Balance method for calculating critical loads
SMART	Simulation Model for Acidification's Regional Trends
SRC	Standardized Regression Coefficient
TFIAM	Task Force on Integrated Assessment Modelling, under CLRTAP/Executive Body/Working Group on Strategies
UN-ECE	United Nations Economic Commission for Europe
VOC	Volatile Organic Compound



# Integrated models for the assessment of air pollution control requirements

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*Johansson, M. 1999. Integrated models for the assessment of air pollution control requirements, Monographs of the Boreal Environment Research No. 13, 1999.*

This study synthesizes the contribution of the author in the development and applications of three Finnish integrated assessment models (IAMs) which were used to evaluate the geographical extent and time development of forest soil acidification and consequent emissions reduction requirements of several air pollutants. The acid rain problem is considered as a serious threat to the environment although acidifying emissions have been reduced by 37% in Europe during 1980–1995. The first model system, the Finnish integrated acidification model HAKOMA, was employed to identify and model the most important factors affecting regional soil acidification and its temporal development. Sensitive regions were highlighted and the relative area of severely acidified forest soils was estimated to be between 57% and 12% in 2040 depending on the emission reduction scenario. The derivation of input data for the model formed an important basis for further work. The second design was based on the critical load integrated model system (CLIM). Critical load describes the long-term tolerance of ecosystems against a pollutant load. The critical loads for forest soils were forwarded to national and international bodies to formulate environmental protection targets for negotiations on emission reductions. A decreasing trend was found for the exceedance of critical loads in Finland, especially after 1980, and scenario calculations predicted 95% protection of ecosystem area by 2010. The third setup, the dynamic integrated model system (DIM) employed the SMART soil acidification model soft linked to other models, such as the DEPUPT model amended to produce site-specific deposition and nutrient uptake histories and scenarios. The results from one forested catchment in southern Finland indicated that only prompt and strict emission reduction controls in Europe were able to prevent further soil acidification at the site. Sulfur and nitrogen deposition, forest clearcut and climate change were found of equal potential importance in affecting the future acidification of the catchment. An uncertainty analysis on forest soil critical loads considering the variability of input parameters pointed out base cation deposition, uptake by forest growth and weathering rate as most important factors. The uncertainty of critical loads in general was  $\pm 30\%$ , which was comparable to the uncertainties of input data. The regional mineral weathering, which is an important inherent soil property for acid neutralization, was re-evaluated on the basis of new geochemical survey data leading to one third smaller critical loads than earlier. A new operational DAIQUIRI deposition model was collaboratively developed to provide deposition scenarios and included improved dispersion estimates for domestic nitrogen emissions. All three integrated assessment model systems proved useful in quantifying factors affecting forest soil acidification at different regional and temporal resolutions with improved insight into potential emission reduction requirements. The models helped to convey important findings to decision-makers and the results were used in formulating national policies.

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**Keywords:** integrated, modeling, assessment, acidification, critical load, forest soil, deposition, emission, sulfur, nitrogen

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# 1 Introduction

## 1.1 Spatial and temporal extent of air pollution problems

The detrimental effects caused by atmospheric pollution have been recognized to be one of the major environmental problems in many countries. Local episodic pollution events pose a threat to human health, and regional long-term harmful effects on ecosystems have been of concern in several countries. The long-range transport of pollutants across country borders has made air pollution a truly international problem. The effects are difficult to mitigate with means other than emission controls at the sources. The situation has resulted in intensive and extensive international cooperation in both scientific research and policy-making. Constructive negotiations have already led to several international agreements on issues such as ozone layer protection, acidification and climate change. The main elements of the air pollutant issue are shown in Fig. 1.

Acid rain has been regarded a major threat during the last decades in Europe and North America, because it has caused detrimental changes in the chemical composition of surface waters and soil solutions (Reuss *et al.* 1987, Tamm and Hallbäck 1988). The concern has motivated large research programs at both national and international levels (Cowling and Nilsson 1995). An important series of international acidification conferences on acid rain has started in 1975 with meetings held every five years (Grönlund *et al.* 1995). International legal instruments have been necessary to enable reduc-

tions of emissions of the most detrimental pollutants. The cooperation between governments has led to the adoption of the Convention on Long-range Transboundary Air Pollution (CLRTAP) in 1979 under the auspices of the United Nations Economic Commission for Europe (UN-ECE). Initially the focus has been on the reduction of acid rain by sulfur emission controls. The activities have later been widened to include nitrogen, volatile organic compounds (VOCs), photochemical oxidants, heavy metals and persistent organic pollutants. The European Union has been preparing its own strategies to combat acidification and ground-level ozone.

Human activities cause various types and amounts of air pollutant emissions. A threat to the receptor may arise when these anthropogenic emissions and the consequent air concentrations and depositions are large compared to the natural background. This is the case for the acid rain problem caused by anthropogenic emissions of sulfur, nitrogen oxides and ammonia (Galloway 1995). The resulting depositions may outweigh natural acidification processes and exceed the long-term tolerance of the ecosystem against acidifying deposition.

The effects of air pollutants vary from direct to indirect, from immediate to accumulated. Prominent air pollution effects have been the direct and indirect impacts of sulfur and nitrogen compounds. Direct effects due to increased concentrations of sulfur and nitrogen oxides may affect plants, animals, human health and materials. Many lakes in northern Europe have been slowly acidified due to the accumulated loading and partly through indirect catchment soil acidification, also in Finland (Rask *et al.* 1995, Henriksen *et al.* 1998). The effects on forests and forest soils in Finland (Hari *et al.* 1984, Nöjd 1990), and especially in central Europe have been under intensive investigation (Ulrich 1983, Matzner and Murach 1995). Excessive nitrogen deposition may also cause unwanted effects due to its eutrophication potential. In general, nitrogen-related effects will gain relative importance due to the decreasing sulfur emissions (Ferrier *et al.* 1995). Acidifying emissions are also connected to ground-level ozone, for which nitrogen oxides and volatile organic compounds are precursors.

The signatories of the CLRTAP have agreed to cooperate on research into the effects of major air pollutants and a large organizational setup has

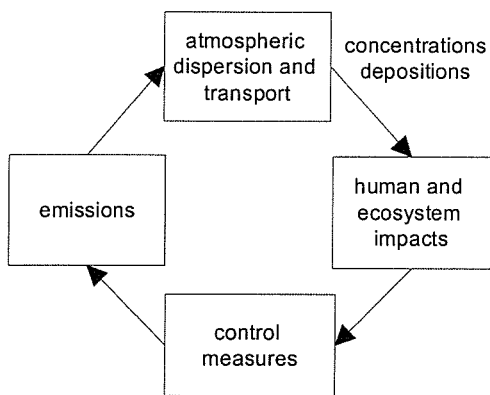


Fig. 1. The main elements of the air pollutant issue.

been operating under this scheme. The coupling of emissions and effects within the acid rain problem is presented in a schematic diagram in Fig. 2. The various feedbacks such as targeted emission reduction requirements are not explicitly shown in the diagram.

Mathematical models and modern computers have enabled simulations of atmospheric and ecosystem processes to estimate the extent of environmental problems due to air pollutant emissions. The temporal and spatial resolution in the models may vary largely depending on the application. Both steady-state (e.g. De Vries *et al.* 1994a, Posch *et al.* 1997c, Warfvinge and Sverdrup 1992) and dynamic models (e.g. Cosby *et al.* 1985, De Vries *et al.* 1989, 1994b, Sverdrup *et al.* 1995) have been developed to predict the acidification of soils, lakes, streams and groundwater. The steady-state methodologies neglect time-dependent processes and finite pools. They are often applied to derive environmental protection targets as basis for policy decisions on emission reduction strategies. Dynamic models are

used to predict responses of ecosystems to changing deposition and land use by including various buffer and adsorption/desorption mechanisms. Thus they enable the assessment of the timing of emission controls.

The international agreements on reductions of air pollutant emissions have often started from a flat percentage reductions as a first step. This approach demands all signatories to reduce national emissions with equal relative amounts, e.g. by 30% (UN-ECE 1996). Further reductions can be based on effectiveness, e.g. best available emission reduction techniques or minimizing environmental effects with funds available. In the effects-oriented approach sensitive geographical areas are identified and mapped and emitters contributing most to the pollutant loading of these areas should be reduced most. If reduction costs are taken into account, cost-effective solutions can be explored as a mathematical optimization problem (e.g. Amann and Klaassen 1995).

An effects-oriented approach may require strict and costly emission reductions if the envi-

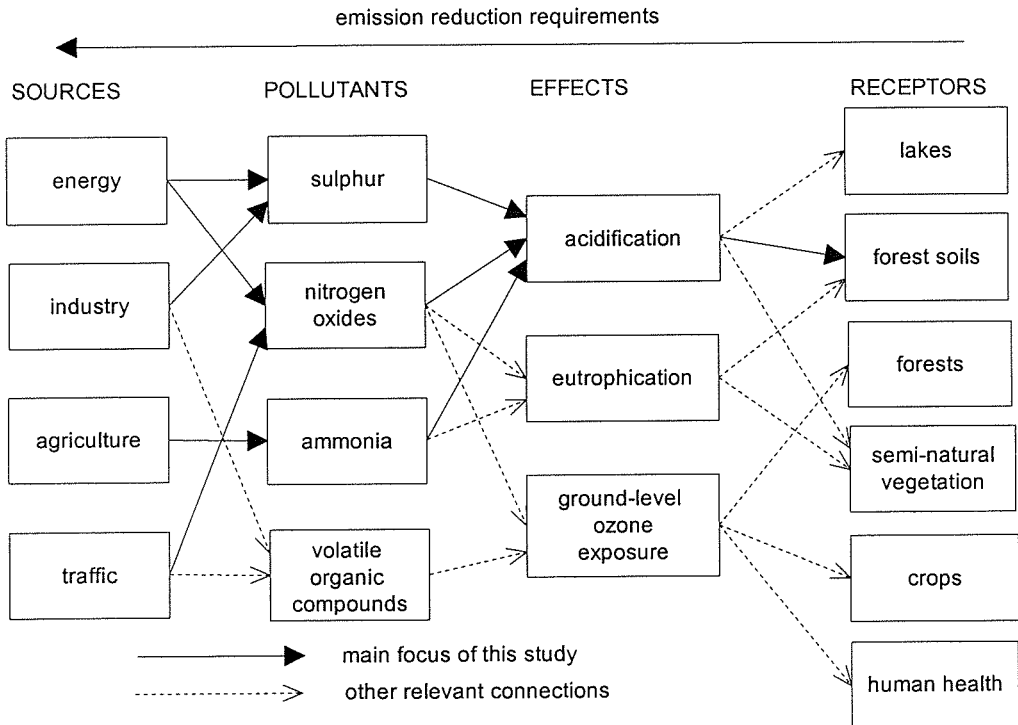


Fig. 2. Schematic diagram of the interrelationships between chemical compounds, their sources, effects and receptors of air pollutants (redrawn after Grennfelt *et al.* 1994).

ronmental targets are very stringent. For example, model studies roughly estimate total costs of about 56000 million ECU per year to further reduce European emissions (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, VOC) from the 1990 level by 2010, according to the current legislation and official reduction plans, however, these measures are still not enough to completely protect ecosystems from acidification and ground-level ozone effects (Amann *et al.* 1998b). Allocation of additional resources to further reduce uncertainties of integrated models would also be cost-effective. As for impact studies, site-specific, regional and even continent wide surveys and monitoring programs on air pollutant effects provide field data, which can be used to assess in detail the ecosystem responses (*e.g.* Henriksen *et al.* 1998, Kleemola and Forsius 1998, Müller-Edzards *et al.* 1997). Studies on monetary benefits of reduced harmful effects have evolved, although dose-response data needed for such studies are often lacking (*e.g.* ExternE 1995).

## 1.2 Concepts in integrated assessment modeling

Many models concentrate on specific segments of the air pollution problem. Emission inventories and projections are made at various levels of spatial and temporal aggregation. Atmospheric models, ranging from local to transboundary resolutions, simulate the chemical reactions, dispersion, concentrations and deposition rates. Economic models may estimate costs related to different emission control techniques or the monetary value of damage. Many models of different types describe the impacts to ecosystems. These models are often developed and used without strong links to other models.

The information of one individual model can serve as input to another model, or they can be coupled together to carry out parallel calculations. A simple example of an integrated model is the linking of a forest growth model to a soil acidification model. An integrated model can be a single model system, such as the RAINS (Regional Acidification INformation and Simulation) model of IIASA (International Institute for Applied Systems Analysis) (Alcamo *et al.* 1990), or a set of individual models connected with each other by a specified input-output interface. Vari-

ous feedback features are usually easier to implement in single model systems. The management of a long chain of interconnected models, which can be partly parallel, may in practice become demanding. Often the development and management of a single integrated model requires considerable resources to achieve meaningful results.

The assessment of an environmental problem usually means a decision-oriented and interactive analysis of existing information. The analysis of existing knowledge needs both collection and interpretation of results, which may be supported with scientific methods. An assessment brings together information in a unified form, often in quantitative terms. The information should be formulated in a way that it can be communicated to and with policy-makers for discussions on related pending decisions. Thus an assessment must be able to pick up relevant conclusions and avoid irrelevant details. This requires knowledge and communication of the probabilities involved or estimates of uncertainty. The assessment itself can be described as a process, whereas models are tools used to assist and support that procedure.

An integrated assessment is the assessment of a specific problem in a wider context with indicated relationships to other issues. Connections to most important details are developed outside the problem. The assessment becomes an overview of the state-of-science, addressing relevant parts of the problem considered. These techniques enable the analysis and evaluation of procedural options on the matter, *i.e.* the policy analysis. However, there are no clear definitions in the literature what elements are necessary or required from an assessment to be labeled 'integrated'. An integrated assessment is essentially team work, bringing together scientists and policy-makers, different methodologies and several disciplines. The main characteristics of integrated assessment could be concentrated in the terms integration, assessment, policy analysis and dialogue.

When an integrated model is used to support an assessment procedure, it may be called an integrated assessment model (IAM). The word 'integrated' can here refer to both 'integrated assessment' and 'integrated model'. An IAM can be a part of the integrated assessment procedure, but an integrated assessment does not necessarily need a mathematical tool or computer models. The goals of integrated assessment and IAM may be well overlapping, but need not to be coincid-

ing. The needs of an integrated assessment will guide the development of a model intended for that procedure.

The connections of various levels of assessment and modeling are schematically illustrated in Fig. 3. Individual models describe the scientific knowledge of a process. These models may be used in the assessment of specific issues. An integrated model includes characteristics of several models. An integrated assessment model may contain all or part of the assessment features of the submodels. In an integrated assessment, the options produced by an IAM will support and affect the policy analysis and finally the decisions.

### 1.3 Integrated models for air pollution

Complex models addressing specific problems can be very effective in answering both scientific and policy questions in the field of their design. However, they may be too complex and data intensive or non-interactive with other models to be operatively used for integrated assessment purposes. Usually, simple models are representative enough and thus most effective in integrated assessment. Existing complex models may also be simplified for integrated modeling purposes (*e.g.* Heyes *et al.* 1996).

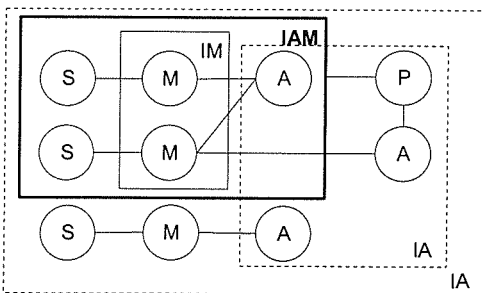
The main driving forces of an environmental problem can be followed through the logical chain of events by integrated modeling. This designates a scenario analysis approach. In the acid rain setting, often the amount and location of emissions change due to the adoption of emission

control measures and the resulting deposition and effects are displayed with proper indicators, such as the ecosystem area under increased risk for damage. The costs for a specified policy to reduce emissions, or a scenario, can be displayed as well. Scenario analysis is an efficient way to quantify and compare factors relevant to the problem and to explore different alternatives to achieve efficient environmental protection.

Another approach is to examine the problem backwards, by optimizing emission reductions to achieve preset targets. In this manner cost-efficient ways are formulated to reduce acidifying emissions to achieve a required overall environmental protection. The optimization is technically challenging due to the complexity of the problem, its mathematical formulation and numerical problems. The results depend on the chosen performance indicators in setting targets, which sometimes may turn out to be unattainable. This leads to the setting of interim targets. Therefore, the interaction between modelers and decision-makers is important to find satisfactory and unambiguous formulation of desired aims and indicators.

The high aggregation levels of spatial, temporal and process descriptions are characteristic for IAMs (Hordijk 1995), the level being determined by the purpose of the models. There is a trade-off between model complexity and regional or temporal applicability (De Vries 1990, 1994). In IAMs the best available detailed estimates on *e.g.* spatial heterogeneity or intra-annual variations of effects can hardly be utilized as such. This emphasizes the need to confirm the performance of aggregated models and identify ranges of applicability and an acceptable level of predictive accuracy (IAEA 1989). The assessment of a single pollutant or effect may be connected to other important pollutants or effects so tightly that is beneficial to include all of them in the model system (*see, e.g.*, Fig. 2). However, this usually demands considerable efforts. Therefore, the relatively high level of aggregation is a prerequisite for the integration of several pollutants and effects in the same assessment. Only carefully designed and restricted aims will lead to an operative integrated modeling system.

The existing integrated assessment models for acid rain (Hordijk 1995, Hordijk and Kroeze 1997a, Hordijk and Kroeze 1997b) include, at a minimum, sources of emissions and an atmospheric module for deposition estimates of at least



**Fig. 3.** A schematic diagram of the possible modeling, assessment and policy-making relationships in the analysis of environmental problems. (S = scientific knowledge, M = model, A = assessment, P = policy analysis, IM = integrated model, IA = integrated assessment, IAM = integrated assessment model.)

one compound, usually sulfur. Most of acid rain IAMs consider environmental impacts and costs of emission controls, but only a few include calculation of benefits. The models can also be characterized by *e.g.* their methodological approach, inclusion of an uncertainty analysis option and the linking method between submodules. In general, emissions are controlled only by end-of-the-pipe technologies. Changes in macroeconomy or energy production structures are exogenous inputs, thus having been defined and calculated outside the IAMs.

The acid rain problem involves of several compounds and effects connected to other issues (Fig. 2). This is notably the case for NO<sub>x</sub>, which plays an important role in acidification, eutrophication and the formation of ground-level ozone. The first protocols on reducing single pollutants have been comparatively straightforward in their integrated assessment modeling and the assumed benefits have outweighed the costs involved. The optimal and fair share of emission reduction burden in different countries becomes an important issue in further emission reductions, when the involved costs have increased (*e.g.* Batterman 1992, Kaitala *et al.* 1992). Other compounds and effects related to the considered environmental problem should be quantified in a comparable way and included in the assessment. Integrated modeling will quickly become complex and lead to a multi-pollutant – multi-effect approach.

Integrated assessment models have supported the work on emission reduction strategies in Europe (Hordijk 1995). The modern approaches are effects-oriented and employ critical loads, the long-term inherent ecosystem tolerance against pollutant load, as basis for environmental protection targets (Hettelingh *et al.* 1995a, Kämäri *et al.* 1992a). The first, and the only one to date, effects-oriented emission reduction protocol supported by IAMs in Europe has been signed under the CLRTAP. In the preparation of the 1994 Sulphur Protocol under CLRTAP (UN-ECE/SP2 1994), the environmental protection target has been to reduce the current sulfur deposition over the long-term ecosystem tolerance, the critical load, by at least 60% all over Europe. In this gap closure approach 1990 has been chosen as base year and the target year for the reduction requirements 2010 with intermediate targets for 2000 and 2005. Three IAMs have been used in evaluat-

ing strategy alternatives: RAINS (Alcamo *et al.* 1990), ASAM (Abatement Strategy Assessment Model) (ApSimon *et al.* 1994) and CASM (Coordinated Abatement Strategy Model) (Gough *et al.* 1995). The models have been capable to separately examine several effects and target settings, and to make optimization for a restricted or full combination of pollutants and effects. In the preparation of second NO<sub>x</sub> protocol under the CLRTAP, the ecosystem area under risk, *i.e.* where the critical loads are exceeded, is chosen as a common basic quantitative indicator to comparing different effects of acidification, eutrophication and ground-level ozone (Amann *et al.* 1998a).

Within the European Union (EU) the ongoing preparation of the National Emission Ceilings (NEC) directive is based on the work of two strategies. In the Community Strategy to Combat Acidification the preliminarily chosen interim environmental protection target is to reduce the ecosystem area with critical load exceedance by at least 50% from the 1990 to 2010 (Amann *et al.* 1998b), although the final aim declared by the European Parliament is to boldly attain the critical loads. In the EU Ozone Strategy, which is closely related to the Acidification Strategy, reductions in ecosystem area with excess exposure and in risks for health impacts and maximum concentration peak cutoffs have been proposed as targets for ground-level ozone. These strategies aim in the preparation of the national emission ceiling directive.

In the present modeling work of both CLRTAP and EU the RAINS model of IIASA is used. Four pollutants (SO<sub>2</sub>, NO<sub>x</sub>, NH<sub>3</sub>, VOC) and four effects (acidification, eutrophication, ground-level ozone impacts on health and vegetation) are simultaneously evaluated. Different computational compensation mechanisms and constraints are analyzed to find alternative cost-effective emission reduction strategies (Amann *et al.* 1998b).

National modeling and other detailed studies are needed in spite of advanced continental integrated model systems. International IAMs may use calculation parameters and data which may not be completely suitable for a country. The results of international IAMs cannot, and do not have to, include such spatial, temporal or sectoral details as often desired by individual countries. To this end national (integrated) modeling is nec-



essary to provide detailed information on input data and calculation parameters to check against the data in international IAMs, to explore in more detail the variation of proposed results within the country and to convey findings and background information for national decision-makers.

In other parts of the world the use of IAMs for direct and extensive policy-making has been less pronounced than in Europe (Hordijk and Kroeze 1997a). In North America, the focus has mostly been on discipline-specific studies of different sectors of the acid rain problem (NAPAP 1991, Rubin 1991). Scenario analyses have been carried out with a set of individual models and the collaboration of different research groups without developing one operational integrated tool. The development of a full IAM has started only recently. In Asia, the rapidly increasing emissions of air pollutants has increased the awareness on the acidity problem. The RAINS model methodology has been applied to Asian conditions (Foell *et al.* 1995, Hettelingh *et al.* 1995b). The environmental impact of acidifying emissions may become a large-scale problem also in Asia and other rapid economic growth areas such as parts of South America (Rodhe *et al.* 1995).

The development of IAMs for acid rain has shown possibilities to take into account several effects simultaneously, including climate change. Linkages between the effects of acid rain and climate change have been presented (Wright and Schindler 1995, Sommaruga-Wögrath 1997, Skjelkvåle and Wright 1998). Simultaneous acidifying and climatic effects on ecosystems have been studied with models, *e.g.* with a regional system consisting of the RAINS model coupled to the IMAGE global change model (Alcamo *et al.* 1995, Posch *et al.* 1996).

#### 1.4 Objectives and structure of the study

This study summarizes the development and application of integrated models in Finland for the assessment of requirements for air pollution control. The aim was to use an effects based approach and estimate the efficiencies of different air pollutant emission reduction strategies, thus giving support to decision-making. The scope is restricted to the assessment of acidification related to forest soils. The transboundary nature of air pollution coupled the work strongly to interdis-

plinary and international research. The overall integration was based on a systematic and modular approach. The most significant factors were identified, best estimates for them were derived and further elaborated beyond the aggregation levels required by the integrated modeling. Uncertainties of parts of the model systems were explored. The research work within this study eventually focused on three distinct integrated model systems sharing partly the same input data and submodels.

The main objective of this study was to develop integrated models, which enable estimates of the extent of air pollution control measures. The subsequent emission reduction requirements were examined by means of scenario analysis. The models quantitatively captured the main processes affecting acidification at levels suitable for integrated modeling. The means to achieve the objectives were:

- to develop and apply integrated assessment models consisting of modules for emissions, atmospheric transport and deposition, forest soil impacts at different spatial and temporal resolutions:
  - regional steady-state (IV)
  - regional dynamic (I, III)
  - site-specific dynamic (VII)
- to derive critical loads as regional environmental protection targets (IV)
- to analyze the input data in further detail and to establish the relative importance of calculation parameters (II, V, VI, VIII)
- to assess the effectiveness and timing of emission reduction measures (III, VII)
- to explore the relation of acidification to other environmental changes (VII)

First, the overall context of the three integrated model systems of this study is presented with a general description of their applications in Finland (chapter 2). Then, the calculation methods and input data are described in detail in chapters 3–5. The sources of emission estimates and means to calculate atmospheric transport and deposition to a receptor are presented (chapter 3). Next, different methods and techniques for impact modeling are discussed. These effect pathways on ecosystems range from steady-state mapping (chapter 4) to dynamic aspects (chapter 5) at both regional and site level. The results from various applications of integrated models are presented in chapter 6. In addition, uncertainties and

soundness of results from integrated models are discussed. Finally, chapter 7 presents conclusions on the use and results from integrated assessment modeling.

Input data, methods and models were continuously updated during the period of this study in order to incorporate the latest scientific findings, especially concerning deposition and critical load maps. The displayed figures, therefore, may not always be comparable to figures in the original articles (I–VIII).

## 2 Development of integrated modeling approaches

### 2.1 The HAKOMA model system

The Finnish integrated acidification model HAKOMA (I, III) was developed to quantify factors relevant to acidification and to find cost-effective emission reduction measures and strategies to decrease acidifying deposition to ecosystems and consequential potential harmful impacts. The structure of the HAKOMA model is shown in Fig. 4a. The basic model system was developed during the Finnish Acidification Research Programme HAPRO (Kauppi *et al.* 1990a, Kauppi *et al.* 1990b, Kenttämies 1991). The different modules and their links in the HAKOMA model were based on the concepts of the RAINS model of IIASA (Alcamo *et al.* 1990). The HAKOMA model consisted of modules for emission inventories and scenarios with emission control technologies and related costs, long-range and mesoscale atmospheric transport and forest soil impacts. The two latter modules were connected to each other with a program interface adapted from the RAINS model source code. Test simulations with the lake module were carried out using depositions from the HAKOMA model. The HAKOMA model was constructed to a user-friendly, distributable software tool for both research and policy purposes, thus loosely following the concept of the RAINS model. Later developments of the model system (after III and Johansson *et al.* 1991) are not covered by this study. The main objectives of the HAKOMA model system were (III):

- to obtain quantitative relationships between

different factors and processes relevant to acidification, to identify the most important factors and the greatest contributors to uncertainties,

- to obtain estimates on the future development of emissions, deposition and impacts on forest soils and lakes, *e.g.* for the design of emission criteria and emission abatement strategies, and
- to collect and present data relevant to acidification in a form which is informative and easy to use.

The emission model has been the first module in the HAKOMA model, containing current and historical inventories of acidifying emissions (sulfur and nitrogen oxides) from Finland and an option to create emission scenarios and abatement strategies. Ammonia emissions have been calculated separately, outside the original emission module (Niskanen *et al.* 1990). Emissions from other countries were obtained from the RAINS model database. The atmospheric module and impact modules were designed after the example of the RAINS model. Long-range transport was computed with the country emission data and unit transfer matrices of EMEP/MSC-W (Co-operative Program for Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe, Meteorological Synthesizing Center - West, in Oslo, Norway). Dispersion of sulfur from domestic sources was calculated with mesoscale transfer matrices prepared by the Finnish Meteorological Institute (I, III). Sulfur was considered the only major acidifying compound in the beginning. The forest soils module was adapted from the RAINS model to enable dynamic simulations of regional forest soil acidification. Effects of several sulfur emission scenarios were assessed and compared to the variation due to input data and method uncertainties. Later, the effects of nitrogen emissions and depositions were demonstrated and the new SMART soil acidification model was tested (Johansson and Savolainen 1991). Lake impacts were assessed in cooperation with the Finnish Environment Institute (formerly the National Board of Waters and the Environment) using the deposition scenarios of the HAKOMA model (III). An optimization module adapted from the RAINS model (Batterman and Amann 1991, Batterman 1992) was later connected to the HAKOMA model system using critical loads of sulfur for forest soils as targets (Johansson *et al.* 1991).

The objectives of the HAKOMA model were achieved to a large extent during the HAPRO program (I, Savolainen and Tähtinen 1990, Niskanen *et al.* 1990, II, III, Kauppi *et al.* 1990a, Kenttämies 1991). The results from regional dynamic soil acidification modeling with various scenarios showed the relative effectiveness of domestic and international emission reductions and pinpointed the importance of different factors contributing to forest soil acidification (I–III). The work also laid ground on further development of critical loads for forest soils as environmental protection targets (Kauppi *et al.* 1990b, Kenttämies 1991). Possibilities for optimization were explored (Johansson *et al.* 1991) and the flexibility of the model system to include a new submodel was demonstrated (Johansson and Savolainen 1991). The results from various modules of the HAKOMA model have later been used in several research studies after the HAPRO program (*e.g.* Kämäri *et al.* 1993, Posch *et al.* 1993a, 1993b, IV, V, Kämäri *et al.* 1995, Tähtinen *et al.* 1997). Support for policy-making was provided in the work of the second Sulfur Committee in Finland (Sulphur Committee II 1993), which used the HAKOMA model results and took note of the effects-based approach but considered deposition levels as suitable indicators for environmental effects.

## 2.2 Critical loads and their exceedances

Critical loads define the maximum pollutant loading which an ecosystem can tolerate in a long time scale and the exceedance is obtained by subtracting critical load from deposition. The methodology for calculating critical loads has been developed in international workshops (*e.g.* Nilsson and Grennfelt 1988, Grennfelt and Thörnelöf 1992) and consequently applied in the countries. The first critical load maps of sulfur for forest soils and surface waters in Finland were presented at the end of the HAPRO program (Kämäri *et al.* 1991). The soil data of the HAKOMA model were used in the mapping of critical load of sulfur for forest soils. The exceedances of critical loads by acidifying deposition were calculated using the modeled depositions from the HAKOMA model (III). The national mapping of critical loads for acidification was later developed further (Kämäri *et al.* 1992b) and harmonized with other

countries (IV). The development of the critical load methodology has been rapid and the guideline suggestions have been adapted to national mapping programs (Hettelingh *et al.* 1991, Downing *et al.* 1993, Posch *et al.* 1995, Posch *et al.* 1997a). The effects of eutrophication by nutrient nitrogen were later included in the calculations (Rosén *et al.* 1992). The calculation and mapping of critical loads is currently carried out at the Finnish Environment Institute (FEI) as a routine activity of the National Focal Center (NFC) for Mapping Critical Loads and Thresholds in Finland as part of the European network (Posch *et al.* 1997a). During the work in this study both methodological and input data improvements were explored, *e.g.* problems in displaying the results (Posch *et al.* 1993b), analysis of uncertainties (V) and improvements of important parameters such as mineral weathering (VI) and modeled depositions (VIII).

Currently, the critical load integrated model system (CLIM) includes modules for emissions, atmospheric transport and deposition and the critical load maps. The emission inventories and scenarios are based on national and international official emission inventories and reduction plans. Atmospheric dispersion and transport descriptions are based on unit transfer matrices for both long-range transboundary pollution and national mesoscale dispersion collected into the DAIQUIRI deposition model (III, VIII). Modeled depositions are used to estimate current and future exceedances of critical loads. The maps of critical loads for lakes and soils as environmental protection targets are calculated using best available input data and harmonized methods, which are based on agreed effects criteria. The current model system is presented in Fig. 4b.

Critical load maps have been used both nationally and internationally in support for policy-making. The Acidification Committee in Finland (Acidification Committee 1998, Syri *et al.* in press) used critical loads as environmental indicators for national and international emission reduction strategy assessment. The ongoing negotiations under CLRTAP for the next protocol on nitrogen oxides and other related compounds are effects-oriented and employ critical loads as environmental protection targets (Posch *et al.* 1997a, Amann *et al.* 1998a). The parallel development of a national emission ceiling directive from the acidification and ozone strategies of

EU is based on the same approach (Amann *et al.* 1998b).

### 2.3 Dynamic model systems

Dynamic acidification models include the temporal aspects of acidification processes. Model applications in Finland have concentrated on site-specific simulations using the SMART (Simulation Model for Acidification's Regional Trends) model (Kämäri *et al.* 1995, Johansson 1995b, Forsius *et al.* 1996, Forsius *et al.* 1997, VIII, Johansson 1997). The regional dynamic modeling with the RAINS soil model was included in the HAKOMA model studies, but only test simulations were carried out with the SMART model in the HAKOMA model system (Johansson and Savolainen 1991).

The modules for the dynamic integrated model system (DIM) are conceptually the same as for critical load calculations: emissions, atmospheric transport and deposition and the dynamic impact model. The important aspect is the need for temporal development of many input parameters. A simple approach is to derive coarse estimates for major parameter changes, *e.g.* a 30% decrease in acidifying sulfur deposition in 2010, to study general responses of the receptor. More detailed estimates or even new submodels are needed to create relationships to more sophisticated emission reduction strategies or forest harvesting schemes. The current model system for site-specific dynamic modeling includes the DAIQUIRI deposition model for present and future depositions based on emission scenarios (VIII) and the soil acidification model SMART (De Vries *et al.* 1989). Other additional models, such as the DEPUPT model reconstructing historical depositions and describing dynamic forest growth, a climate generator and hydrological models, were soft linked to the basic modules (VII). The schematic diagram for dynamic integrated model system is presented in Fig. 4c.

Dynamic modeling is in general used to study in detail different processes related to acidification of soils and surface waters and to provide information on temporal responses to changes in major driving forces such as acidifying deposition. Applications in Finland ranged from regional with simple dynamic models (I, II) to site-specific with more complex models coupled to

other dynamic modules (VII). The use of dynamic model in decision support has not yet reached maturity (Forsius *et al.* 1997).

### 2.4 Characteristics of different approaches

The three main integrated model systems presented in this study are the Finnish integrated acidification model HAKOMA, the critical load integrated model system (CLIM) and the dynamic integrated model system (DIM), mainly for site-specific applications. These approaches share common methods, models and input data. All model systems are deterministic and mainly process-oriented. This choice enabled adequate information detail with appropriate regional applicability (De Vries 1990), however, the aggregation level of input parameters depended on specific applications. All systems were designed to be used interactively with a personal computer.

The three systems also differ from each other in many aspects. The HAKOMA model was devised to be a fully integrated single model system and it had a user-friendly interface. However, the full integration was operative only for deposition and soil acidification calculations. The emission module was soft linked as a stand-alone module producing required output for later calculations. The critical load integrated model system was designed as a decision support tool. The soft linked system consisted of separate computer programs with data exchange through compatible input-output files. Dynamic integrated modeling was based on the use of existing separate models soft linked to each other. The model characteristics are displayed in schematic diagrams in Fig. 4 and technical details are summarized in Table 1.

## 3 Derivation of emission and deposition values

### 3.1 Emissions

#### 3.1.1 General

Emission estimates are required in proper spatial and temporal detail for deposition models and integrated assessment purposes. The modeling of

atmospheric dispersion and transport benefits from as detailed emission estimates as possible, concerning the spatial distribution, temporal and chemical aspects and effective emission heights. The spatial and temporal resolutions of emissions should be in harmony with the atmospheric dispersion and transport model employed. Emission source categories are useful for sectoral allocation of reduction requirements, but they are not necessary for impact assessment purposes. Usually an-

nual total country emissions are used in IAMs in Europe.

### 3.1.2 Finland

The national register on air pollution permits of the environment administration has been the basis or a reference database for most Finnish emission estimates for stationary sulfur and nitrogen oxides sources (*e.g.* I, Savolainen and Tähtinen 1990).

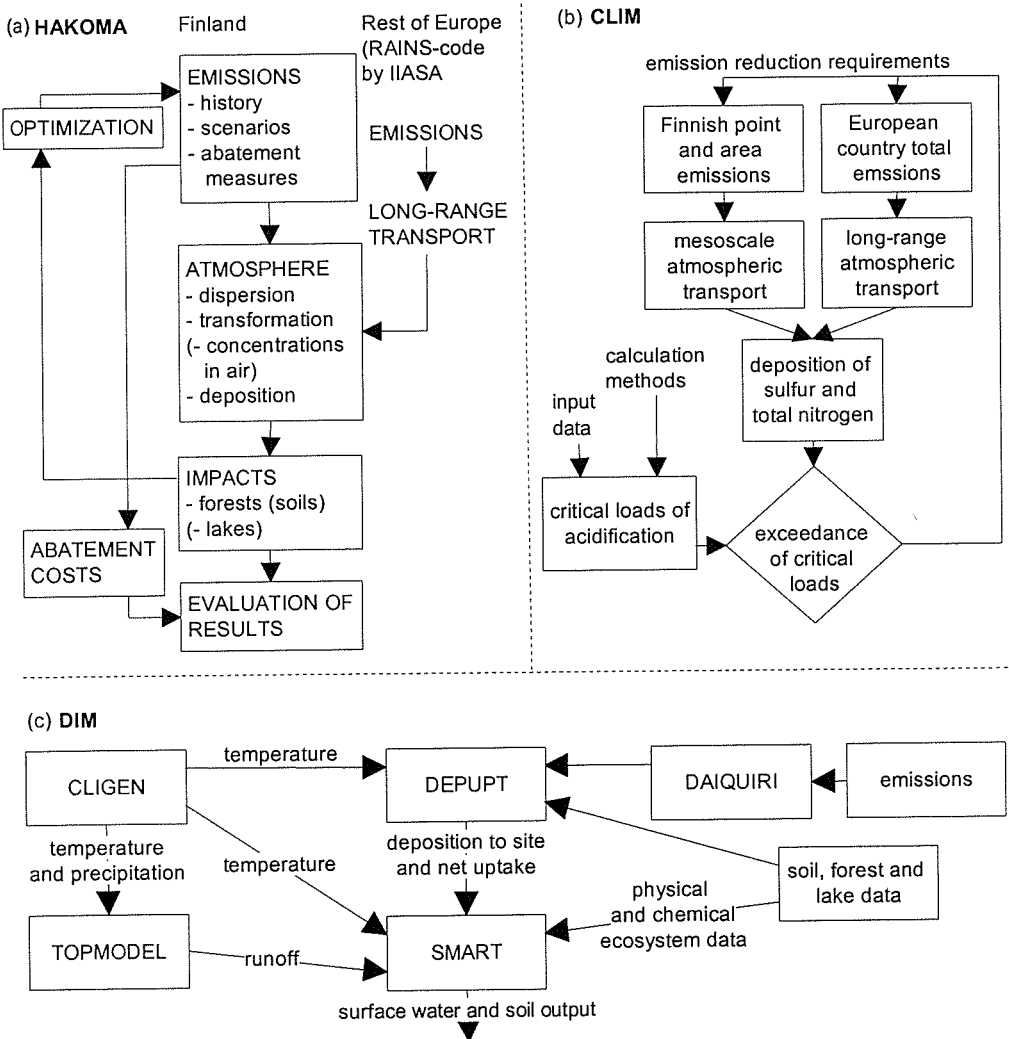


Fig. 4. Model diagrams for (a) the Finnish integrated acidification model HAKOMA (redrawn after I, III), (b) the critical load integrated model system (CLIM) (IV, Kämäri *et al.* 1992b) and (c) the dynamic integrated model system (DIM) (VII).

**Table 1.** Technical details of the three integrated model systems summarized in this study including temporal and spatial resolutions: Finnish integrated acidification model HAKOMA, critical load integrated model system (CLIM) and dynamic integrated model system (DIM) (I–IV, VII, VIII).

	HAKOMA	CLIM	DIM
<b>Emissions:</b>			
– pollutants <sup>1</sup>	SO <sub>2</sub> , NO <sub>x</sub> , NH <sub>3</sub>	SO <sub>2</sub> , NO <sub>x</sub> , NH <sub>3</sub>	SO <sub>2</sub> , NO <sub>x</sub> , NH <sub>3</sub>
– method	calculated	input	input
– emission sources <sup>2</sup>			
– domestic	fuel/activity	country total	country total
– long-range	country total	country total	country total
– spatial resolution <sup>3</sup>			
– domestic	ps/as	ps/as	ps/as
– long-range	country total	country total	country total
– time step	annual	annual	annual
<b>Transfer matrices for deposition<sup>4</sup>:</b>			
– pollutants	S (later NO <sub>y</sub> , NH <sub>x</sub> <sup>5</sup> )	S, NO <sub>y</sub> , NH <sub>x</sub>	S, NO <sub>y</sub> , NH <sub>x</sub>
– spatial resolution (S,N) <sup>6</sup>			
– mesoscale	0.25° × 0.125°	0.25° × 0.125°	0.25° × 0.125°
– long-range	150 km × 150 km	150 km × 150 km	150 km × 150 km
– time period <sup>7</sup>			
– time step	annual	annual	annual
– mesoscale S	ave (10 a)	ave (10 a)	ave (10 a)
– mesoscale N	ave+annual (1 a)	ave+annual (2 a)	ave+annual (2 a)
– long-range	ave+annual (4 a)	ave+annual (11 a)	ave+annual (11 a)
<b>Impacts<sup>8</sup>:</b>			
– effects	acid (S, later N)	acid (S,N), eutr (N)	acid (S,N), eutr (N), others
– receptors	soil	soil, water	soil, water
– method	dynamic, steady-state	steady-state	dynamic
– time step	annual	annual	annual/monthly
– spatial	site, 0.25° × 0.125°	0.25° × 0.125°	site, 0.25° × 0.125°
<b>Integration<sup>9</sup>:</b>			
IAM method <sup>10</sup> :	full/soft simulation, optimization (S)	soft simulation	soft simulation
Period <sup>11</sup> :	1950–2040	steady-state	1800–2100

Abbreviations and explanations (for further details refer to text in relevant chapters):

<sup>1</sup> SO<sub>2</sub> = sulfur oxides, NO<sub>x</sub> = nitrogen oxides, NH<sub>3</sub> = ammonia, N = all nitrogen species

<sup>2</sup> domestic = Finland, long-range = rest of Europe; for sulfur the areas nearby Finland (Estonia, St. Petersburg and Leningrad Area, Karelian Republic, Murmansk Area) were treated as domestic sources

<sup>3</sup> ps = (large) point source, as = area sources aggregated into municipality level

<sup>4</sup> transfer matrices describe the atmospheric dispersion, transport and deposition; NO<sub>y</sub> = nitrogen oxides,

NH<sub>x</sub> = ammonia

<sup>5</sup> the transfer matrices for N in the HAKOMA model were different from those of the DAIQUIRI model

<sup>6</sup> mesoscale = mesoscale transfer matrices based on Finnish deposition models for S and N; 0.25° × 0.125° = the calculation grid of 0.25° longitude × 0.125° latitude covering the whole Finland and used by the National Focal Center for Mapping Critical Loads and Thresholds; 150 km × 150 km = the 150 km × 150 km European calculation grid of the EMEP/MSC-W long-range transport deposition model

<sup>7</sup> ave = averaged linear transfer matrices of several meteorological years (mesoscale in Finland: S statistics from 1972–1981, N: simulations in 1990 and 1993; long-range by EMEP/MSC-W: simulations in 1985–1995)

<sup>8</sup> acid = acidification, eutr = eutrophication caused by nutrient N, soil = forest soils, water = surface waters

<sup>9</sup> full/soft = links between modules fully integrated or via output-input transfer

<sup>10</sup> method of integrated modeling can be based on scenario simulation or optimization methods

<sup>11</sup> analysis period used for most simulations; for critical loads exceedances can be calculated using deposition from any year desired

The data on point sources are supplemented by estimates on other emission sources such as traffic, domestic heating and agriculture (Melanen and Ekqvist 1997).

The emission module of the HAKOMA model (I, Savolainen and Tähtinen 1990) describes national annual sulfur emissions from 44 emission areas with three emission height classes in Finland. The emission point in each area is located at the biggest source in the area, with other emissions in the vicinity aggregated to it. The first annual nitrogen emissions have been estimated at municipality level, resulting in about 450 emission points in Finland (III). Historical emissions back to 1950 and future scenarios have been calculated at the same spatial resolution.

Emission data have been available from other sources, too (Melanen and Ekqvist 1997, Grönroos *et al.* 1998, Acidification Committee 1998, VIII). The official CORINAIR air emission inventory for Finland is maintained at the Finnish Environment Institute (FEI). The inventory is based on the methodology of the European Environment Agency of the European Union. The CORINAIR air emission inventory for 1990 has provided the basis for some of the emission estimates used in this and other studies (VIII, Amann *et al.* 1998a, 1998b).

### 3.1.3 Areas nearby Finland

Spatially detailed data on sulfur emissions from areas nearby Finland were used in calculating depositions. Early sulfur emission estimates from Soviet Union areas nearby Finland in the HAKOMA model were rough estimates only (Kulmala 1989, I, Savolainen and Tähtinen 1990, III, Johansson *et al.* 1991). The estimates were refined with Russian official data covering the years 1980–1994, based on data exchange with the Atmosfera institute in St. Petersburg. The emission areas concerned are St. Petersburg and Leningrad area, Karelian Republic and Murmansk area. Some more detailed data were supplied by the Finnish Meteorological Institute (FMI) (Tuovinen *et al.* 1993, Kämäri *et al.* 1995). Estonian sulfur emission estimates employed are based mainly on studies made at FMI (Kivivasara 1994). Later estimates collected at FMI or Technical Research Centre of Finland were not implemented in the inventories used in this study (Häkkinen *et al.* 1995, Tähtinen *et al.* 1997). The

uncertainty of the emissions from areas nearby Finland should be taken into account when evaluating model computed depositions.

### 3.1.4 Rest of Europe

Emissions from outside Finland have been available from mainly two sources: EMEP/MSC-W center for long-range transboundary air pollution and the RAINS model. Emissions are reported as one-year totals for countries or other specified emission areas.

EMEP/MSC-W reports country-level official emission inventory data for European countries. The data are based on official submissions to the UN-ECE. They are cross-checked with the country-specific CORINAIR air emission inventory data, if available, which should coincide with official country totals (EMEP/MSC-W 1998). However, the emission totals of the countries are not necessarily consistently calculated and may retain significant uncertainties. The historical, current and future emissions according to officially announced current reduction plans (CRP) are reported. Data are available from 1980 and 1985–1996 and future emissions according to CRP from 2000, 2005 and 2010. The latest spatial distribution of emissions within the emission area and two emission heights are used in the atmospheric transport model simulations of the EMEP/MSC-W.

The RAINS model of IIASA has its own emission calculation module (RAINS 1998), which uses the energy balance of a country as input. The aggregation level and methodology within the emission module is directed at scenario development and the application of various control technologies and related costs. The emission module has been calibrated to reproduce the 1990 emissions for countries where official CORINAIR air emission inventory results are available. Several scenarios derived with the RAINS model of IIASA were used in Finnish applications, especially the effects of current national and European Union legislation (current legislation, CLE) and the maximum technically feasible reduction measures (MFR) scenarios.

The relative importance of different acidifying compounds and their temporal development are shown in Fig. 5 for both Europe and Finland (EMEP/MSC-W 1998) in moles of charge or equivalents (Miinusmaa 1990). The development

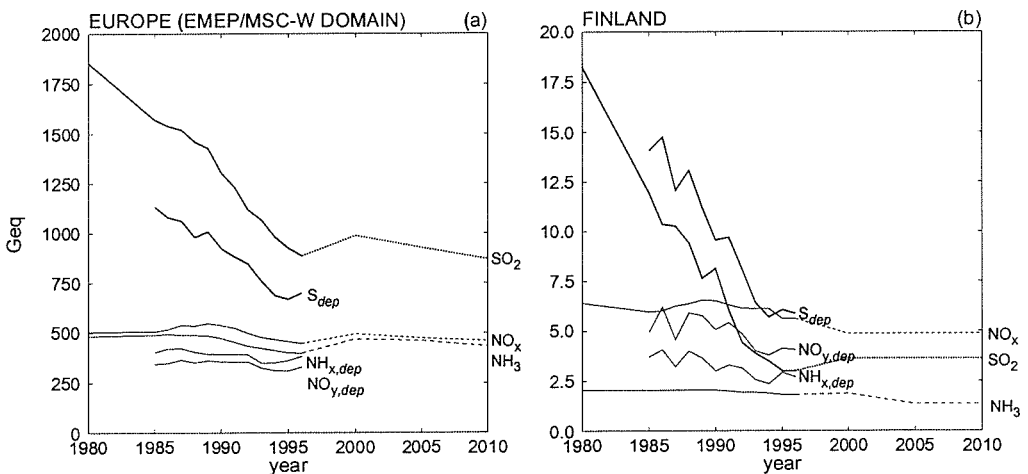
of total sulfur emission and deposition in Europe, roughly equivalent to the EMEP/MSC-W modeling domain and including the sea areas, has been sharply decreasing since 1980. The changes in nitrogen compounds have not been very large and their contribution to the acid rain problem is still at most equal to that of sulfur alone. The sulfur emissions reduction in Europe and in Finland during 1980–1995 was 50% and 84%, respectively, and for total nitrogen 14% and 12%, respectively. All acidifying emissions were reduced 37% in Europe and 61% in Finland. Finnish emissions and depositions are about 1% of those of Europe. The sulfur emissions and depositions in Finland have declined substantially, more than on the average in Europe. For nitrogen compounds a slightly decreasing trend is visible. During 1985–1995 the sulfur deposition has decreased in Europe and in Finland 41% and 57%, respectively, and the nitrogen deposition 10% and 18%, respectively. The country imports more sulfur and ammonia than it exports, while the flux is opposite for nitrogen oxides. It is evident that nitrogen has become more important than earlier. Note, that the deposition values for 1996 are not directly comparable with data from earlier years due to an extension of the calculation domain and new type of meteorological data (EMEP/MSC-W 1998), however, these values in Fig. 5 were modified for compatibility by EMEP/MSC-W.

## 3.2 Atmospheric transport and deposition

### 3.2.1 Linearized source-receptor relationships

Models for the transport of air pollutants describe processes in the atmosphere (dispersion, transport, chemical and physical transformation, deposition) for different chemical compounds. The practical outputs of the models are air concentrations and depositions to surfaces, which are compared to measurements to gauge the performance of the model (EMEP/MSC-W 1998). These models include a mathematical description of chemical and physical processes and they require spatially and temporally detailed meteorological input data and long computing times on powerful computers. Their applicability to integrated assessment is restricted, where repeated deposition calculations due to various emission scenarios are needed. In terms of working time and ease-of-use, there is a need for tools of a more operational nature, which are capable of connecting emissions to depositions via simplified descriptions.

Linear relationships with a certain accuracy can be established between emissions and depositions from the simulation results of detailed atmospheric models. A transfer coefficient describes the deposition or concentration due to a



**Fig. 5.** The temporal development of emissions (1980–2010) in Finland and Europe and depositions (1985–1995) of sulfur and nitrogen compounds to Finland and to Europe (or within the EMEP/MSC-W calculation domain) (EMEP/MSC-W 1998). Future depositions are based on the current reduction plans (CRP) reported by countries to the UN-ECE. Both emissions (SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub>) and depositions (S<sub>dep</sub>, NO<sub>y,dep</sub> and NH<sub>x,dep</sub>) are displayed as moles of charge, or equivalents. The values for 1996 depositions were modified for compatibility by EMEP/MSC-W for this figure.



unit emission source at a given receptor. A transfer matrix is a collection of these coefficients. An example is a matrix derived from EMEP/MSC-W modeling results describing the transport from emissions of a country to grid cells of about  $150 \text{ km} \times 150 \text{ km}$  covering the whole of Europe (for definition of that grid system see Saltbones and Dovland 1983, Barrett and Berge 1996, Posch *et al.* 1997a).

A transfer matrix can be assumed to describe the dispersion within an area of sufficiently homogeneous climatology and surface properties. Thus, such a matrix can be centered at any single emission point to produce a deposition field around that point. The total deposition is formed by adding up these separate fields due to each emission point at different geographical locations. An example of this approach is the sulfur mesoscale transfer matrix in the HAKOMA model (I, III).

The average deposition in the cells of the calculation grid is assumed to vary linearly according to changes in the emissions, *i.e.* a change in emissions results in equal relative change in deposition. The uncertainty in the assumption on linearity depends on the compound, emission levels and the amount of other interacting chemicals in the atmosphere. Linearity can be assumed to hold for variations within acceptable limits (Alcamo and Bartnicki 1987). However, the nonlinearity issues should be recognized, especially under extreme reductions of emissions.

The linear relationship can represent a single meteorological year or an average of several simulation years. The effect of interannual meteorological variability is reduced in the average matrix, which makes it appealing to use it for future deposition scenario calculation.

In the following, the use of linearized transfer matrices is described for different spatial resolutions:

- i) the long-range transport at European level, which employs country-to-grid transfer matrices in the  $150 \text{ km} \times 150 \text{ km}$  grid, and
- ii) the mesoscale range, which uses point-to-grid transfer matrices for emission sources in Finland (sulfur and nitrogen) and areas nearby (sulfur only) in the basic national grid of  $52 \times 88$  cells, each  $0.25^\circ$  longitude  $\times$   $0.125^\circ$  latitude in size (approximately  $14 \text{ km} \times 14 \text{ km}$  in southern Finland at  $60^\circ\text{N}$ ).

### 3.2.2 Long-range transfer matrices

Extensive modeling of long-range transport of air pollutants in Europe has been carried out at the EMEP/MSC-W center under the Convention on Long-range Transboundary Air Pollution (CLRTAP) (EMEP/MSC-W 1998). The major modeling scheme relevant for integrated assessment in Europe has been the Lagrangian trajectory acid deposition and ozone models with  $150 \text{ km} \times 150 \text{ km}$  grid resolution. EMEP/MSC-W has provided data on annual country total emissions and resulting modeled depositions in the  $150 \text{ km} \times 150 \text{ km}$  grid separately due to emissions from each country. These data were converted into unit transfer matrices by dividing the deposition in a grid cell originating from a specific country emission by that emission value. These linear relationships were averaged over the available simulation years and used for estimating scenarios for future total sulfur and nitrogen depositions based on country emission estimates. Matrices for four meteorological years were available in the first Finnish assessments with the HAKOMA model (I–III). Results for eleven years (1985–1995) were used (Berge 1997) in later studies (VII, VIII).

The deposition to an  $150 \text{ km} \times 150 \text{ km}$  grid cell is an average value for that area. The within-grid variation is not available. The filtering factor  $\phi$  was used to redistribute the deposition between open and forested area of the grid cell maintaining the mass deposited (Kauppi *et al.* 1986, I–III, V). Results on redistributing the deposition according to land use and consequent varying deposition velocity has been presented (Erismann *et al.* 1994, 1995). Modeled depositions at the  $50 \text{ km} \times 50 \text{ km}$  of the Eulerian model of EMEP/MSC-W have been available for preliminary studies only recently (Jakobsen *et al.* 1997). An aggregated deposition estimate limits studies on ecosystem effects in areas where steep deposition gradients occur. In this study the deposition to a receptor point has been interpolated with inverse square distance weighing from four surrounding  $150 \text{ km} \times 150 \text{ km}$  grid cells. This procedure inevitably leads to slight underestimation of depositions peaks within the grid cell but avoids sharp deposition changes at grid cell borders.

### 3.2.3 Mesoscale transfer matrices

The mesoscale transfer matrices for sulfur used in the HAKOMA model (I, III) have been derived at FMI using a mesoscale sulfur model (Nordlund *et al.* 1985, Nordlund and Tuovinen 1988). The model is of a hybrid type containing Eulerian, Lagrangian, and statistical features and it employed synoptic meteorological observations over the period 1972–1981. The transfer matrices have been calculated at FMI for 14 km × 14 km grid cells for four climatological zones in Finland and for three effective emission heights. A comparison of interpolated sulfur deposition from the HAKOMA model to calculated sulfur fluxes in surface waters has shown a good general agreement except for Eastern Lapland (Forsius 1992).

The first illustrative estimates on nitrogen mesoscale transfer matrices were introduced in the HAKOMA model, based on experimental data on ammonia submitted by the FMI (III). First, the deposition field has been calculated by using all emissions and then, another field without emissions from one grid cell in central Finland. However, these first estimates were not implemented for the early use in the HAKOMA model.

Operational mesoscale transfer matrices for nitrogen compounds were added later into the HAKOMA model (Johansson and Savolainen 1991). One transfer matrix for both nitrogen oxides and ammonia was derived. The primary data covering the whole country had been calculated at FMI as a grid-to-grid description describing the deposition of one emission grid cell to all surrounding receptors. The 150 km × 150 km grid size was considered too coarse for mesoscale descriptions in the HAKOMA model, even when the dispersion from each emission grid cell was available. Therefore, all grid-to-grid calculations from all Finnish emission grid cells were averaged and the resulting data were interpolated into the national 0.25° longitude × 0.125° latitude grid system consistent with the HAKOMA model data management. This average transfer matrix was separately centered to all emission points to produce deposition fields, which then were summed for total deposition. Both transfer matrices assumed a single average emission height for emissions. Since for ammonia the emission estimates had a large uncertainty at that time and the near-source deposition was known to be signifi-

cant, 20% of emissions were assumed to remain in the emission cell and the internal deposition structure of that cell was refined with a S-shaped power function. Due to the uncertainty entailed in these modifications the nitrogen depositions were used for approximating regional deposition levels and not accurate site-specific values (V).

A new deposition model called DAIQUIRI (Deposition, AIr QUality and Integrated Regional Information) was developed as a collaborative effort for both the critical load and dynamic modeling needs (VIII). The model incorporated the latest long-range transport matrices from EMEP/MSC-W data and the mesoscale transfer matrices calculated at FMI. The major improvement was made in the preparation of new nitrogen mesoscale transfer matrices, for which the primary data and linearization have been calculated at FMI. The modification and extension of matrices and other software development was done at FEI as a team effort. Currently, the DAIQUIRI model covers sulfur, nitrogen oxides and ammonia deposition estimates at different spatial and temporal resolutions due to emissions from selectable areas under one computer program. The mesoscale transfer matrices for sulfur are the same as in the HAKOMA model (III). For nitrogen compounds matrices for two years (1990 and 1993) are currently available. In addition, the wet and dry fractions of mesoscale nitrogen depositions are included. Transfer matrices for long-range transport are available for 1985–1995 and are based on the EMEP/MSC-W Lagrangian model (Berge 1997). Estimates for historical total depositions for sulfur and nitrogen in Europe were made available from the DEPUPT model data (VII). Future total depositions are calculated by adding the depositions due to both mesoscale and long-range transfer matrices with emissions corresponding to the chosen scenario. The schematic diagram of the DAIQUIRI model is presented in Fig. 6.

The new mesoscale transfer matrices for nitrogen oxides and ammonia for the DAIQUIRI model (VIII) have been derived at FMI using results of the regional air quality model based on the FINOX model (Hongisto 1992). First, the deposition has been calculated with actual full domain emissions and boundary concentrations. Second, new calculations have been made after eliminating a single emission source from the center of the calculation area, in this case in cen-

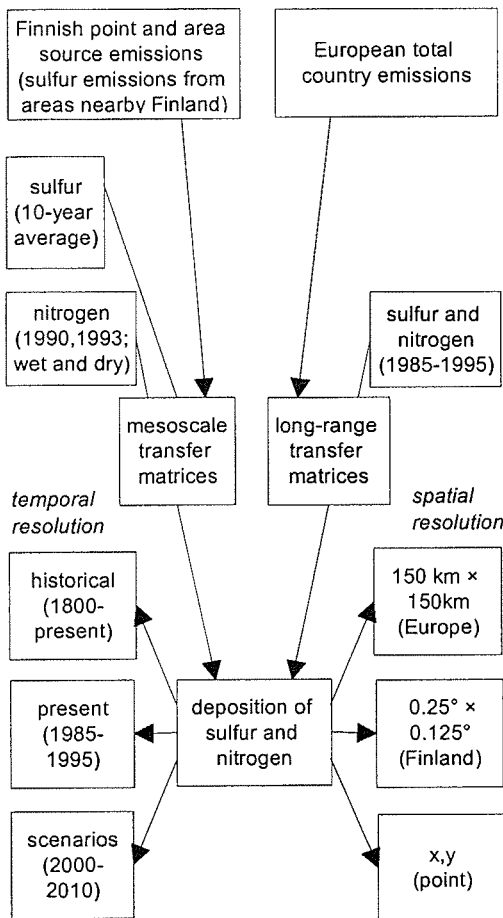


Fig. 6. Schematic diagram of the DAIQUIRI deposition model (VIII).

tral Finland. The difference between these two deposition fields gives the annual unit deposition caused by this one source. This indirect method gives a more reliable estimate on dispersion in current chemical atmosphere than by using a unit source in the absence of other emissions. However, no increases due to some nonlinearities in nitrogen chemistry in the latter deposition field were allowed after eliminating one source. The matrices for nitrogen oxides were prepared for three different emission height classes, but only one was considered for ammonia.

The overall regional deposition patterns of both nitrogen compounds were quite similar with the FMI regional air quality model and DAIQUIRI models (VIII). The DAIQUIRI model overestimated  $\text{NO}_y$  depositions near coastlines

and for  $\text{NH}_x$  it calculated higher near-source depositions. The total depositions modeled with the FMI regional air quality model and the DAIQUIRI model were compared against measured annual values of 1990 from measuring stations of the FMI (Leinonen and Junto 1991). The measurements include wet deposition and only a fraction of dry deposition reaching the collector, whereas the models attempt to estimate total deposition by separately calculating wet and dry deposition. The correlation between measured and deposition modeled with the DAIQUIRI model was found good at all deposition levels for inland stations (VIII). The linear coefficient gives an indication of the amount of dry deposition not present in precipitation measurements. At coastal measuring stations no clear correlation between measurements and modeled depositions was detected. This should be taken into account when using the results of the current model version. As the calculated deposition values include a considerable long-range transported share based on the EMEP/MSC-W model results, the discrepancy reflects the tendency of the EMEP model to underestimate components in precipitation in northern Europe in general and at sites exposed to marine conditions or due to underestimation of anthropogenic emissions in the Baltic Sea area from ships (Sandnes and Styve 1992, Kangas *et al.* 1994). Future comparison will become easier through the implementation of separate transfer matrices for wet and dry depositions.

An example of the evolution of modeled nitrogen depositions is shown in Fig. 7. First, nitrogen depositions due to the old emission estimates of 257 Gg  $\text{NO}_x$  and 43 Gg  $\text{NH}_3$  from Finland for 1990 (HAKOMA 1992, Niskanen *et al.* 1990) were calculated with old transfer matrices from the HAKOMA model and new transfer matrices of the DAIQUIRI model. In general, the HAKOMA model tends to distribute the deposition further from emission sources and gives lower overall deposition levels than the DAIQUIRI model. For both depositions  $\text{NO}_y$  and  $\text{NH}_x$ , the DAIQUIRI model gives clearly higher short-range deposition than the HAKOMA model and even more so for  $\text{NO}_y$ . This can be partly explained by the simplified method to derive transfer matrices in the HAKOMA model. Note, that the old  $\text{NO}_x$  emissions for the DAIQUIRI model were allocated to three emission height classes according to the relative distribution of the up-

dated emissions. If all old emissions had been in the highest class, the distribution of  $\text{NO}_y$  deposition from the DAIQUIRI model would have been similar to that of the HAKOMA model but with deposition twice as high in southern Finland. Next, updated emission estimates of 274 Gg  $\text{NO}_x$  and 38 Gg  $\text{NH}_3$  for 1990 (VIII, Grönroos *et al.* 1998) were used as input to calculate deposition with the DAIQUIRI model. The changes in emission estimates had a clear effect on depositions, especially for  $\text{NO}_y$ . The comparison suggested that both the location and the estimated quantity of emissions can have as large impacts on modeled depositions as the representative performance of the atmospheric dispersion description. Note, that different sources of information may contain different emission values, *e.g.* reported  $\text{NO}_x$  and  $\text{NH}_3$  emissions in 1990 varied from 300 and 35 Gg (UN-ECE 1997, EMEP/ MSC-W 1998) to 276 and 40 Gg (Amann 1998b), respectively. The former estimates are officially submitted current country data and the latter values are based on Finnish CORINAIR air emission inventory data for 1990, which have become outdated. The modelled depositions have in general improved due to better emission estimates and new revised mesoscale transfer matrices.

### 3.2.4 Deposition histories

Dynamic acidification models generally need historical deposition estimates in their calibration. Historical data on sulfur emissions and depositions from 1880–1991 are available in the 150 km  $\times$  150 km grid (Mylona 1993, 1996). Despite some uncertainties, *e.g.* the use of only one meteorological year in deposition modeling, the results are currently the best estimate for the spatial resolution of historical deposition across Europe. Nitrogen history used in model applications are based on the aggregated European nitrogen emission history and estimates compiled for other studies (Alveteg *et al.* 1998b, Sverdrup *et al.* 1995, Asman *et al.* 1988).

These data were compiled and made operational in the DEPUPT model taking into account measurements and forest characteristics of a site (VII). The histories were also delivered into the DAIQUIRI model (VIII), however, without the forest filtering effect. This enabled rough esti-

mates on changes in historical depositions without obtaining separate emission inventories from the past. Although the uncertainties on emission and deposition histories for nitrogen are still considerable, the description of historical depositions were included in the model systems of this study (VII).

### 3.2.5 Site-specific estimates

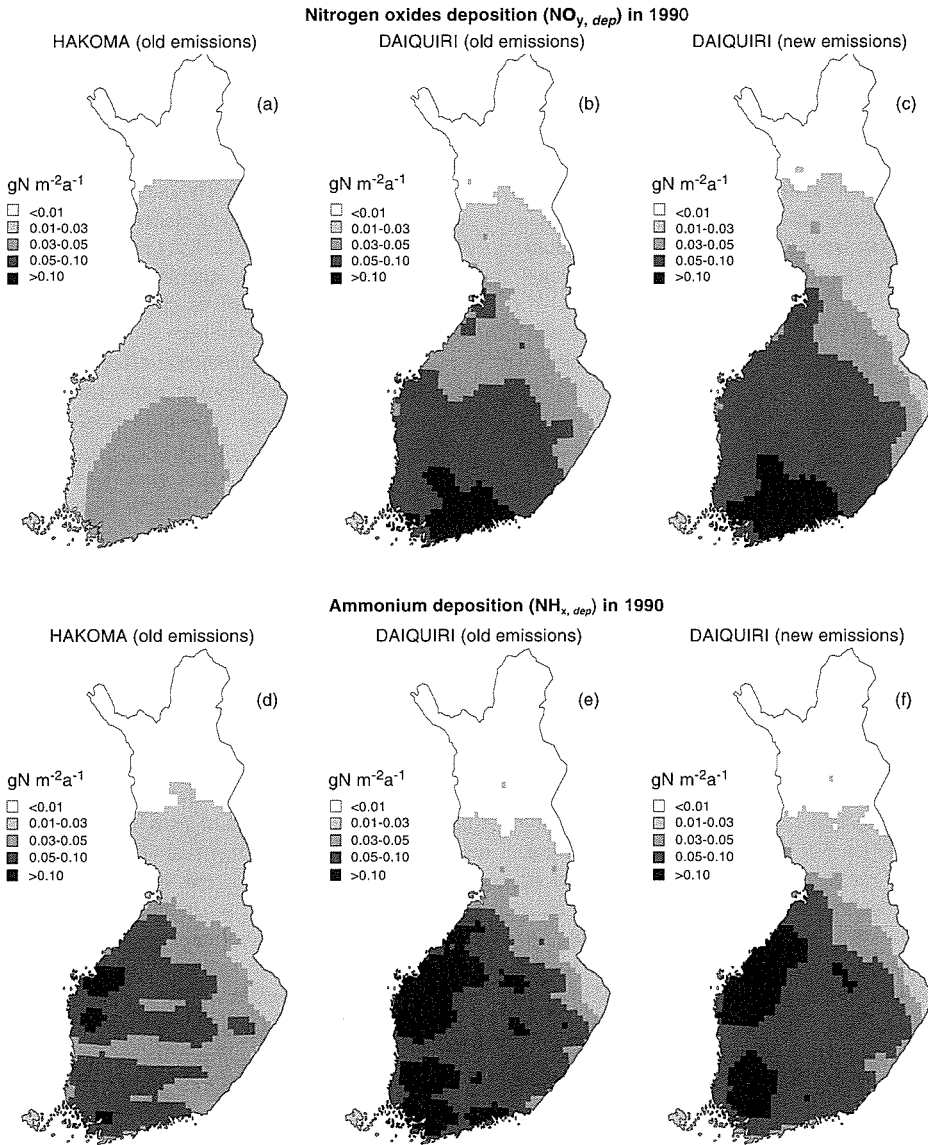
The DAIQUIRI model can provide time series of total sulfur and nitrogen depositions using the same historical data as the DEPUPT model. The spatial resolution can be chosen from the 150 km–150 km grid covering Europe, the national  $0.25^\circ \times 0.125^\circ$  grid covering Finland or any specific location in Europe. In the last case, the deposition value is interpolated from the chosen grid to a given specific coordinate representing, *e.g.* a small catchment. The model predicts total depositions, the value of which may differ from measured wet only, bulk or throughfall values. The local characteristics due to *e.g.* emissions, meteorological conditions, land cover *etc.* may additionally contribute to the differences.

When the modeled depositions are employed at the site level, for better representativity the modeled depositions should be calibrated to available measurements. Algorithms have been developed to estimate site-specific deposition and nutrient uptake input data for dynamic soil acidification models (Alveteg *et al.* 1998b), such as the DEPUPT model (VII).

## 4 Steady-state mass balance method

### 4.1 Critical loads

The definition for critical load is ‘the highest deposition of acidifying compounds that will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function’ (UBA 1996, Nilsson and Grennfelt 1988). The indicators and criteria are to be defined according to receptor, type of effect, acceptable changes *etc.* The critical loads concept has been originally developed and used as a policy tool by the Canadian Government in the early 1980s (EPRI 1991). In the late 1980s it has been discussed and evaluated in Europe within the Convention on Long-range Transboundary Air Pollu-



**Fig. 7.** Nitrogen oxides and ammonium deposition due to Finnish emissions using (a, d) the transfer matrices of the HAKOMA model and (b, c, e, f) the new matrices derived for the DAIQUIRI model for the year 1990 with old emission estimates of (a, b) 257 Gg  $\text{NO}_x$  and (d, e) 43 Gg  $\text{NH}_3$  and updated emissions estimates of (c) 274 Gg  $\text{NO}_x$  and (f) 38 Gg  $\text{NH}_3$ .

tion (CLRTAP) and sponsored by the Nordic Council of Ministers (Nilsson and Grennfelt 1988). The quantification has led to relatively simple environmental protection target values, which have been used for emission reduction negotiations under CLRTAP (Bad Harzburg 1989, Hettelingh *et al.* 1995a). Integrated models have

been used to optimize emission reductions to achieve desired or maximum protection of ecosystem and/or least overall emission control costs in Europe (Alcamo *et al.* 1990, ApSimon *et al.* 1994, Gough *et al.* 1995). The first protocol based on effects, or critical loads, was the Sulphur Protocol signed by 28 countries in Oslo in June 1994

(UN-ECE/SP2 1994). Current negotiations within CLRTAP and EU for combined reductions of emissions causing acidification, eutrophication and ground-level ozone are also supported by integrated assessment models (Amann *et al.* 1998a, 1998b). The incorporation of several pollutants (sulfur, nitrogen oxides, ammonia, volatile organic compounds) and effects has resulted in a very complex model and policy assessment, known as the multi-pollutant – multi-effect approach (Fig. 2).

Critical load maps for Europe have been calculated at the National Focal Center of each country or by the Coordination Center for Effects (CCE) if a national contribution was lacking. Updated and improved data have been compiled annually by the Coordination Center for Effects and reported in biannual status reports (Hettelingh *et al.* 1991, Downing *et al.* 1993, Posch *et al.* 1995, Posch *et al.* 1997a). The calculation methods and input data derivation have been harmonized through guidelines in the status reports and official manuals (UBA 1996).

Target loads based on other than scientific reasons may be derived from critical loads to be used as final or interim targets for emission reductions. The calculation of critical loads is based on scientific hypotheses and excludes subjective aspects as much as possible. It attempts to describe the inherent tolerance of an ecosystem against a pollutant load. However, some subjective decisions have been made on what ecosystems to protect and in the selection of appropriate threshold for risk. A safety margin can be introduced to the critical load based on *e.g.* uncertainty analyses. In a conservative approach the target load may thus become smaller than the original critical load. If some damage is acceptable, the target load may be higher than the original critical load. In practice, it may not be possible to reduce the emissions and consequently the depositions to the levels of critical loads. For example, some critical loads may be very near to background deposition values. Decisions have to be made to agree on an appropriate target load, possibly using not only scientific but also political, economical and social grounds. In addition, interim target loads may be set taking into account the potential response time of ecosystems before achieving the chemical conditions of the critical load in the ecosystems.

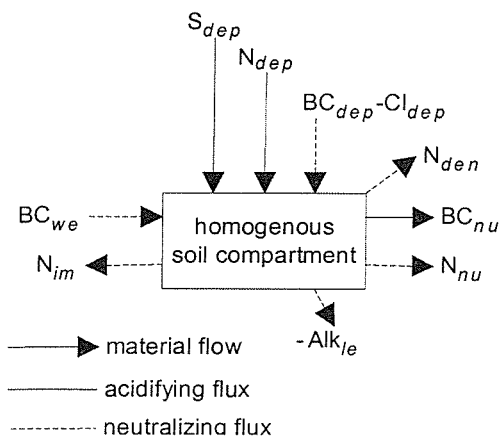
## 4.2 Effects criteria

The suggested method to calculate critical loads is the steady-state mass balance (SMB) method (UBA 1996). The method is presented in detail in Annex A. The major material fluxes considered are shown in Fig. 8. In steady-state, the acidifying inputs to the ecosystem are balanced by neutralizing sources and export of acidity from the system. The level of alkalinity leaching is based on a chemical criterion, which is chosen to avoid of harmful effects in ecosystems.

The criterion for detrimental effects on forest vitality is the critical molar ratio 1.0 of base cation concentration to aluminum concentration in soil solution,  $([Bc]_{molar} : [Al]_{molar})_{crit}$  of the tree rooting zone,  $(Bc : Al)_{crit}$  (Eq. A4 in Annex A). Aluminum is toxic to plants in high concentrations by inhibiting root growth and functioning and ultimately plant growth and performance. The primary toxic effects are considered to be competition with base cations for uptake sites in the apoplast and interference with the integrity of the plasma membrane. The base cations are assumed to provide protection for the fine roots and the vitality of the tree, if they are relatively abundant in the soil solution. The threshold value of 1.0 has been suggested by Ulrich (1983) and supporting evidence has been provided later from several studies mainly from laboratory bioassays (Meiwes *et al.* 1986, Sverdrup and Warfvinge 1993), though they may include uncertainties (Løkke *et al.* 1996, Jongmans *et al.* 1997). The criterion does not take into account the absolute levels of cation concentrations.

## 4.3 Mapping procedure

The critical loads for forest soils were mapped to represent the sensitivity of various forest site types and regions in Finland (Kämäri *et al.* 1992b). The critical loads were calculated for 1057 sites, where geochemical input data for weathering rate calculations were sampled. The 1057 sites covered the whole of Finland rather evenly with an average sampling density of one per 300 km<sup>2</sup>. Most of other major input data were available in the national 0.25° × 0.125° grid or they were constants. These data were allocated or interpolated to the 1057 calculation points.



**Fig. 8.** Material and acidity fluxes considered in the determination of the critical loads for forest soils:  $S_{dep}$ ,  $N_{dep}$ ,  $BC_{dep}$ ,  $Cl_{dep}$  denote deposition of sulfur, nitrogen, base cations and chloride,  $N_{nu}$  and  $BC_{nu}$  the net uptake by forest growth,  $BC_{we}$  the mineral weathering of base cations and  $-Alk_{le}$  the acidity leaching.

The regional maps should not be used for estimates at small sites *e.g.* via interpolation. The display of critical load data at province level may be too detailed (Savonen and Johansson 1996). Critical loads for individual sites must be calculated using data from or specifically estimated for that area.

#### 4.4 Derivation of input data

##### 4.4.1 Auxiliary data and constants

The derivation of Finnish input data for the calculation of critical loads for forest soils was one of the key tasks of this study (II, IV, VI, VIII). The countrywide mapping work required databases with sufficiently complete geographical coverage. The derived data described the input variables either directly (*e.g.* the measured and interpolated  $BC_{dep}$ ) or through well-defined transfer functions (*e.g.* net nutrient uptake calculated with ETS-dependent biomass growth and element concentration in biomass). Recommendations for input data derivation and treatment have been given in the mapping manual and other guidelines (UBA 1996, Posch *et al.* 1995). Most of the input data were derived on a national grid of  $52 \times 88$  cells, where each cell is  $0.125^\circ$  longitude  $\times$   $0.25^\circ$  latitude (approximately  $14 \text{ km} \times 14 \text{ km}$  in south-

ern Finland at  $60^\circ\text{N}$ ) and the lower left corner is at  $19^\circ\text{E}$ ,  $59.5^\circ\text{N}$ .

The main calculation parameters needed are presented in Annex A and Fig. 8. The critical load calculation points were located at sites where the mineral weathering was computed. Other data, if not constant, were allocated to individual calculation points from the national  $0.25^\circ \times 0.125^\circ$  grid with either taking the value of the cell where the point was located (net nutrient uptake, sulfur and nitrogen deposition as described in chapter 3) or using the inverse distance interpolation method with three closest measured values (base cation deposition). It was important to retain a balance between the significance of an input parameter to the results and efforts made to estimate it in spatial and quantitative detail. More sophisticated interpolation techniques currently in use, such as kriging (Suutari *et al.* 1999), were not available for this study.

The effective temperature sum (ETS) was needed in the computation of mineral weathering and net nutrient uptake. It denotes the accumulated heat during the year, obtained by subtracting the threshold temperature  $5^\circ\text{C}$  from the mean temperature of the month and summing up over the year. The ETS values for Finland have been calculated on the national  $0.25^\circ \times 0.125^\circ$  grid by the Finnish Forest Research Institute (Ojansuu and Henttonen 1983, Henttonen and Mäkelä 1988). The modeled values were interpolated to the grid using climatic data for the period 1951–1980 at weather stations maintained by the Finnish Meteorological Institute and taking into account latitude, height above sea level and distance to the sea. The ETS is a long-term average, but spatially it can retain uncertainty when the interpolation grid cell is far from weather stations or when local effects, *e.g.* topography or large inland water areas, affect the result (Henttonen 1991).

Data on the regional distribution of tree species were needed for the net nutrient uptake estimates. The fraction of forest soils on peat land was used to determine denitrification factors. The geographical distribution for tree species and forest site types were derived from the results of the 7<sup>th</sup> National Forest Inventory (NFI-7) conducted during 1977–1984. The data were provided by the Finnish Forest Research Institute. The forest inventory data were converted from the forest survey grid ( $8 \text{ km} \times 8 \text{ km}$  squares covering most of Finland, a different and sparser grid had been

used in northern Finland) to the national  $0.25^\circ \times 0.125^\circ$  grid (II, Johansson 1988). The procedure allocates the fraction of each square falling in a cell of the national  $0.25^\circ \times 0.125^\circ$  grid. The primary data describe for each cell a fraction of area dominated by each tree species, *i.e.* no mixed forests were assumed to exist. Primary forest inventory data on forest site types were categorized as well and poorly growing forests on both mineral and peat soils with forest site types 1–8. For practical use, the forest site type information was aggregated into three distinct groups of rich and poor mineral soils and peat soils (II).

Denitrification  $N_{den}$  was assumed proportional to the net incoming nitrogen. The fraction denitrified was related to the soil type by linearly interpolating between a low value of 0.1 for podzolic mineral soils and a value of 0.8 for peat soils (De Vries *et al.* 1993, Posch *et al.* 1997c).

The net precipitation values for earlier studies (I–III) were extracted from the RAINS soil model data for Europe (Alcamo *et al.* 1990). For the mapping of critical loads and site-specific dynamic model applications (IV, V, VII) runoff values for Finland were obtained from a digitized runoff map for 1961–1975 (Leppäjärvi 1987).

The gibbsite equilibrium constant was derived from values found in Nordic field studies (Sverdrup *et al.* 1990). The constant describes the lumped dynamics between aluminum and proton concentration interactions in soil. Most European soils do not contain gibbsite in detectable amounts, but operationally the aluminum concentration is estimated from an assumed equilibrium with a solid phase (Sverdrup *et al.* 1990). Thus the gibbsite model employed is not based on a generally dominant real mechanism and other substances such as organic acids may affect aluminum activity in soils. The use of gibbsite constant is one source of uncertainty in the model applications. The first values used were  $10^{8.5}$  and  $10^{8.7} \text{ mol}^{-2} \text{ l}^2$  adapted from the RAINS soil model and the SMART model defaults, respectively (I–III, Kauppi *et al.* 1986, De Vries *et al.* 1989). The average value  $10^{8.3} \text{ mol}^{-2} \text{ l}^2$  was later chosen for the calculations of Finnish forest soils based on Scandinavian values (Sverdrup *et al.* 1990, Kämäri *et al.* 1992b, IV, V, VI). In dynamic modeling the values may have been adjusted during the calibration procedure, *e.g.*  $10^{8.0} \text{ mol}^{-2} \text{ l}^2$  in VII.

The immobilization of nitrogen  $N_{im}$  was con-

sidered a long-term average constant value for Finnish forest soils (Rosén *et al.* 1992). In this study  $N_{im}$  included the fixation of nitrogen, which in general is rather negligible in Finland. The immobilization term should describe the situation of nitrogen saturation, where the availability of inorganic nitrogen is in excess of the total combined plant and microbial nutritional demand, and will be discharged in leaching (Grennfelt and Thörnclöf 1992). The currently measured nitrogen immobilization may be an overestimate, since the ecosystems may not yet be nitrogen saturated. Different ranges for  $N_{im}$  have been suggested: from 0 to  $3 \text{ kgN ha}^{-1} \text{ a}^{-1}$  where the higher value is for systems which are nitrogen limited or with C-N-ratio  $> 25$  (Grennfelt and Thörnclöf 1992), from 2 to  $5 \text{ kgN ha}^{-1} \text{ a}^{-1}$  for long-term natural net immobilization including fixation (Downing *et al.* 1993) and from 0.5 to  $1 \text{ kgN ha}^{-1} \text{ a}^{-1}$  (UBA 1996). This parameter was not employed in IV, V. The value in VI was  $2 \text{ kgN ha}^{-1} \text{ a}^{-1}$  (Downing *et al.* 1993) and in current mapping the value of  $1 \text{ kgN ha}^{-1} \text{ a}^{-1}$  is employed (UBA 1996).

#### 4.4.2 Weathering

The weathering of soil minerals releasing base cations ( $BC_{we}$ ) is the long-term inherent soil property that buffers acidification. Several methods can be used to estimate weathering rates, *e.g.*,

(i) A bedrock map can be used to derive a coarse classification, as was done in the early version of the RAINS model (Kauppi *et al.* 1986, Alcamo *et al.* 1990, UNESCO 1972).

(ii) Weathering rate estimates can be based on input-output budgets of elements, for example within a single drainage basin (Paces 1985, Starr *et al.* 1998). The difference between measured incoming base cation deposition and the sum of net uptake by vegetation and leaching can be assigned to the mineral weathering of base cations.

(iii) Complex weathering models have been introduced to describe actual weathering processes and responses to environmental conditions in more detail, including *e.g.* soil pH changes. The PROFILE model (Warfvinge and Sverdrup 1992) is rather data intensive and based on site mineralogy and other specific soil profile data such as soil texture.

(iv) An approach based on zirconium variations in the soil profile was introduced in Sweden (Olsson and Melkerud 1991, Olsson *et al.* 1993),



which is suitable for estimating historical weathering rates after deglaciation for granite bedrock areas, also in Finland.

The applicability of data intensive methods is restricted by the need to regionalize in the critical load mapping. More in-depth studies (*e.g.* Starr *et al.* 1998) carried out at specific sites provide new data for comparison, calibration and validation purposes, *i.e.* to find out the range of conditions where the results may be applied with acceptable accuracy. In the following, the three major phases of weathering rate estimation in Finland are described (VI).

The first simulations with the HAKOMA soil module (I–III) used the relative regional distribution of weathering rates taken from the soil module of the RAINS model (Kauppi *et al.* 1986). It was based on the dominant bedrock type described in the geological map of Europe (UNESCO 1972). In Finland, seven different bedrock types were displayed. Each grid cell of  $1.0^\circ$  longitude  $\times$   $0.5^\circ$  latitude was assigned to one of four predetermined weathering rate categories, for which the bedrock types had been classified. The absolute values were scaled with the help of Swedish ion budget studies in various catchments (Sverdrup and Warfvinge 1988) and the results of Finnish lake modeling (Kämäri 1988).

The zirconium method describing the historical weathering of base cations (Olsson and Melkerud 1991, Olsson *et al.* 1993, Melkerud *et al.* 1992) is based on studies of major and trace elements in several Swedish soil profiles. The amount of Zr in the resistant mineral zircon ( $\text{ZrSiO}_4$ ) is assumed to be uniform throughout all horizons of a soil profile since the last deglaciation. Zr concentrations in the upper layers have relatively increased with time due to losses of less resistant material due to weathering. The present Zr concentrations in different soil horizons have thus been used to calculate the absolute losses of soil material and other elements. The estimated depth of the weathered zone inferred from changes in Zr concentration varies from site to site. The average depth of weathering was 50 cm with a range from 20 cm to one meter (Olsson and Melkerud 1991). The element losses correlated well with the product of the effective temperature sum and the total concentration of the element in the coarse fraction ( $< 2.0$  mm) of the C-horizon. However, the extrapolation of the regression line outside, especially towards the low end

of the line, may result in uncertain weathering rates.

The second weathering rate estimate in Finland was based on computations using the zirconium method. The calculations were done at 1057 sites in Finland, where geochemical data from the fine fraction ( $< 0.063$  mm) were readily available on total element contents in till, the dominant Finnish forest soil type (Koljonen 1992). The third estimate was carried out for a restricted set of sites using the coarse fraction ( $< 2.0$  mm) concentrations, for which the method was originally derived. Thus a re-analysis was carried out in Geological Survey of Finland for the 618 sites from which samples were left in the store. The correlations for only Ca and Mg were applied in weathering computations. The values for Na and K were considered too uncertain to be used. The sodium concentrations were relatively low and the release of potassium caused by weathering of K-feldspar is still quite limited in young Finnish podzol soil profiles.

The spatial distribution of the concentrations was similar for the two different fractions. Calcium and magnesium tend to become enriched in the fine fraction of till and therefore the weathering rate estimates based on this fraction were an overestimation. The correlation between the two fractions was derived using data from whole country. This correlation was used to estimate the fine fraction content to sites where only measured coarse fraction was available. This may have resulted in uncertainties in Lapland, where no re-sampling or re-analyzing was possible and where pre-glacial regolith and older till layers are present.

Weathering rates were estimated for divalent base cations (Ca+Mg) for 1057 sites, and 618 sampling locations were used for the comparison between the fine and coarse fractions analyzed (VI, Tarvainen 1995). The weathering rate map conformed to the same general spatial pattern as the total concentration maps. The total divalent base cation weathering rate using the coarse fraction data, calculated separately for each element and then summed, was about 65% compared to the values obtained from using the fine fraction data. One of the most distinct areas, showing the largest decrease in the weathering rate, was in the extreme south-east of the country, coinciding with the rapakivi granite area, where the divalent weathering was already rather low. In the areas

that were submerged after last glaciation, the uppermost till layer of the soil profile may have altered, mainly due to leaching, and thus differ from the parent till in the C-horizon.

The data sheets for geochemical total concentrations did not include information on the top soil type or above-ground properties such as tree species or forest site type at the sampling sites. Although there is a connection between weathering and soil texture (Sverdrup 1990) there were no quantitative data available to scale the weathering rate obtained from the zirconium method for different soil types. More information may be available in the future from site-specific studies in Finland. The equations derived from the Swedish field studies have been compared to three other methods in the Integrated Monitoring catchment of Hietajärvi in Finland, where it gave values for Ca+Mg weathering between a low estimate from the PROFILE model and a high one from the zirconium method based on measured concentrations and an input-output budget (Starr *et al.* 1998).

#### 4.4.3 Nutrient uptake

The growth of vegetation and trees consumes base cation and nitrogen from available sources as total uptake ( $BC_{tu}$  and  $N_{tu}$ ). These nutrients can be taken up directly by canopy exchange from deposition, from mineralized litter or from the tree rooting zone originating from the mineral weathering or the cation exchange pool. The fluxes can be large and vary considerably inter- and intra-annually.

The term net nutrient uptake ( $BC_{nu}$ ,  $N_{nu}$ ) in this study denotes the uptake and accumulation of nutrients (Ca, Mg, K, N) by those tree compartments, which at some point in time will be removed from the ecosystem, *e.g.* via harvesting. This results in a permanent loss of nutrients. In areas or countries where whole-tree harvesting is practiced in managed forests, the nutrients in stem, branches and needles must be accounted for. In Finland the current harvesting practices generally utilize the tree trunk only (stem over bark). For non-managed areas, *e.g.* natural parks, no permanent nutrient losses are experienced. In the calculation of critical loads only managed forests were considered.

The annual average nutrient uptake by forest growth is calculated from the average annual vol-

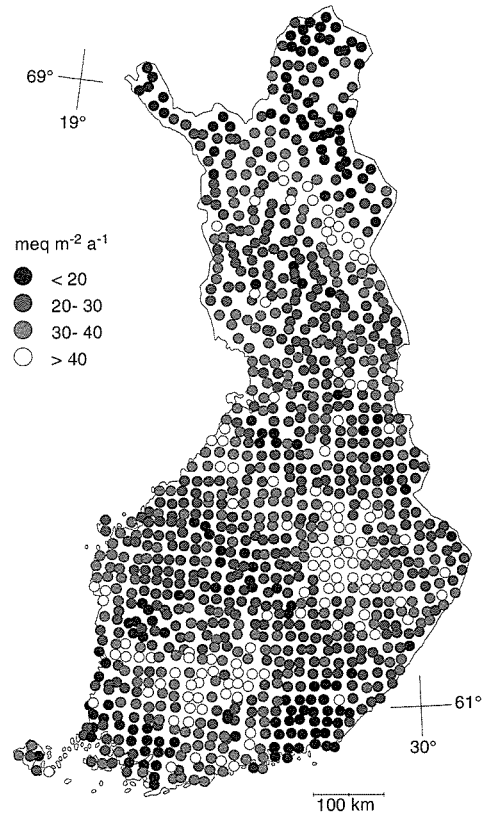


Fig. 9. The divalent base cation weathering of minerals,  $BC_{we} = (Ca+Mg)_{we}$ , calculated with the zirconium method equations (Olsson and Melkerud 1991, Olsson *et al.* 1993) and geochemical data from the coarse fraction (VI).

ume growth and the nutrient concentrations in the biomass. First, the average annual forest growth in Finland was estimated spatially for each tree species (deciduous, spruce, pine) separately. The calculation was based on the provision of tabular national forest inventory data and manipulation examples (Kuusela 1977). The total forest area and total annual volume growth for three tree species were given separately for the 21 forest management districts with their ETS values. The ETS describes the climatological effect on growth by integrating the length and intensity of the growing period, and in general it is sufficient to characterize forest growth in Finland. In each district and for each tree species separately the total annual growth was divided by the forest area. The resulting data were plotted as function of district-specific ETS values. A logistic function was fitted

separately for all tree species with a non-linear regression method (Press *et al.* 1986). Only the value in Ahvenanmaa (Åland) was treated separately. The functions derived this way enabled the average annual growth estimate for each tree species for any location in Finland provided that the ETS for the point was known. The use of these results retain uncertainty due to *e.g.* old inventory data, changes in forest age structure, other factors such as the amount of stones in soil affecting local growth conditions, differences in forest densities for tree species and exclusion of site fertility, though this is partly reflected by tree species. Second, the biomass densities of each tree species (Mälkönen 1975, Rosén 1982) were used to convert average annual volume growth to mass increments. Third, the annual average nutrient increment in biomass was calculated using the data on nutrient contents in biomass. These data were based on field measurements (Mälkönen 1975, Olsson *et al.* 1993, Rosén 1982) and provided concentration for each tree species and tree compartment. Some of these data were dependent on the latitude. For critical loads, only nutrient uptake by stem over bark was considered, reflecting the most common harvesting practice in Finland. For total nutrient uptake, required by *e.g.* the DEPUPT model computing dynamic uptake by above-ground growth, data for concentrations in branches and needles were additionally used.

#### 4.4.4 Base cation deposition

Base cation deposition can neutralize acidifying deposition. Alkaline emitters include both natural sources, *e.g.* soil dust, and anthropogenic activities, *e.g.* fly ash from energy production. The characterization of the sources and atmospheric transport has proven to be quite difficult. There have been only few studies on base cation deposition effects, since base cations were not considered to be primary tracers for environmental problems. Therefore, current deposition estimates rely heavily on measurements.

In the acidification studies, the effective net flux of the base cation deposition is of interest. Part of the elements originate from the sea. The marine base cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{K}^+$ ,  $\text{Na}^+$ ) are accompanied and compensated by marine anions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ). They are assumed to have no net effect when passing through the soil. The fraction of marine components in measurements can be

calculated by a so-called seasalt correction using a trace element. Usually Na is used, which generally has no other sources. The sea-salt corrected base cation deposition is denoted with an asterisk,  $Bc_{dep}^* = \text{Ca}_{dep}^* + \text{Mg}_{dep}^* + \text{K}_{dep}^*$ .

First-regional estimates of sea-salt corrected base cation deposition were made on the basis of monthly bulk precipitation measurements conducted during 1972–1981 at 38 measuring stations over Finland (Järvinen 1986, I). The dense network with good spatial coverage over Finland and long time series supported the use of these data in comparison with the relatively few stations of the FMI. The values were assigned to represent the year 1977 and were kept constant onwards. The effect of forest filtering was included to describe the increased deposition to forests and forest soils compared to open land (II, Kauppi *et al.* 1986).

Currently the base cation deposition used in the critical load assessment (V, Posch *et al.* 1997a) is calculated from the bulk precipitation measuring network of FEI using the average of three most recent data years. Currently the years 1991–1993 are still used since the general deposition level has not changed significantly. The network consists of several stations (39 stations were in operation during 1991–1993) where monthly bulk samples are analyzed for total content of base cations and other components. First, a three-year average for each compound is calculated to each station using monthly values. The station values are interpolated to the national  $0.25^\circ \times 0.125^\circ$  grid using an inverse distance weighing from the three nearest stations. The interpolation is made separately for each compound and after the sea-salt correction with Na as tracer. The map of  $Bc_{dep}^* - \text{Cl}_{dep}^*$  is obtained by summing the sea-salt corrected base cations together and subtracting sea-salt corrected chloride.

Although the measurements of base cation deposition were quite reliable as such, there were some methodological difficulties on their contribution to critical load calculations. First, earlier some assumptions were made on the physiologically active fraction of base cation deposition (II, III). Second, part of the measured base cation deposition may come from other than external sources to the ecosystem. The internal element cycling of a forest may affect bulk collectors and measurement results do not represent the external input only (Anttila 1990, II, III). Therefore, in II

the consistent proportion (30%) of wet to bulk deposition measurements of FMI and Järvinen (1990), respectively, was used to scale the spatial deposition map derived from the denser network of Järvinen (1990). Third, the base cation deposition reaching the soil in the forest should enter critical load calculation. When assessing the net flux of both wet and filtered dry fractions to forest soil under the canopy, the separation of the effect of internal cycling from the measurements is difficult. The availability of both wet (or bulk) and throughfall measurements will facilitate the estimates on canopy leaching. A quantitative method to estimate wet and dry deposition and canopy exchange from bulk and throughfall measurements has been suggested by Alveteg *et al.* (1998b) and applied in the DEPUPT model (VIII). Fourth, stations with clear anthropogenic influence of nearby agricultural and particle (*e.g.* Estonian oil-shale burning) sources were in some cases excluded to obtain an effective, long-term steady-state deposition (Kämäri *et al.* 1991). Fifth, the FEI bulk precipitation measuring network is regionally comprehensive, especially when compared to the stations operated under various programs with the FMI. However, the long sampling period of one month may have introduced larger uncertainties than short sampling periods of *e.g.* EMEP stations. Other monitoring results in Finland (*e.g.* Leinonen and Junnto 1991) have suggested lower base cation deposition levels.

Since the future levels of base cation deposition were very difficult to estimate, the values entering critical load calculations were based on latest averaged measurements available. However, levels can become much lower. In dynamic model applications, some additional assumptions were introduced (VII). The non-marine component of base cations was assumed to be partly connected to anthropogenic activities reflected by measurement data during a period of considerable emission reductions (Hedin *et al.* 1994). Large sulfur emitters have been substantial particle emitters, for which the first reduction techniques normally are particle-removing electrical filters. Therefore, the temporal changes of base cation deposition were assumed to partly follow the historical variation of sulfur deposition.

## 5 Dynamic modeling techniques

### 5.1 Dynamic soil acidification models

The temporal development of soil acidification can be assessed with dynamic acidification models, which have been widely applied to specific sites and regionally (Jenkins *et al.* 1990, De Vries 1994, Sverdrup *et al.* 1995, NAPAP 1991). Model applications demonstrate the inherent sensitivity of different soils and their chemical responses to anthropogenic stresses and environmental variables, such as the acidifying deposition, forest harvesting and temperature changes.

The requirements for input data are higher for dynamic models than for the steady-state approaches, such as critical load calculations. The level of process descriptions and input data should be compatible with the purpose and the temporal and spatial resolution of the application. Input variables such as nutrient uptake by forest growth can often be modeled more accurately at a specific site where measurement data are more readily available than at a regional level. A dynamic model applied on a regional grid can at best describe the average dynamics within a grid cell.

The models should be calibrated to observations to obtain more reliable values of output variables. In practice, there are often less measured variables than calculation parameters with well-defined values. In addition, time series of observation are generally not available for long-term calibration and validation. Therefore, the uncertainties in absolute results may increase when moving further away from the calibration point, although relative changes may retain a higher accuracy. Validation efforts of dynamic acidification models have been carried out both regionally and at specific sites (*e.g.* Cosby *et al.* 1989, Posch *et al.* 1989a). This study does not report direct validation activities but comments on other studies using the same models.

In this study, both regional and site-specific applications of dynamic forest soil acidification models are presented. The first one is the modification and regional application of the RAINS soil model for Finland (I–III). The second one is a site-specific application of the SMART model in a system of several models providing input to the SMART model (VII). Compared to the steady-state methods dynamic modeling gives information on the changes in the significance of different

calculation parameters, the point of time when distinct chemical processes dominate, the responses and delays to changing acidifying fluxes, the duration and degree of recovery from acidification and the timing and amount of measures required to prevent an unwanted degree of acidification.

## 5.2 Regional dynamic modeling of soil acidification

### 5.2.1 The RAINS soil model

The soil model in the early version of the RAINS model (Alcamo *et al.* 1990) was used in a regional dynamic model application in Finland (I–III). The model source code was modified and integrated into the HAKOMA model structure.

The RAINS soil model is based on the buffer range concept introduced by Ulrich (1983). The main factors considered are the  $H^+$  deposition derived from the atmospheric transport of sulfur emissions, base cation deposition, the carbonate and base cation contents in soil, mineral weathering of base cations, dissolution and precipitation of aluminum and nutrient uptake tree growth. The model assumes a depletion of the base cation pool by leaching with mobile sulfate ions. Several of the guidelines in De Vries *et al.* (1989) were applied (II, III), since for applications at a regional scale there is a need to minimize input data requirements and complexity.

Most soils with a base saturation above 5% buffer primarily by cation exchange (Ulrich 1983), which is the case for Finland in general (Tamminen and Starr 1990). Base saturation ( $BS$ ) describes the ratio of base cations in the exchange pool in soil ( $BC_{exch}$ ) to the total cation exchange capacity of soil,  $CEC$ , which roughly equals  $(BC+Al+H)_{exch}$ . Soils shift to the aluminum buffer range at low base saturation values. After the depletion of exchangeable base cations, aluminum is mainly responsible for acid neutralization in soil solution. This was assumed to have an effect on the vitality of forests and was thus used as an indicator of an increasing risk for harmful effects.

### 5.2.2 Regional input data derivation

The RAINS soil model was run for each cell of the national  $0.25^\circ \times 0.125^\circ$  grid over Finland (I–

III). For each cell, input data were derived for most important input variables: sulfur and base cation deposition, mineral weathering and base cation uptake.

Only sulfur deposition was used as an acidifying input in the RAINS soil model. It was assumed that forests gathered more sulfur deposition than open areas (Kauppi *et al.* 1986). The effectiveness of the forest to increase dry deposition was described with the filtering factor,  $\phi$ . The factor value  $\phi = 2$  was used in the calculations.

Base cation deposition was interpolated from a countrywide measuring network (Järvinen 1990) and scaled to represent the external input to forest ecosystems (see section 4.4.4).

The spatial distribution of base cation weathering was adapted from the RAINS soil model estimates for the whole of Europe in a grid of  $1.0^\circ$  longitude  $\times$   $0.5^\circ$  latitude, approximately  $56 \text{ km} \times 56 \text{ km}$  in southern Finland at  $60^\circ\text{N}$  (see section 4.4.2). The distribution was also checked with the bedrock map (UNESCO 1972). The absolute values were adjusted with the results from budget studies and model simulations (Sverdrup and Warfvinge 1988, Kämäri 1988).

Net nutrient uptake by forest growth leads to losses of base cations in soil. These amounts were estimated using Swedish uptake estimates (Rosén 1988), biomass density and the nutrient contents in biomass in Finland (Mälkönen 1975) and annual average forest growth. Forest growth was estimated for three groups (poor and rich mineral soils, peat soils) of forest site types (Vuokila 1987, Kuusela 1977). Literature data for uptake values were used and the regional variability of uptake was considered latitude-dependent (III). Therefore this approach was simpler than the method later used for deriving input for steady-state critical loads and dynamic modeling (see section 4.4.3).

The amount of exchangeable base cations in soils was estimated using results from various field studies, which enabled the derivation of relationships between forest site types and soil base cation content (II). The neutralizing store in forest soil is the base cation pool, or the base cation exchange capacity,  $BC_{exch}$ . It denotes the amount of cations attached to the exchange sites in the soil matrix. The pool can be depleted or recharged over time, giving a dynamic buffering property to the soil.

The base cation exchange capacity for differ-

ent forest soils were examined with two sets of survey data provided by the Finnish Forest Research Institute (Tamminen and Starr 1990). Statistical parameters were sought on  $BC_{exch}$  and the total cation exchange capacity,  $CEC$ , for each forest site type group (poor and rich mineral soils, peat soils). The first data set comprised of 27 samples from an intensive study with measurements of  $BC_{exch}$  and  $CEC$  for the whole soil profile. The data were then aggregated for the top 50 cm soil layer. The sampling points covered southern and central Finland. The second data set consisted of values from an extensive survey of 1246 plots. The data were pre-calculated for humus and 30 cm mineral top soil. The plots were mainly from southern Finland. The results from the two data sets on  $BC_{exch}$  were compared to each other and against forest site types. The distribution of  $BC_{exch}$  values for each forest site type was relatively wide and there were overlaps between forest site types. The results, however, suggested that a forest site type satisfactorily described the base cation content in soil. Therefore,  $BC_{exch}$  was correlated to the forest site type, for which the spatial distribution for Finland was extractable from the data of the 7<sup>th</sup> National Forest Inventory (II, Johansson *et al.* 1990).

An aggregation to three major groups of forest soils with  $BC_{exch}$  and  $CEC$  values was made for the simulations: rich and poor mineral soils and peat soils (II). The base cation values for peat lands was derived from the results of another survey (Westman 1981). The values were assumed to describe the forest soil status in the year 1987, when the intensive forest soil survey was mainly carried out.

### 5.3 The SMART model

#### 5.3.1 Model structure

The SMART model (De Vries *et al.* 1989, Posch *et al.* 1993c) was developed to improve the relatively simple RAINS soil module. The purpose was to estimate long-term chemical changes in soil and soil water in response to changes in atmospheric deposition. The model is specifically designed for applications on a regional scale. The outputs include soil base saturation and the concentrations of the major anions and cations in soil solution and runoff water. Apart from the net up-

take of nitrogen and base cations in harvested plants and the net nitrogen immobilization in the forest floor, the influence of the nutrient cycle (foliar exudation, foliar uptake, litterfall, mineralization and root uptake) is not taken into account. A lake water module has been developed for the SMART model to enable the calculation of ion concentrations in lake water and to include in-lake processes (Kämäri *et al.* 1995).

The SMART model is calibrated to observations by adjusting relevant input data or model parameters within estimated uncertainty ranges. The variables adjusted may affect flow rates or stores. First the inputs of monovalent ions ( $Cl^-$ ,  $Na^+$ ,  $K^+$ ) and sulfur are determined so that the modeled output are in accordance with the measurements. Second, the nitrogen fluxes are matched by adapting nitrogen deposition and uptake values. Then, variables affecting soil base saturation and divalent base cation concentration are adjusted, including the base cation deposition, the weathering rate, uptake, ion exchange constants and partial pressure of  $CO_2$  in soil. Lastly, the pH and the surface water alkalinity are fitted by the partial pressure of  $CO_2$  in water or possibly readjusting some earlier quantified parameters.

The SMART model has been used regionally over Europe in studies to estimate possible temporal responses of soil acidification (*e.g.* De Vries *et al.* 1994b, Hettelingh and Posch 1994) and in site-specific applications in several catchments over Europe, comparing it with two other soil acidification models (Forsius *et al.* 1998). In Finland the model has been applied on several sites (*e.g.* Kämäri *et al.* 1995, Forsius *et al.* 1998, Johansson 1997).

#### 5.3.2 Finnish applications

To study the effects of deposition, climate change and forest harvesting at the Rudbäcken catchment in southern Finland (VII) some changes were made in the original SMART model. All mass balances and the full charge balance were considered, although originally monovalent base cations were assumed to be balanced by chloride. Since the stream discharge of organic nitrogen was significant at the Rudbäcken catchment, an empirical equation was added to describe the leaching of organic nitrogen. Furthermore, an exponential temperature effect on the immobilization rate  $N_{im}$  was introduced in order to allow an assessment of

nitrogen leaching due to climate change scenarios.

The DEPUPT model was originally developed to provide consistent input for comparing three soil acidification models at Integrated Monitoring sites in Europe (Forsius *et al.* 1998). This model simulates the nutrient uptake due to forest growth and includes a simple description of the main nutrient circulation in the forest (foliar exudation, foliar uptake, litterfall, mineralizable material). The DEPUPT model was applied in (VII) to provide dynamic deposition and uptake input data for the SMART model.

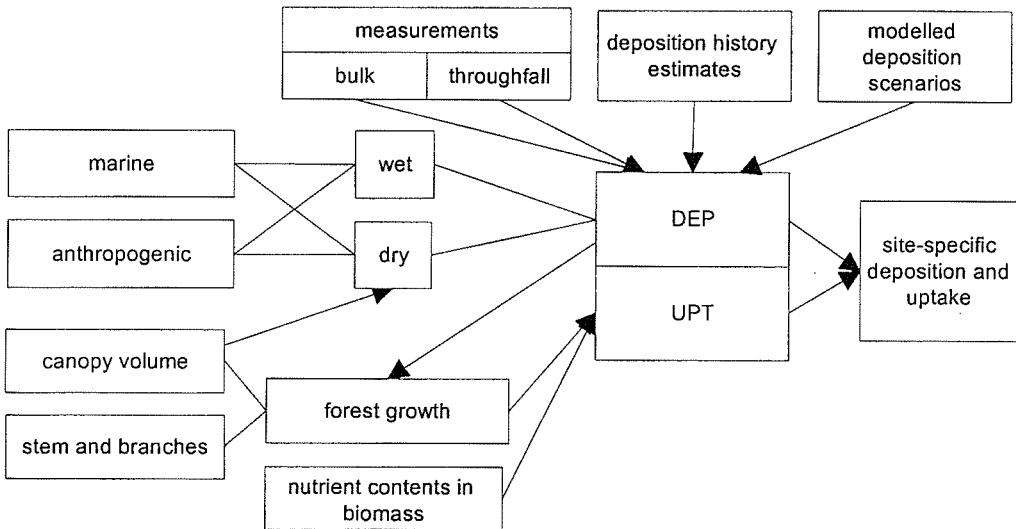
**5.3.3 Site-specific input data derivation**

Site-specific dynamic modeling usually requires temporally detailed input data. If measurements are not available, data must be extracted from regional databases or expert opinion. Part of the input data required in dynamic model applications are common to those used in the steady-state critical load calculations. In addition, values determining soil characteristics are required. A list of input data for the SMART model can be found in Posch *et al.* (1993c).

The derivation of input data for the SMART model application to the Rudbäcken catchment in southern Finland (VII) is presented in the follow-

ing. Physical and chemical soil variables, such as soil depth,  $BC_{exch}$  and  $CEC$ , were based on measurements and lumped into averages representing the whole Rudbäcken catchment. The weathering rate was estimated from the regional mapping (VI) and was thus considered rather uncertain for this site.

The temporal development of depositions and nutrient uptakes were derived with the DEPUPT model, which was calibrated to relevant measurements from the site. The method in the DEPUPT model has evolved from the MAKEDEP model (Alveteg 1998, Alveteg *et al.* 1998a, 1998b), which was developed to provide input data for the SAFE model (Warfvinge and Sverdrup 1992). The method was modified and applied for simulations of several sites of the UN-ECE Integrated Monitoring Program (Johansson 1995a, Johansson *et al.* 1996, Forsius *et al.* 1996, Forsius *et al.* 1997) and other studies (VII, Johansson 1997). The nutrient uptake by forest growth is calculated for the simulation period using current forest properties and management history. Deposition and growth are estimated simultaneously, because it is assumed that canopy volume affects the amount of dry deposition and that the growth is in some cases limited by available nitrogen from deposition. The calculation scheme of the DEPUPT model is presented in Fig. 10.



**Fig. 10.** The DEPUPT model calculation scheme for the derivation of deposition histories and scenarios as well as dynamic uptake due to forest growth.

In the deposition module of the DEPUPT model (VII), measured bulk and throughfall depositions are used to determine the anthropogenic and marine fraction and wet and dry component for each acidifying and neutralizing compound. The total dry deposition component for sulfur, chloride and sodium was assumed to be the difference between throughfall and bulk deposition. For other base cations (Ca, Mg and K) the sodium filtering factor was used. The filtering factor was defined as the ratio of throughfall to bulk deposition. Nitrogen deposition was assumed to filter according to the smallest filtering factor from sulfur, sodium or chloride. The wet marine components were estimated with sea-salt correction factors using sodium as a tracer. The deposition not allocated to marine or anthropogenic origin was assigned to canopy exchange.

The historical and future developments of depositions were assigned separately to the disaggregated components. The wet marine deposition component was assumed to remain constant over time. Anthropogenic deposition components depended on deposition histories, calibrated to presently measured values. Sulfur deposition during 1880–1991 was available for each 150 km × 150 km grid cell (Mylona 1993, 1996) and back to 1790 from Sverdrup *et al.* (1995). Average nitrogen histories for the whole of Europe were based on the SAFE model and MAKEDEP model applications (Alveteg *et al.* 1998b, Sverdrup *et al.* 1995, Asman *et al.* 1988). The non-marine deposition component of base cations was assumed to be partly connected to anthropogenic activities reflected by measurement data during a period of considerable emission reductions (Hedin *et al.* 1994), and was here assumed to follow the historical sulfur deposition curve. All dry deposition components were affected by forest filtering, which was assumed to depend linearly on the modeled tree canopy volume.

In the forest growth and uptake module of the DEPUPT model (VII) the uptake due to forest growth was based on the biomass density, element content and the annual increment of each tree compartment (stem over bark, branches, needles). Potential annual growth and standing volume were calculated from a logistic curve (Alveteg *et al.* 1998b) but calibrated to the observed actual volume and annual growth of the forest at the site concerned. If nitrogen from the atmosphere and mineralization was not enough to

satisfy the modeled potential annual growth, the mass increment was reduced equally for all above-ground compartments.

An additional feature was introduced to the DEPUPT model in to describe the temperature-dependent forest yield (VII). The climate change effects on temperature were calculated with the climate generator (Carter *et al.* 1995). The annual potential forest growth at the site was assumed to depend on the effective temperature sum (ETS). The relative changes of the mean annual temperature predicted by the climate generator were assumed to implicate equal relative changes for the average ETS value. Thus the potential forest growth value was modified annually according to relative variations of the temperature.

## 6 Applications of integrated models

### 6.1 Critical loads and their exceedances

#### 6.1.1 Maps of critical loads and their exceedances

The mapping of critical loads for forest soils and surface waters in Finland was first reported in 1990 for the acidifying sulfur (Henriksen *et al.* 1990, Kauppi *et al.* 1990a, Kenttämies 1991). Next, critical loads were calculated for acidifying sulfur and nitrogen depositions. Later, updates of critical loads for both acidification and eutrophication included forest soils and surface waters (Kämäri *et al.* 1992b) and provisional data on critical levels for ozone exposure were presented (Posch *et al.* 1997a). The maps have been used for scientific (*e.g.* IV, V, VI), policy-supporting (*e.g.* Kämäri *et al.* 1992b, Acidification Committee 1998) and popular dissemination (*e.g.* Savonen and Johansson 1996) purposes. The most recent update of Finnish data have been included in the European database used in negotiations on further emission reductions (Amann *et al.* 1998a, 1998b).

The critical load data may be presented in a map form in many different ways, depending on the specific purpose. The display format of the results must be chosen carefully to ensure correct interpretation and transparency of the data (Posch *et al.* 1993b). For the scientific community it is desirable to retain as much information as possible in the display of the results, whereas clarity



and simplicity of presentation are crucial for the understanding and acceptance by decision-makers.

First, a proper grid scale for the map is chosen. Then, values of all calculation points in a grid cell are ordered into a cumulative distribution function (CDF), from which the desired statistical descriptor can be easily computed. For critical loads the 5<sup>th</sup> percentile of the CDF is routinely used for grid map displays. It represents the most sensitive fraction of ecosystems and eliminates potential outliers at the low end of the CDF. The exceedances of critical loads are accordingly displayed in the grid maps as the 95<sup>th</sup> percentile of the CDF of point exceedances. Generally, the critical load point values are weighed with the respective ecosystem areas.

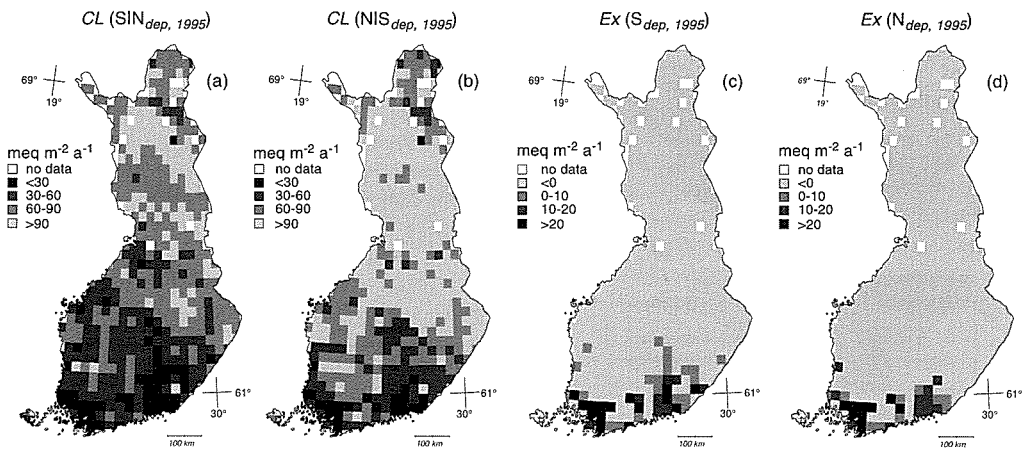
The critical loads of acidifying deposition can be separately presented for sulfur and nitrogen, which both contribute to acidifying deposition. Since the ecosystem does not distinguish between acidity due to sulfur or nitrogen, the critical load for one compound can be obtained by first fixing the other one. The standard procedure is to use current depositions to fix critical loads of one compound. The notation  $CL(SIN_{dep,1995})$  denotes the conditional critical load for sulfur assuming the 1995 nitrogen deposition,  $N_{dep,1995}$ . The allocation method is explained in more detail in Annex A.

Depositions for the integrated assessment

modeling for critical loads were calculated with the DAIQUIRI model. The exceedance, or the excess deposition over critical loads, was calculated by subtracting the critical load from the respective deposition, e.g.  $Ex(S_{dep,1995}) = S_{dep,1995} - CL(SIN_{dep,1995})$ . The maps of critical loads of acidity allocated for sulfur and nitrogen and their exceedances in Finland using modeled deposition of 1995 are shown in Fig. 11. The map in Fig. 11a can roughly be compared with the first preliminary critical load map for forest soils (Kauppi *et al.* 1990a). It reveals a shift in sensitivity from north to the south of the country, probably due to decreased base cation deposition. However, the overall sensitivity level is the same.

An alternative way to express the exceedances of critical loads of acidifying deposition was introduced in IV. The effect from the excess was given as the resulting molar base cations to aluminum ratio in soil solution, ( $Bc:Al$ ), in addition to the usual excess deposition value in acidifying equivalents. This indicator gave supplementary information on the degree of risk by indicating areas where the ratio differs one order of magnitude from the original protection target.

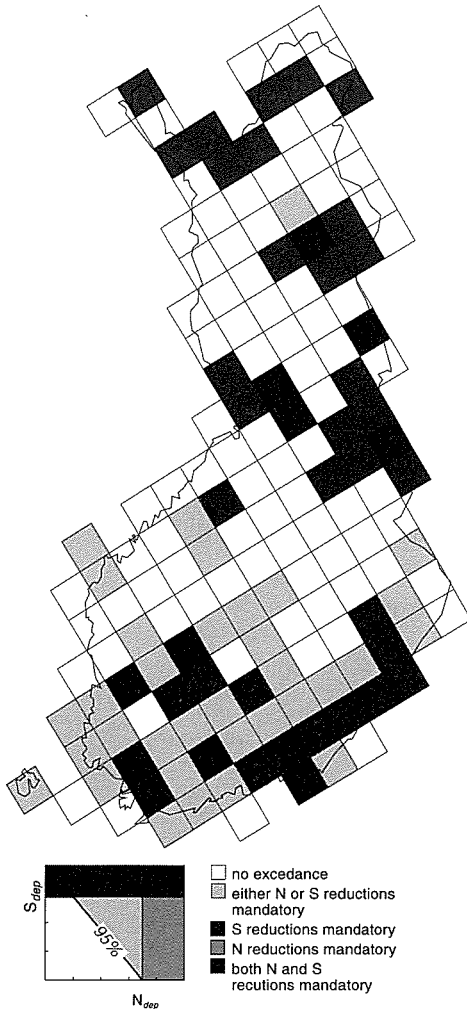
Due to the non-uniqueness of separate critical loads of sulfur and nitrogen, as explained in Annex A and shown in Fig. A1, an infinite number of possible combinations exist to reduce excess sulfur and nitrogen deposition. The reduction needs can be defined with the method explained



**Fig. 11.** Conditional critical loads of acidifying deposition for forest soils allocated to (a) sulfur ( $CL(SIN_{dep,1995})$ ) with fixed nitrogen deposition and (b) nitrogen ( $CL(NIS_{dep,1995})$ ) with fixed sulfur deposition, and the respective exceedances of (c) sulfur ( $Ex(S_{dep,1995})$ ) and (d) nitrogen ( $Ex(N_{dep,1995})$ ), using modeled sulfur and total nitrogen depositions of 1995 from the DAIQUIRI model.

in Annex A and shown in Fig. A2. Five cases exist with different reduction requirements to achieve non-exceedance of the critical loads in the grid cell as shown in Fig. 12 (Posch *et al.* 1997a). Critical loads for both forest soils and lakes and modeled deposition in 1995 in a 50 km × 50 km grid were used in the calculations. The results indicate that sulfur is still an important acidifying compound and in many areas further reductions of sulfur deposition are mandatory.

Preliminary maps of critical loads for acidity



**Fig. 12.** Five cases of reduction requirements of exceedances of critical load to achieve protection of 95% of Finnish lakes and forest soils against acidification in each 50 km × 50 km grid cell at present (1995) sulfur and nitrogen depositions (Posch *et al.* 1997a).

were used as environmental protection targets for finding cost-effective ways to reduce sulfur emissions in Finland and areas nearby (Johansson *et al.* 1991). Emission reduction costs for Finland and areas nearby depended both on the fraction of the critical load of acidification allocated to sulfur and the general reduction level in other European countries. The results suggested that the attainment of critical loads require considerable reductions outside Finland and the most stringent loads were very difficult to achieve with sulfur reductions only.

The temporal development of the exceedances of critical loads for sulfur  $Ex(S_{dep})$  during 1960–2010 is shown in Fig. 13. Deposition data for 1960–1985 were from the HAKOMA model (III, HAKOMA 1992) and for 1990–1995 from the DAIQUIRI model (VIII). These deposition values were used to calculate the development of  $CL(SIN_{dep})$  (NMI 1995, Acidification Committee 1998). The two emission scenarios were calculated separately (Amann *et al.* 1998a). In 1980 more than one fourth of forest soil critical loads in Finland were exceeded. The exceedances decreased about 74% till 1995, the fastest development taking place in the 1990s. This was mostly due to the declining domestic sulfur emissions but also a result of reduced long-range transport. The relative importance on imported pollution in Finland remained at over 80% for sulfur and nitrogen oxides and for ammonia about two thirds, although a slightly decreasing trend was noticeable for nitrogen compounds (EMEP/MSW-W 1998). Trends of exceedances of critical loads of sulfur in Europe have been presented by Berge (1997) and  $Ex(S_{dep})$  has reduced to less than half during 1980–1994. The values in Fig. 13 were also compared with the results of dynamic acidification models describing the forest soil area under a chosen indicator for harmful effects, such as a low base saturation,  $BS$ , or molar  $Bc-Al$ -ratio,  $(Bc:Al)$ , in soil solution. The exploratory results for Finland (II) and Europe (De Vries *et al.* 1994b) indicated a prompt response only to very large deposition reductions.

There are several advantages in the use of critical load maps as environmental protection targets and basis for emission reduction strategies. The method considers and quantifies the most important variables affecting the acidifying and neutralizing fluxes, making the calculation simple and transparent. The data for these param-

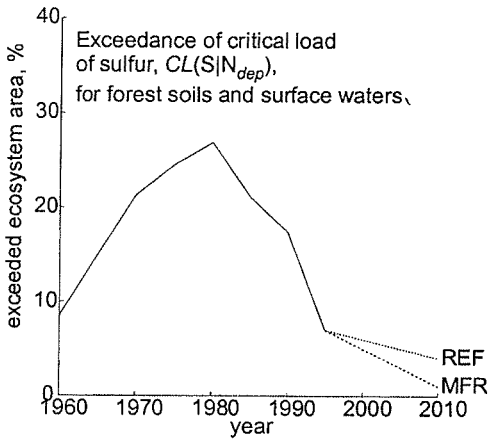


Fig. 13. The temporal development of exceedances of critical loads of sulfur for forest soils and lakes in Finland in 1960–2010 with two future deposition scenarios (REF and MFR).

eters are generally available. The calculation methods are harmonized for all European countries and they are documented. The relatively simple concept of critical loads is conveyable to decision-makers. The steady-state values also supply sound targets for optimization calculations.

There are also disadvantages in the use of critical loads. The critical load method inevitably simplifies ecosystem processes. Relevant, consistent and accurate input data must be available from large regions for successful mapping. In spite of harmonized methods countries easily stick to their research traditions and idiosyncracies. There are differences in methods for monitoring, derivation and interpretations of input data. In addition, there are several ways to technically implement the steady-state mass balance (SMB) method. In spite of the difficulties, harmonizing the mapping of critical loads in Europe has been both successful and useful. The concept of exceedances of critical load by total acidifying deposition being allocated to sulfur and nitrogen reduction requirements with certain assumptions may not be transparent to policy-makers. The comparison of critical loads, which are long-term steady-state targets, to observed harmful effects is difficult, since dynamics is ignored and several other exogenous factors influence ecosystem functioning. The timing of deposition reductions cannot be determined. In the optimization calculations, the critical loads cannot be achieved in practice even with

maximum technically feasible emission control measures, which leads to compromises in formulating interim targets and performance indices.

A large research program on external costs of energy production (ExternE 1995) has employed available dose-response relationships, including critical loads, in evaluating the potential monetary losses through harmful environmental effects. One of the relationships is based on the mathematical correlation between total exceedances of critical loads and defoliation severity classes at country level in Europe (Kuylenstierna and Chadwick 1994). This is coupled with results from Sweden (Söderberg 1991) relating defoliation to growth decrements observed in forest inventories. In spite of the good statistical relationships the uncertainties are rather large and do not necessarily support a causal relationship between exceedances of critical loads and growth decrement. The applicability of these relationships to Finnish conditions were evaluated and suggested to be treated as indicative only (Pingoud *et al.* 1999).

Attempts have been made to complement steady-state critical load studies with dynamic model simulations. The SMB method has been compared with dynamic model equations both theoretically and through examples at specific sites providing insights into the two approaches (Posch *et al.* 1997b, Posch and De Vries in press). The responses of European soils to emission reduction measures have been studied (De Vries *et al.* 1994b) and the timing of the reduction measures at some Integrated Monitoring sites has been assessed (Forsius *et al.* 1997). The lack of satisfactory input data, difficulties in interpreting some dynamic calculation variables and relatively complex model management hamper to date the operative use of regional dynamic modeling in emission reduction studies. In spite of the uncertainties of the critical load approach it still performs favorably over other methodologies as a basis for evaluating effects-oriented cost-optimal emission reduction requirements.

### 6.1.2 Uncertainty analyses

Uncertainty analysis is a useful method to estimate the most important factors of a model, whether the model structure itself, the described processes or parameters and their variability. The consequences of the uncertainties in the model in-

puts are quantified for the model outputs. There are two fundamentally different types of uncertainty. First, the stochastic variability due to the nature of the phenomenon (*e.g.* in a measured value) and second, the incomplete knowledge about the items concerned (*e.g.* the formulation of a mathematical model and its representativity to the underlying processes). Several uncertainty analyses of acidification models have concentrated on applications at specific sites (*e.g.* Beck 1987, Kros *et al.* 1993). Analyses on the regional variability have been done on both dynamic and steady-state approaches (De Vries 1994, Posch *et al.* 1997c, Barkman 1998), though they have mostly dealt with water quality (*e.g.* Hettelingh *et al.* 1992, Posch *et al.* 1993a, Kämäri *et al.* 1993). Some analyses have focused on the uncertainty and robustness of the atmospheric part of the acid rain problem (Alcamo and Bartnicki 1987, Batterman and Amann 1991, Watanabe and Ellis 1993). The regional variation of the important parameters are often hard to evaluate intuitively, however, methods to overcome this problem have been suggested (Hettelingh 1990, Hettelingh *et al.* 1992).

The uncertainty analysis of critical loads for forest soils was carried out in Finland (V). The central part of this uncertainty analysis was the assessment of the effect of individual input parameters for the model and associated uncertainties, that is, the first above-mentioned fundamental type of uncertainty. Depending of the examined parameter, the uncertainty was assumed to arise from poor measurement quality (*e.g.* total potassium content in biomass), erroneous interpretation of the observed phenomenon (*e.g.* base cation bulk deposition) or lack of knowledge (*e.g.* site-specific forest filtering factors). Other potential sources of uncertainty in modeling were the chemical criteria and the model structure. The value of the criterion for harmful effects, the molar Bc-Al-ratio 1.0 in soil solution, was taken as given and not included. The model structure was also assumed to sufficiently describe the observed system and was not considered uncertain in this analysis.

First, an uncertainty analysis was carried out for the equations used in critical load calculations. Each input parameter was given a range from its minimum and maximum values found in the country. The purpose was to find potential extremes of output values. Then, an uncertainty

analysis was carried out using the standard critical load mapping data for forest soils. Each input parameter at each calculation point was given a *a priori* uncertainty range.

The distributions of parameter errors or uncertainties should preferably be derived from observations. However, for many of the parameters results from measurements producing such data were not available. The often used assumption of normally distributed errors was in general difficult to justify due to lack of supporting data. Another often used assumption of a uniform probability distribution within a range seemed counterintuitive from data that were available. Therefore, symmetric triangular distributions were chosen to portray the distribution of parameter value errors. Other types of ranges, such as a uniform probability distribution, were not considered. The uncertainty in the parameter values was expressed in terms of the coefficient of variation. The min-max range for the assumed symmetric triangular distribution can then be derived from it.

The quantification of correlations of some input parameters, *e.g.* the co-deposition of sulfate and ammonium, was not possible due to the lack of sufficient data. In general, when the input parameter values are assumed independent from each other, the resulting uncertainty distribution produces a more pessimistic result than in the case of correlations.

The input parameters included in the analysis were given individual uncertainty ranges from literature values or with best estimates. The uncertainty of the weathering rate was separately calculated by varying the effective temperature sum (ETS) and total soil contents of calcium and magnesium with their estimated uncertainties. Similar calculation was done for the nutrient uptake affected by variations in the ETS, biomass density and nutrient contents in the biomass. Uncertainties were directly assigned to depositions of sulfur, nitrogen oxides, ammonia and base cations, filtering factor for sulfur deposition, runoff and the area fraction of each tree species.

A Monte Carlo approach was chosen for the analysis, since a complete assessment of the uncertainty (evaluating *e.g.* distributions) was not readily achieved by analytical manipulations of the SMB method. The software package UNCSAM (UNCertainty analysis by Monte Carlo SAMpling techniques) developed at the National Institute of Public Health and the Envi-

ronment (RIVM) in the Netherlands has been designed for the uncertainty analysis of complex models (Janssen *et al.* 1992, Heuberger and Janssen 1994).

Conventional statistical descriptors such as the standard deviation, percentiles and cumulative distributions, were calculated to display the uncertainties of the model outputs, here the critical loads and their exceedances. The contribution of the input parameters to the uncertainty of the model results were assessed by linear regression and correlation analysis of the model outputs on the corresponding input parameters. If the linear regression was appropriate, *i.e.* if the  $R^2$  of regression (here denoted as the coefficient of determination, COD) is near to 1, then the standardized regression coefficients (SRC) measure the fraction of the uncertainty in the model output which is contributed by the various input parameters (Heuberger and Janssen 1994). The SRC is especially suited for uncorrelated input parameters, which was also the assumption used in this analysis.

First, an uncertainty analysis was performed at one virtual plot using the range of possible input values that were found in Finland as the uncertainty range of the specific parameter. This gave the output range from all possible combinations of available calculation parameters. Some combinations characterized real sites in Finland, when their parameter values were allowed to vary within a ten-percent range. The COD was high for all output variables. Therefore, the SRC could be used to find out the most important parameters.

The relative ranking according to SRC of some input parameters is shown in Table 2. The most important value explained on the average 40–50% of the resulting uncertainty, the second about 20%, the third 10% and the rest under ten percent. The three most important variables were the sulfur deposition, the weathering rate and the base cation deposition.

Next, the uncertainty analysis was carried out regionally over the whole of Finland using individually determined uncertainty range for each calculation variable considered. The results for the regional analysis are presented in Fig. 14. It gives the cumulative distribution functions both in the 150 km × 150 km grid and aggregated for the whole country. The uncertainty ranges are expressed as the mean with standard deviation and as median with a 90% confidence interval. The critical load CDF had an uncertainty, expressed with standard deviation, of about ±30%. The critical loads did not differ much spatially, but the standard deviation was smaller in the north than in the south. The country aggregated CDF showed a lower limit on the left due to the assumption concerning the minimum base cation leaching (see Eq. A2 in Annex A). The exceedance uncertainties were highest in the south, where the acidifying deposition was also highest. Even within one grid cell the uncertainty varied significantly.

Some statistics of the most important parameters for critical load and exceedance calculations were compiled, based on the ranking of the three most important parameters at each site. The most

**Table 2.** The relative ranking of some input parameters according to SRC (V).  $BC_{nu}$  and  $N_{nu}$  stand for the net uptake of the most important tree species.

	$CL(S+N)$	$Ex(S+N)$	$CL(S)$	$Ex(S)$	$CL(N)$	$Ex(N)$
$BC_{we}$	1	2	1	2	3	2
$BC_{dep}^*$	2	3	2	3	5	3
$BC_{nu}$	3	5	5	5	6	4
$N_{nu}$	4	7	7	7	7	7
$S_{dep}$	6	1	3	1	1	1
$NO_{y,dep}$	5	4	4	4	4	5
$NH_{x,dep}$	7	6	6	6	2	6

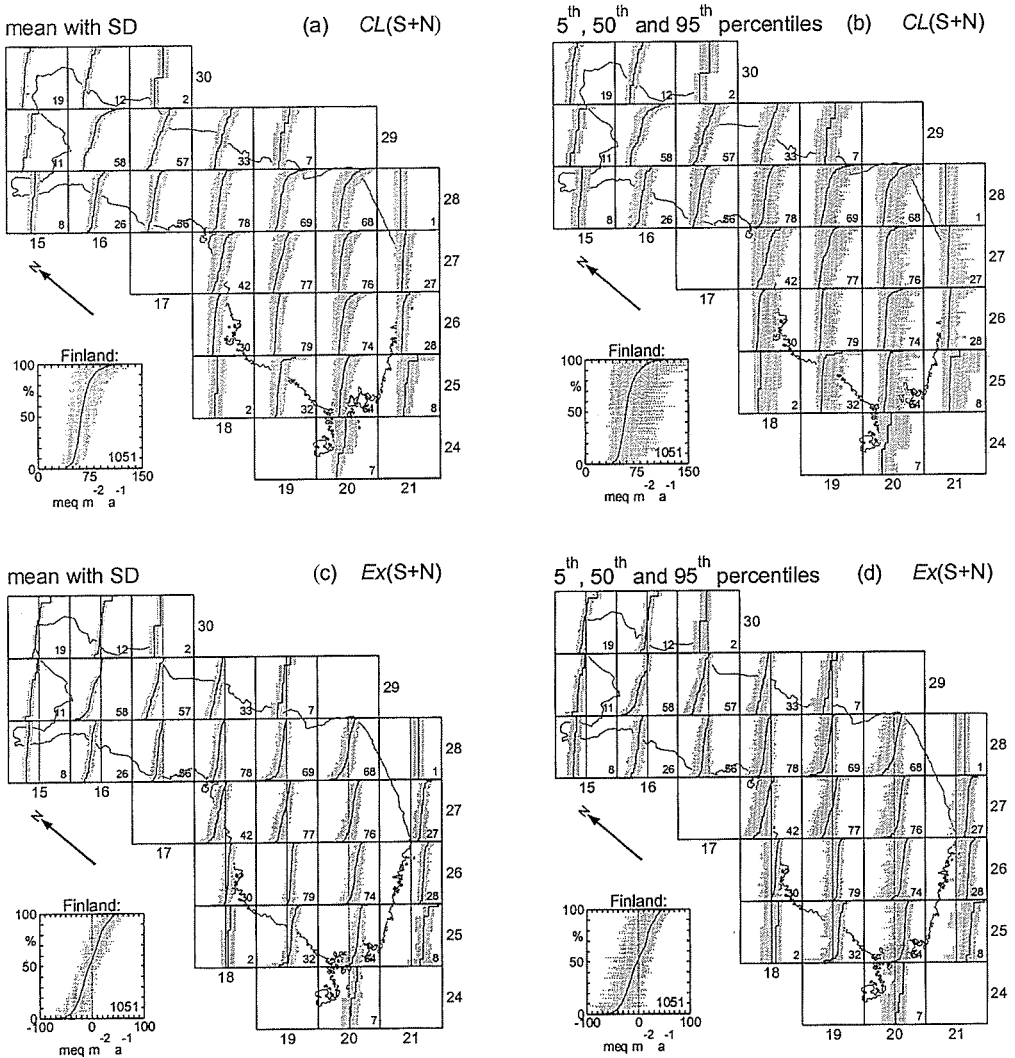


Fig. 14. The uncertainty bands for the cumulative distribution functions of critical loads of sulfur and nitrogen (a, b) and their exceedances (c, d). The uncertainties are presented both by mean with standard deviation (a, c) and by the 5<sup>th</sup>, 50<sup>th</sup> and 95<sup>th</sup> percentiles (b, d) with grey shaded bands. The results are displayed as cumulative distribution functions in each of the grid cells covering Finland and also for the whole country in the lower left corner of the map. The number of calculation points in the lower right corner of each grid cell.

influential parameter in the whole country was base cation deposition, which was higher in the data years 1986–88 than currently. The next ones were the base cation and nitrogen net uptakes by deciduous trees. The growth rate of deciduous trees was high and the nutrient contents in the biomass are relatively high, therefore, even a small area of deciduous trees may affect the re-

sults. The fourth most important parameter was the weathering rate. The critical loads and their exceedances for nitrogen were also influenced by the filtering factor of deciduous forests.

The spatial distribution pattern of the most influential factors was shortly examined. For the critical load of sulfur and nitrogen the most important factor in some southern parts of the country was nitro-

gen net uptake, in most of the southern and central Finland base cation uptake, and in Lapland base cation deposition along with the weathering rate.

None of the input parameters were assigned a correlation. This has led to a possible overestimation of the uncertainties. For example, the uncertainty of the net uptake values probably represented an upper limit, since the input parameters were varied independently of each other, although they are highly correlated.

The importance of the critical chemical criterion for harmful effects, the critical molar Bc:Al-ratio 1.0, was not included in this uncertainty analysis. Although the applicability of the value 1.0 to actual forest growth has not been thoroughly confirmed for Finnish conditions (Nissinen and Ilvesniemi 1990), the constant value provided a systematic approach and comparable results with international mappings. The involvement of other criteria, *e.g.* a critical limit for the aluminum concentration, would have shown more sensitivity of the effects criterion to other input parameters like runoff. The effect of the criterion has been shown to be of importance only when the base cation leaching,  $BC_{le}$  (see Eq. A2 in Annex A) is very small (Posch *et al.* 1997a). The analysis of the most influential parameters for critical loads in general may also divert the attention from the decisive ecosystems determining the regional critical loads, namely the most sensitive 5<sup>th</sup> percentile of the calculation points. The interest should thus be rather focused on these low critical load values only.

The results of the regional uncertainty analysis on the steady-state mass balance model (V) suggested that the resulting uncertainties of the critical loads were not considerably higher than the uncertainties in the input parameters. The fraction of ecosystems with certain exceedance or non-exceedance was relatively small at national level, when the certainty was defined within mean  $\pm$  standard deviation, but clearly larger in individual 150 km  $\times$  150 km grid cells. The outcome implied that due to the variability of ecosystem properties and deposition loading it would be unreasonable to use single targets for large regions or the whole country.

The effect of different weathering rate estimates on critical loads were shortly examined (VI). This parameter is very important in the SMB method (Eqs. A1 and A2 in Annex A). The weathering rate estimates have developed from qualita-

tive to quantitative techniques, or from bedrock acidity index to use of climatological and geochemical data. The differences due to the change of techniques resulted more in spatial than absolute level differences. The inclusion of new soil geochemical data from the coarse fraction instead of the fine fraction changed the absolute levels more than the spatial distribution. The differences affected more the critical loads of sulfur, since critical loads of nitrogen are more independent of the weathering rate. Very low weathering rates, and consequently critical loads, were more uncertain than higher ones. This was due to the regression equation of the zirconium method derived from Swedish field data, which did not cover areas of very low total soil concentrations and cold climate, or low effective temperature sums.

Uncertainty analyses have been carried out on the Swedish forest soil data (Barkman *et al.* 1995, Barkman 1998). The results suggest that the variation due to input data uncertainty is rather independent of the number of calculation points in the cell, the shape of the CDF remains the same and the variation is within tens of percents.

The Coordination Center for Effects has carried out a regional uncertainty analysis using the critical load database compiled from data from National Focal Centers (Downing *et al.* 1993, Posch *et al.* 1997a). The results show that in most cases the main base cation fluxes play a major role and  $BC_{we}$  is most important of them. In areas where net base cation input  $BC_{le}$  is higher than 10 meq m<sup>-2</sup> a<sup>-1</sup>  $Alk_{le}$  is not significant in explaining the uncertainty of  $CL_{max}(S)$  and  $CL_{max}(N)$ .

### 6.1.3 Applicability of critical loads

The chemical criterion used for forest soils, the critical molar Bc:Al-ratio 1.0 in the soil solution of the tree rooting zone,  $(Bc:Al)_{crit}$ , has been derived from laboratory studies. Some studies have reported only the Ca:Al-ratio. A compilation of these results has been made by Sverdrup and Warfvinge (1993) and the reference to Finnish conditions can be derived from laboratory experiments with acidification effects on seedlings by Nissinen and Ilvesniemi (1990). The use of this single criterion for critical load mapping has been criticized by Løkke *et al.* (1996), however, no better indicators or new interpretations suitable for regional scale applications have been suggested.

The critical load defines the long-term steady-

state pollutant load, which would not lead to harmful effects. For forest soils, the requirement for long-term steady-state conditions leads to assessment periods longer than the normal forest rotation length, which is decades to a couple of hundred years depending on the region and tree species. Therefore, the dynamics of the nutrient uptake via forest growth was not included in critical load calculations, but as an annual average only. In addition, the formulation of SMB with the criterion of critical molar Bc:Al-ratio 1.0 implies that the neutralizing capacity of the exchangeable base cation pool in the soil has been depleted to a very low base saturation of about 5% (De Vries 1994, Posch *et al.* 1997b). Therefore, the critical load values are not directly comparable with currently observed harmful effects.

A comparison of three types of models has been carried out using data from the Solling spruce site (De Vries *et al.* 1995). The critical loads of sulfur obtained with the dynamic and integrated models were close to the steady-state model calculations with the SMB method. The result lends confidence to the simplified critical load mapping carried out in Finland and on the European scale.

In southern Norway a statistical correlation has been found between spatial patterns of defoliation and the exceedance of critical loads after excluding potential effects of natural factors (Nelleman and Frogner 1994). The correlations between coniferous forest defoliation and anthropogenic stress factors have been explored using ten years of forest monitoring data at the European level (Müller-Edzards *et al.* 1997), but no clear relationships have been found between observed crown thinning and acid deposition or critical loads. No clear relationships have been found between depositions or critical loads of sulfur and nitrogen and the average defoliation corrected for the stand age in Finland (Müller-Edzards *et al.* 1997). Uncertainties in these studies include the steady-state assumptions in critical loads, therefore, areas exceeding critical loads do not have to correlate with currently observed effects. An exceedance rather indicates a risk for harmful effects in the long term. Depending on the pollutant load history of a site, the period before harmful effects become apparent may be from years to decades as suggested by results of dynamic model applications (Kämäri *et al.* 1995).

A conservative way to determine critical loads

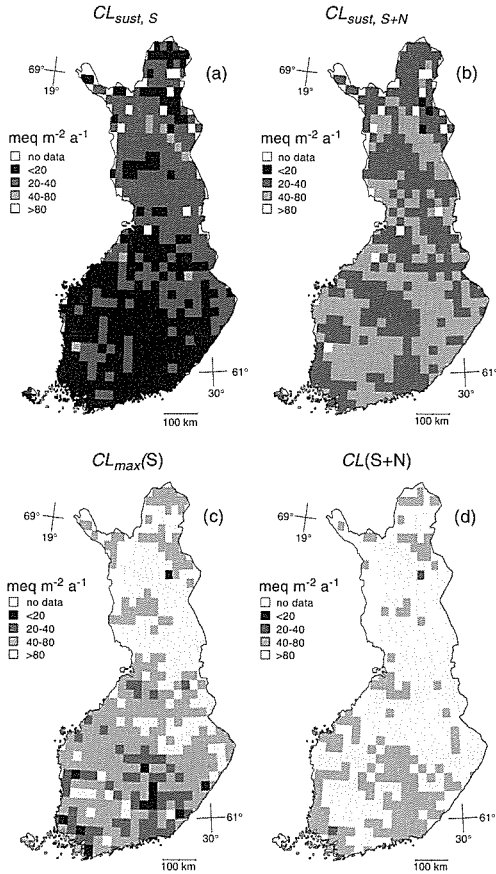
was considered by completely leaving out the alkalinity leaching  $Alk_{le,crit}$  in the SMB method. Consequently, the criterion of critical molar Bc:Al-ratio 1.0 in soil solution,  $(Bc:Al)_{crit}$ , is not used. The approach implies that no change compared to the current situation is allowed. First, only base cation fluxes are considered. The alkalinity leaching in Eq. A10 is zero and  $CL_{max}(S) = BC_{le}$ . The right hand side of Eq. A2 in Annex A will then determine the maximum sustainable sulfur load  $CL_{sust,S} = Bc_{dep}^* - Cl_{dep}^* + BC_{we} - BC_{nu} = BC_{le}$ , which is required to be non-negative, and shown in Fig. 15a. This map can be compared with Fig. 15c, which displays the maximum critical load of sulfur,  $CL_{max}(S) = BC_{le} - Alk_{le,crit}$  (Eq. A10 in Annex A). The values of  $CL_{sust,S}$  are about one third or half of  $CL_{max}(S)$ . Actually the approach of  $CL_{sust,S}$  was originally used to calculate the first preliminary critical loads (Kauppi *et al.* 1990a). Only a few input variables were needed for this formulation and it was possible to allocate more efforts in refining these major fluxes. Comparison of Fig. 15a to the map in Kauppi *et al.* (1990a) suggests the same overall sensitivity level, however, currently the northern parts of the country appear more sensitive, possibly due to different weathering rate estimates. Next, the nitrogen fluxes were included. The alkalinity leaching was set zero in Eq. A7 in Annex A describing  $CL(S+N)$  and thus  $CL_{sust,S+N} = N_{nu} + N_{im} + N_{den} + BC_{le}$ , where  $N_{den}$  was calculated using  $N_{dep,1995}$ . This sustainable critical load is shown in Fig. 15b. For comparison, the critical load for acidifying sulfur and nitrogen was calculated with Eq. A7 and mapped in Fig. 15d. The comparison of Fig. 15b to 15d illustrates that sustainable critical loads of sulfur and nitrogen were about one half of the critical loads including alkalinity leaching. The estimation on sustainable critical loads indicated that environmental protection targets could be well below currently calculated critical loads if no alkalinity leaching were allowed and subsequently the depletion of base cation pool were not accepted.

## 6.2 Regional dynamic modeling

### 6.2.1 RAINS soil model simulations

The RAINS soil model was applied regionally to evaluate potential acidification of forest soils un-





**Fig. 15.** Sustainable critical loads for forest soils excluding the alkalinity leaching and allowing no change in soil chemical status. The first one (a)  $CL_{sust,S}$  included only base cation fluxes and the second one (b)  $CL_{sust,S+N}$  additionally incorporated nitrogen fluxes. For comparison, (c) maximum critical load of sulfur  $CL_{max}(S)$  and (d) critical load for sulfur and nitrogen  $CL(S+N)$  are presented. The 5<sup>th</sup> percentile of each grid cell is shown.

der various deposition scenarios in Finland (I–III). The analysis was carried out with the HAKOMA model comprising the main aspects of an integrated assessment for effects of acidification (III). The purpose was to give indications on relative changes in the output variables, pH and soil base saturation, due to variations of the driving force, *i.e.* the sulfur deposition.

The simulations were initialized for the year 1987, when the soil input data had been sampled. The time step used was one year and the simulation was run to 2040. The initialization of the model served as an implicit calibration of the model to

measurements. Different sulfur deposition scenarios applied were based on three energy use scenarios for Finland (reference, natural gas, energy conservation) and three alternatives to install sulfur emission controls (no controls, mandated plans by the Finnish government, maximum feasible technical reductions) (Savolainen and Tähtinen 1990). For the rest of Europe four different emission abatement strategies were assumed: a decrease of 30% by 1993 from the levels of 1980 (I) and no controls, current reduction plans or maximum feasible technical reductions (II, III).

For the simulation results the soil base saturation, *BS*, was chosen as a primary indicator on the acidity status of forest soils. The base saturation value 5% corresponds to a soil solution pH value of about 4.2, an acidity value implying an increased risk of forest damage (Kauppi *et al.* 1986). First, the final soil status was studied (II). In the year 2040 most affected areas appeared near significant sulfur emission sources or were influenced by the long-range transboundary deposition. Other clearly affected areas had low weathering rates, which was the most important parameter determining the spatial sensitivity pattern. The relative effects of extreme deposition scenarios (from no controls to maximum reductions) were noticeable. The comparison of the early dynamic soil simulation results (I) to maps of critical loads for sulfur and associated exceedances (Fig. 11) indicate similar sensitivity patterns in general. This was expected, since the calculation methods and input data are partly same.

Second, the characteristics of the temporal development were examined (II, III). The potential effects were described with the percentage of forest soil area in Finland falling under a base saturation of 5%. Five different combinations of Finnish and European sulfur emission scenarios were compared. The results are shown in Fig. 16. The outcome indicated that 57% of the soils would belong to the risk category in 2040 without emission controls and 12% in the case of applying the maximum feasible technical reductions in all countries. The worst case, where no reduction measures are installed, showed continuously increasing soil acidification, although the rate slightly slowed down after 2020. For other scenarios the rate was either increasing or stabilizing within the time span studied. The effectiveness of emission reductions in and outside Finland was clearly noted, but Finnish reductions alone had only a limited effect.

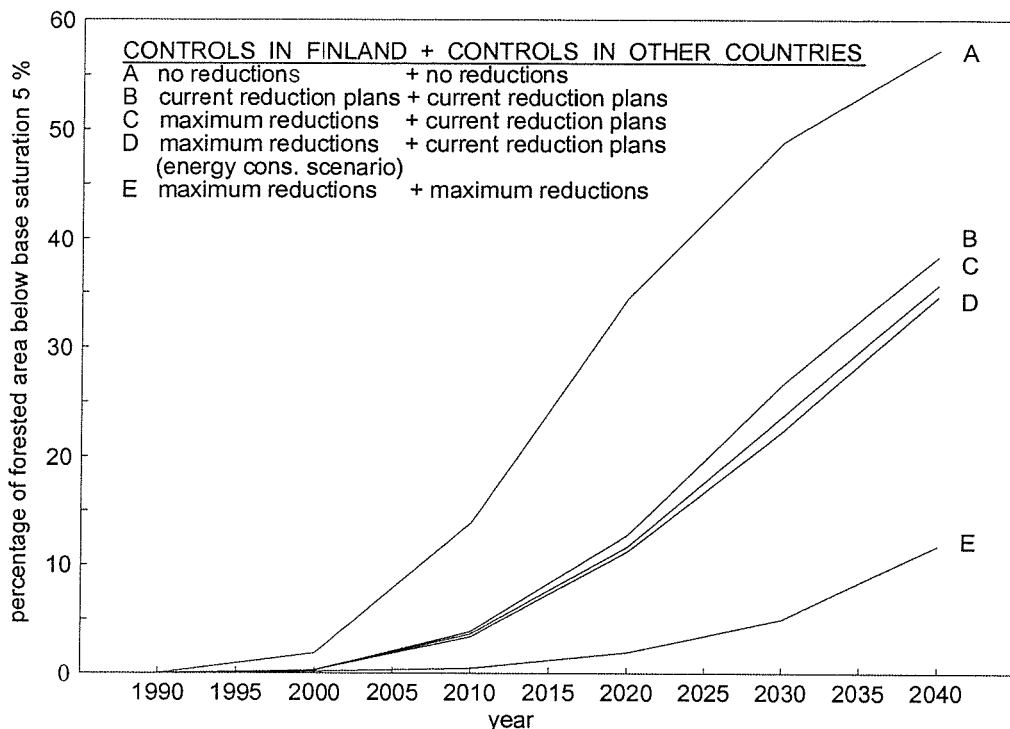


Fig. 16. Estimated temporal development for the percentage of the total forested area in Finland below the soil base saturation 5%. Five combinations of emission scenarios for Finland and the other European countries were considered.

### 6.2.2 Analyses on uncertainty and applicability

A number of test simulations were performed to clarify the importance of different input parameters in the regional application of the RAINS soil model (II). The factors considered in the sensitivity analysis were forest filtering effect, base cation deposition, weathering rate, net removal of base cations due to harvesting and a more detailed classification of the exchangeable base cation content in soils. The regional simulations were based on the chemical soil status in 1987 at the time of a large-scale soil sampling. This was considered as a joint initialization and calibration procedure. The results were not, however, compared to other observations due to lack of sufficient data and the relatively large spatial uncertainties involved. Therefore the emphasis was put on the evaluation of relative differences of various future scenarios rather than the absolute values of the results. Thus the results were compared against the reference case in 2040 corresponding

to the current emission reduction plans.

The filtering factor  $\phi$  for sulfur was changed to 1.6 (Ivens 1988) instead of the default 2.0 for coniferous forests (Kauppi *et al.* 1986), later 1.8 (Alcamo *et al.* 1990). The lower filtering factor resulted in still continuing but slower acidification of forest soils. The effect was marginal compared to the uncertainty of other parameters.

The base cation deposition was originally relatively high in southern Finland and thus helped to neutralize the high acidifying loads. Reducing the base cation deposition to half resulted in some local acidified areas in Lapland. The southwestern part of the country became strongly acidified. Doubling cation deposition lead to slower acidification than in the reference case, with mostly local effects left. The latter possibility was much more improbable in practice than the first one due to the observed decreasing trend in base cation deposition.

The weathering rate was one of the most difficult parameters to quantify and validate. Using half of the original weathering rate values re-

sulted in a rapid deterioration of soils in most of Finland. Doubling the rate caused only very few and restricted areas to lose the cation buffering capacity compared to the reference values, and large areas even showed increased soil base saturation.

The classification of soil base cation exchange capacity was refined by considering five classes instead of three. This was done by splitting each mineral soil class into two and choosing corresponding average calculation values from the soil survey data. The use of five classes produced a larger area of soils below base saturation value of 5% in the south compared to results with three classes. The refinement of soil classes may improve the overall uncertainty of the simulations. However, the three groups seemed to describe well enough the overall trend (Johansson 1987, II).

In the case where no net removal of base cations through harvesting would occur, the model predicted that only a small fraction of forest soils would acidify considerably. The positive effect was most evident in the southern parts of the country. This situation would depict the conditions of *e.g.* nature conservation areas where no harvesting is allowed. In the early simulations the effect of nitrogen was not considered, which would have mostly canceled out the positive effect of no base cation removal.

The relative importance of the different factors is shown in Table 3. The modeling results seemed to be sensitive to the quantification of many parameters. The most influential parameter was the weathering rate followed by the base cation deposition. The analysis was performed in an early stage of regional dynamic modeling in Finland and it covered only the few most important parameters affecting forest soil acidification.

An uncertainty analysis was performed for the RAINS soil model on the European scale (Posch *et al.* 1985, 1989b). The results stressed the importance of knowing the chemical status of the soil at the time of model initialization. Uncertainties prevail in the standard procedure for dynamic soil acidification model calibration, when the soil solution is assumed to have been in equilibrium with deposition in pre-acidification times (1800–1900) and the following historical depositions, uptakes and other parameters have to be estimated (Cosby *et al.* 1985, Sverdrup *et al.* 1995). The simulations may also start already from an acidified unnequilibrium situation (I, II, De Vries *et al.* 1994b). The starting time of model simulations depends on the numerical solution methods of the model and model characteristics (Posch 1998).

In regional dynamic modeling some of the input data which in reality are time-dependent are intentionally described as long-term average values, such as the net uptake due to forest growth. The uncertainty at a single site would be significant for a given point in time, since the stage of forest growth is not described correctly. In regional long-term applications this is not considered a problem, because the purpose is to study relative contributions of different input parameters. In addition, the temporal description entails uncertainties in itself, possibly distorting the results even more than the use of average values. In some dynamic regional modeling the temporal development of net uptake by forest growth, for example, has been taken into account (Kurz *et al.* 1998).

A semi-regional SMART model application was carried out for ten selected small catchments in south-eastern Finland (Johansson 1995b) as part of a research program to estimate the need for neu-

**Table 3.** Results of the sensitivity analysis in terms of the forest soil area with base saturation smaller than 5% in the year 2040 (III). The results are given in percentage of the total forest soil area and as relative units compared with the reference case.

reference case	38%	1.00
filtering factor $\phi = 1.6$ (reference 2.0)	35%	0.92
half base cation deposition	46%	1.21
double base cation deposition	18%	0.46
half weathering rate	57%	1.48
double weathering rate	11%	0.27
more detailed base cation classification	43%	1.12
no base cation removal by harvesting	13%	0.35

tralization of surface waters in Finland (Iivonen and Kenttämies 1994). The potential need and effectiveness of liming compared to changes in deposition was assessed. A partly automatic calibration routine was developed for the SMART model using prescribed input parameter uncertainty ranges to attain acceptable reproduction of current observations. The results suggested that for all selected lakes the acidification trend was similar, which could be reversed only with significant emission reductions. The neutralizing effect from lake liming was temporary and lasted some years only. The results indicated new ways in performing and presenting regional dynamic simulations, although definite conclusions from a restricted set of lakes should be drawn with care (Stoddard *et al.* 1998). The semi-automatic model calibration provided new insights for improvements of input data or process descriptions.

The performance of the RAINS soil model and the more complex SMART model have been tested using soil chemistry data from southern Sweden (Posch *et al.* 1989a). Both of these models have been designed to generate overviews on regional acidification rather than site-specific developments. The first soil samples date back to 1949–1950 and samples had been retaken in 1984–1985. Validation results have shown that the change in the amount of exchangeable base cations is predicted fairly well, while the changes in soil pH could hardly be reproduced by the RAINS soil model, but reasonably well by the SMART model. These conclusions supported the use of soil base cation indicators rather than pH in applications of the RAINS soil model. Studies on the substantiation of regional dynamic performance of other acidification models are available (*e.g.* Kämäri *et al.* 1989, Kämäri 1990). Possibilities for forthcoming validation studies in Finland could be based on data from intensive monitoring and mapping programs on forest soils (De Vries *et al.* 1998) and small catchments (Kleemola and Forsius 1998, Mannio and Vuorenmaa 1995).

The results of the regional dynamic modeling study showed an increasing risk for forest soil acidification and revealed the most important input parameters in different regions in Finland. The importance of input variables changed also in time. The analysis included a constant range of uncertainty and no correlations between the variables, therefore a more careful estimation is needed before definite conclusions can be made.

The results showed that a relatively simple model with regional input data and proper indicators on soil acidification can provide useful insight to estimate spatial and temporal allocation of emission reduction requirements.

### 6.3 Site-specific dynamic modeling

#### 6.3.1 SMART model applications

A small forested catchment in southern Finland, Rudbäcken, was chosen to study on the relative importance of climate change, acidic deposition and forest harvesting on soil and streamwater chemistry (VII). The evaluation was carried out with a linked model system. It employed the climate generator (CLIGEN), the DEPUPT model, the hydrological TOPMODEL and the SMART model. The model system is presented in Fig. 4c. The emphasis was to assess long-term effects from different combinations of scenarios on the three driving forces.

The models were calibrated to measured values of runoff and annual dynamics of the groundwater level (TOPMODEL), observed soil and streamwater chemistry (SMART) and the measured deposition and growth stage of the forest (DEPUPT). Daily time series of precipitation and temperature were generated with a climate generator (CLIGEN) assuming the maximum climate change scenario developed for the Finnish Research Programme on Climate Change (Carter *et al.* 1995). The consequent average annual (and monthly) runoff values for the SMART model were then produced with the TOPMODEL using the temperature and precipitation values generated with the climate generator.

The DEPUPT model was used to create histories and future scenarios of total deposition and nutrient uptake. Modeled depositions were related to values measured at the site. The future depositions were calculated with the DAIQUIRI model (VIII). The base case deposition scenario A included the current reduction plans from the 1994 Sulphur Protocol signed under the CLRTAP (Barrett *et al.* 1995) and present deposition levels for nitrogen compounds. The total average depositions to the catchment soils are shown in Fig. 17a. The considerable emission reductions scenario C indicated maximum feasible reductions for sulfur (Cofala and Schöpp 1995) and a flat

30% emission reduction for nitrogen oxides (denoted here MFR'). Forest growth and clearcutting effects were simulated with a modified growth submodel. The hypothetical catchment clearcutting in the year 2000 was assessed to demonstrate the extreme potential effects of complete forest felling in scenario B. The total nutrient uptakes by forest growth, excluding nutrients released from litterfall, of scenario B are shown in Fig. 17. The decreasing filtering effect of forest canopy on deposition is noticeable after the clearcut in 2000. The effect of the high climate scenario on the modeled growth was taken into account with changes in the potential annual growth dependent on ETS, which was correlated to average annual temperature in scenarios D and E. Changes in temperature resulted in large increases in both the

annual growth and the final forest volume.

The catchment simulation results from different scenarios were examined with acidity-related output values of base saturation, *BS* (Fig. 18), streamwater pH, base cation and nitrate fluxes. Scenario A, in which current reduction plans for emissions and no change in the present climate are assumed, provided the reference level. The model results indicated that soil acidification would stop only with the considerable emission reductions scenario C. The clearcut scenario B first resulted in improving the *BS* due to reduced base cation uptake in the starting phase of forest growth but later leading to continuing decline of *BS*. The magnitude of clearcutting results were comparable to those of the C scenario, which assumed considerable cuts in sulfur and nitrogen oxides emissions, especially those of sulfur. The consequent deposition decline at the site was smaller than relative emission reductions. The benefit from applying maximum emission reductions as soon as possible was reflected in the quick stabilization of *BS*. Increased soil acidification was indicated for the two climate change scenarios D and E. This was caused by both higher base cation uptake related to increased forest growth under generally warmer climate and the more efficient filtering of deposition as a result of increased canopy volume. In this case the base cation uptake by forest growth became an increasingly important contributor to soil acidification. In addition, the modeled higher level of nitrate ( $\text{NO}_3^-$ ) output in streamwater resulted in an increased leaching loss of base cations from the soil pool. In scenario E nitrate already comprised a significant proportion of the outflux of strong acid anions by the year 2050, thus contributing to both soil and water acidification and downstream eutrophication (Wright and Hauhs 1991).

The effects of the three main driving forces (climate, deposition and clearcutting/nutrient uptake) showed complex interactions, which were not always intuitively evident beforehand. Climate change, considerable emission reductions, and total catchment clearcutting were almost equally significant for effects related to soil acidification. No scenario resulted in sustainable development or recovery of the catchment soil in the view of examined parameters. Considerable emission reductions would result after the reduction measures in either a slight improvement (streamwater pH) or near-stabilization (soil base saturation) compared

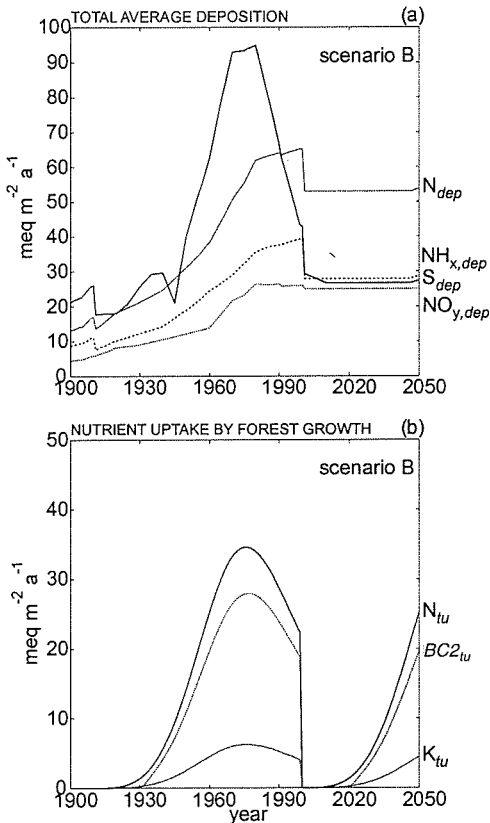


Fig. 17. Simulated (a) total average deposition of sulfur and nitrogen compounds to catchment soil and (b) total uptake of base cations, where  $BC2 = \text{Ca} + \text{Mg}$ , and nitrogen by forest growth excluding nutrients released from litterfall. Both figures represent scenario B of forest clearcut in the year 2000.

with the current situation. The most negative response on these variables was predicted for the maximum climate change scenario, in which a decreased nitrogen immobilization in the soil was assumed. Although for these scenarios clearly increasing nitrate leaching was predicted, the nitrogen process description used may not have fully entailed proper quantifications for the soils considered. The lack of long-term empirical data from the site did not allow the relation of model predictions to actual ecosystem responses.

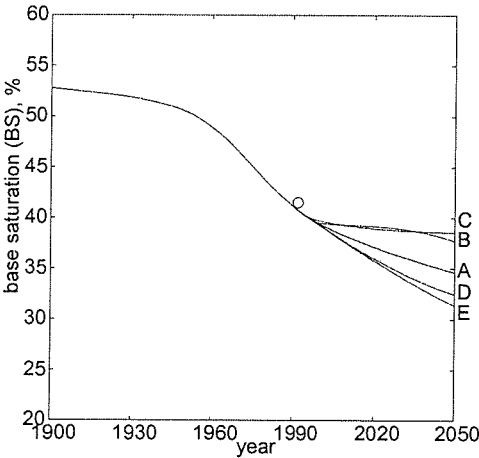
The SMART model simulations were carried out also with monthly time steps for the Rudbäcken site to assess intra-annual responses to changes in atmospheric deposition and forest growth and effects of snow melt (Johansson 1997). The model was first calibrated using annually averaged observations (VII) and then run with monthly input data. In general, the SMART model was able to simulate rather well monthly leaching values, however, more differences were found in concentrations and in pH. At this site, the intra-annual variation of modeled deposition or uptake seemed to have a minor effect on the results of the examined leaching values. The results proved simulations of intra-annual acidification processes feasible with the SMART model. The finer temporal resolution requires more elaborate calibration to measurements and better site-spe-

cific seasonal input data, thus limiting the use of high temporal resolution.

**6.3.2 Analyses on uncertainty and applicability**

Uncertainty in dynamic modeling of environmental impacts is inevitable. Complex environmental systems can hardly be well described in the traditional physico-chemical sense (Hornberger and Spear 1981). In catchment-scale applications, the data hold uncertainty at two levels: measurements of the input data and the interpretation of regional aggregated properties based on discrete samples. In practice there are always more unknown model parameters than there are factors with measured or well-defined values from *e.g.* literature. This retains inherent uncertainty in the model performance. The procedure of the model calibration to observed values provides insight which parameters are most important for the specific site. Although the model coincides with current measurements, the uncertainty for historical and future values may be considerable. Therefore, the predicted relative changes of simulated outputs are more important in the interpretation of results than the absolute values.

The calibration of the SMART model to observed  $\text{NO}_3^-$  output fluxes for the Rudbäcken site (VII) proved to be difficult. It was possible to get modeled values close to observed ones only by using C-N-ratio as high as 36 in the model, close to the observed maximum value in the soil profile (38). This obviously reflects the current limitations in modeling nitrogen dynamics (*e.g.* Rasmussen *et al.* 1995) and makes the estimation of the timing of possible nitrogen leaching increases uncertain. However, increasing trends in  $\text{NO}_3^-$  stream output fluxes have been observed in two forested catchments with long-term monitoring, located only some 30 km from Rudbäcken (Lepistö 1995), suggesting an increasing importance of nitrogen saturation for forested ecosystems in this region. The new nitrogen process descriptions on organic nitrogen leaching and temperature-dependent mineralization introduced to the SMART model (VII) retained uncertainty. The results indicated that the current nitrogen process description in the model may not account for all important factors. Further analysis on the model performance could be possible in the future, since part of the catchment was harvested



**Fig. 18.** Average catchment soil base saturation for five different scenarios (VII): A: base case (CRP,  $T_{ave}$ ,  $Q_{ave}$ ), B: forest clearcut (A+clearcut in 2000), C: considerable emission reductions (A+MFR'), D: climatological changes (CRP,  $T_{high}$ ,  $Q_{high}$ ,  $growth(T)$ ), E: temperature-sensitive nitrogen processes (D+N<sub>im</sub>(T)).

and some sampling has been done over the transition period.

The derivation of deposition and forest growth scenarios with the DEPUPT model was found to be sensitive to the calibration values at the Rudbäcken site (VII). The fitting of modeled depositions to values averaged over the observation period was relatively straightforward. Several separate measurement years may introduce additional difficulties for the calibration, if there are large variations in emission levels and local meteorology. The calibration procedure would clearly benefit from more detailed comparisons and method development.

The description of the forest growth was very general in the DEPUPT model. It probably could not reflect the variations of the forest cycle at the site due to missing information of forest history. The calibration of the DEPUPT model was difficult when the nitrogen deposition levels were extremely low and being a limiting factor for forest growth. The calibration to presently observed annual growth rate and standing volume may result in growth underestimations after clearcutting in general. This would, however, play a significant role only if such variations were large or occurred at times affecting the model calibration to observations. The new additional simple description of the temperature dependency of forest growth was an illustration of potential effects of temperature to growth rate increase. The linkage was made at an annual level, assuming that the average temperature is reflected in the ETS and the annual growth period. Such relationship may overestimate the variation in potential growth.

The analysis for intra-annual variations at the Rudbäcken site (Johansson 1997) was directed at exploring possibilities to describe seasonal variations in the catchment. After calibration to annual data and using input from monthly observations of deposition the model predictions were fair. The best explained variables were related more to hydrology-related variables such as leaching. Using annual modeled deposition allocated to monthly estimates according to general observed relative variations did not achieve better results, limiting current possibilities for scenario simulations. The further use of the SMART model for intra-annual simulations was found useful to study changes of short duration only if sufficiently detailed monitoring data were available for the model calibration.

The SMART model was applied to small catchments in northeast Lapland to demonstrate ecosystem responses for different assumptions of sulfur deposition and adsorption in soil (Kämäri *et al.* 1995). The high emissions near the Russian border result in large deposition gradients to the southwest direction and the depositions have high uncertainty at the Christmas Lakes (Joulujärvet). The SMART model was calibrated to present soil and water quality data using best estimates for historical deposition patterns for the period 1890–1990 first by assuming a high sulfur deposition in combination with sulfate adsorption and second a lower present deposition with no adsorption. Higher and lower bounds for present sulfur deposition attained were between 0.4 and 0.8 gS m<sup>-2</sup>a<sup>-1</sup>, falling within the range from nearby measurements (Järvinen and Vänni 1990, Derome *et al.* 1992) and deposition models (III, Tuovinen *et al.* 1993). Neither measurements nor models have clearly revealed the real deposition to soils at the site. The discrepancies may be due to uncertainty in emission estimates or difficulties of dry deposition estimates in both measurements and models, assuming that other input variables were accurate enough and the process descriptions in the SMART model were adequate for the site. The results suggested that the soil models are sensitive to the identification and quantification of influential parameters and processes and that cautious interpretation is required. However, the general trends and relative differences from various scenarios and assumptions proved to give consistent and thus useful indications on the responses.

A full validation of a dynamic model analyzing long-term responses over decades or even centuries is practically impossible. The validation procedure can rather be a stepwise procedure of gaining credibility through confirming research results. As to the SMART model or related modeling approaches, indications on the efficiency have been acquired by applications to paleolimnological data (Wright *et al.* 1986, Huttunen *et al.* 1990), historical soil data (Posch *et al.* 1989a) and intercomparisons between several soil and surface water models (Cosby *et al.* 1985, Warfvinge *et al.* 1992). Application and performance of other acidification models at regional levels have been reported (see, *e.g.*, Kämäri *et al.* 1989, Kämäri 1990). An analysis with two Dutch models, NUCSAM and RESAM, with daily and

annual time steps, respectively, calibrated and validated at a German site concluded that the uncertainties due to neglected seasonal variation are rather small (Kros *et al.* 1995). Comparisons of other models to historical values of soil chemistry can be found (Falkengren-Grerup *et al.* 1987, Sverdrup *et al.* 1995). An extensive comparison of several models on hydrology, soil acidification and forest growth was carried out with data from Solling site (Grinsven 1995) with an important conclusion, that complex models did not deliver any better results than simple models.

The conclusions from site-specific dynamic integrated model simulations revealed the feasible and useful linkage of models that describe processes affecting acidification. The SMART model was used to estimate the relative importance of different driving forces affecting acidification and the importance of intra-annual effects and specific input parameters to the long-term dynamics of acidification. The results showed the necessity to improve current knowledge on nitrogen process descriptions, to further study the different interconnected factors affecting acidification and the importance to include temporal aspects in emission reduction requirements.

#### 6.4 Attributes of integrated assessment models

The research on effects of acid rain has been pursued during the last two decades. Reported damage and predicted future risks have led to consider further emission reductions on an effects-based approach substituting more traditional flat rate reductions or sole implementation of best available technologies. The large spatial and temporal scales treated in complex integrated assessment models (IAMs) and increasing emission control costs support further evaluation of uncertainties involved. *E.g.* large-scale effects on forests have been analyzed with the help of monitoring data to evaluate the ecosystem responses to changing emissions (Skelly and Innes 1994, Müller-Edzards *et al.* 1997). In general, IAMs have been able to grasp the wide scale of factors affecting the acid rain problem in an informative way to provide support for policy-making.

The attributes of the three national integrated assessment model systems presented in this study are shown in Fig. 19 in terms of time, space and

the ease of integrability. According to the experiences, the easiest integrable have been regional steady-state critical loads (IV) and site-specific dynamical model simulations (VII). The extension of dynamic modeling to regional applications has increasing spatial uncertainty in input data and lack of suitable observations for model calibration (I, II). However, the latter weakness may be redeemed by forthcoming intensive monitoring (De Vries *et al.* 1998) and other data. Critical loads are usually calculated for large regions. They have hardly been separately calculated for the use at specific sites only, where the temporal aspects of input data for steady-state calculations may pose interpretational difficulties (Posch *et al.* 1997b). Although critical loads are a valuable tool in quantifying and displaying parameters relevant to acidification, they hardly contribute to the development and elaboration of issues on dynamic modeling, whereas the opposite has generally been the case.

The validation of models is an important issue, especially when they are used as a means to evaluate large-scale and/or complex physical processes and in supporting decision-making (Oreskes *et al.* 1994, Konikow and Bredehoeft 1992). In practice, terms verification, validation and confirmation are often (mis)used interchangeably. Their pragmatic definitions in the context of integrated assessment models can be presented as follows (see, *e.g.*, IAEA 1989). Verification of a model shows that the concepts, mathematical representation, numerical solutions and computer codes are correctly implemented and the model (or the code) behaves as intended. This is usually done by comparing results from analytical solutions to numerical ones. In validation the model predictions are compared with observations and experiments independent from model development. The predictive accuracy should be acceptable over the range of conditions of model applications. As to confirmation, empirical observations can be interpreted as deductive consequences of a theory or scientific law. These observations then confirm the theory, law or model performance. The more in number and diversity the confirming observations are obtained, the more probable it is that the conceptualization embodied in the model is not flawed.

One may agree that verification does not justify model behavior outside the range and realm of analytical solutions, and that a validated model



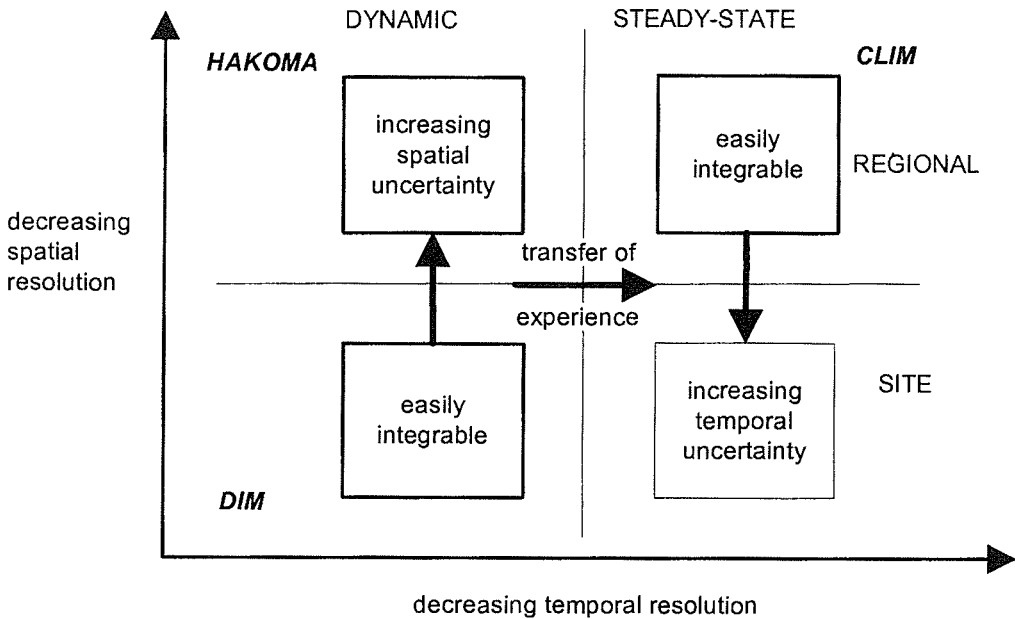


Fig. 19. Attributes of the three integrated assessment model (IAM) systems presented in this study in terms of time, space and the ease of integrability. The abbreviations denote the three IAMs of this study: Finnish integrated acidification model HAKOMA, critical load integrated model system (CLIM) and dynamic integrated model system (DIM).

may any time be invalidated with new observations that contradict model predictions. In this case it may be appropriate to accept that confirming observations do not demonstrate model veracity but support its probability for accurate performance (Oreskes *et al.* 1994). In spite of these philosophical and practical difficulties to evaluate a single model or an integrated model system performance, the use of models has its advantages. Models and model systems can be used as representations of complicated interactions and processes of the problem as a quantifying and guiding tool to explore differences between models of different resolutions and complexity, to illustrate areas for further study, to provide insight through sensitivity analyses and to portray the range of certainty of the results for the user.

The efforts to confirm the three integrated model systems in this study included the examination of sensitivity and uncertainty aspects for the main components of the IAMs. The confirmation of critical loads with observations on visible or measurable damage is practically impossible, due to the steady-state approach of the calculation method. The critical load method portrays dy-

namical and ever-changing ecosystem properties in a simple steady-state approach which is conveyable to policy-makers and useful in the preparation of emission reduction strategies based on long-term ecosystem protection targets. The dynamic soil acidification models were calibrated to current measurements in site-specific and regional simulations. The confirmation was restricted due to lack of long-term observations.

## 7 Conclusions

The acid rain problem is considered as a serious threat to the environment requiring innovative scientific research and policy analysis to find and implement appropriate solutions. Models have been used to clarify the importance of factors affecting acidification and to find out practical counteracting measures. The national integrated models presented in this study demonstrated the conversion of key findings into practical assessment tools. In addition to promoting scientific research, these model systems helped to convey necessary information to decision-makers inter-

actively. These tools were directly used for formulating national policy and provided the Finnish data for international negotiations.

Three integrated model systems were applied in this study. The first was the Finnish integrated acidification model HAKOMA, for which the module linkages and impact modules were developed. Forest soil acidification was assessed with regional dynamic soil model simulations. Sensitive regions in Finland, where soil base saturation decreases under 5% were identified. The forest soil area belonging to this risk category was estimated to be between 57% and 12% in 2040 depending on the emission reduction scenarios in Finland and in Europe. The optimization of sulfur emission reductions cost-effectively in Finland and areas nearby to attain environmental protection targets based on critical loads indicated the importance and cost-effectiveness to reduce long-range transboundary pollution.

The critical load integrated model (CLIM) system was developed for national and international applications, where critical loads were used as environmental protection targets for formulating efficient emission reduction strategies. Critical loads for forest soils were mapped according to internationally agreed methods. The maps provided a comparatively simple concept to formulate environmental protection targets. The exceedances of critical loads of sulfur in Finland have currently decreased to about 7% of ecosystem area. The decrease of exceedance is difficult with domestic emission reduction measures only, since Finland still imports a relatively large fraction of its acidifying deposition, about 80% for sulfur and nitrogen oxides and 67% for ammonium. Scenario calculations indicate that a 95% protection of ecosystem area will be attained in 2010 with current reduction plans and legislation in Europe.

Dynamic integrated model systems (DIM) were used to study in more detail the relevant calculation parameters and processes and the temporal responses to driving forces, *i.e.* acidifying deposition, forest growth and climate change, at one forested catchment in southern Finland. The development of the new DEPUPT model enabled the dynamic description of deposition and forest growth. The results clarified the complex interactions of major input parameters and the three driving forces were found to potentially have equal quantitative importance on soil acidifica-

tion. The results indicated that only prompt and strict emission reduction controls in Europe would prevent further acidification.

The basic input data for the model systems were first obtained for regional dynamic modeling in the HAKOMA model. They were later modified and updated for the critical load mapping. The most significant input variables, mineral weathering, base cation deposition and nutrient uptake, were originally derived within this study. The transfer matrices in the HAKOMA model and the later DAIQUIRI model were developed and made operational as a collaborative effort using results from detailed models for the atmospheric transport of acidifying compounds. New emission inventories and new nitrogen transfer matrices in the DAIQUIRI model improved estimates of the pollutant loads both regionally and for specific sites. The adoption of a method to generate historical and future deposition and nutrient uptake estimates with the DEPUPT model provided new useful input data for site-specific dynamic soil acidification modeling.

Uncertainty analyses due to input data variability were carried out on forest soil critical loads and dynamic responses at regional level. The uncertainty of critical loads was in general  $\pm 30\%$  and comparable with estimated input parameter uncertainties. The most influential factors for exceedances of critical loads in Finland were base cation deposition, nutrient uptake by forest growth and base cation weathering rate. The uncertainty in critical loads due to incomplete knowledge was demonstrated through the re-evaluation of base cation weathering rate on the basis of new geochemical data. The resulting critical loads of sulfur were about one third lower than estimated earlier. It was concluded that there was more uncertainty in modeling absolute values than comparing different scenarios to each other, where the relative changes, *e.g.* ecosystem area under risk, were of interest.

The confirmation of the model systems in this study was assessed mainly on the basis of other studies using the same or similar methods and models. Dynamic models were either initialized with current observations or calibrated to reproduce them. In spite of the simplicity of the critical load approach based on steady-state mass balance method, more useful methods or criteria were not found as basis for estimating emission

reduction requirements. For illustrative purposes maps of sustainable critical loads were derived, assuming no alkalinity leaching and no further changes of soil base cation pool. These sustainable critical loads were about one third to half from the standard critical loads and would lead to stricter environmental protection targets than at present.

The integrated model systems presented in this study proved to be functional tools in assessing acidification and relations to other environmental problems. The integration of results from several disciplines into a common framework provided new insight on the most important parameters and links between processes affecting forest soil acidification. The extension of model systems to a multi-pollutant – multi-effect approach increased the complexity of the data and model management and interpretation of results.

The three different integrated assessment models (IAMs) and their applications presented in this study suggested their acceptance and feasibility in supporting national effects-oriented policy-making. The input data and model performance were at an adequate level for that purpose. This was notably the case for the critical load approach enabling a deep discourse between scientists and decision-makers in the survey of environmental protection targets and the extent of emission reductions required for the non-exceedance of critical loads. National integrated models enabled the necessary evaluation, checking and information transfer within Finland on the scenario analyses of international IAMs. Although acidifying sulfur and nitrogen emissions have been reduced in Europe 37% during 1980–1995, in Finland acidifying emissions 61% and notably sulfur emissions 84%, the acid rain problem requires further assessment.

The practical development and maintenance of an IAM always requires considerable resources. A fully integrated single software tool, such as the RAINS model and to some extent the HAKOMA model, is usually a long-term and resource-demanding task. Another approach is to creatively soft link existing models, as was done in the critical load and dynamic integrated model systems in this study. The latter approach requires a strong and clear systems analytical point of view on the integration scheme. It proved efficient and flexible but required more coordination and management. Additional limitations include the availabil-

ity of appropriate models to cover all needs, the careful design to embody compatible input-output interfaces in the models and the long-term financial support for developing an integrated model system. The lack of a common user interface and the need to manage a variety of different models and input data limit the use of soft linked integrated model systems mainly to research purposes.

The three integrated model systems presented in this study were not able to address all details and novel considerations within the field. The model results and uncertainty analyses pointed out some important areas for further improvement:

- further uncertainty analysis and confirmation of the model systems is needed to reduce uncertainty in the demands on more costly emission reduction measures and to gain more confidence in the model predictions,
- additional studies on the dynamics of acidification, including the effects of the timing of emission reductions, to provide more insight in the attainment of environmental targets defined through steady-state critical load methods,
- supplementary elaboration of some input parameters is desired, such as the temporal development of base cation deposition and a simple yet realistic dynamic forest growth model,
- strong efforts are required in evaluating the increasingly important nitrogen processes, which are currently not well understood and inadequately described in existing integrated models,
- the steps taken toward a multi-pollutant – multi-effect approach, which links several pollutants and effects within the same modeling framework, need continuous support if they are to be maintained operative and extended to account for climate change or other important environmental issues.

## Yhteenveto

Ilmansaasteiden haitallisista vaikutuksista erityisesti happamoituminen on ollut viime vuosikymmeninä vakava ympäristöongelma Euroopassa ja Pohjois-Amerikassa. Vaikka happamoittavien rikki- ja typpiyhdisteiden päästöt ovat Euroopassa laskeneet 37% vuosina 1980–1995, on päästöjä

pienentämiseen edelleen tarvetta. Mallijärjestelmien avulla voidaan arvioida laajoille alueille kulkeutuvien ilmansaasteiden vaikutuksia sekä laskea päästöjen rajoituskustannuksien perusteella tehokkaita strategioita kansainvälisten päästövähennyssovimusten pohjaksi. Yhdennettyjä arviointimalleja käytetään tällä hetkellä sekä kaukokulkeutumissopimuksen uuden pöytäkirjan että Euroopan unionin päästökattodirektiivin valmistelussa. Ne perustuvat useiden ilmansaasteiden ja useiden vaikutusten yhtäaikaiseen tarkasteluun ja päästövähennysten kustannustehokkaaseen optimointiin.

Tässä tutkimuksessa käsitellään kolmea Suomessa kehitettyä kansallista yhdennettyä arviointimallia ja niiden käyttöä metsämaan happamoitumisen maantieteellisen laajuuden, ajallisen kehityksen ja erilaisten päästövähennysten vaikutusten arviointiin. Mallijärjestelmiin koottiin happamoitumisen kannalta olennaiset kuvaukset happamoittavista rikin, typenoksidien ja ammoniakkitypen päästöistä, kulkeutumisesta ilmakehässä, laskeumista sekä ekosysteemien vasteista. Eri osakuvausten tarkkuus ratkaistiin kulloistenkin tutkimustavoitteiden perusteella.

Ensimmäinen mallijärjestelmistä oli happamoitumisen kokonaismalli HAKOMA. Siihen liitettyllä vaikutusmallilla arvioitiin metsämaiden happamoitumiseen vaikuttavat tärkeimmät tekijät, niiden ajallinen vaihtelu sekä herkimmat alueet. Simulointien perusteella pitkälle happamointuneiden metsämaiden osuus oli 12–57% vuonna 2040 päästöjen vähennysstrategioista riippuen. Mallikehityksen yhteydessä kerättyjä lähtöaineistoja käytettiin hyväksi myös seuraavissa mallijärjestelmissä.

Toinen yhdennetty arviointimallijärjestelmä perustui kriittisten kuormitusten kartoitustyöhön. Kriittinen kuormitus kuvaa ekosysteemin saaste-kuormituksen sietokykyä pitkällä aikavälillä. Kuormitusarvot laskettiin ajasta riippumattomalla massatasapainomenetelmällä, joka on kansainvälisesti sovitettu laskentatapa. Metsämalle laskettu ja happamoittavan laskeuman kriittisiä kuormituksia käytettiin sekä kansallisten että kansainvälisten suojelutavoitteiden muodostamiseen. Suomessa kriittisten kuormitusten ylitys on pienentynyt erityisesti vuoden 1980 jälkeen. Suomen ja muiden Euroopan maiden päästörajoituksia koskevien suunnitelmien ja lainsäädännön perusteella kriittistä kuormitusta ei laskelmien mukaan enää ylitetä 95% ekosysteemien pinta-alasta Suo-

nessa vuonna 2010.

Kolmantena mallijärjestelmänä oli dynaamisen yhdennetty mallijärjestelmä, jossa maaperän happamoitumiskehitystä kuvaava SMART-malli kytkettiin useisiin muihin malleihin. Tulokset yhdeltä eteläisen Suomen metsävaluma-alueelta osoittivat, että vain pikaiset ja suuret päästövähennykset Euroopassa riittävät pysäyttämään kyseisen valuma-alueen maaperän happamoitumisen. Happamoittavan laskeuman, metsän hakkuiden ja ilmastomuutoksen mahdolliset vaikutukset valuma-alueen maaperän happamoitumiskehitykseen arvioitiin yhtä suuriksi.

Metsämaiden kriittisille kuormituksille tehty epävarmuustarkastelu osoitti tärkeimmiksi laskeutamuuttujiksi emäskationilaskeuman, metsän kasvuun sitoutuvat ravinteet sekä mineraalien rapautuminen maaperästä. Kriittisten kuormitusten lukuarvojen epävarmuudeksi laskettiin yleisesti  $\pm 30\%$ , mikä oli samansuuruinen kuin laskeutamuuttujien arvioidut epävarmuudet. Mineraalien rapautumisesta vapautuvien emäsravinteiden määrä laskettiin uudelleen käyttäen hyväksi uutta geokemiallista kartoitustietoa, mikä johti kriittisten kuormitusten lukuarvojen yleiseen pienemiseen noin kolmanneksella. Yhteistyönä kehitetty DAIQUIRI-malli rikki- ja typpilaskeumien arviointiin sekä uudet päästötiedot tarkensivat Suomen typpipäästöjen leviämiskuvausta sekä laskeumien kehitysarvioita. Valuma-aluekohtaisia happamoittavia laskeumia ja ravinteidenottoa kuvattiin laaditulla DEPUPT-mallilla, joka ottaa huomioon metsän kasvun ja päästöjen muutosten aikakehityksen.

Kaikki tutkimuksessa kehitetyt kolme yhdennettyä arviointimallijärjestelmää osoittautuivat käyttökelpoisiksi, kun kuvattiin metsämaiden happamoitumiseen vaikuttavia määrällisiä tekijöitä sekä happamoittavien päästöjen vähennystarpeita. Mallijärjestelmien erilaiset tekniset toteutukset osoittivat, että yksittäinen kokonaismalli oli yhtenäinen ja helppokäyttöinen, mutta hankala ylläpitää. Toisiinsa liitettyjen erillisten mallien muodostama järjestelmä oli tutkimuskäytössä nopea ja tehokas, huolimatta vaativasta mallintamistietojen yhteenkokoamisesta ja tarkistuksesta. Mallien luotettavuus perustuu havaintoihin, joihin malleja on sovitettu tai joihin tuloksia on verrattu, epävarmuusanalyysiin sekä samojen mallien muualla tehtyihin vertailu- ja todentamistarkasteluihin. Tässä työssä käytetyt kansalliset kolme yhdennettyä arviointimallijärjestelmää vä-

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Matti Johansson

## References

- Acidification Committee 1998. Happamoitumistoimikunnan mietintö (Report of the Acidification Committee), Ministry of the Environment, Edita Ltd, Helsinki, Finland, *The Finnish Environment* 219, 182 pp. (In Finnish with English summary.)
- Alcamo J. & Bartnicki J. 1987. A framework for error analysis of a long-range transport model with emphasis on parameter uncertainty. *Atmospheric Environment* 21(10): 2121–2131.
- Alcamo J., Krol M. & Posch M. 1995. An integrated analysis of sulphur emissions, acid deposition and climate change. *Water, Air and Soil Pollution* 85: 1539–1550.
- Alcamo J., Shaw R. & Hordijk L. (eds.) 1990. *The RAINS model of acidification. Science and strategies in Europe*, Kluwer Academic Publishers, Dordrecht, The Netherlands, 402 pp.
- Alveteg M. 1998. Dynamics of forest soil chemistry. Doctoral thesis, Department of Chemical Engineering II, Lund University, Lund, Sweden, *Reports in Ecology and Environmental Engineering* 3:1998, 198 pp.
- Alveteg M., Walse C. & Sverdrup H. 1998a. Evaluating simplifications used in regional applications of

- the SAFE and MAKEDEP models. *Ecological Modelling* 107: 265–277.
- Alveteg M., Walse C. & Warfvinge P. 1998b. Reconstructing historic atmospheric deposition and nutrient uptake from present day values using MAKEDEP. *Water, Air and Soil Pollution* 104: 269–283.
- Amann M., Bertok I., Cofala J., Gyarfas F., Heyes C., Klimont Z., Makowski M., Schöpp W. & Syri S. 1998a. Cost-effective control of acidification and ground-level ozone. Part C: Acidification and eutrophication scenarios. Fifth interim report prepared for the 21st meeting of the UN/ECE Task Force on Integrated Assessment Modelling. International Institute for Applied Systems Analysis, Laxenburg, Austria.
- Amann M., Bertok I., Cofala J., Gyarfas F., Heyes C., Klimont Z., Makowski M., Schöpp W. & Syri S. 1998b. Cost-effective control of acidification and ground-level ozone. Part A: Methodology and databases, Part B: Emission control scenarios. Sixth interim report to the European Commission DG–XI. International Institute for Applied Systems Analysis, Laxenburg, Austria.
- Amann M. & Klaassen G. 1995. Cost-effective strategies for reducing nitrogen deposition in Europe. *Journal of Environmental Management* 43: 289–311.
- Anttila P. 1990. Characteristics of alkaline emissions, atmospheric aerosols and deposition. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 111–134.
- Aoyama K., Katoh K., Murano T., Paces T. & Taguchi Y. (eds.) 1997. *Proceedings of the international congress of acid snow and rain*, 6–8 Oct 1997, Niigata, Japan, 738 pp.
- ApSimon H., Warren R.F. & Wilson, J.J.N. 1994. The abatement strategies assessment model – ASAM: Applications to reductions of sulphur dioxide emissions across Europe. *Atmospheric Environment* 28: 649–663.
- Asman W.A.H., Drukker B. & Janssen A.J. 1988. Modelled historical concentrations and depositions of ammonia and ammonium in Europe. *Atmospheric Environment* 22(4): 725–735.
- Bad Harzburg 1989. Methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded, draft manual. UN-ECE/CLRTAP Workshop on Mapping, 6–9 Nov 1989, Bad Harzburg, Federal Republic of Germany.
- Barkman A. 1998. Critical loads - assessment of uncertainty. Doctoral thesis, Department of Chemical Engineering II, Lund University, Sweden, *Reports in Ecology and Environmental Engineering* 1:1998, 170 pp.
- Barkman A., Warfvinge P. & Sverdrup H. 1995. Regionalization of critical loads under uncertainty. *Water, Air and Soil Pollution* 85: 2515–2520.
- Barrett K. & Berge E. (eds.) 1996. *Transboundary Air Pollution in Europe, Part 1: Estimated dispersion of acidifying agents and of near surface ozone*. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MSC-W Report 1/96*, 152+xxx pp.
- Barrett K., Seland Ø., Foss A., Mylona S., Sandnes H., Styve H. & Tarrason L. 1995. European transboundary acidifying air pollution. Ten years calculated fields and budgets to the end of the first Sulphur Protocol. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MSC-W Report 1/95*, 71 pp.+appendices.
- Batterman S. 1992. Optimized acid rain abatement strategies using ecological goals. *Environmental Management* 16(1): 133–141.
- Batterman S. & Amann M. 1991. Targeted acid rain strategies including uncertainty. *Journal of Environmental Management* 32: 57–72.
- Beck M.B. 1987. Water quality modeling: A review of the analysis of uncertainty. *Water Resources Research* 23(8): 1393–1442.
- Berge E. (ed.) 1997. *Transboundary air pollution in Europe. Part 1: Emissions, dispersion and trends of acidifying and eutrophying agents*. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/ MSC-W Report 1/97*, 108 pp.+addendum/corrigendum.
- Carter T., Posch M. & Tuomenvirta H. 1995. SILMUSCEN and CLIGEN – user's guide. Helsinki, Finland, *Publications of the Academy of Finland* 5/95.
- Cofala, J. & Schöpp, W. 1995. Assessing future acidification in Europe. Current state of the RAINS model development. Note prepared for the 15<sup>th</sup> meeting of the UN-ECE Task Force on Integrated Assessment Modelling, May 95, The Hague, The Netherlands. International Institute for Applied Systems Analysis, Laxenburg, Austria.
- Cosby B.J., Hornberger G.M., Galloway J.N. & Wright R.F. 1985. Modeling the effects of acid deposition: Assessment of a lumped parameter model of soil water and streamwater chemistry. *Water Resources Research* 21: 51–63.
- Cosby B.J., Hornberger G.M. & Wright R.F. 1989. Estimating time delays and extent of regional deacidification in southern Norway in response to several deposition scenarios. In: Kämäri J., Brakke D., Jenkins A., Norton S.A. & Wright R.F. (eds.), *op.cit.*, pp. 151–166.

- Cowling E. & Nilsson J. 1995. Acidification research: Lessons from history and visions of environmental futures. *Water, Air and Soil Pollution* 85: 279–292.
- Derome J., Lindroos A.-J., Niska K., & Välikangas P. 1992. Kokonaislaskeuma Lapissa vuonna 1990–1991 (Bulk deposition in Finnish Lapland during July 1990 to June 1991), In: Kauhanen H. & Varmola, M. (eds.), *The Lapland Forest Damage Project*, Interim report. Finnish Forest Research Institute, Finland, *Research Paper* 413: 39–48. (In Finnish with English abstract.)
- De Vries W. 1990. Philosophy, structure, and application methodology of a soil acidification model for the Netherlands. In: Kämäri J. (ed.), *op.cit.*, pp. 3–21.
- De Vries W. 1994. Soil response to acid deposition at different regional scales. Field and laboratory data, critical loads and model predictions. Doctoral thesis, Agricultural University, Wageningen, The Netherlands, 487 pp.
- De Vries W., Kros J. & Voogd J.C.H. 1994a. Assessment of critical loads and their exceedance of Dutch forests using a multi-layer steady-state model. *Water, Air and Soil Pollution* 76: 407–448.
- De Vries W., Posch M. & Kämäri J. 1989. Simulation of the long-term response to acid deposition in various buffer ranges. *Water, Air and Soil Pollution* 48: 349–390.
- De Vries W., Posch M., Oja T., Van Oene H., Kros H., Warfvinge P. & Arp P.A. 1995. Modelling critical loads for the Solling spruce site. *Ecological Modelling* 83: 283–293.
- De Vries W., Posch M., Reinds G.J. & Kämäri J. 1993. Critical loads and their exceedance on forest soils in Europe. DLO The Winand Staring Center for Integrated Land, Soil and Water Research, Wageningen, The Netherlands, *Report* 58 (revised version), 77 pp.
- De Vries W., Reinds G.J., Deelstra H.D., Klap J.M. & Vel E.M. (eds.) 1998. Intensive monitoring of forest ecosystems in Europe. UN-ECE/CLRTAP/International Cooperative Programme on Assessment and Monitoring of Air Pollution Effects on Forests and EU Scheme on the Protection of Forests against Atmospheric Pollution, Forest Intensive Monitoring Coordinating Institute, The Netherlands, *Technical Report* 1998, 193 pp.
- De Vries W., Reinds G.J., Posch M. & Kämäri J. 1994b. Simulation of soil response to acidic deposition scenarios in Europe. *Water, Air and Soil Pollution* 78: 215–246.
- Downing R., Hettelingh J.-P. & De Smet P. (eds.) 1993. *Calculation and mapping of critical loads in Europe. Status Report 1993*. Coordination Center for Effects, RIVM Report 259101003, Bilthoven, The Netherlands, vi+163 pp.
- EMEP/MS-C-W 1998. Transboundary air pollution in Europe. Part 1: Estimated dispersion of acidifying and eutrophying compounds and comparison with observations. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MS-C-W Report* 1/98, 150 pp.
- EPRI 1991. The concept of target and critical loads. Electric Power Research Institute, Ecological Studies Program, Environment Division, EN-7318 Research Project 2799-1, *Topical Report*.
- Erismann J.W., Potma C., Draaijers G.P.J., Van Leeuwen E.P. & Van Pul W.A.J. 1995. A generalized description of the deposition of acidifying pollutants on a small scale in Europe. *Water, Air and Soil Pollution* 85: 2101–2106.
- Erismann J.W., Van Pul A. & Wyers P. 1994. Parametrization of surface resistance for the quantification of atmospheric deposition of acidifying pollutants and ozone. *Atmospheric Environment* 28(16): 2595–2607.
- ExternE 1995. Externalities of energy. Vol. 2 Methodology. Prepared by ETSU & Metroeconomica, UK. EUR 16521 EN, European Commission, DG XII, Brussels, Belgium, 571 pp.
- Falkengren-Grerup U., Linnemark, N. & Tyler, G. 1987. Changes in soil acidity and cation pools of south Sweden soils between 1949 and 1985. *Chemosphere* 16: 2239–2248.
- Ferrier R.C., Jenkins A., Cosby B.J., Helliwell R.C., Wright R.F. & Bulger A.J. 1995. Effects of future N deposition scenarios on the Galloway region of SW Scotland using a coupled sulphur and nitrogen model (*MAGIC-WAND*). *Water, Air and Soil Pollution* 85: 707–712.
- Foell W., Green C., Amann M., Bhattacharya S., Carmichael G., Chadwick M., Cinderby S., Haughland T., Hettelingh J.-P., Hordijk L., Kuylenstierna J., Shah J., Shrestha R., Streets D. & Zhao D. 1995. Energy use, emissions and air pollution abatement strategies in Asia. *Water, Air and Soil Pollution* 85: 2277–2282.
- Forsius 1992. Acidification of lakes in Finland: Regional estimates of lake chemistry and critical loads. National Board of Waters and the Environment, Helsinki, Finland, *Publications of the Water and Environment Research Institute* 10, 37 pp.
- Forsius M., Alveteg M., Bak J., Guardans R., Holmberg M., Jenkins A., Johansson M., Kleemola S., Rankinen K., Renshaw M., Sverdrup H. & Syri, S. 1997. Assessment of the effects of the EU acidification strategy: Dynamic modelling on integrated monitoring sites. First Results. Finn-

- ish Environment Institute, Edita Ltd, Helsinki, Finland, 40 pp.
- Forsius M., Alveteg M., Jenkins A., Johansson M., Kleemola S., Lükewille A., Posch M., Sverdrup H., Syri S. & Walse C. 1996. Dynamic model applications at selected ICP-IM sites. In: Kleemola S. & Forsius M. (eds.), *5<sup>th</sup> annual report 1996*, UN-ECE, CLRTAP, International Cooperative Programme - Integrated Monitoring. Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment* 27: 10–24.
- Forsius M., Alveteg M., Jenkins A., Johansson M., Kleemola S., Lükewille A., Posch M., Sverdrup H. & Walse C. 1998. MAGIC, SAFE and SMART model applications at Integrated Monitoring sites: Effects of emission reduction scenarios. *Water, Air and Soil Pollution* 105: 21–30.
- Galloway J.N. 1995. Acid deposition: Perspectives in time and space. *Water, Air and Soil Pollution* 85: 15–24.
- Gough C.A., Chadwick, M.J., Biewald, B., Kuylenstierna, J.C.I., Bailey P.D. & Cinderby S. 1995. Developing optimal abatement strategies for the effects of sulphur and nitrogen deposition at European scale. *Water, Air and Soil Pollution* 85: 2601–2606.
- Grasman J. & Van Straten G. (eds.) 1994. *Predictability and nonlinear modelling in natural sciences and economics*, Kluwer Academic Publishers, Dordrecht, The Netherlands, 653 pp.
- Grennfelt P., Hov Ø. & Derwent D. 1994. Second generation abatement strategies for NO<sub>x</sub>, NH<sub>3</sub>, SO<sub>2</sub> and VOCs. *Ambio* 23(7): 425–433.
- Grennfelt P., Rodhe H., Thörnelöf E. & Wisniewski J. (eds.) 1995. Acid reign '95? Proceedings from the 5<sup>th</sup> international conference on acidic deposition: science & policy, Volume 1: Conference statement, plenary papers. 26–30 Jun 95, Göteborg, Sweden. Reprinted from *Water, Air and Soil Pollution* 85(1): 1–295.
- Grennfelt P. & Thörnelöf E. (eds.) 1992. Critical loads for nitrogen – a workshop report. Report from a workshop on 6–10 Apr 92 at Lökeberg, Sweden, organized by the Nordic Council of Ministers and the CLRTAP, *Nord* 1992:41, 428 pp.
- Grinsven H.J.M. (ed.) 1995. Modelling water, carbon and nutrient cycles in forests: Application of 16 simulation models to a spruce stand at Solling, Germany. Special issue, Proceedings of a workshop held in Leudsen, The Netherlands, 10–14 May 1993. *Ecological Modelling* 83: 1–283.
- Grönroos J., Nikander A., Syri S., Rekolainen S. & Ekqvist M. 1998. Maatalouden ammoniakkipäästöt. Osa 1: Päästöt ja niiden kehittyminen. Osa 2: Päästöjen vähentäminen ja vähentämiskustannukset (Agricultural ammonia emissions in Finland. Part 1: Emission assessment. Part 2: Emission reduction and reduction costs). Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment* 206, 65 pp. (In Finnish with English abstract.)
- Häkkinen A.J., Markkanen K., Hongisto M., Kartastenpää R., Milyaev V., Yasensky A.N., Kuznetsov V.I., Kopp I.Z. & Kivivasara J. 1995. Emissions in St. Petersburg, the Leningrad Area and Karelia. In: Tolvanen M., Anttila P. & Kämäri J. (eds.), Proceedings of the 10<sup>th</sup> World Clean Air Congress, Espoo, Finland, 28 May–2 June 1995, Vol 1. Emissions and control, The Finnish Air Pollution Prevention Society, Abstract 103, 3 pp.
- HAKOMA 1992. The distribution version of the HAKOMA model software. Sulfur and nitrogen depositions, soil acidification results display and sulphur optimization modules. February 1996, Technical Research Centre of Finland, Espoo, Finland.
- Hari P., Arovaara H., Raunemaa T. & Hautojärvi A. 1984. Forest growth and the effects of energy production: A method for detecting trends in the growth potential of trees. *Canadian Journal of Forest Research* 14: 437–440.
- Hedin L.O., Granat L., Likens G.E., Buishand T.A., Galloway J.N., Butler T.J. & Rodhe, H. 1994. Steep declines in atmospheric base cations in regions of Europe and North America. *Nature* 367: 351–354.
- Henriksen A., Kämäri J., Posch M., Lövblad G., Forsius M. & Wilander A. 1990. Critical loads to surface waters in Fennoscandia. Intra- and inter-grid variability of critical loads and their exceedances. Nordic Council of Ministers, Nord 1990:124, Reclamo, Oslo, Norway, *Miljørapport* 1990:17, 43 pp.
- Henriksen A., Skjelkvåle B.L., Mannio J., Wilander A., Harriman R., Curtis C., Jensen J.P., Fjeld E. & Moiseenko T. 1998. Northern European lake survey, 1995, Finland, Norway, Sweden, Denmark, Russian Kola, Russian Karelia, Scotland and Wales. *Ambio* 27(2): 80–91.
- Henttonen H. 1991. Kriging in interpolating July mean temperatures and precipitation sums. University of Jyväskylä, Jyväskylä, *Reports from the department of statistics* 12/1991.
- Henttonen H. & Mäkelä A. 1988. Estimation of local values of monthly mean temperature, effective temperature sum and precipitation sum in Europe. International Institute for Applied Systems Analysis, Laxenburg, Austria, *IASA Working Paper* WP-88-061, 20 pp.
- Hettelingh J.-P. 1990. Uncertainty in modeling regional environmental system. The generalization



- of a watershed acidification model for predicting broad scale effects, Doctoral thesis, Free University, Amsterdam, The Netherlands and International Institute for Applied Systems Analysis, Laxenburg, Austria, *IASA Research Report* RR-90-3, 224 pp.
- Hettelingh J.-P., Downing R. & De Smet P. (eds.) 1991. Mapping critical loads for Europe. UNECE/CLRTAP, Coordination Center for Effects, RIVM, Bilthoven, The Netherlands, *Technical report No. 1*, viii+86 pp.+appendices.
- Hettelingh J.-P., Gardner R. & Hordijk L. 1992. A statistical approach to the regional use of critical loads. *Environmental Pollution* 77: 177–183.
- Hettelingh J.-P. & Posch M. 1994. Critical loads and a dynamic assessment of ecosystem recovery. In: Grasman J. & Van Straten G. (eds.), *op. cit.*, pp. 439–446.
- Hettelingh J.-P., Posch M., De Smet P.A.M. & Downing R.J. 1995a. The use of critical loads in emission reduction agreements in Europe. *Water, Air and Soil Pollution* 85: 2381–2388.
- Hettelingh J.-P., Sverdrup H. & Zhao D. 1995b. Deriving critical loads for Asia and Europe. *Water, Air and Soil Pollution* 85: 2565–2570.
- Heuberger P.S.C. & Janssen P.H.M. 1994. UNCSAM: A software tool for sensitivity and uncertainty analysis of mathematical models. In: Grasman J. & Van Straten G. (eds.), *op. cit.*, pp. 362–376.
- Heyes C., Schöpp W., Amann M. & Unger S. 1996. A reduced-form model to predict long-term ozone concentrations in Europe. International Institute for Applied Systems Analysis, Laxenburg, Austria, *IASA Interim Report* WP-96-12, 58 pp.
- Hongisto M. 1992. A simulation model for the transport, transformation and deposition of oxidized nitrogen compounds in Finland. 1985 and 1988 simulation results. Finnish Meteorological Institute, Helsinki, Finland, *Finnish Meteorological Institute Contributions* 9.
- Hordijk L. 1995. Integrated assessment models as a basis for air pollution negotiations. *Water, Air and Soil Pollution* 85: 249–260.
- Hordijk L. & Kroeze C. 1997a. Integrated assessment models for acid rain. *European Journal of Operational Research* 102: 405–417.
- Hordijk L. & Kroeze C. 1997b. Integrated assessment models for acid rain. In: Aoyama K., Katoh K., Murano T., Paces T. & Taguchi Y. (eds.), *op. cit.*, pp. 714–727.
- Hornberger G. M. & Spear R.C. 1981. An approach to the preliminary analysis of environmental systems. *Journal of Environmental Management* 12: 7–18.
- Huttunen P., Kenttämies K., Liehu A., Liukkonen M., Nuotio T., Sandman O. & Turkia J. 1990. Palaeoecological evaluation of the recent acidification of susceptible lakes in Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op. cit.*, pp. 1071–1090.
- IAEA 1989. Evaluating the reliability of predictions made using environmental transfer models. International Atomic Energy Agency, Vienna, Austria, *Safety Series* 100, 106 pp.
- Iivonen P. & Kenttämies K. (eds.) 1994. Happamoituneiden vesistöjen kalkitus Suomessa. Neutralointiseurantaryhmän loppuraportti (Liming of acidified surface waters in Finland. The final report of the coordination group for neutralization), National Board of Waters and the Environment, Helsinki, Finland, *Publication of the Water and Environment Administration A* 204, 74 pp. (In Finnish with English abstract.)
- Ivens W. 1988. Atmospheric deposition of sulfur and base cations to European forests. International Institute for Applied Systems Analysis, Laxenburg, Austria, *IASA Working Paper* WP-88-101.
- Jakobsen H.A., Jonson J.E. & Berge E. 1997. The multi-layer Eulerian model: Model description and evaluation of transboundary fluxes of sulphur and nitrogen for one year. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MSC-W Report 2/97*, 50 pp.
- Janssen P.H.M., Heuberger P.S.C. & Sanders R. 1992. UNCSAM 1.1: A software package for sensitivity and uncertainty analysis. Manual. National Institute of Public Health and Environmental Protection, Bilthoven, The Netherlands, *Report* 959101004.
- Järvinen O. 1986. Laskeuman laatu Suomessa 1971–1982 (Deposition measurements in Finland 1971–1982), National Board of Waters and the Environment, Helsinki, Finland, *Mimeograph Series* 408, 142 pp. (In Finnish).
- Järvinen O. & Vänni, T. 1990. Bulk deposition chemistry in Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op. cit.*, pp. 151–165.
- Jenkins A., Whitehead P.G., Musgrove T.J. & Cosby B.J. 1990. A regional model of acidification in Wales. *Journal of Hydrology* 116: 403–416.
- Johansson M. 1988. *Rikkipäästöjen aiheuttama metsämaan happamoituminen* (Forest soil acidification due to sulphur emissions), M.Sc. thesis, Helsinki University of Technology, Technical Physics, Espoo, Finland, 83 pp. (In Finnish with English abstract.)
- Johansson M. 1995a. Derivation of deposition and uptake scenarios. In: *Annual synoptic report 1995*,

- Chapter 4: Dynamic model applications to selected ICP-IM catchments, UN-ECE, CLRTAP, International Cooperative Programme - Integrated Monitoring Programme Centre, Finnish Environment Agency, Helsinki, Finland.
- Johansson M. 1995b. Happamoitumis-kalkitusmallin alueellinen sovitus Kaakkois-Suomessa (Regional application of acidification-liming-model in southeastern Finland), In: Iivonen, P. and Kenttämies, K. (eds), *op.cit.*, pp. 36–37. (In Finnish).
- Johansson M. 1997. Modelling acidification of a forested site using different time steps. In: Aoyama K., Katoh K., Murano T., Paces T. & Taguchi Y. (eds.), *op.cit.*, pp. 433–438.
- Johansson M., Alveteg M., Walse C. & Warfvinge, P. 1996. Derivation of deposition and uptake scenarios. In: Knoflacher M., Schneider J. & Soja G. (eds.), *International workshop on exceedance of critical loads and levels*. Spatial and temporal interpretation of elements in landscape sensitive to atmospheric pollutants, Vienna 22–24 Nov 95, Federal Environment Agency, Vienna, Austria, *Conference Papers* BD.15/VOL.15, pp. 318–324.
- Johansson M. & Savolainen I. 1991. Regional estimation of future forest soil acidification. In: Pulkkinen E. (ed.), *op.cit.*, pp. 303–309.
- Johansson M., Savolainen I. & Tähtinen, M. 1990. Comparison of forest soil acidification estimates in different energy use and emission control scenarios. In: Fenhann, J., Larsen, H., Mackenzie, G.A. and Rasmussen, B. (eds.), *Environmental models: Emissions and consequences*, Elsevier, The Netherlands, pp. 449–459.
- Johansson M., Tähtinen M. & Amann M. 1991. Optimal strategies to achieve critical loads in Finland. In: Kainlahti, E., Johansson, A., Kurki-Suonio, I. & Geshwiler, M. (eds.), *Energy and environment 1991*, Proceedings from 1991 international symposium on energy and environment, 25–28 Aug 1991, Espoo, Finland, American Society of Heating, Refrigerating and Air-Conditioning Engineers, Inc., Atlanta, USA, pp. 341–345.
- Jongmans A.G., Van Breemen N., Lundström U., Van Hees P.A.W., Finlay R.D., Srinivasan M., Unestam T., Giesler R., Melkerud P.-A. & Olsson M. 1997. Rock-eating fungi. *Nature* 389: 682–683.
- Kaitala V., Pohjola M. & Tahvonen O. 1992. Transboundary air pollution and soil acidification: A dynamic analysis of an acid rain game between Finland and USSR. *Environmental and Resource Economics* 2: 161–181.
- Kämäri J. 1988. Regional lake acidification: Sensitivity and dynamics. Doctoral thesis, Department of Limnology, Faculty of Agriculture and Forestry, University of Helsinki, Helsinki, Finland.
- Kämäri J. (ed.) 1990. *Impact models to assess regional acidification*. International Institute for Applied Systems Analysis, Kluwer Academic Publishers, Dordrecht, The Netherlands, 310 pp.
- Kämäri J., Amann M., Brodin Y.-W., Chadwick M., Henriksen A., Hettelingh J.-P., Kuylenstierna J., Posch M. & Sverdrup H. 1992a. The use of critical loads for the assessment of future alternatives to acidification. *Ambio* 21(5): 377–386.
- Kämäri J., Brakke D., Jenkins A., Norton S.A. & Wright R.F. (eds.) 1989. *Regional acidification models: Geographic extent and time development*, Springer-Verlag, Berlin Heidelberg, 306 pp.
- Kämäri J., Forsius M., Johansson M. & Posch M. 1992b. Happamoittavan laskeuman kriittinen kuormitus Suomessa (Critical loads for acidifying deposition in Finland), Ministry of the Environment, Helsinki, Finland, *Report* 111/1992, 59 pp. (In Finnish with English summary and figure captions.)
- Kämäri J., Forsius M. & Posch M. 1993. Critical loads of sulphur and nitrogen for lakes II: Regional extent and variability in Finland. *Water, Air and Soil Pollution* 66: 77–96.
- Kämäri J., Forsius M., Posch M., Johansson M., Kauppi P. & Savolainen I. 1991. Critical concentrations and loads. In: Kenttämies, K. (ed), *op.cit.*, pp. 36–38.
- Kämäri J., Posch M., Kähkönen A.-M. & Johansson M. 1995. Modeling potential long-term responses of a small catchment in Lapland to changes in sulfur deposition. *Science of the Total Environment* 160/161: 687–701.
- Kangas L., Junto S., Laurila T. & Nordlund G. 1994. Comparison of EMEP model predictions and observations in Finland. Helsinki, Finland, *Publications on Air Quality* 19, 59 pp.
- Kauppi P., Anttila P., Karjalainen-Balk L., Kenttämies K., Kämäri J. & Savolainen I. (eds.) 1990a. Happamoittuminen Suomessa. HAPRO:n loppuraportti (Acidification in Finland. Report of the HAPRO programme), Valtion painatuskeskus, Helsinki, Finland, *Ympäristöministeriön ympäristönsuojeluosaston sarja A/89/1990*, 89 pp. (In Finnish, translated in English: Kenttämies 1991.)
- Kauppi P., Anttila P. & Kenttämies K. (eds.) 1990b. *Acidification in Finland*, Springer-Verlag, Berlin Heidelberg, 1237 pp.
- Kauppi P., Kämäri J., Posch M., Kauppi L. & Matzner E. 1986. Acidification of forest soils: Model development for analyzing impacts of acidic deposition in Europe. *Ecological Modelling* 33: 231–253.

- Kenttämies K. (ed.) 1991. *Acidification research in Finland*, Review of the results of the Finnish Acidification Research Programme (HAPRO) 1985–1990, Ministry of the Environment, Environmental Protection Department, Governmental Printing Centre, Helsinki, Finland, *Brochure 39/1991*, 48 pp.
- Kivivasara J. 1994. *Eestin päästökartoitus vuodelle 1991 – päästömallin kehittäminen* (An emission inventory in Estonia for year 1991 – construction of the emission model), M.Sc. thesis, Helsinki University of Technology, Espoo, Finland, 129 pp. (In Finnish with English abstract.)
- Kleemola S. & Forsius M. (eds.) 1998. 7<sup>th</sup> annual report 1998. UN-ECE, CLRTAP, International Co-operative Programme – Integrated Monitoring, Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment 217*, 44 pp.
- Koljonen T. (ed.) 1992. *The Geochemical Atlas of Finland. Part 2: Till*, Geological Survey of Finland, Espoo, Finland, 218 pp.
- Konikow L.F. & Bredehoeft J.D. 1992. Ground-water models cannot be validated. *Advances in Water Resources 15*(1): 75–83.
- Kros J., De Vries W., Janssen P.M.H. & Bak C.I. 1993. The uncertainty in forecasting trends of forest soil acidification. *Water, Air and Soil Pollution 66*: 29–58.
- Kros J., Groenenberg J.E., De Vries W., & Van Der Salm C. 1995. Uncertainties in long-term predictions of forest soil acidification due to neglecting seasonal variability. *Water, Air and Soil Pollution 79*: 353–375.
- Kulmala A. 1989. Sulphur dioxide emissions in the northwestern regions of USSR. Finnish Meteorological Institute, Helsinki, Finland, *Interim Report*.
- Kurz D., Alveteg M. & Sverdrup H. 1998. Acidification of Swiss forest soils. Development of a regional dynamic assessment. Swiss Agency for the Environment, Forests and Landscape, Berne, Switzerland, *Environmental Documentation, Air/Forests 89*.
- Kuusela, K. 1977. Suomen metsien kasvu ja puutavaralajirakenne sekä niiden alueellisuus vuosina 1970–1976. (Increment and timber assortment structure and their regionality of the forests of Finland in 1970–1976), Finnish Forest Research Institute, Helsinki, Finland, *Folia Forestalia 320*, 31 pp. (In Finnish with English summary.)
- Kuylenstierna J. & Chadwick M. 1994. Relationships between forest damage and acidic deposition in Europe. Report for the ITE/ETSU/EC study, Stockholm Environment Institute, 57 pp.
- Leinonen L. & Junto S. 1991. *Results of air quality at background stations, July–December 1990*, Finnish Meteorological Institute, Helsinki, Finland.
- Lepistö, A. 1995. Increased leaching of nitrate at two forested catchments in Finland over a period of 25 years. *Journal of Hydrology 171*: 103–123.
- Leppäjärvi R. (ed.) 1987. *Hydrological Yearbook 1981–1983. Publications of the Water Research Institute Finland 66*, 238 pp.
- Løkke H., Bak J., Falkengren-Grerup U., Finlay R.D., Ilvesniemi H., Nygaard P.H. & Starr M. 1996. Critical loads of acidic deposition for forest soils: Is the current approach adequate? *Ambio 25*(8): 510–516.
- Mälkönen E. 1975. Annual primary production and nutrient cycle in some Scots pine stands. Helsinki, Finland, *Communicationes Instituti Forestalis Fenniae 84*, 87 pp.
- Mannio J. & Vuorenmaa J. 1995. Regional monitoring of lake acidification in Finland. *Water, Air and Soil Pollution 85*: 571–576.
- Matzner E. & Murach D. 1995. Soil changes induced by air pollutant deposition and their implication for forests in Central Europe. *Water, Air and Soil Pollution 85*: 63–76.
- Meiwes K.J., Khanna P.K. & Ulrich B. 1986. Parameters for describing soil acidification and their relevance to the stability of forest ecosystems. *Forest Ecology and Management 15*: 161–179.
- Melanen M. & M. Ekqvist (eds.) 1997. Suomen ilmapäästöt ja niiden skenaariot, SIPS-projekti, Tietojärjestelmän tietopohja ja alustavia tuloksia (Finland's air emissions and their scenarios, First report of the SIPS project), Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment 131*, 48 pp. (In Finnish with English abstract).
- Melkerud P.-A., Olsson M. & Rosén K. 1992. Geochemical atlas of Swedish forest soils. Swedish University of Agricultural Sciences, Uppsala, Sweden, *Reports in Forest Ecology and Forest Soils 65*, 85 pp.
- Miinusmaa H. 1990. Elektrolyyttisistä kylvivistä. In: Äpy L. (ed.), *Kootut Äpyt*, WSOY, Porvoo, Finland, pp. 188–189. (In Finnish.)
- Müller-Edzards C., De Vries W. & Erisman J.W. 1997. Ten years of monitoring forest condition in Europe. Studies on temporal development, spatial distribution and impacts of natural and anthropogenic stress factors. Technical background report. United Nations Economic Commission for Europe, European Commission, 386 pp.
- Mylona S. 1993. Trends of sulphur dioxide emissions, air concentrations and depositions of sulphur in Europe since 1880. Meteorological Synthesizing

- Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MSC-W Report 2/93*, 35 pp.+appendices.
- Mylona S. 1996. Sulphur dioxide emissions in Europe 1880–1991 and their effect on sulphur concentrations and depositions. *Tellus* 48B: 662–689.
- NAPAP 1991. National acid precipitation assessment program, 1990 integrated assessment report. The U.S. national acid precipitation program, the NAPAP Office of the Director, Washington, D.C., 520 pp.
- Nelleman C. & Frogner T. 1994. Spatial patterns of spruce defoliation: Relation to acid deposition, critical loads, and natural growth conditions in Norway. *Ambio* 23(4–5): 255–259.
- Nilsson J. & Grennfelt P. (eds.) 1988. *Critical loads for sulphur and nitrogen*, Report from a workshop, 19–24 March 1988, Skokloster, Sweden, Nordic Council of Ministers, *Miljørapport* 1988:15.
- Niskanen R., Keränen S. & Pipatti R. 1990. Ammonia emissions in the 1980s. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 31–39.
- Nissinen A. & Ilvesniemi H. 1990. Effects of acid deposition on exchangeable cations, acidity and aluminium solubility in forest soils and soil solution. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 287–304.
- NMI 1995. Nordiske Miljøindikatorer, Høringsutkast. Utarbeidet for Nordisk miljøovervåkings- og data-gruppe. Nordisk Ministerråd, 93 pp.+appendices.
- Nöjd P. 1990. Detecting forest growth responses to environmental changes – a review of Finnish studies. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 507–522.
- Nordlund G., Lumme E., Pietarinen M. & Tuovinen J.-P. 1985. Laskentamenetelmä rikin laskeumaosuusien erittämiseksi (Calculation method for estimating sulphur deposition). Finnish Meteorological Institute, Helsinki, Finland, *Report* 115. (In Finnish).
- Nordlund G. & Tuovinen J.-P. 1988. Modelling long-term averages of sulphur deposition on a regional scale. In: *Proceedings of the WMO conference on air pollution modelling and its applications (vol. III)*, 19–24 May 1986, Leningrad USSR. Technical report WMO/TD 187, pp. 53–61.
- Ojansuu R. & Henttonen H. 1983. Kuukauden keskilämpötilan, lämpösumman ja sademäärän paikallisten arvojen johtaminen Ilmatieteen laitoksen mittautiedoista (Estimation of local values of monthly mean temperature, effective temperature sum and precipitation sum from the measurements made by the Finnish Meteorological Office), *Silva Fennica* 17(2): 143–160. (In Finnish with English summary.)
- Olsson M. & Melkerud, P.-A. 1991. Determination of weathering rates based on geochemical properties of the soil. In: Pulkkinen, E. (ed.) *op.cit.*, pp. 69–78.
- Olsson M., Rosén K. & Melkerud P.-A. 1993. Regional modelling of base cation losses from Swedish forest soils due to whole-tree harvesting. *Applied Geochemistry*, Special Issue 2: 189–194.
- Oreskes N., Shrader-Frechette K. & Belitz K. 1994. Verification, validation, and confirmation of numerical models in the earth sciences. *Science* 263: 641–646.
- Paces T. 1985. Sources of acidification in Central Europe estimated from elemental budgets in small basins. *Nature* 315: 31–36.
- Pingoud K., Mäilä H., Wihersaari M., Hongisto M., Siitonen S., Lehtilä A., Johansson M., Pirilä P. and Otterström T. 1999. ExternE national implementation Finland. Technical Research Centre of Finland, Espoo, Finland, *VTT Publications* 381.
- Posch M. 1998. Averaging and other simplifications in dynamic soil models. In: *Report of the workshop on data analysis for modelling and assessment of biogeochemical effects of air pollution in temperate ecosystems*, 8–11 Oct 97, CIEMAT, Madrid, pp. 61–67.
- Posch M., De Smet P.A.M., Hettelingh J.-P. & Downing (eds.) R.J. 1995. *Calculation and mapping of critical thresholds in Europe, status report 1995*. Coordination Center for Effects, RIVM, Bilthoven, The Netherlands, iv+198 pp.
- Posch M. & De Vries, W. in press. Derivation of critical loads by steady-state and dynamic soil models. In: Langan S.J. (ed.), *The impact of nitrogen deposition on natural and semi-natural ecosystems*. Kluwer.
- Posch M., Falkengren-Grerup U. & Kauppi P. 1989a. Application of two soil models to historical soil chemistry data from Sweden. In: Kämäri J., Brakke D., Jenkins A., Norton S.A. & Wright R.F. (eds.), *op.cit.*, pp. 241–252.
- Posch M., Forsius M. & Kämäri J. 1993a. Critical loads of sulphur and nitrogen for lakes I: Model description and estimation of uncertainty. *Water, Air and Soil Pollution* 66: 173–192.
- Posch M., Hettelingh J.-P., Alcamo J. & Krol M. 1996. Integrated scenarios of acidification and climate change in Asia and Europe. *Global Environmental Change* 6(4): 375–394.
- Posch M., Hettelingh J.-P., De Smet P.A.M. & Downing R.J. (eds.) 1997a. *Calculation and mapping of critical thresholds in Europe, status report 1997*. Coordination Center for Effects, RIVM,

- Bilthoven, The Netherlands, iv+164 pp.
- Posch M., Johansson M. & Forsius M. 1997b. Critical loads and dynamic models. In: Kleemola S. & Forsius M. (eds.), *6<sup>th</sup> annual report 1997*, UNECE, CLRTAP, International Cooperative Programme - Integrated Monitoring. Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment* 116, pp. 13–23.
- Posch M., Kauppi L. & Kämäri J. 1985. Sensitivity analysis of a regional scale soil acidification model. International Institute for Applied Systems Analysis, Laxenburg, Austria, *IIASA Collaborative paper* CP-85-45, 33 pp.
- Posch M., Kauppi L. & Kämäri J. 1989b. Sensitivity analysis of a regional scale soil acidification model. *Syst.Anal.Model.Simul.* 6: 451–473.
- Posch M., Kämäri J., Forsius M., Henriksen A. & Wilander A. 1997c. Exceedance of critical loads for lakes in Finland, Norway, and Sweden: Reduction requirements for acidifying nitrogen and sulfur deposition. *Environmental Management* 21(2): 291–304.
- Posch M., Kämäri J., Johansson M. & Forsius M. 1993b. Displaying inter- and intra-regional variability of large-scale survey results. *Environmetrics* 4(3): 341–352.
- Posch M., Reinds G.J. & De Vries W. 1993c. SMART - A simulation model for acidification's regional trends: Model description and user manual. National Board of Waters and the Environment, Helsinki, Finland, *Mimeograph Series* 477, 43 pp.
- Press, W.H., Flannery, B.P., Teukolsky, S.A. & Vetterling, W.T. 1986. *Numerical recipes. The art of scientific computing*. New York, Cambridge University Press, 818 pp.
- Pulkkinen E. (ed.) 1991. *Environmental geochemistry in northern Europe*. Geological Survey of Finland, Espoo, Finland, *Special Paper* 9, 321 pp.
- RAINS 1998. The Regional Acidification Information and Simulation model, version 7.2, International Institute for Applied Systems Analysis, Laxenburg, Austria.
- Rask M., Mannio J., Forsius M., Posch M. & Vuorinen P.J. 1995. How many fish populations in Finland are affected by acid precipitation? *Environmental Biology of Fishes* 45: 51–63.
- Rasmussen L., Hultberg H. & Cosby B.J. 1995. Experimental studies and modelling of enhanced acidification and recovery. *Water, Air and Soil Pollution* 85: 89–99.
- Reuss J.O., Cosby B.J. & Wright R.F. 1987. Chemical processes governing soil and water acidification. *Nature* 329: 27–32.
- Rodhe H., Grennfelt P., Wisniewski, J., Ågren C., Bengtsson G., Johansson K., Kauppi P., Kucera V., Rasmussen L., Rosseland B., Schotte L. & Selldén G. 1995. Acid Reign '95? - Conference summary statement. *Water, Air and Soil Pollution* 85: 1–14.
- Rosén K. 1982. Supply, loss and distribution of nutrients in three coniferous forest watersheds in central Sweden. Swedish University of Agricultural Sciences, Uppsala, Sweden, *Reports in Forest Ecology and Forest Soils* 41.
- Rosén K. 1988. Effects of biomass accumulation and forestry on nitrogen in forest ecosystems. In: Nilsson J. & Grennfelt P. (eds.), *op.cit.*, pp. 269–293.
- Rosén K., Gundersen, P., Tegnhammar, L., Johansson, M. & Frogner, T. 1992. Nitrogen enrichment of Nordic forest ecosystems. *Ambio* 21(5): 364–368.
- Rubin E.S. 1991. Benefit-cost implications of acid rain controls: An evaluation of the NAPAP integrated assessment. *J. Air Waste Manage. Assoc.* 41(7) 914–921.
- Saltbones J. & Dovland H. 1986. Emissions of sulphur dioxide in Europe in 1980 and 1983. Norwegian Institute for Air Research (NILU), Lillestrøm, Norway, *EMEP/CCC Report* 1/86, 32 pp.
- Sandnes H. & Styve H. 1992. Calculated budgets for airborne acidifying components in Europe, 1985, 1987, 1988, 1989, 1990 and 1991. Meteorological Synthesizing Centre West, The Norwegian Meteorological Institute, Oslo, Norway, *EMEP/MSW Report* 1/92.
- Savolainen I. & Tähtinen M. 1990. Sulphur dioxide and nitrogen oxide emission scenarios to 2040. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 1–19.
- Savonen K. & Johansson M. 1996. Rikki- ja typpilaskeuman kriittinen kuormitus Hämeessä 1990 (Critical loads for sulphur and nitrogen deposition in 1990 in the Province of Häme, southern Finland), Regional Environmental Agency of Häme, Hämeenlinna, Finland, *Regional Environmental Publications* 8, 26 pp. (In Finnish with English abstract.)
- Skelly M.J. & Innes J.L. 1994. Waldsterben in the forests of central Europe and eastern North America: Fantasy or reality? *Plant Disease* 78(11): 1021–1032.
- Skjelkvåle B.L. & Wright R.F. 1998. Mountain lakes; Sensitivity to acid deposition and global climate change. *Ambio* 27(4): 280–286.
- Söderberg U. 1991. The relation between increment and defoliation for Scots pine and Norway spruce in Sweden. IUFRO and ICP-Forest workshop on monitoring air pollution impact on permanent

- sample plots, data processing and results interpretation, 2–6 Sep 1991, Prahaticce, CSFR, pp. 119–127.
- Sommaruga-Wögrath S., Koinig K.A., Schmidt R., Sommaruga R., Tessadri R. & Psenner R. 1997. Temperature effects on the acidity of remote alpine lakes. *Nature* 397: 64–67.
- Starr M., Lindroos A.-J., Tarvainen T. & Tanskanen H. 1998. Weathering rates in the Hietajärvi Integrated Monitoring catchment. *Boreal Environment Research* 3: 275–285.
- Stoddard J.L., Driscoll C.T., Kahl J.S. & Kellogg J.H. 1998. Can site-specific trends be extrapolated to a region? An acidification example from the Northeast. *Ecological Applications* 8(2): 288–299.
- Sulphur Committee II 1993. Report of the Sulphur Committee II. Ministry of the Environment, Helsinki, Finland, *Committee report* 1993:6, 211 pp. (In Finnish with English summary.)
- Suutari R., Johansson M. & Tarvainen T. 1999. Aineistojen alueellistaminen kriging-menetelmällä ympäristömallintamisessa (Regionalising data with kriging method in environmental modelling), Finnish Environment Institute, Edita Ltd, Helsinki, Finland, *The Finnish Environment* 268, 47 pp. (In Finnish with English abstract.)
- Sverdrup H. 1990. *The kinetics of base cation release due to chemical weathering*. Lund University Press, Lund, Sweden, 246 pp.
- Sverdrup H., De Vries W. & Henriksen A. 1990. Mapping critical loads: A guidance manual to criteria, calculations, data collection and mapping. Nordic Council of Ministers, Copenhagen, Denmark, *Miljørapport* 1990:14.
- Sverdrup H. & Warfvinge P. 1988. Assessment of critical loads of acid deposition on forest soils. In: Nilsson J. & Grennfelt P. (eds.), *op.cit.*, pp. 81–129.
- Sverdrup H. & Warfvinge P. 1993. The effect of soil acidification on the growth of trees, grass and herbs as expressed by the (Ca+Mg+K)/Al ratio. Department of Chemical Engineering II, Lund University, Lund, Sweden, *Reports in Ecology and Environmental Engineering* 2:1993, 177 pp.
- Sverdrup H., Warfvinge P., Blake L. & Goulding K. 1995. Modelling recent and historic soil data from the Rothamsted experimental station, UK, using SAFE. *Agriculture Ecosystems and Environment* 53: 161–177.
- Syri S., Johansson M., Grönroos J. & Ekqvist M. in press. Assessing the effects of domestic and international energy scenarios and emission reduction strategies on acidification in Finland. *Environmental Modeling & Assessment*.
- Tähtinen M., Lehtilä A., Pipatti R., Wistbacka M. & Savolainen I. 1997. Economic reduction of acidifying deposition in Finland by decreasing emissions in Finland, Estonia and Russia. *The Science of the Total Environment* 204: 177–192.
- Tamm C.O. & Hallbäck L. 1988. Changes in soil acidity in two forest areas with different acid deposition: 1920s to 1980s. *Ambio* 17(1): 56–61.
- Tamminen P. & Starr M. 1990. A survey of forest soil properties related to soil acidification in southern Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *op.cit.*, pp. 235–251.
- Tarvainen T. 1995. The geochemical correlation between coarse and fine fractions of till in southern Finland. *J Geochem. Exploration* 54: 187–198.
- Tuovinen J.-P., Laurila T., Lättilä, H., Ryaboshapko, A., Brukhanov, P. & Korolev, S. 1993. Impact of the sulphur dioxide sources in the Kola peninsula on air quality in northernmost Europe. *Atmospheric Environment* 27: 1379–1395.
- UBA 1996. Manual on methodologies and criteria for mapping critical loads/levels and geographical areas where they are exceeded. Texte 71-96, Umweltbundesamt, Berlin, Germany, 144+lxiv pp.
- Ulrich B. 1983. Soil acidity and its relations to acid deposition. In: Ulrich B. & Pankrath J. (eds.) *Effects of accumulation of air pollutants in forest ecosystems*. Proc. Workshop. Göttingen, FRG, May 16–19, 1982. D. Reidel Publishing Company, Dordrecht, The Netherlands, pp. 127–146.
- UN-ECE/SP2 1994. Protocol to the 1979 Convention on Long-range Transboundary Air Pollution on further reductions of sulphur emissions. Document ECE/EB.AIR/40, New York and Geneva, 106 pp.
- UN-ECE 1996. 1979 Convention on long-range transboundary air pollution and its protocols. United Nations Economic Commission for Europe, New York and Geneva.
- UN-ECE 1997. Anthropogenic emissions of air pollutants (1980–2010) in the ECE region. EB.AIR/GE.1/1997/3. United Nations Economic Commission for Europe, Geneva, Switzerland.
- UNESCO 1972. International geological map of Europe and the Mediterranean region. Bundesanstalt für Bodenforschung, Hannover and Unesco, Paris.
- Vuokila Y. 1987. *Metsänkasvatuksen perusteet ja menetelmät* (Fundamentals and methods of forestry), WSOY, Porvoo, Finland. (In Finnish.)
- Warfvinge P., Holmberg M., Posch M. & Wright R.F. 1992. The use of dynamic models to set target loads. *Ambio* 21(5): 369–376.
- Warfvinge P. & Sverdrup H. 1992. Calculating critical loads at acid deposition with PROFILE - a steady state soil chemistry model. *Water, Air and Soil Pollution* 63: 119–143.

- Watanabe T. & Ellis H. 1993. Robustness in stochastic programming models. *Applied Mathematical Modelling* 17: 547–554.
- Westman C.J. 1981. Fertility of surface peat in relation to the site type and potential stand growth. Society of Forestry in Finland, Helsinki, Finland, *Acta Forestalia Fennica* 172, 77 pp.
- Wright R.F., Cosby R.J., Hornberger G.M. & Galloway, J.N. 1986. Comparison of paleolimnological with MAGIC model reconstructions of water acidification. *Water, Air and Soil Pollution* 30: 367–380.
- Wright R.F. & Hauhs M. 1991. Reversibility of acidification: Soils and surface waters. In: Last F.T & Watling R. (eds.), *Acidic deposition: Its nature and impacts*, The Royal Society of Edinburgh, pp. 169–191.
- Wright R.F. & Schindler W. 1995. Interaction of acid rain and global changes: Effects on terrestrial and aquatic ecosystems. *Water, Air and Soil Pollution* 85: 89–99.

## ANNEX A. Formulation of the steady-state mass balance (SMB) method for forest soils

### Total acidity

The equations currently used in the critical load mappings are presented in the following due to their rapid development and changes during this study and, in addition, their overall importance. The equations here apply for forest soils only, those for the lakes differ in some respects (Posch *et al.* 1997c). Critical loads of acidity of sulfur and nitrogen are derived from mass and acidity balance considerations. The following acidity balance is assumed for the sum of sulfur and nitrogen depositions (Kämäri *et al.* 1992c, Posch *et al.* 1995, 1997a, 1997c):

$$S_{dep} + N_{dep} = N_{nu} + N_{im} + N_{den} + BC_{le} - Alk_{le} \quad (A1)$$

where  $S_{dep}$  = the total sulfur deposition to forest,  $N_{dep}$  = the total nitrogen deposition to forest soil,  $N_{nu}$  = nitrogen net uptake by growth,  $N_{im}$  = the immobilization of nitrogen in soils,  $N_{den}$  = nitrogen denitrified in soils,  $BC_{le}$  = the base cation leaching and  $Alk_{le}$  = the alkalinity (acid neutralizing capacity) leaching. Base cation leaching is given by

$$BC_{le} = BC_{dep}^* - Cl_{dep}^* + BC_{we} - BC_{nu} \quad (A2)$$

where  $BC_{dep}^*$  = the sea-salt corrected base cation deposition,  $BC_{we}$  = the base cation weathering of soil minerals,  $BC_{nu}$  are the net growth uptake of base cations. Since the interest is in the anthropogenic emissions, from which the atmospheric transport models normally calculate acidifying depositions, the non sea-salt deposition of physiologically active base cations should be used in exceedances of critical loads. They are calculated by using Na as tracer, and  $BC_{dep}^* = Bc_{dep}^* = Ca_{dep}^* + Mg_{dep}^* + K_{dep}^*$ . Consequently, throughout the text it is implicitly assumed that  $S_{dep} = S_{dep}^*$ . Note, that  $BC_{nu} = Bc_{nu}$  and in Finland  $BC_{we} = (Ca + Mg)_{we}$ .

It is required that  $Bc_{dep}^* - Cl_{dep}^* + BC_{we} \geq BC_{nu}$ . In addition, a limiting concentration, below which trees can no longer extract nutrients from soil solution, is set to a precautionary value of  $[Bc]_{min} = 2 \mu\text{eq l}^{-1}$  (UBA 1996, Posch *et al.* 1995). If necessary the weathering rate, which is considered the least certain of the inputs for  $BC_{le}$ , is corrected to meet the requirement  $BC_{le,min} = Q [Bc]_{min}$ , where  $Q$  = runoff.

The alkalinity leaching is

$$Alk_{le} = HCO_{3,le} + RCOO_{le} - H_{le} - Al_{le} \quad (A3)$$

where RCOO is the sum of organic anions,  $H_{le}$  = hydrogen ion leaching and  $Al_{le}$  = aluminum ion leaching.

The critical aluminum leaching is defined with the threshold criterion of harmful effects on tree root functioning, critical molar ratio of base cations to aluminum in soil solution, and calculated by

$$Al_{le,crit} = (Bc:Al)_{crit} (Bc_{dep} + BC_{we} - BC_{nu}) \quad (A4)$$

where  $(Bc:Al)_{crit}$  = critical molar ratio of base cation concentration to aluminum concentration in soil solution,  $([Bc]_{molar} : [Al]_{molar})_{crit}$  of the tree rooting zone. The threshold value 1.0 is used to indicate risk for detrimental effects. Note, that here the base cation deposition  $Bc_{dep}$  instead of  $Bc_{dep}^*$  is used, since all base cations, except Na, contribute to the protection of plant roots against Al independent of their origin.

The critical hydrogen leaching is obtained solving

$$Al_{le,crit} / Q = K_{gibbs} (H_{le,crit} / Q)^3 \quad (A5)$$

where  $K_{gibbs}$  = the gibbsite equilibrium constant (the value  $10^{8.3} \text{ mol}^{-2} \text{ l}^2$  has been used in Finland). Using the critical leaching values for hydrogen and aluminum we get for critical alkalinity leaching for acid forest soils, where  $HCO_{3,le}$  and  $RCOO_{le}$  can be neglected,

$$Alk_{le,crit} = -H_{le,crit} - Al_{le,crit} \quad (A6)$$



When the critical alkalinity leaching has been specified, Eq. A1 can be written as

$$CL(S+N) = CL(S) + CL(N) = N_{nu} + N_{im} + N_{den} + BC_{le} - Alk_{le,crit} \quad (A7)$$

The net input of nitrogen deposition, left after net uptake and immobilization, independent of its form, is assumed to acidify the forest soil.  $N_{den}$  can be described as a fixed value or modeled dependent on the net input of nitrogen into the ecosystem. If the dependency is linear, as is assumed in applications in Finland, we get

$$N_{den} = f_{den} (N_{dep} - N_{nu} - N_{im}) \quad (A8)$$

where  $N_{dep} < N_{nu} + N_{im}$  (otherwise,  $N_{den} = 0$ ) and  $f_{den}$  is the fraction of denitrified nitrogen, smaller than or equal to one and dependent on ecosystem properties ( $0.1+0.7 \cdot$  fraction of peat land). We then have

$$S_{dep} + (1 - f_{den}) N_{dep} = (1 - f_{den}) (N_{nu} + N_{im}) + BC_{le} - Alk_{le} \quad (A9)$$

It can be seen from this equation, that when  $N_{dep} = N_{nu} + N_{im}$  the nitrogen fluxes do not contribute to acidification.

The maximum critical load for sulfur, when nitrogen sinks cannot compensate deposited sulfur acidity, is

$$CL_{max}(S) = BC_{le} - Alk_{le,crit} \quad (A10)$$

If all deposited nitrogen is consumed by the nitrogen sinks in the soil and there is no excess nitrogen to contribute to acidification, then the threshold value is called the minimum critical load for nitrogen

$$N_{dep} \leq N_{nu} + N_{im} = CL_{min}(N) \quad (A11)$$

When  $N_{dep} \leq CL_{min}(N)$ , the maximum allowable deposition for sulfur is defined with  $CL_{max}(S)$ . The maximum allowable nitrogen deposition, in case of zero sulfur deposition, is solved from Eq. A7

$$CL_{max}(N) = N_{nu} + N_{im} + (BC_{le} - Alk_{le}) / (1 - f_{den}) = CL_{min}(N) + CL_{max}(S) / (1 - f_{den}) \quad (A12)$$

#### The critical load function

Both sulfur and nitrogen depositions can contribute to the acidification of an ecosystem, and therefore no unique critical loads of sulfur or acidifying nitrogen can be specified. These critical loads are rather characterized by three quantities:  $CL_{max}(S)$ ,  $CL_{min}(N)$  and  $CL_{max}(N)$ . They together define the critical load function shown in Fig. A1. In case the eutrophication effects are included, the  $CL_{nur}(N)$  line may or may not vertically cut through the area for acidification effect. For every pair of deposition ( $S_{dep}$ ,  $N_{dep}$ ) lying on the function or below it (the grey-shaded area) neither critical loads of sulfur or nitrogen are exceeded. As illustrated by the point E and paths to points Z1, Z2 and Z3, the non-exceedance can be reached by reducing a single component or a combination. It is impossible to define a unique exceedance in the sense of quantifying the amount of sulfur and nitrogen to be reduced for non-exceedance. In practice external factors such as the costs of emission reduction measures of either sulfur or nitrogen compounds will determine the pathway to non-exceedance.

#### Conditional critical loads of sulfur and nitrogen

If one is interested in reductions of only one of the two pollutants, a unique critical load can be derived which is consistent with the presented formulations. If emission reductions deal with sulfur only, a unique critical load of sulfur for a fixed nitrogen deposition can be derived from the critical load function. This conditional critical load of nitrogen,  $CL(S|N_{dep})$ , is

$$CL(S|N_{dep}) = \begin{cases} 0, & \text{if } N_{dep} \geq CL_{max}(N) \\ [CL_{max}(N) - N_{dep}] / \alpha, & \text{if } CL_{min}(N) < N_{dep} < CL_{max}(N) \\ CL_{max}(S), & \text{if } N_{dep} \leq CL_{min}(N) \end{cases} \quad (A13)$$

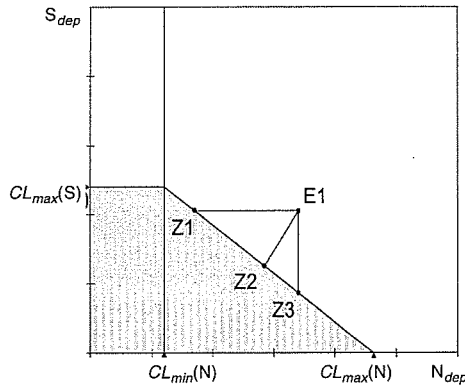


Fig. A1. Critical load function for sulfur and acidifying nitrogen (redrawn from Posch *et al.* 1997a).

where  $\alpha = [CL_{max}(N) - CL_{min}(N)] / CL_{max}(S)$ . In an analogous manner a conditional critical load of nitrogen,  $CL(N|S_{dep})$ , for a fixed sulfur deposition  $S_{dep}$  can be computed as

$$CL(N|S_{dep}) = \begin{cases} CL_{min}(N), & \text{if } S_{dep} \geq CL_{max}(S) \\ CL_{max}(N) - \alpha S_{dep}, & \text{if } S_{dep} < CL_{max}(S) \end{cases} \quad (A14)$$

#### Exceedances of critical loads

The exceedance of critical load of acidifying total deposition is the sum of sulfur and nitrogen depositions minus the critical load given by Eq. A9

$$Ex(S_{dep}, N_{dep}) = (1 - f_{den}) N_{dep} + S_{dep} - [(1 - f_{den})(N_{nu} + N_{im}) + BC_{le} - Alk_{le}] \quad (A15)$$

The critical load is not exceeded, when the acidity balance of Eq. A15 is non-positive for a given pair of deposition of sulfur and nitrogen. There are many combinations of S and N depositions which fulfill the non-exceedance requirement, and thus a unique critical load value cannot be defined. To examine exceedances analytically, the critical load function has been defined. At this line, the non-exceedance is achieved. Using conditional critical loads, the exceedances are obtained simply by subtracting the conditional critical loads from the deposition.

#### Historical definition of sulfur-induced acidity

The critical load of acidity as defined above was not directly applicable to sulfur reduction negotiations preceding the signing of the 1994 Sulphur Protocol. A fraction of the allowable load was assigned to sulfur using the assumptions, that (i) the fraction of actual sulfur deposition of the total acidifying deposition is used to separate the fraction of critical load for sulfur, and (ii) total nitrogen deposition contributes to acidification when not taken up or immobilized. This leads to the sulfur factor  $\sigma$

$$\sigma = S_{dep} / (S_{dep} + N_{dep} - N_{nu} - N_{im}) \quad (A16)$$

At the time of applying this concept, the nitrogen denitrification process was not considered of importance, and thus it did not enter the calculations. Using the sulfur factor, we get the critical deposition of sulfur and nitrogen

$$CD(S) = \sigma CL_{max}(S) \quad (A17)$$

$$CD(N) = N_{nu} + N_{im} + (1 - \sigma) CL_{max}(S) \quad (A18)$$

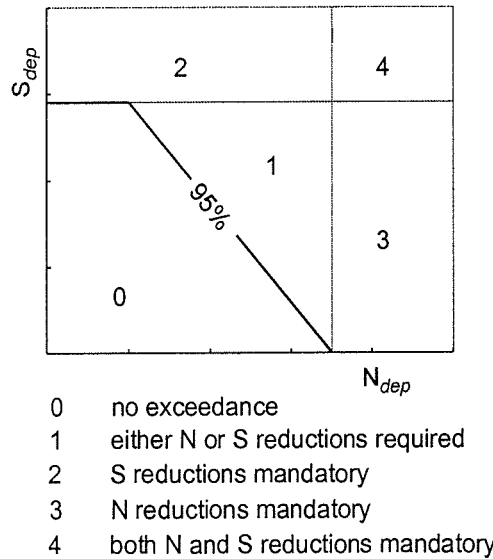
The calculation of  $CD(S+N)$  is trivial. The exceedance of the critical deposition of sulfur and nitrogen from Eqs. A17–A18 is simply

$$Ex(S) = S_{dep} - CD(S) \tag{A19}$$

$$Ex(N) = N_{dep} - CD(N) \tag{A20}$$

*Deposition reduction requirements*

The necessary reductions for excess sulfur and nitrogen depositions can be determined with the help of Fig. A1 and A2. Fig. A1 shows the critical load function characterized by three quantities  $CL_{max}(S)$ ,  $CL_{min}(N)$  and  $CL_{max}(N)$  derived from ecosystem properties. When a deposition pair of sulfur and nitrogen lies on this function, the critical load is not exceeded. When comparing different combinations of the sulfur and nitrogen depositions five cases can arise as shown in Fig. A2: 0) the deposition pair lies below the critical load function and there is no exceedance, 1) reductions of  $S_{dep}$  and  $N_{dep}$  are interchangeable and non-exceedance can be reached by either  $S_{dep}$  or  $N_{dep}$  reduction alone, although both can also be used, 2) reductions of  $S_{dep}$  are mandatory and possibly leading to option 1, 3) reductions of  $N_{dep}$  are mandatory and possibly leading to option 1, and 4) reductions of both  $S_{dep}$  and  $N_{dep}$  are required, at least to some extent.



**Fig. A2.** Five cases for reduction requirements of exceedances of critical loads to achieve protection of 95% of ecosystems in each grid cell against acidification due to sulfur and nitrogen depositions (redrawn after Posch *et al.* 1997a).

When calculating information for a grid cell a representative critical load function or so-called protection isoline is constructed. There are several ways to achieve this. The points defining the critical load function can be derived as the 5<sup>th</sup> percentile of all respective ecosystem values in the cell. This simple approach has been used for producing Fig. 12 with Finnish data. Alternatively, all individual critical load functions can be handled simultaneously and calculate the 5<sup>th</sup> percentile isoline at different angles of the quadrant. The resulting isoline is not necessarily convex but always monotonously decreasing. The major difference to the previous approach is a more accurate and correct treatment between cases 0 and 1. This approach has been used by the Coordination Center for Effects in creating European maps for reduction requirements and producing so-called protection isolines for emission reduction optimization purposes of the IAM groups (Posch *et al.* 1997a).



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#### Book chapters:

Tamminen P. & Starr M. 1990. A survey of forest soil properties related to soil acidification in Southern Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 237–251.

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