CRITICAL LOADS OF SULFUR AND NITROGEN FOR LAKES II: REGIONAL EXTENT AND VARIABILITY IN FINLAND

JUHA KÄMÄRI, MARTIN FORSIUS and MAXIMILIAN POSCH
Water and Environment Research Institute, P.O. Box 250, SF-00101 Helsinki, Finland

(Received October 3, 1991; revised January 20, 1992)

Abstract. The concept of critical loads has been developed to assist in the design of environmentally sound abatement strategies for the emissions of acidifying compounds. In this paper the critical loads of S and N for lakes in Finland are computed and mapped, based on methods presented in an accompanying paper. The employed steady-state mass balance model allows the simultaneous evaluation of the reductions required of S and N deposition exceeding these critical loads. Special emphasis has been put on the presentation of the spatial variability and the uncertainty of the critical loads and their exceedances. The derived critical loads of S and N for lakes in Finland show a substantial spatial variability. The highest exceedance of critical loads is presently estimated in the south-east of the country, where up to 80% of the lakes show an exceedance of the critical loads of S. The evaluation of two emission scenarios shows that only "maximum feasible reductions" would be sufficient for protecting most Finnish lakes from the impacts of acidic deposition. The results of this study form a basis for setting national targets for emission reductions in Finland.

1. Introduction

Due to the increasing concern over the effects of anthropogenic air pollutant depositions in large areas of the northern hemisphere, governments are looking for scientific means of planning effective and efficient emission controls. The critical load concept has been found valuable in the context of developing and implementing future control strategies for transboundary air pollutants. The critical load for atmospheric inputs of S and N acidity to surface waters has been defined in a variety of ways at different scientific and administrative workshops. Dose-response relationships form the basis for the definitions, the critical load being exceeded when the load causes harmful effects to the receptor. In the most widely used definition a critical load is 'a quantitative estimate of the loading of one or more pollutants below which significant harmful effects on specified sensitive elements of the environment are not likely to occur according to present knowledge' (Nilsson and Grennfelt, 1988).

The purpose of determining critical loads is to set goals for future deposition rates of acidifying compounds such that the environment is protected. The critical load concept offers a basis for developing emission reduction strategies which are both internationally accepted and environmentally compatible. Critical loads have to be determined separately for different receptors, such as soils and lakes. The critical load for a particular receptor will vary from site to site, depending on its inherent sensitivity. Criteria for an 'unacceptable change' are set in relation to effects on terrestrial and aquatic indicator organisms. The goal should be to

Water, Air, and Soil Pollution **66**: 77–96, 1993.

protect the environment to such an extent that a critical chemical component or combination of components (e.g. ANC, pH, Al/Ca ratio) stay above or below a value that does not cause a harmful response in the selected biological indicator.

There is, unfortunately, no well defined water quality value above or below which an aquatic ecosystem can be said to be protected from the harmful effects of acidic deposition, such as decline and extinction of plant and animal populations. Acidic deposition and the consequent decline in surface water pH and alkalinity and the increase in inorganic Al is positively correlated to a reduced number of taxonomic groups of aquatic flora and fauna (e.g. Henriksen *et al.*, 1989; Schindler *et al.*, 1989; Kelso *et al.*, 1990). The definition of criteria for what might be considered as harmful is, however a matter of subjective evaluation, and society has to decide what it wants to protect.

Quantitative estimates of critical loads (e.g. Henriksen and Brakke, 1988; Henriksen *et al.*, 1990a; Sverdrup and Warfvinge, 1990; Forsius *et al.*, 1992), together with pollutant deposition values, can be used to identify areas where present deposition exceeds the critical load values. Coupling this information with atmospheric transport models enables one to determine how much and where emission reductions are required in order to reduce the deposition below the critical load (Alcamo *et al.*, 1990). Earlier assessments of critical loads for surface waters (Nilsson, 1986; Nilsson and Grennfelt, 1988) suggest that acidic deposition in various sensitive locations should be very low, less than 20 meq m^{-2} yr^{-1}, in order not to harm, for example, fish production. This means that the acidifying deposition should be reduced almost to background values in order to protect every sensitive stream and lake. In practise, such a reduction will not always be possible, and therefore decisions will have to be made about an 'acceptable level of damage'. A deposition value that might include some acceptable damage is related to political decisions and is refered to as a 'target load'. From the ecological point of view, a target load should be set equal to or lower than the critical load, however, for economic reasons it will probably be set higher in many cases. There is a major conceptual difference between a critical load and a target load. Scientific constructs, including models to calculate critical loads and time patterns of ecosystem responses, are considered as scientific hypotheses that are in many respects 'value free'. Standards, like target loads, should be derived from scientific evidence, but they also take into account human values and preferences.

In 1979 the United Nations Economic Commission for Europe (ECE) adopted the Convention of Long Range Transboundary Air Pollution under which protocols to control SO_2, NO_X and VOC emissions have been drawn up. An international agreement on reducing emissions of NO_X was signed in November 1989 in Sofia. This agreement was still based on flat rate emission reductions. However, the agreement does specify that subsequent international agreements should be based on generally accepted critical loads. A 'Task Force for Mapping the Critical Loads and the Areas where the Critical Loads are Exceeded' was set up after the Sofia meeting with the aim of mapping critical loads in the ECE-countries. This Task

Force compiled a manual with definitions, methods and instructions for the preparation of maps showing critical loads for airborne S and N (ECE, 1990), and a Coordination Centre for Effects oversees the production of harmonized maps of critical loads for Europe and North America. Although maps on such a large scale are an important step for multilateral negotiations, it is evident that mapping results on an European scale do not provide sufficient resolution for national assessments and determination of target loads.

The objective of this paper is to evaluate the critical loads of Finnish lakes, to assess the areas where critical loads are exceeded, and to provide estimates on how large reductions of N and/or S depositions are required in order to protect these ecosystems. To accomplish this, extensive research has been conducted for the development of methods for calculating critical loads of S and potentially acidifying N and for the collection and derivation of input data. These procedures are described in detail in an accompanying paper by Posch *et al.* (1992). In this paper, special emphasis also has been put on the presentation of critical loads, their spatial variability as well as the uncertainty in the results due to uncertain inputs and model parameters. This information on regional distributions of critical loads and on the amount they are exceeded at individual sites forms a basis for setting national targets for emission reductions in Finland.

2. Materials and Methods

2.1. Model description

The expressions for critical loads of S and potentially acidifying N compounds were derived from mass balance considerations (Posch *et al.*, 1992). In steady-state conditions, when no acidification of the system is permitted, the sources of alkalinity must balance the input of acidity. This implies that dynamic processes with a limited maximum capacity, such as adsorption/desorption of sulfate and cation exchange, are neglected in the derivation of long-term critical loads (see Sverdrup and Warfvinge, 1990; de Vries and Kros, 1991).

The critical load is determined by both terrestrial and in-lake processes. The critical load of non-marine S, $CL(S)$, is given by

$$CL(S) = f\mathrm{S}_{upt} + \sigma(1 + rs_S/Q)\mathrm{BC}_{le,crit} \tag{1}$$

with

$$\mathrm{BC}_{le,crit} = \mathrm{BC}^{\star}_{dep} + \mathrm{BC}_{w} - \mathrm{BC}_{upt} - \mathrm{Alk}_{crit} \tag{2}$$

where f is the fraction of forests in the catchment, S_{upt} is the uptake of S in the catchment, r is the lake:catchment area ratio , s_S is the in-lake net mass transfer coefficient for S and Q is the annual runoff. $\mathrm{BC}_{le,crit}$ is a critical original level of base cation leaching, $\mathrm{BC}^{\star}_{dep}$ is the base cation deposition (the star indicating the non-marine fraction), BC_w denotes the weathering of base cations, Alk_{crit} is the critical alkalinity leaching. The sulfur factor σ ($0 \leq \sigma \leq 1$) accounts for the fact that also the deposition of N compounds has to be balanced by base cations.

A similar equation was derived for potentially acidifying N deposition. Assuming that all deposited ammonium is completely taken up and/or nitrified, the critical load for N, $CL(N)$, is given by

$$CL(N) = f\mathrm{N}_{upt} + (1 - \sigma)(1 + rs_N/Q)\mathrm{BC}_{le,crit} \tag{3}$$

where N_{upt} is the net growth uptake of N in the catchment and s_N is the in-lake net mass transfer coefficient for nitrate.

It is assumed that the lake catchments have been in steady-state relative to deposition inputs during pre-acidification times. The quantity $\mathrm{BC}^{\star}_{dep} + \mathrm{BC}_w - \mathrm{BC}_{upt}$ can, therefore, be estimated by determining the pre-acidification leaching of base cations ($Q[\mathrm{BC}]^{\star}_0$) from the catchment area by employing an extension of a well-known steady-state model (Henriksen, 1984). In this model the change in the leaching of base cations (Ca, Mg, Na, K) is related to long-term changes in inputs of strong acid anions by the so-called F-factor, which describes the degree of watershed neutralization (Brakke *et al.*, 1990; Henriksen *et al.*, 1990a; Posch *et al.*, 1992):

$$[\mathrm{BC}]^{\star}_0 = [\mathrm{BC}]^{\star}_t - F([\mathrm{SO}_4^{2-}]^{\star}_t + [\mathrm{NO}_3^-]_t - [\mathrm{SO}_4^{2-}]^{\star}_0 - [\mathrm{NO}_3^-]_0), \tag{4}$$

where the subscriptions 0 and t refer to the original and present concentrations, respectively. The relationship between F and the original base cation concentration is described by a non-linear function:

$$F = 1 - \exp(-[\mathrm{BC}]^{\star}_0/B) \tag{5}$$

where B is a parameter to be estimated from available data.

Acid Neutralization Capacity (ANC), defined as non-marine base cations minus strong acid anions, appears to be a suitable chemical criterion for sensitive indicator organisms in surface waters (Lien *et al.*, 1989; Henriksen *et al.*, 1990a, b). The critical original level of base cation leaching for a selected indicator organism is therefore given by

$$\mathrm{BC}_{le,crit} = Q([\mathrm{BC}]^{\star}_0 - [\mathrm{ANC}]_{limit}) \tag{6}$$

where $[\mathrm{ANC}]_{limit}$ is the tolerance criterion for the aquatic organisms considered. This equation has been used earlier to calculate critical loads of S for lakes in Norway and in the whole of Fennoscandia (Henriksen *et al.*, 1990a, b).

2.2. Input Data and Estimation of Uncertainty

There is uncertainty associated with many of the input data and with the determination of proper values of several of the model parameters used in the calculations. Because this uncertainty should be reflected also in the estimated critical loads, probability distributions were assigned to all those input values and parameters considered uncertain. These included runoff, the N uptake by forests, the mass transfer coefficient for sulfate and nitrate retention, background sulfate concentration, the *F*-factor and the ANC-limit. The shapes of these distributions and the methods for deriving them are described in Posch *et al.* (1992). A distribution of critical loads for each lake was derived by running the model 1000 times, each time selecting a set of input values from the given probability distributions.

The water chemistry variables $[BC]^*_t$, $[SO_4^{2-}]^*_t$ and $[NO_3^-]_t$, and the lake:catchment area ratios were obtained from a lake survey conducted during fall overturn in the year 1987, and designed to be representative for the whole of Finland (Forsius *et al.*, 1990a, b; Kämäri *et al.*, 1991). The base cations were analyzed by flame-AAS, sulfate by ion chromatography and nitrate by a colorimetric method. The lake sample represents ca. 2% of the total number of lakes in the size range 0.01 to 10 km^2 in southern and central Finland, and 5% of the lakes with surface areas 0.1 to 10 km^2 in the northern regions. For the northern part of the country 480 additional lakes sampled during the fall overturn of 1988 and 1989 by the Lapland Water and Environment District were included. For these lakes sulfate was determined by a colorimetrical method that can be used for clearwater lakes dominating in northern Finland. The total number of lakes used for the critical load calculations was 1450. The spatial distribution of the data set reflects the actual lake density in the different regions.

Annual runoff was obtained from a national runoff map available at the Water and Environment Research Institute. From this map a runoff value has been assigned to each of the 2268 1/4 ° × 1/8 ° grids covering Finland (grid area ≈ 14 × 14 km^2 at 60 °N). The runoff Q for calculating the ion flux for a particular lake was then obtained by bilinear interpolation from the four nearest grid values. The values used in the calculations were allowed to vary 20% around this value, following a symmetric triangular distribution.

The mass transfer coefficient for sulfate (s_S) was assumed to follow a triangular distribution, where the most probable value (0.5 m yr^{-1}) and its uncertainty range (0.2 to 0.8 m yr^{-1}) was obtained from a retention model calibration by Baker and Brezonik (1988). Also for the nitrate mass transfer coefficient (s_N) a triangular distribution was assumed, the corresponding modal value and its uncertainty range being 5 m yr^{-1} and 2 to 8 m yr^{-1}, respectively (Dillon and Molot, 1990).

Survey data for water chemistry and fish status has been used for assessing the ANC-limit for fish in Norway. According to this data an ANC-limit of 20 μeq L^{-1} seems to be suitable for the determination of critical loads (Lien *et al.*, 1989; Henriksen *et al.*, 1990b). Hence, in the critical load calculations presented in this paper the ANC-limit was varied around this value following a triangular distribution with a minimum value of 0 and a maximum value of 40 μeq L^{-1}.

The pre-acidification sulfate concentration was estimated by linear regression between $[SO_4^{2-}]^*_t$ and $[BC]^*_t$ from 61 lakes located in northwestern Finland receiving only minor S deposition. Based on the derived regression equation, for a given $[BC]^*_t$-value $[SO_4^{2-}]^*_t$-values were sampled from a Gaussian distribution (see Posch *et al.*, 1992). For Finland we assume that the pre-acidification nitrate concentration $[NO_3^-]_0$ is negligible, and therefore set to zero.

An empirical probability distribution was derived for the scaling factor B, which is used for estimation of the F-factor (see Figure 4 in Posch *et al.*, 1992). The distribution was estimated from a data set for 27 lakes for which paleolimnological acidification estimates and water quality data were available (Huttunen *et al.*, 1990).

The net uptake of S, S_{upt}, in the catchment area was considered negligible and set to zero. The average net uptake of N by forests, N_{upt}, was calculated from forest growth and the N concentration in stemwood and bark, derived from national forest inventory results (Johansson and Savolainen, 1990). Forest growth was estimated as a function of effective temperature sum and forest site type. Information on the N concentration in stemwood and bark was available from studies on three Finnish stands (Mälkönen, 1974). Again, the value for a particular catchment was obtained by interpolation. To account for the large uncertainty of this parameter, N uptake was allowed to vary uniformly between 70 and 130% of its interpolated value in the calculations of the critical loads. The fraction of forests f was obtained from forest inventory data provided by the Finnish Forest Research Institute. The map in Figure 1a shows the product fN_{upt} for each of the 2268 grids covering Finland.

Estimates of present and future deposition of S as well as reduced and oxidized N compounds were obtained from model calculations of the Finnish acidification model HAKOMA (Johansson *et al.*, 1990). The emission submodel of HAKOMA covers 1) emissions of SO_2 from energy production, energy use, transportation and industrial processes (Savolainen and Tähtinen, 1990) in Finland and nearby areas of Russia, 2) emissions of NO_x from combustion processes in Finland, 3) and ammonia emissions from livestock manure and artificial fertilizers (Niskanen *et al.*, 1990). Deposition (wet+dry) estimates were calculated by HAKOMA for each of the 1/4 ° latitude by 1/8 ° longitude grids by atmospheric transport models. The present deposition of S and the sum of oxidized and reduced N compounds is shown in Figure 1b and 1c.

The sulfur factor σ accounts for the fraction of cations balancing S input and $1 - \sigma$ for the fraction balancing acidifying N input. Therefore its magnitude depends on the fraction of S of the total net acidity input. Recalling that $S_{upt} = 0$ we therefore have

$$\sigma = \frac{S^{\star}_{dep}}{S^{\star}_{dep} + N_{dep} - fN_{upt}} \, . \tag{7}$$

This formulation implies that the critical load depends on the deposition to the ecosystem (see Posch *et al.*, 1992). Figure 1d depicts the σ-values calculated from the present depositions (Figures 1b and 1c). A σ-value less than 0.5 shows a dominance by the potentially acidifying N deposition, and values greater than 0.5 by S deposition.

Two example scenarios for future depositions were introduced using the HAKOMA-model by assuming alternative abatement strategies for the emissions. The

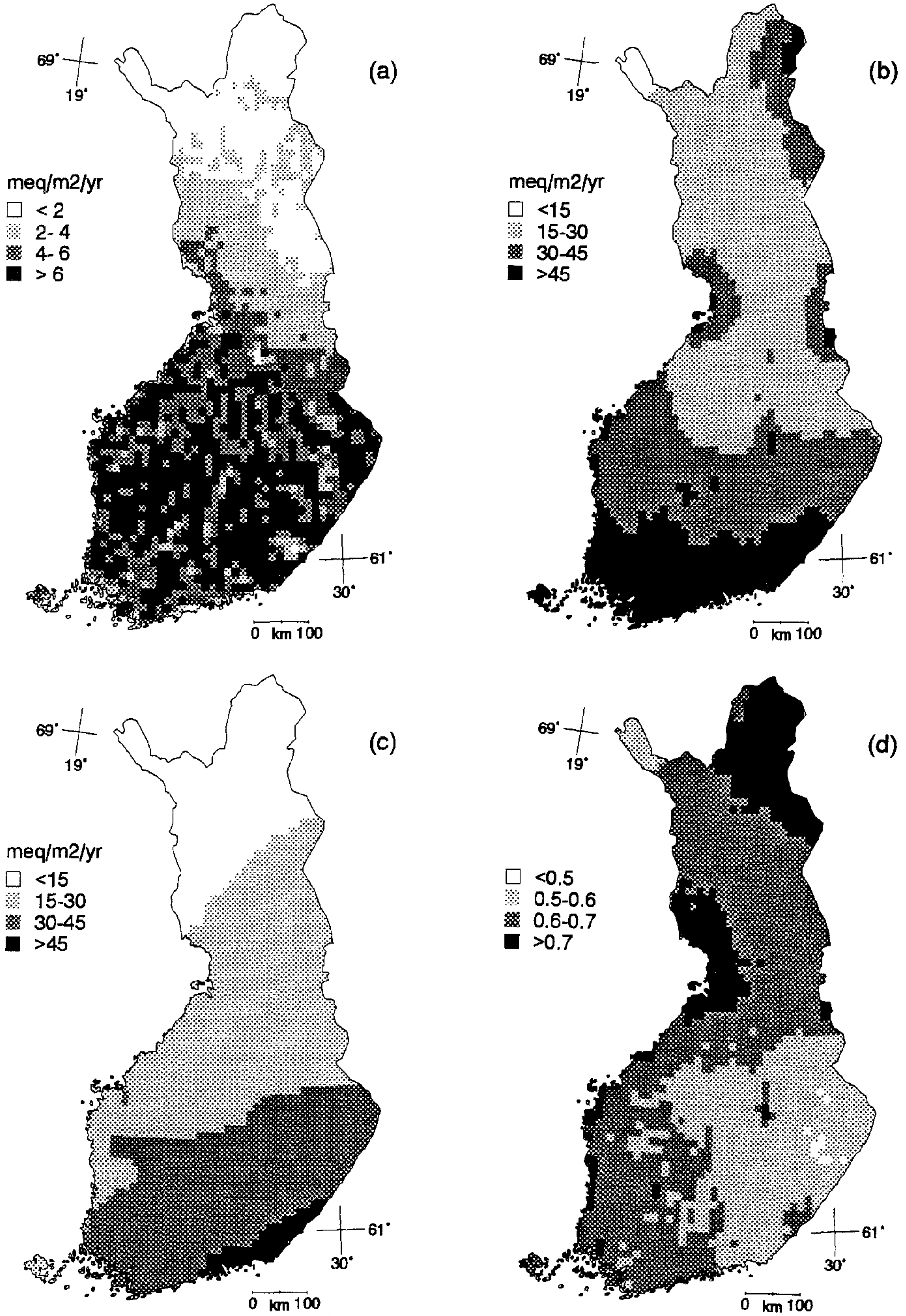

Fig. 1. Maps of Finland showing grid averages of (a) the net uptake of N by forests, (b) the present deposition of S, (c) the present deposition of N, and (d) the sulfur factor σ, i.e. the fraction of S of the total net acidifying deposition.

TABLE I

Changes in emission (in %) of SO_2, NO_x and ammonia relative to the 1980 level in Finland, nearby areas of Russia+Estonia, Eastern Europe and Western Europe for two scenarios: 'Current Reduction Plans' (CRP) and 'Maximum Feasible Reductions' (MFR). (Note that the figures for the N compounds for Eastern Europe include those from nearby Russia+Estonia)

	CRP			MFR		
	SO_2	NO_x	NH_3	SO_2	NO_x	NH_3
Finland	−63	+28	+8	−75	−36	0
Nearby Russia+Estonia	−58	–	–	−70	–	–
Eastern Europe	−12	+1	+30	−78	−62	0
Western Europe	−22	−12	0	−85	−58	−30

'Current Reduction Plans' scenario (CRP) assumes control measures that are based on current plans of Finland and other countries to reduce their emissions. For S the reduction percentages assumed are 63 and 58% for Finland and nearby areas of Russia+Estonia, respectively. For other countries contributing to the S deposition of Finland the reduction percentages assumed vary between 50 and 65% from the 1980 level. In the 'Maximum Feasible Reductions' scenario (MFR) strong measures to reduce emissions are assumed. For Finland and other countries the reductions assumed for S generally vary between 80 and 90%. The changes in emissions assumed in the two scenarios are presented in Table I. In Figure 2 the S and N deposition resulting from these scenarios are shown.

3. Results

For the presentation of the results of our critical load calculations a compromise had to be found between two extremes: with an oversimplified display of a few numbers for the whole country important information would be lost, e.g., about the regional variability of the critical loads, while presenting the information for each individual lake at its geographical location would make the inference of average regional values practically impossible. Therefore grids with a sidelength of 150 km covering the whole country have been chosen as the basic unit for aggregation. In Figures 3 to 6 these grids are plotted in polar stereographic projection, and the north-direction is indicated by an arrow. This coordinate system is refered to as the EMEP-grid, since it is the basic grid of the 'Co-operative Programme for Monitoring of the Long Range Transmission of Air Pollutants in Europe' (EMEP) and it is also used by the UN-ECE in its exercise of mapping of critical loads in Europe.

In Figure 3 the cumulative frequency distributions of the critical load of S and N for lakes within each EMEP-grid in Finland are displayed for present deposition

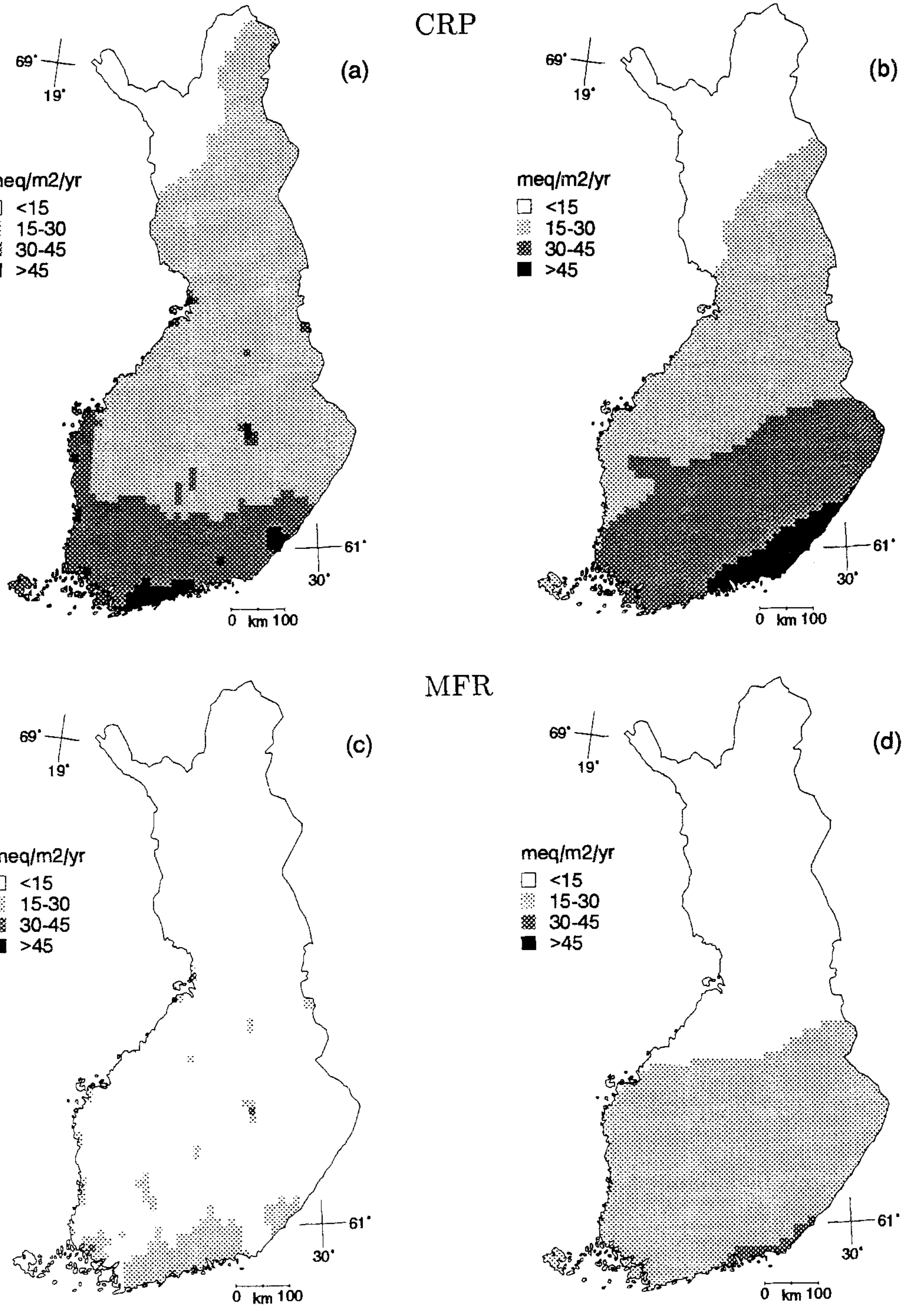

Fig. 2. Maps of Finland displaying the S (a,c) and N (b,d) deposition due to the 'Current Reduction Plans' (CRP) and 'Maximum Feasible Reductions' (MFR) scenarios.

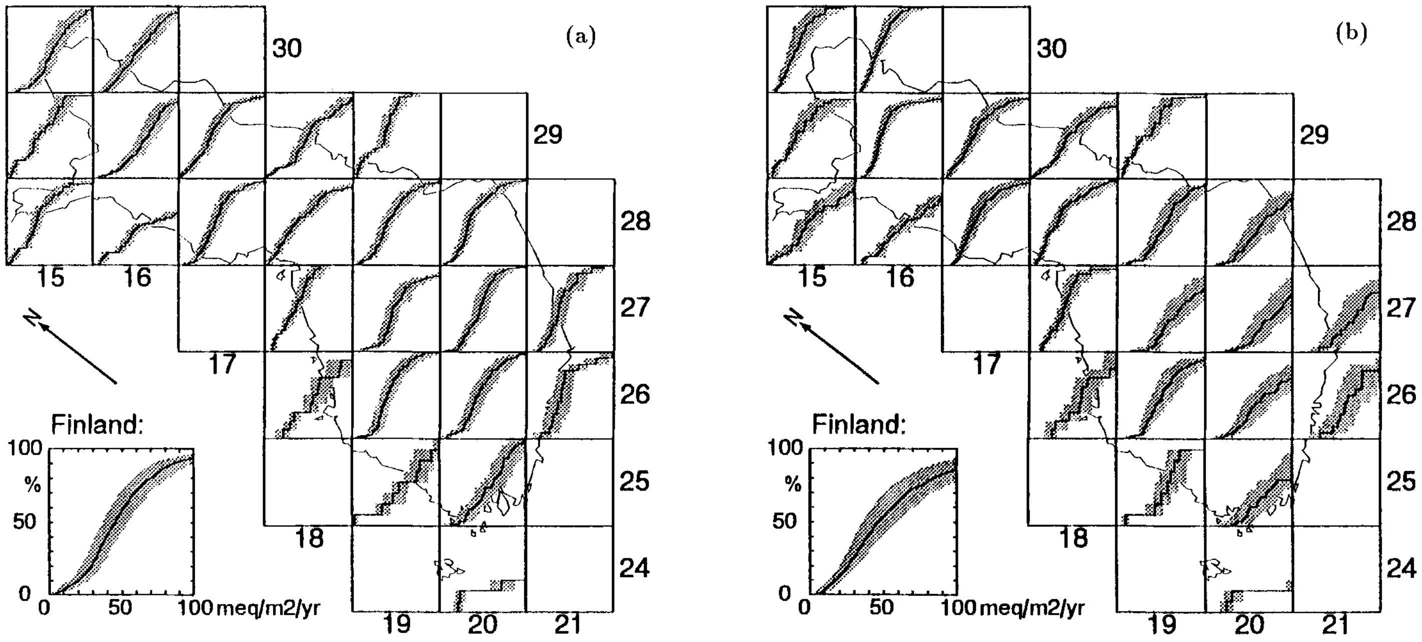

Fig. 3. The critical load of S (a) and N (b) for lakes displayed as cumulative distribution functions in each EMEP-grid covering Finland. The grey bands delineate the uncertainty (between 5th and 95th percentile) while the black line shows the median values. The scale and units of each grid are as shown for the summary display in the lower left corners. This summary display shows the cumulative distribution for all lakes.

values. The grey shaded area around each cumulative distribution function depicts the uncertainty bands within which 90% of the calculated critical loads lie. The critical load values are obtained by randomly drawing input data and parameter values from the probability distributions described in the previous section, running the steady-state mass balance model for 1000 times and saving the 5th, 50th (median) and 95th percentile critical load values for each lake. These values are then used to plot the cumulative distribution functions: The uppermost curve of the uncertainty band depicts the cumulative distribution of the 5th percentile critical loads, the lower curve the cumulative distribution of the 95th percentile, and the black line in the middle of each uncertainty band the cumulative distribution of the median critical loads. In some grids, parts of the uncertainty bands are narrow or only lines. In these cases (e.g. grid 20/24) the actual distribution of the critical load is bimodal and there are no lakes between some values of critical loads. In some cases the uncertainty bands are fairly wide (e.g. grid 21/26) indicating a high degree of uncertainty in the results due to the uncertainty in the input data.

The results show a wide variation in the sensitivity to acidifying pollutants within and between the EMEP-grids and also between larger regions. Within a grid the critical load of S or N may vary from zero to more than 100 meq m^{-2} yr^{-1}. Moreover, the shapes of the cumulative distribution functions vary from region to region. In some grids there is a large number of sites with high sensitivity, shown by a steep lower end of the distribution function, but still some very resistant sites, reflected by a very flat and long uppermost tail of the distribution. The most sensitive regions are located in the northwestern parts of the country, were the critical loads of both S and N are below 20 meq m^{-2} yr^{-1} in a large percentage of the lakes (Figure 3a, b).

The amount by which the critical loads are exceeded are obtained by comparing the critical load of S and N to the respective present S and N deposition. A negative exceedance refers to receptors where the critical load is not exceeded. In Figure 4 the exceedances of the critical loads due to present deposition values are shown as cumulative distribution functions, but in this case as 'inverse' distributions, i.e., 100% minus the cumulative percentage of the exceedance. This has the advantage that the percentage of lakes where the critical load is exceeded can directly be read as the value where the distribution function meets the vertical axis. Note, that the uncertainty bands in Figure 4 reflect the uncertainty of the critical loads, not those of the deposition values. The highest exceedance is presently estimated for south-eastern Finland due to the high present-day loads. In this region up to 80% of the lakes show an exceedance of the critical loads. In Lapland, the critical load is generally exceeded in only 10 to 30% of the lakes. The amount by which the critical loads are presently exceeded reach 40 meq m^{-2} yr^{-1} for S and 20 meq m^{-2} yr^{-1} for N.

It should be kept in mind that the critical loads as well as the exceedance figures are calculated for the present S and N deposition values. If the deposition patterns change, new values for both the critical loads and their exceedances have to be computed. This is due to the non-linear dependence of the critical loads and the

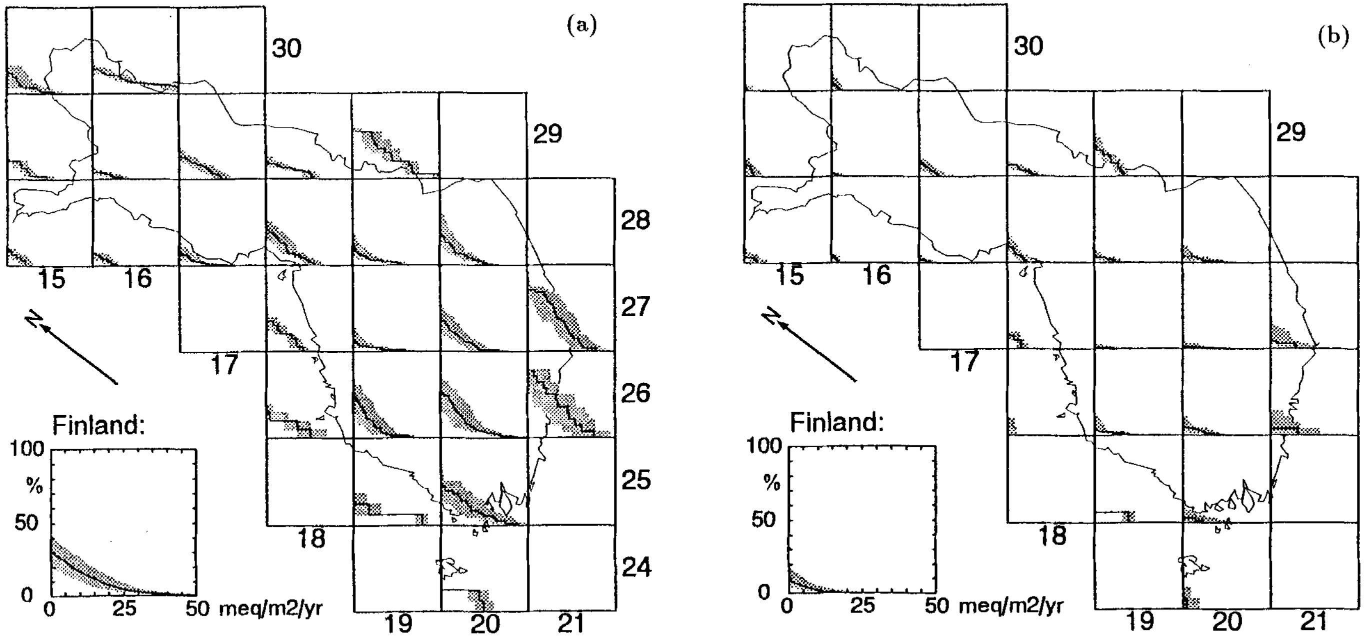

Fig. 4. The exceedance of the critical load of S (a) and N (b) for lakes displayed as 'inverse' cumulative distribution functions in each EMEP-grid covering Finland. The grey bands delineate the uncertainty (between 5th and 95th percentile) while the black line shows the median values. The scale and units of each grid can be seen from the summary display in the lower left corners.

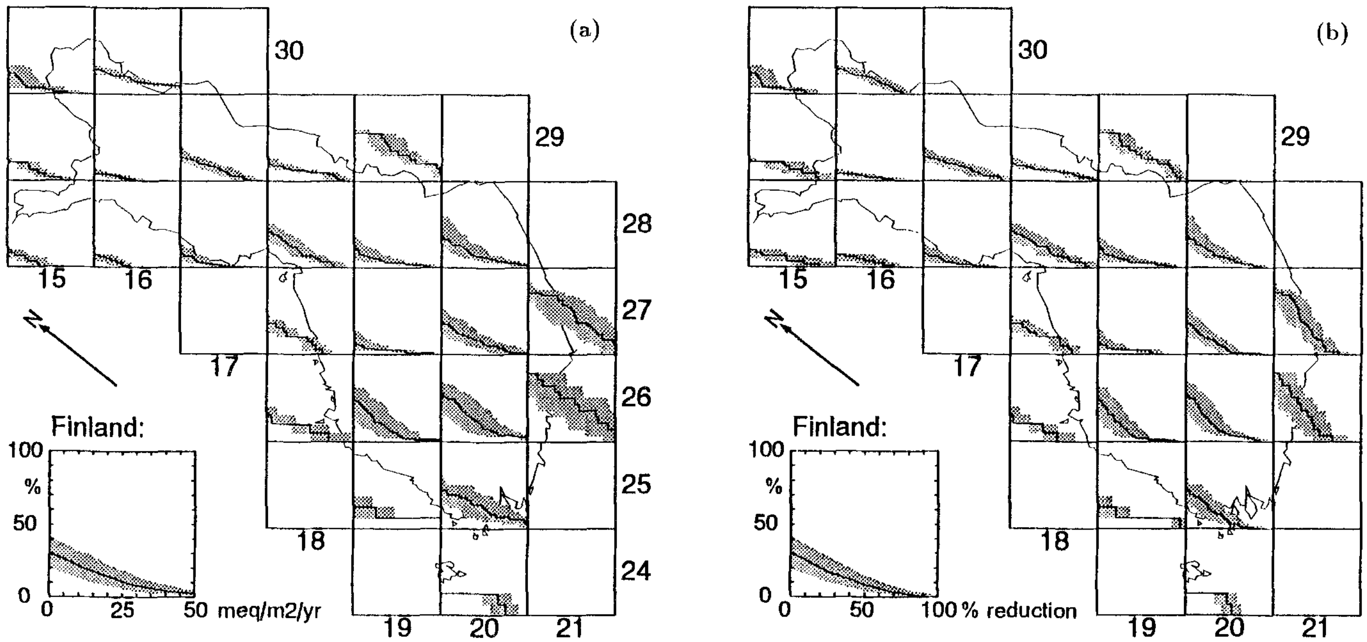

Fig. 5. Required total (S+N) deposition reductions to achieve zero exceedance of both S and N critical loads for lakes, displayed as 'inverse' cumulative distribution functions in each EMEP-grid covering Finland (a) in absolute amounts (meq m^{-2} yr^{-1}) and (b) expressed as percentage of the present total deposition values. The grey bands delineate the uncertainty (between 5th and 95th percentile) while the black line shows the median values. The scale and units of each grid can be seen from the summary display in the lower left corners.

exceedances on the prevailing deposition pattern via the sulfur factor σ. Therefore, the deposition reductions required to avoid exceedance for both S or N, cannot be read from Figure 4, they only give an *indication* of the required reductions. However, as detailed in Posch *et al.* (1992), the total reduction required, i.e. the reduction in the sum of S and N deposition in order not to have any exceedance, can be computed in relation to the present or any deposition levels. These results are shown in Figure 5. The amount by which the present total deposition has to be reduced in order to have zero exceedance is displayed as inverse cumulative distribution functions (Figure 5a). This amount is compared to the present deposition levels and expressed as percentages of reduction required from present deposition levels in Figure 5b.

Three types of regions can be identified from Figures 5a and 5b with respect to the response of the catchments to reductions in loading. First, there are grids in southeastern Finland (e.g. grids 21/26 and 21/27) where a reduction in deposition is required for more than 70% of the lakes: a feasible reduction of about 70% from the present level, can protect all the lakes, because the inverse cumulative distribution falls steeply from high values to zero. Northern Finland (e.g. grids 17/29 and 18/29) and the southwestern coast (e.g. grid 19/25) represent a completely opposite type of regions: only a small portion of the lakes require reduction in acidifying deposition, but the reduction required is significant, even more than 90% of the present deposition levels. Moreover, in some areas (e.g. grid 19/29), for a quite high fraction of the lakes the critical load is exceeded and in addition the reduction percentage required is high, more than 90%. This implies that there are some very sensitive lakes requiring high reductions, in addition to insensitive ones. The last case is the most positive one: the critical load is exceeded only for a small percentage of lakes and a reduction of some 50% in total acidifying deposition protects all of them. Examples of these regions are the Åland Islands (grid 20/24) and western Finland (grids 18/26 and 17/28).

Two emission-deposition scenarios described earlier were selected to illustrate the use of the steady-state mass balance model for analyzing reduction strategies. The percentage of lakes where the critical loads of S and N are exceeded under the two scenatios are shown in Figure 6 along with present conditions. The median percentages of exceedance is shown as bars in each grid, and the associated uncertainty (5th and 95th percentile) as an error-bar on top of each bar. These figures indicate that the acidification status of the Finnish lakes can be significantly improved with current reduction plans and even more so with the maximum feasible reductions. Particulary, for southeastern Finland, where the percentage of lakes exceeding the critical load of S is largest, the maximum feasible reductions are sufficient to protect most of the lakes from the impact of acidic deposition. However, even with these strong measures of reducing S and N emissions, still some 4% of the lakes in the whole of Finland show an exceedance of the critical load of S.

For some grids (e.g. 19/26 in Figure 6a) the uncertainty in the percentage of lakes exceeding the critical load is fairly high (up to 40%). In other grids (e.g.

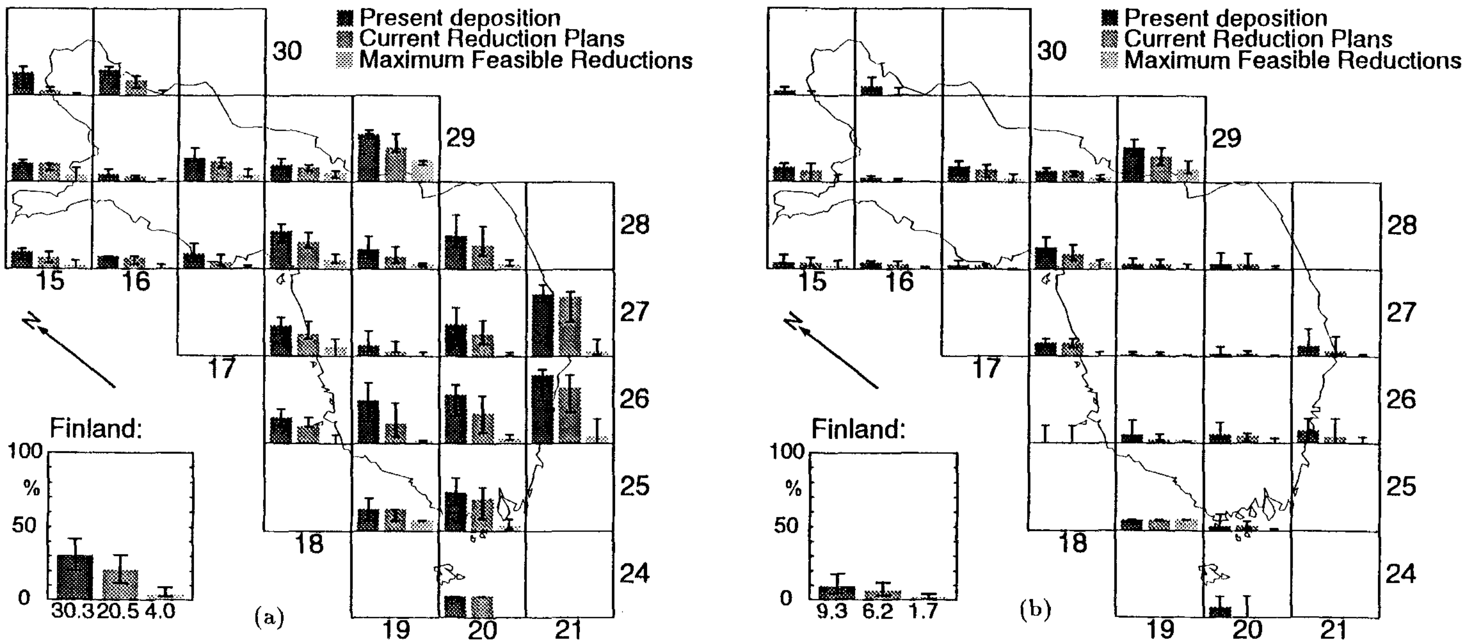

Fig. 6. The percentage of lakes exceeding the critical loads of S (a) and N (b) for present depositions and two deposition scenarios. The shaded bars in the EMEP-grids indicate the median value for that grid and the small error-bars indicate the range between the 5th and 95th percentile. In the lower left corners the results for the whole of Finland are displayed.

grid 20/24) there is no uncertainty shown by the error-bars. This can be explained by the shape of the distribution around zero exceedance: in the grid the distribution of the critical load of S (Figure 3a) and its exceedance (Figure 4a) are bimodal. Therefore, for some 25% of the lakes the critical load of S is presently clearly exceeded and for the rest of the lakes the critical load is clearly not exceeded. As a consequence, there is no uncertainty in the percentage of lakes with critical load exceedance even though the critical loads are uncertain.

4. Discussion

To date, international emission control policies have followed the so-called flat rate reductions, i.e. countries have agreed to reduce emissions by the same percentage. Critical load maps can be used to direct abatement measures to those areas where they are needed the most. Work is in progress to use knowledge of the spatial patterns of critical loads as basis for international negotiations aiming at agreements on air pollution control. The first phase of the international program for mapping critical loads has concentrated on mapping critical loads of acidity and S. So far only little emphasis has been put on the role of N in critical load estimates for surface waters. In the paper by Posch *et al.* (1992) a method for simultaneously calculating critical loads of acidifying S and N for surface waters has been presented for the first time. This paper demonstrates the use of these methods for mapping critical loads of S and N for lakes by using regional survey data.

In an earlier study (Henriksen *et al.*, 1990a) the same national lake survey data were used for evaluating the critical loads of S for Fennoscandian lakes. In that study the critical loads of S were calculated by the water chemistry method (Henriksen *et al.*, 1990b). The critical loads of S shown in Henriksen *et al.* (1990a) are on the average higher than those presented in Figure 3a: the median critical load of S for Finland is 45 meq m^{-2} yr^{-1} in Figure 3a and about 65 meq m^{-2} yr^{-1} in Henriksen *et al.* (1990a). This has several reasons: (i) due to the simultaneous consideration of S and N in our model formulation only part of the base cation leaching, quantified by the sulfur factor σ, is available for buffering S inputs; (ii) the in-lake sulfate retention has not been taken into account in Henriksen *et al.* (1990a); (iii) the dependence of the *F*-factor on the base cation concentration is modeled differently; and (iv) the background sulfate is estimated from Finnish data in this paper.

When linking regional critical load distributions to optimization models, all acidifying substances have to be considered simultaneously. As shown in Posch *et al.* (1992), critical loads of S and N depend on catchment properties but also on the ratio of S to N deposition. Therefore, analyzing only one substance at a time may lead to biased results. Secondly, considerable savings may be obtained by optimizing S and N control options simultaneously, because a reduction of S deposition may also reduce the exceedance of the N critical load.

Knowledge on the level of uncertainty associated with the model results is an essential element in assessing the model outputs for formulating policy options. In calculations of regional critical loads and their exceedances using steady-state mass balance models uncertainty is associated with the model assumptions and the lake-specific parameters of the mass balance model, the input data due to measurement errors, the interpolation of grid-values to lake-specific values as well as the relationships between lake chemistry and aquatic biota (e.g. Brakke and Henriksen, 1989; Jones *et al.*, 1990; Forsius *et al.*, 1992; Posch *et al.*, 1992). In addition, the representativeness of the modelled lake population is a source of uncertainty when aiming at national or regional estimates. In this study, most of the lake chemistry data used was obtained from a lake survey that was designed to be representative for the whole of Finland, and therefore this source of uncertainty (i.e. the regionalization error) is probably the least significant and was not considered in the uncertainty analysis.

The deposition estimates of S and N compounds used in this study (Johansson *et al.*, 1990) are based on model calculations and include also dry deposition. However, there are obviously large uncertainties included in both the emission data and transport model calculations, as well as in the interpolations of catchment-specific deposition values from regional averages (e.g. Savolainen and Tähtinen, 1990; Jones *et al.*, 1990). Dry deposition may vary greatly also within small areas depending on local fluxes of air masses, watershed area, watershed vegetation and lake hydrology (Norton *et al.*, 1988). The deposition estimates for the present situation as well as the two future scenarios were taken as given, and no attempt has been made to estimate their uncertainty.

To account for the uncertainty in the model parameters and input data wide uncertainty bands have been assigned as ranges or distributions for the Monte Carlo sampling procedure. These ranges were designed to reflect both the uncertainty of the parameters and the spatial and temporal variation in the input data. Despite the rather wide ranges used for the inputs, the output distributions displayed on the EMEP-grids (see Figures 3 to 6) show in most cases fairly narrow uncertainty bands for the critical loads and their exceedances. The uncertainty bands computed for the different grids reflect the uncertainty in the model parameters and inputs at the individual sites. Highest uncertainties were calculated for the critical loads of N for the lakes in southeastern Finland. This is partly due to the fact that the knowledge on parameters and inputs with respect to N is least documented and accurate.

Special emphasis was devoted in this project for developing ways to present critical loads and to account for the variability within and between grids. Cumulative distributions have proven useful in presenting spatially varying information. Any percentile of the critical loads can be read from the cumulative distributions, and practically no information is lost in displaying results in such way. This means that the actual shares of different ecosystems can easily be read from the figures. Often, results on regional variability of critical loads are given as shaded grids (e.g. Hettelingh *et al.*, 1991) with the drawback that only one value is chosen to represent the whole grid. Even this value cannot be displayed as such, but the shading represents only a class with predefined class ranges. In case of a distribution a percentile (or other statistical characteristic) has to be chosen for display. Due to the nature of critical loads low percentiles should be preferred, implying that a larger share of the ecosystem will be protected, but still the question remains, which percentile out of the low ones to choose for further assessments. Cumulative distributions allow the examination of the entire population of critical loads without having to pre-determine the percentiles of interest.

The results in this paper show a high degree of variability of critical loads within individual grids and between grids. This intra- and intergrid variability reveals the existence of highly sensitive surface waters to acidic deposition in most parts of Finland. In the international negotiations for air pollution control, target loads will be set nationally. These target loads can be easily evaluated on the basis of the results on intra-grid variability. The fraction of lakes where critical loads are exceeded can directly be read from the cumulative distribution functions. In fact, already in establishing the national target loads, the intra-grid variability should be carefully considered in order to account for the ecological variability within grids. The inter-grid variability of the critical loads suggests that it is unreasonable to set a single target load for all regions of a country. Regional differences should be taken into account by evaluating the spatial distribution of the critical loads and their exceedances.

5. Conclusions

Critical loads are now recognized as a means for providing a scientific basis for international negotiations on emission reduction strategies. The critical loads of S and N for lakes in Finland show a great variability on both small and large spatial scales. The most sensitive areas are located in the north-western and eastern parts of the country. The highest exceedance of critical loads is presently estimated for southern Finland where the acidic load is highest. In the south-eastern part of the country up to 80% of the lakes show an exceedance of the critical loads of S. However, an adoption of an abatement strategy based on maximum feasible reductions would be sufficient for protecting most Finnish lakes from the detrimental impact of acidic deposition. The high variability of critical loads suggest that it is unreasonable to set a single target load for the whole country. Therefore, results of regional critical load calculations should be linked to optimization models in order to develop abatement strategies for S and N emissions which are both cost-effective and ecologically sustainable.

Acknowledgements

We thank Matti Johansson from the Technical Research Centre of Finland for providing the deposition scenarios and N uptake data and the Finnish Forest

Research Institute for the forest inventory data. The Finnish Ministry of Environment is gratefully acknowledged for the financial support of this study.

References

Alcamo, J., Shaw, R., and Hordijk, L. (eds.): 1990, *The RAINS Model of Acidification – Science and Strategies in Europe*, Kluwer Acad. Publ., Dordrecht, The Netherlands.

Baker, L. A. and Brezonik, P. L.: 1988, *Wat. Resour. Res.* **24**, 65.

Brakke, D. F. and Henriksen, A: 1989, in J. Kämäri, D. F. Brakke, A. Jenkins, S. A. Norton, and R. F. Wright, *Regional Acidification Models: Geographic Extent and Time Development*, Springer, Berlin, p. 45.

Brakke, D. F., Henriksen, A., and Norton, S. A.:1990, *Verh. Internat. Verein. Limnol.* **24**, 146.

De Vries, W. and Kros, J.: 1991, *Assessment of Critical Loads and the Impact of Deposition Scenarios by Steady State and Dynamic Soil Acidification Models*, The Winand Staring Centre for Integrated Land, Soil and Water Research, Wageningen, The Netherlands, Report 36.

Dillon, P.J. and Molot, L. A.: 1990, *Biogeochemistry* **11**, 23.

ECE: 1990, *Draft Manual for Mapping Critical Levels/Loads.* Prepared by the Task Force on Mapping, Umweltbundesamt, Berlin.

Forsius, M., Malin, V., Mäkinen, I., Mannio, J., Kämäri, J., Kortelainen, P., and Verta, M.: 1990a, *Environmetrics* **1**, 79.

Forsius, M., Kämäri, J., Kortelainen, P., Mannio, J., Verta, M., and Kinnunen, K.: 1990b, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland,* Springer, Berlin, p. 751.

Forsius, M., Kämäri, J., and Posch, M.: 1992, Critical Loads for Finnish lakes: Comparison of three steady-state models. *Env. Poll.* (in press).

Henriksen, A.: 1984, *Verh. Internat. Verein. Limnol.* **22**, 692.

Henriksen, A. and Brakke, D. F.: 1988, *Environ. Sci. Technol.* **22**, 8.

Henriksen, A., Lien, L., Rosseland, B. O., Traaen, T. S., and Sevaldrud, I. S.: 1989, *Ambio* **18**, 314.

Henriksen, A., Kämäri, J., Posch, M., Lövblad, G., Forsius, M., and Wilander, A.: 1990a, *Critical Loads to Surface Waters in Fennoscandia – Intra- and Inter-Grid Variability of Critical Loads and Their Exceedance*, Miljørapport 1990:17. Nordic Council of Ministers, Copenhagen, Denmark.

Henriksen, A., Lien, L., and Traanen, T.S.: 1990b, *Critical Loads for Surface Waters – Chemical Criteria for Inputs of Strong Acids*, Norwegian Institute for Water Research, Oslo, Acid Rain Research Report 22/1990.

Hettelingh, J.-P., Downing, R. J., and de Smet, P. A. M. (eds.): 1991, *Mapping Critical Loads for Europe*, CCE Technical Report No. 1, National Institute of Public Health and Environmental Protection, Bilthoven, The Netherlands.

Huttunen, P., Kenttämies, K., Liehu, A., Liukkonen, M., Nuotio, T., Sandman, O., and Turkia, J.: 1990, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland*, Springer, Berlin, p. 1071.

Johansson, M. and Savolainen, I.: 1990, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland*, Springer, Berlin, p. 253.

Johansson M., Kämäri, J., Pipatti, R. Savolainen, I., Tuovinen J. -P., and Tähtinen, M.: 1990, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland*, Springer, Berlin, p. 1171.

Jones, M. L., Minns, C. K., Marmorek, D. R., and Heltcher, K. J.: 1990, *Can. J. Fish. Aquat. Sci.* **48**, 599.

Kämäri, J., Forsius, M., Kortelainen, P., Mannio, J., and Verta, M.: 1991, *Ambio* **20**, 23.

Kelso, J. R. M., Shaw, M. A., Minns, C. K., and Mills, K. H.: 1990, *Can. J. Fish. Aquat. Sci.* **47**, 644.

Lien, L., Henriksen, A. Raddum, G. G., and Fjellheim, A.: 1989, *Critical loads for surface waters – Fish and invertebrates* (in Norwegian). Norwegian Institute for Water Research, Oslo, Report 2373.

Mälkönen, E.: 1974, *Comm. Inst. For. Fenniae* **84** (4), 1.

Nilsson, J. (ed.): 1986, *Critical Loads for Sulphur and Nitrogen*, Miljørapport 1986:11, Nordic Council of Ministers, Copenhagen, Denmark.

Nilsson, J. and Grennfelt, P. (eds.): 1988, *Critical Loads for Sulphur and Nitrogen*, Miljørapport 1988:15,

Nordic Council of Ministers, Copenhagen, Denmark.
Niskanen, R., Keränen, S., and Pipatti, R.: 1990, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland*, Springer, Berlin, p. 31.
Norton, S. A., Kahl, J. S., Brakke, D. F., Brewer, G. F., Haines, T. F., and Nodvin, S. C.: 1988, *Sci. Tot. Environ.* **72**, 183.
Posch, M., Forsius, M., and Kämäri, J.: 1993, Critical Loads of sulfur and nitrogen for lakes I: Model description and estimation of uncertainty, *Water, Air, and Soil Pollut.* **66**, 173.
Savolainen, I. and Tähtinen, M.: 1990, in P. Kauppi, P. Antilla and K. Kenttämies (eds.), *Acidification in Finland*, Springer, Berlin, p. 1.
Schindler, D. W., Kasian, S. E. M., and Hesslein, R. H.: 1989, *Environ. Sci. Technol.* **23**, 573.
Sverdrup, H. and Warfvinge, P.: 1990, *Water, Air, and Soil Pollut.* **52**, 71.