

# ENVIRONMENTAL AUDITING

## Exceedance of Critical Loads for Lakes in Finland, Norway, and Sweden: Reduction Requirements for Acidifying Nitrogen and Sulfur Deposition

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**ABSTRACT** / The main objectives of this study were to identify the regions in Fennoscandia where the critical loads of sulfur (S) and acidifying nitrogen (N) for lakes are exceeded and to investigate the consequences for deposition reductions, with special emphasis on the possible trade-offs between S and N deposition in order to achieve nonexceedance. In the steady-state model for calculating critical loads and their exceedances, all relevant processes acting as

sinks for N and S are considered. The critical loads of N and S are interrelated (defining the so-called critical load function), and therefore a single critical load for one pollutant cannot be defined without making assumptions about the other. Comparing the present N and S deposition with the critical load function for each lake allows determination of the percentage of lakes in the different regions of Fennoscandia where: (1) S reductions alone can achieve nonexceedance, (2) N reductions alone are sufficient, and (3) both N and S reductions are required but to a certain degree interchangeable. Secondly, deposition reduction requirements were assessed by fixing the N deposition to the present level, in this way analyzing the reductions required for S, and by computing the percentage of lakes exceeded in Finland, Norway and Sweden for every possible percent deposition reduction in S and N, in this way showing the (relative) effectiveness of reducing S and/or N deposition. The results showed clear regional patterns in the S and N reduction requirements. In practically the whole of Finland and the northern parts of Scandinavia man-made acidification of surface waters could be avoided by reducing S deposition alone. In the southern parts of Sweden some reductions in N deposition are clearly needed in addition to those for S. In southern Norway strong reductions are required for both N and S deposition.

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Critical loads have been widely accepted in Europe as a basis for negotiating control strategies for transboundary air pollution as evidenced by the signing of the Second Sulphur Protocol (UN/ECE 1994) in Oslo in June 1994. The United Nations Economic Commission for Europe's (UN/ECE) Executive Body on Long-range Transboundary Air Pollution has set up the Task Force on Mapping Critical Levels/Loads under its Working Group on Effects. Critical load data from individual countries are collected, mapped, and reported by the Coordination Center for Effects, located at the National Institute of Public Health and the Environment (RIVM)

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in Bilthoven, the Netherlands (e.g., Downing and others 1993). Since 1985 the Nordic Council of Ministers has supported and funded workshops and research with the objective to assist the Task Force on Mapping in establishing a scientific basis for calculating and mapping critical loads.

The scientific discussion on critical loads started at a workshop organized by the Nordic Council of Ministers in 1986 in Sundvollen, Norway (Nilsson 1986), and provided, for the first time, estimates for critical loads of sulfur and nitrogen for forest soils, groundwaters, and surface waters. The first workshop on critical loads held under the auspices of the UN/ECE was organized in 1988 by the Nordic Council of Ministers at Skokloster, Sweden, and provided the still-valid definition of a critical load as "the quantitative estimate of an exposure to one or more pollutants below which significant

harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (Nilsson and Grennfelt 1988).

As the role of nitrogen in the acidification of soils and surface waters gained increasing attention at the end of the 1980s in both the scientific and policy arena, a workshop was organized by the Nordic Council of Ministers and the US Environmental Protection Agency on that topic in Copenhagen in 1988 (Malanchuk and Nilsson 1989). The purpose of that workshop was to review the state of science on the role of nitrogen in the acidification of the environment. The foundation for the actual mapping of critical loads in the ECE countries was laid in a UN/ECE workshop held in 1989 in Bad Harzburg (Germany), resulting in a draft manual for mapping critical levels and loads (ECE 1990). Furthermore, in a workshop on critical loads for nitrogen organized by the Nordic Council of Ministers in Lökeberg (Sweden) in 1992, recommendations for deriving critical loads of nitrogen and their exceedances were elaborated (Grennfelt and Thörnelöf 1992). Although the concept of critical loads has found wider political acceptance in Europe, research in this field is also carried out in North America. For a critical load assessment framework in the United States see, e.g., Strickland and others (1993), Hunsacker and others (1993), Hicks and others (1993), and Holdren and others (1993); and for Canadian lakes, see Jeffries and Lam (1993).

In a Nordic project on inter- and intraregional variability of critical loads (Henriksen and others 1990a, 1992), data for assessing the critical loads of sulfur acidity for lakes in Finland, Norway, and Sweden were, for the first time, brought together in a single data base and mapped in a common format. The results of the mapping exercise were consistent: the gradients, both in deposition and in critical loads, continued smoothly across country borders. The "variability project" was followed by a project aiming at deriving critical loads of both N and S, thus providing the means for assessing reduction strategies and target loads for both pollutants simultaneously (Henriksen and others 1993). The outcome of this project forms the basis of the this paper.

The most important question concerning the response of catchments to airborne N loading has to do with the mobility of N. The change in the functioning of the N cycle from a virtually closed internal cycle to an open cycle where the excess inorganic N is leached from the system, is often referred to as N saturation (see, e.g., Aber and others 1989, Stoddard 1994). The fate of nitrate is key in determining the effects of the N cycle on soil acidification. If nitrification rates or supply

rates of inorganic N from external sources exceed the ecosystem's assimilation rate, nitrate leaching occurs, resulting in an increased proton production (ecosystem acidification) and manifesting itself in higher soil and lake water concentrations of aluminum, increased base cation export in runoff or a decline in runoff alkalinity. While there are cases in which nitrate leaching is caused by an imbalance between nitrification and assimilation rates (e.g., Van Miegroet and Cole 1984), most cases are attributed to an excess N supply from atmospheric sources (Van Breemen and others 1982). Nitrate leaching from a catchment can therefore be used as an indicator of the acidifying role of N compounds in surface waters.

An assessment of nitrate leaching based on water chemistry data from lakes in Finland, Norway, and Sweden showed that the median of the leached nitrate was above 10 meq/m<sup>2</sup>/yr only in southern Norway and southern Sweden, whereas elsewhere in Fennoscandia it was mostly at background levels (Henriksen and others 1993). In 95% of all the lakes, the leaching of nitrate was less than 20% of that of sulfate, although the deposition of these compounds is of the same order of magnitude. According to the data, it is evident that at present N compounds contribute to surface water acidification only in the southern parts of Norway and Sweden.

This paper presents a simple steady-state model for the simultaneous computation of critical loads of acidifying N and S and their exceedances for lakes, based on the charge balance principle. This model, together with data bases on catchment characteristics, water chemistry, and depositions for about 4500 lakes in Finland, Norway, and Sweden is then used to assess the reduction requirements for N and S deposition in the Nordic countries. While originally designed for the analysis of critical loads in the Nordic countries, the same model is also used to provide input to the forthcoming negotiations of a new nitrogen protocol in Europe.

### Critical Load Model

There are two major approaches for determining the critical load for surface waters: process-oriented models and empirical models. Process-oriented models attempt to develop mathematical descriptions for the mechanisms underlying the cause-effect relationships between acidic deposition and water quality. Models of this type can be either dynamic simulation models (e.g., Cosby and others 1985, De Vries and others 1989) or steady-state models based on mass and charge balance calculations (e.g., Sverdrup and Warfvinge 1990, Posch and others 1993). In steady-state conditions, when no

acidification of the system is permitted, the sources of alkalinity must balance the input and production of acidity. Dynamic processes, such as cation exchange and the adsorption/desorption of sulfate, should not be considered in the derivation of critical loads, since they are a temporary phenomena. For example, the soil solution will reach a new equilibrium with the sulfate at the adsorption complex under critical loads, irrespective whether saturation is reversible or not. It is the timing (time lags) of the release/storage of sulfate that depends on the adsorption/desorption characteristics of the soil and the way the transition from present to critical loads is effected (see, e.g., Reuss and Johnson 1986). This transition can last for decades and can be of great interest for timing abatement strategies, but it can be only described with dynamic models; it can not be incorporated into a critical load definition, which considers only the long-term ( $t \rightarrow \infty$ ) steady-state situation.

Two assumptions and simplifications are made concerning the behavior of N in the terrestrial part of the catchment: (1) The effect of nutrient cycling between plants and soil is ignored, and (2) the leaching of ammonium is negligible, implying a complete uptake and/or nitrification in the root zone (Sverdrup and others 1990, De Vries and Kros 1991). This is true for practically all nonagricultural areas in the Nordic countries (see Kämäri and others 1992). Furthermore, it is assumed that immobilization, reduction, and uptake of sulfate in the catchment soils are negligible.

We consider only catchments with negligible inputs of N due to agricultural activities, since agriculturally dominated catchments are unlikely to be acidified. Considering the sources and sinks of the major elements in the terrestrial catchment area draining to the lake and the retention processes in the lake itself, one obtains from the charge balance of the lake water (Kämäri and others 1992, Posch and others 1993, Henriksen and others 1993):

$$N_{dep} + S_{dep} = fN_{upt} + (1 - r)(N_{imm} + N_{den}) + rN_{ret} + rS_{ret} + BC_{le} + Alk_{le} \quad (1)$$

where  $N_{dep}$  and  $S_{dep}$  are the total deposition of N and S,  $f$  is the fraction of forested land in the catchment area,  $N_{upt}$  is the net growth uptake of N by the forests,  $r$  is the lake:catchment area ratio,  $N_{imm}$  is N immobilized in the terrestrial catchment,  $N_{den}$  is N lost by denitrification,  $N_{ret}$  and  $S_{ret}$  are the in-lake retention of N and S,  $BC_{le}$  is the nonmarine base cation leaching, and  $Alk_{le}$  is the leaching of alkalinity.

All the above quantities (except  $f$  and  $r$ ) are in equivalents (moles of charge) per unit area and time. While N uptake is limited to the (managed) forest area

fraction  $f$ , N immobilization and denitrification are assumed to occur in the whole terrestrial catchment area (fraction  $1 - r$ ). Note, that  $N_{imm}$  stands for the long-term net immobilization of N, including fixation and the export of organic N out of the catchment.

Immobilization and growth uptake of N are assumed independent of the N deposition, whereas denitrification depends on the available N. A simple description for the rate of denitrification is (De Vries and others 1994):

$$N_{den} = \begin{cases} f_{de}(N_{dep} - N_{imm} - N_{upt}) & \text{for forested land} \\ f_{de}(N_{dep} - N_{imm}) & \text{for open land} \end{cases} \quad (2)$$

where  $f_{de} \leq 1$  is a constant denitrification fraction. This equation is based on the assumption that immobilization and growth uptake are faster processes than denitrification.

Similarly, the in-lake retention of N is assumed proportional to the net input of N to the lake

$$rN_{ret} = \rho_N [N_{dep} - fN_{upt} - (1 - r)(N_{imm} + N_{den})] \quad (3)$$

The retention factor  $\rho_N$  is modeled by a kinetic equation (Kelly and others 1987):

$$\rho_N = \frac{s_N}{s_N + z/\tau} = \frac{s_N}{s_N + Q/r} \quad (4)$$

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

If sufficient information were available, the sources and sinks of base cations, especially the weathering rates of the catchment soils, could be estimated using data for soil mineralogy (Sverdrup 1990). An alternative approach—and the one used in this paper—is to estimate the net base cation leaching from water quality data by the steady-state model introduced by Henriksen (1984). In this formulation the base cation leaching is approximated by the preacidification leaching of base cations ( $Q[BC]^*$ ) from the catchment area, which is estimated from the present leaching of base cations and the long-term changes in the inputs of strong acid anions using the so-called  $F$  factor (Henriksen 1984, Brakke and others 1990, Posch and others 1993):

$$[BC]^*_t - [BC]^*_0 = F([SO_4^{2-}]^*_t + [NO_3^-]_t - [SO_4^{2-}]^*_0 - [NO_3^-]_0) \quad (5)$$

where the subscripts 0 and  $t$  refer to the original (background) and present concentrations, respectively. Replacing the alkalinity leaching by  $Q[ANC]$  and inserting the expressions for denitrification (equation 2), in-lake N retention (equations 3 and 4), and an analogous equation for the in-lake S retention [ $rS_{ret} = \rho_S S_{dep}$

and  $\rho_S = s_S / (s_S + Q/r)$  into equation 1 yields, after rearranging terms:

$$a_N N_{dep} + a_S S_{dep} = b_1 N_{upt} + b_2 N_{imm} + Q([\text{BC}]_0^* - [\text{ANC}]) \quad (6)$$

where the dimensionless constants  $a_N$ ,  $a_S$ ,  $b_1$ , and  $b_2$  are all smaller than 1 and are obtained from the lake/catchment properties as:

$$a_N = [1 - f_{de}(1 - r)](1 - \rho_N) \quad (7a)$$

$$a_S = 1 - \rho_S \quad (7b)$$

$$b_1 = f(1 - f_{de})(1 - \rho_N) \quad (7c)$$

$$b_2 = (1 - r)(1 - f_{de})(1 - \rho_N) \quad (7d)$$

$a_N$  and  $a_S$  can be interpreted as the fractions of N and S deposition, respectively, that contribute to acidification.

Inserting values for N and S deposition into equation 6 yields the concentration of ANC in the lake; conversely, this equation can be used to derive critical loads for N and S, if one can relate  $[\text{ANC}]$  to ‘‘harmful effects’’ of the chosen indicator organism. For a variety of aquatic organisms (fishes) thresholds for  $[\text{ANC}]$  have been determined (Lien and others 1996; see also below). Inserting one of these  $[\text{ANC}]_{limit}$ s, we obtain the following relationship between the critical load of N,  $CL(N)$ , and the critical load of S,  $CL(S)$ :

$$a_N CL(N) + a_S CL(S) = b_1 N_{upt} + b_2 N_{imm} + L_{crit} \quad (8)$$

where we have introduced the following abbreviation:

$$L_{crit} = Q([\text{BC}]_0^* - [\text{ANC}]_{limit}) \quad (9)$$

These critical loads of N and S are limited by the following constraints:

$$CL(N) \leq (b_1 N_{upt} + b_2 N_{imm} + L_{crit}) / a_N =: CL_{max}(N) \quad (10)$$

and

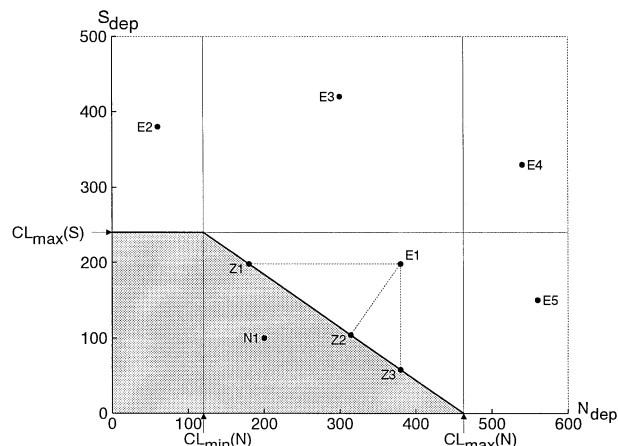
$$CL(S) \leq L_{crit} / a_S =: CL_{max}(S) \quad (11)$$

Furthermore, if

$$N_{dep} \leq (b_1 N_{upt} + b_2 N_{imm}) / a_N =: CL_{min}(N) \quad (12)$$

all N is consumed by uptake and immobilization and S can be considered alone.

Figure 1 illustrates the relationship between the deposition and critical load given by the above equations. The thick lines indicate all possible pairs of critical loads of N and S acidity, and this has been named the critical load function (the tilted line reflects the linear relationship of equation 8; the horizontal line for  $N_{dep} \leq CL_{min}(N)$  is due to the constraint given by equation 12).



**Figure 1.** The relationship between the N and S depositions and critical loads (units are  $\text{meq}/\text{m}^2/\text{yr}$ ). The case for nonexceedance (N1), the five cases for exceedance (E1–E5), and the three cases for critical loads (Z1–Z3) are described in the text.

Note that in the above formulation individual critical loads of N and S are not (and cannot be) specified; each pair of depositions ( $N_{dep}$ ,  $S_{dep}$ ) fulfilling equation 8 (and the constraining equations 10–12), i.e., lying on the critical load function, are called critical loads. As the exceedance of the critical loads, we define the difference between the left- and right-hand side of equation 8 after inserting  $N_{dep}$  and  $S_{dep}$ , i.e.,

$$Ex(N_{dep}, S_{dep}) = a_N N_{dep} + a_S S_{dep} - b_1 N_{upt} - b_2 N_{imm} - L_{crit} \quad (13)$$

If  $Ex \leq 0$ , the point ( $N_{dep}$ ,  $S_{dep}$ ) lies in the grey area in Figure 1 (or on the critical load function) and we have nonexceedance; if  $Ex > 0$ , it lies outside (e.g., points E1–E5 in Figure 1) and we have exceedance of critical loads. Note, however, that a positive exceedance value is not necessarily the amount by which N or S has to be reduced to achieve nonexceedance. This can be easily seen from the example in Figure 1: starting from E1 and reducing  $N_{dep}$  one reaches the point Z1, i.e., nonexceedance without reducing S; on the other hand one can reach nonexceedance solely by reducing  $S_{dep}$  until reaching Z3. Finally, with a smaller reduction of both  $N_{dep}$  and  $S_{dep}$  one can reach nonexceedance as well (e.g., point Z2). In practice, external factors, such as costs of emission reductions, will determine which path to follow to reach the critical load function and thus non-exceedance.

## The Input Data

The input data for calculating and mapping critical loads and their exceedances were obtained largely from national lake surveys carried out in Finland (Forsius

and others 1990, Kämäri and others 1991), Norway (Henriksen and others 1988), and Sweden (Bernes 1991), originally conducted to assess the acidification status of surface waters in the respective countries. All the surveys concentrated on small (mostly headwater) lakes, which are not directly influenced by human activities such as agriculture and industry. Although the design of the surveys was different in each country, we consider them representative for the purpose of deriving critical loads. Only the analysis of a common Nordic lake survey, conducted in the three countries in fall 1995, can reveal any possible inconsistencies.

#### Selection of Lakes/Catchments

The surveys of 1450 lakes conducted in Finland during 1987–1989 form the basis for calculating critical loads. The spatial distribution of the sampled lakes is proportional to the actual lake density in different regions, and therefore no data were available for a few grids with a very low number of lakes. A statistical survey of 970 lakes was carried out in 1987 (Forsius and others 1990, Kämäri and others 1991), and during the years 1987–1989, 480 additional lakes were surveyed by the Lapland Water and Environment District.

For Norway, a grid net of  $1/4^\circ$  longitude by  $1/8^\circ$  latitude was used as the basis for the selection of lakes and rivers. One lake or stream was selected from 1:50,000 maps to be representative for each grid, based on expert judgment. Three sources of data were used when selecting a site for each grid: First, lakes sampled during the 1000-lake survey of 1986 were used whenever possible. Second, additional lakes in southern Norway were sampled in fall of 1989 and 1990 for those grids not covered by the 1000-lake survey. Third, for the northern countries, data for small streams, collected by the Norwegian Geological Survey in the mid-1980s, were used. The total number of surface waters used for the calculations was 2305.

All Swedish lakes with an area  $>1$  ha constituted the basis for selection for the 1990 lake survey (Bernes 1991). Some exceptional lake types were excluded (e.g., waterbodies receiving sewage water). In order to obtain a reasonable geographic distribution, the lake register was stratified by county, and within each county the lakes were classified into four size classes 0.01–1, 1–10, 10–100 and  $>100$  km<sup>2</sup>. From each size class a pre-defined percentage of lakes was randomly selected for sampling. If there were less than 40 lakes in a size class, all lakes were sampled. In addition, an extra set of 40 lakes per county was selected randomly. For the latter group, the decision for sampling was left to the county authorities. A total of 4018 lakes was sampled between the end of January and beginning of May 1990, representing winter conditions. For the calculations pre-

sented in this paper, a statistically representative subset of 760 lakes, which were analyzed for N compounds and not influenced by liming, has been used.

#### Water Quality Data

The methods for calculating critical loads of N and S require information on the present and preindustrial lake water concentrations of base cations and acid anions. The present-day information can be found in the above survey data bases. Standard analytical methods have been used on all samples (Forsius and others 1990, Hovind 1991). The background (preacidification) sulfate concentration (in microequivalents per liter) was estimated from the relationship between  $[\text{SO}_4^{2-}]_i^*$  and  $[\text{BC}]_i^*$  from 251 Finnish, Norwegian, and Swedish lakes located in northern regions, receiving the lowest acidic deposition in these countries:

$$[\text{SO}_4^{2-}]_0^* = 0.078[\text{BC}]_i^* + 18.75 \quad (14)$$

$(N = 251, r = 0.66)$

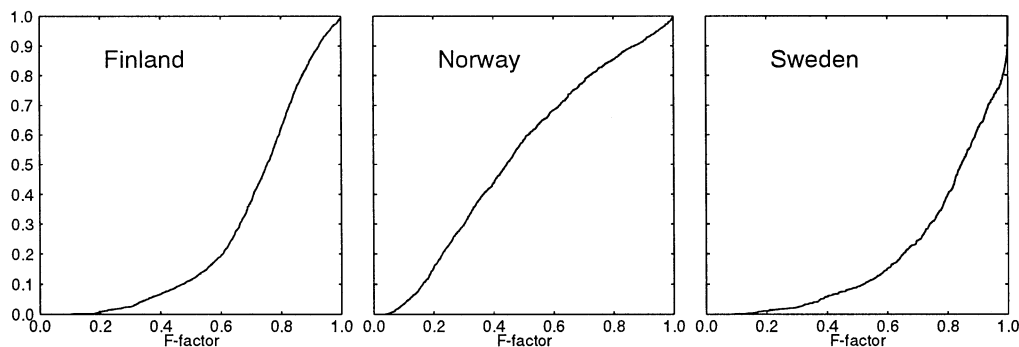
This equation suggests that there is an atmospheric background contribution of sulfate and a geological contribution that is proportional to the concentration of base cations (see Henriksen and others 1990a). However, although these lakes are located in remote northern areas, they may nevertheless receive slightly elevated levels of acidic deposition. It was therefore decided to use the mean value of  $[\text{SO}_4^{2-}]_0^*$  (given by equation 14) minus one standard deviation (see Posch and others 1993). Only  $[\text{SO}_4^{2-}]_0^*$ -values less than the present-day sulfate concentrations  $[\text{SO}_4^{2-}]_i^*$  were accepted. The preacidification nitrate concentration was assumed to be zero,  $[\text{NO}_3^-]_0 = 0$ .

#### F factor

The value of  $F$  in equation 5 is a function of the base cation concentration, ranging from near zero in dilute lakes to one in lakes with high concentrations of base cations (Brakke and others 1990, Marmorek and others 1990). In this study we have used the continuous nonlinear relationship between  $F$  and the background base cation concentration derived by Posch and others (1993):

$$F = 1 - \exp(-[\text{BC}]_0^*/B) \quad (15)$$

where  $B$  is a scaling factor. Inserting this expression for  $F$  into equation 5 gives a nonlinear equation for  $[\text{BC}]_0^*$ , which is solved by an iterative procedure. The parameter  $B$  requires the knowledge of the preacidification status of a representative sample of lakes. An expression for  $B$  has previously been derived from paleolimnological estimates and water quality data of 27 Finnish lakes



**Figure 2.** Cumulative distribution functions of the  $F$  factors for Finland (1450 lakes), Norway (2305 lakes), and Sweden (760 lakes).

(see Posch and others 1993). The median value of this distribution,  $131 \mu\text{eq/liter}$ , was used for the calculations in this paper. Since  $F$  depends on the original base cation concentration,  $F$  is not time-dependent, but a unique property of the catchment. In Figure 2 the cumulative distribution functions of the  $F$  factors in each of the countries are shown. It should be mentioned here that other formulations of the  $F$  factor (e.g., Brakke and others 1990) render it time-dependent, and this time dependence of the  $F$  factor has been studied using dynamic models (Wright and others 1991).

#### ANC Limit

Acid neutralizing capacity (ANC), defined as the difference between nonmarine base cations and strong acid anions, appears to be a suitable chemical criterion for sensitive indicator organisms in surface waters (Henriksen and others 1990b, Lien and others 1996). Fish was chosen as the biological indicator for selecting the critical ANC limit. Extensive fish status and water chemistry data have been collected during the Norwegian 1000-lake survey in 1986 (Henriksen and others 1988, 1989), and these data have been used to calculate the probability for fish damage at different levels of ANC (Lien and others 1996). According to these data, an  $[\text{ANC}]_{\text{limit}}$  of  $20 \mu\text{eq/liter}$  seems to be appropriate for calculating the critical load for the most common freshwater fish (trout, perch, and arctic char), and this value was used also in the present study. One should be aware, however, that the natural ANC in lakes can be below  $20 \mu\text{eq/liter}$  in areas with granitic and gneissic bedrock and thin soil cover. For such lakes the ANC limit is set to the present ANC of the lake.

#### Mass Transfer Coefficients

Due to a lack of data, the net mass transfer coefficient for sulfate ( $s_s$ ) was taken from a retention model calibration to mass balance data of 11 lakes located in North America and northern Europe (Baker and

Brezonik 1988). The mean value of  $s_s$  for these lakes,  $0.5 \text{ m/yr}$ , was used for the critical load calculations. The value for  $s_N$ ,  $5 \text{ m/yr}$ , was obtained from a similar study of 12, mostly Canadian, lakes (Dillon and Molot 1990).

#### Lake-to-Catchment-Area Ratios

For Finnish lakes, both lake and catchment areas were estimated from topographical maps (1:50,000), and for the Norwegian 1000-lake survey lake and catchment areas were estimated from 1:50,000 topographical maps; for the remaining Norwegian lakes the median ratio ( $r = 0.072$ ) of the 1000-lake survey data was used. For Sweden, the ratio was estimated from a regression between lake and catchment areas for 3343 lakes located in different parts of the country. The regression equations for the Finnish, Norwegian, and Swedish data sets are given in Henriksen and others (1993).

#### Runoff

The annual runoff for Finnish lakes was obtained by interpolation from a national runoff map (Leppäjärvi 1987). For Norwegian lakes, the mean annual runoff values were read manually from national runoff maps for 1931–1960 (National Board for Water and Electricity 1987). For Swedish lakes the runoff values were taken from a digitized runoff map (Tryselius 1971).

#### Nutrient Uptake by Forests

Only the net growth uptake, i.e., the amount removed from the catchment by harvesting, enters the critical load calculations. For Finland, the net uptake of N by forests ( $N_{\text{upt}}$ ) was computed from tree growth and the N concentration in stemwood and bark, derived from national forest inventory results (Johansson and Savolainen 1990). The fraction of forests ( $f$ ) was obtained from forest inventory data provided by the Finnish Forest Research Institute.

In Norway values for  $N_{\text{upt}}$  are based on inventory

results provided by the Norwegian Forest Research Institute (NISK). The information on forest coverage was obtained from a data base of the Norwegian Institute for Air Research (NILU).

For Sweden  $N_{upt}$  values for each catchment were interpolated from a data base of 21,000 forest sites (Rosén and others 1992). The forest fraction  $f$  was obtained from a data base of Lövblad and others (1992).

#### Denitrification and N Immobilization in Soils

It was assumed that the denitrification fractions are related to the soil types in the catchments. In deeply drained podzolic soils, which dominate in northern Europe, denitrification values are generally low. However, high values may occur in areas with peatsoils (Klemedtsson and Svensson 1988, De Vries and others 1994). Therefore, the average denitrification fraction for each catchment was approximated by the following linear relationship:

$$f_{de} = 0.1 + 0.7f_{peat} \quad (16)$$

where  $f_{peat}$  is the fraction of peatlands in the catchment area.

For Finland, information on  $f_{peat}$  was available for the 970 catchments surveyed in 1987. For the remaining Finnish lakes, located mainly in Lapland, the peatland fraction was estimated from chemical oxygen demand (COD) data by linear regression (Henriksen and others 1993). From the Finnish data base, a nonlinear relationship between  $f_{peat}$ , latitude (as substitute for temperature), and total organic carbon (TOC) was established, from which the  $f_{peat}$  was derived from TOC values of Norwegian and Swedish lakes (see Henriksen and others 1993).

For the long-term immobilization of N in forest soils ( $N_{imm}$ ), a constant value of 2 kg N/ha/yr was used. This value represents the lower end of the range suggested for European critical load calculations (Downing and others 1993).

#### Deposition

For Finland, deposition estimates of S and reduced and oxidized N compounds for the year 1990 were obtained from model calculations with the Finnish acidification model HAKOMA for each of the 1/4° longitude by 1/8° latitude grids covering Finland (Johansson and others 1990). Base cation deposition estimates (bulk precipitation) were obtained from a national monitoring network (Järvinen and Vänni 1990). Catchment-specific values were interpolated from the three nearest stations.

For Norway and Sweden, land-use weighted deposi-

tion of S, total N, and base cations estimated by Lövblad and others (1992) were used. Total deposition had been calculated as the sum of measured wet deposition and estimated dry deposition on 50 × 50-km<sup>2</sup> grid squares. Dry deposition of S and N was estimated by multiplying interpolated monitored air concentrations with annual mean deposition velocities (dependent on land cover) in each grid.

#### Results and Discussion

As shown above, unique critical loads of N and S that depend on ecosystem properties alone cannot be specified. Furthermore, the quantity defined as the critical load exceedance (equation 13) is *not* the reduction requirement for S and/or N. The dependence of the retention processes (which are different for S and N) on the actual deposition values do not allow us to express the reduction requirements for S and N in a unique way. Displaying the constraints (equations 10–12) limiting the critical loads of N and S (and their respective exceedances) is one way of roughly illustrating the lakes' sensitivity to acidifying deposition, but these extreme quantities offer only limited insight into the actual reduction requirements. A more informative way is to display the minimum and maximum amount of the combined S + N deposition reduction required to achieve nonexceedance, which illustrates the range within which one has the choice of allocating reductions of S and/or N deposition. However, this range can be fairly large, as shown for Finland by Downing and others (1993).

In this paper we study the reduction requirements by first identifying which pollutant has to be reduced and which one can be reduced in order to reach critical loads (nonexceedance). This is investigated first by classifying each lake according to the present N and S deposition and its critical load function (cases N1 and E1–E5 in Figure 1). Second, the amount by which S deposition has to be reduced in order to reach critical loads is calculated by fixing the deposition level of N at present levels. Finally, we investigate the relative effectiveness of S and N reductions to protect the lake ecosystems in Finland, Norway, and Sweden. This exercise is performed for the present (1990) depositions and for the likely situation in the year 2010, i.e., after the implementation of the Second Sulfur Protocol (UN/ECE 1994).

The question of which pollutant has to be reduced to reach critical loads is investigated by comparing the present S and N deposition for each lake with its critical load function. As shown in Figure 1, six cases can occur: (1) either S reductions alone or N reductions alone can

lead to nonexceedance (E1), (2) only S reductions can achieve zero exceedance (E2), (3) S reductions are mandatory, but they can partially be substituted by N reductions (E3), (4) both N and S reductions are needed to reach nonexceedance (E4), (5) like E3, but with the roles of N and S reversed (E5), and (6) no reductions are needed (N1). Note, that there is no N case equivalent to E2, since there is no sulfur process comparable to the uptake and immobilization of N.

In Figure 3A the percentage of the different cases (except for case E5, which occurs less than 1% in each country) are plotted for each EMEP grid, a coordinate system in polar stereographic projection with grid cells of  $150 \times 150 \text{ km}^2$  used by the Cooperative Programme for Monitoring and Evaluation of the Long-Range Transmission of Air Pollutants in Europe (EMEP) for modeling the long-range transport of sulfur and nitrogen in Europe. The results show a clear regional pattern in the exceedance. The critical loads are exceeded in about 19%–42% of the lakes in Fennoscandia (depending on country), with the greatest proportions in the southernmost parts of Norway and Sweden, where up to 100% of the lakes exceed critical loads in some grids. In most grids in southern Finland up to 50% of the lakes exceed the critical loads. In the northern parts of Fennoscandia the critical loads are exceeded significantly only in the northeastern parts of Finland and Norway. In the rest of the Fennoscandian region the percentage of lakes exceeding the critical loads is generally less than 20%.

In Figure 3B, the bars depict the percentages of lakes in the four exceedance classes, now as the percentage of the total number of exceeded lakes. From this map the character of the exceedance and the pollutant causing the exceedance are better visible. It is evident that for large parts of Finland and for the northern parts of Sweden and Norway the exceedance is mainly caused by S deposition, and nonexceedance in these regions can be obtained by reducing S deposition alone (cases E1–E3). In particular, the northern parts of Fennoscandia require S reductions only (case E2). Mandatory reductions in N deposition are required only in a quite limited area in southern Sweden and in southern and central Norway. In these regions the critical loads are exceeded to such an extent that mandatory reductions in both S and N are required for a large percentage of lakes (30%–100%) in order to achieve nonexceedance (case E4). The regions where strong reductions are required for N deposition coincide with regions where the N concentrations are elevated (e.g., Henriksen and Brakke 1988). In Finland only 2% and in Sweden 18% of the exceeded study lakes (implying <1% and <5% of all lakes in Finland and Sweden, respectively) require a

mandatory reduction in N deposition in order to reach critical loads. As a consequence, most lakes of Finland and Sweden can be protected by reducing S deposition alone: The summary bars of Figure 3B illustrate that 76% of the exceeded lakes in Finland, 28% in Norway, and 64% in Sweden require a mandatory reduction in S deposition (case E2 or E3). The largest proportion of exceeded lakes in Norway (56%) require a mandatory reduction for both S and N. In addition, a significant proportion (17%–23%) of the exceeded lakes in the Nordic countries can be restored by any combination of reductions in S and N depositions (case E1). This case is fairly significant in southern Finland and Sweden (up to 40%), leaving a broad range for optimizing emission reductions between S and N.

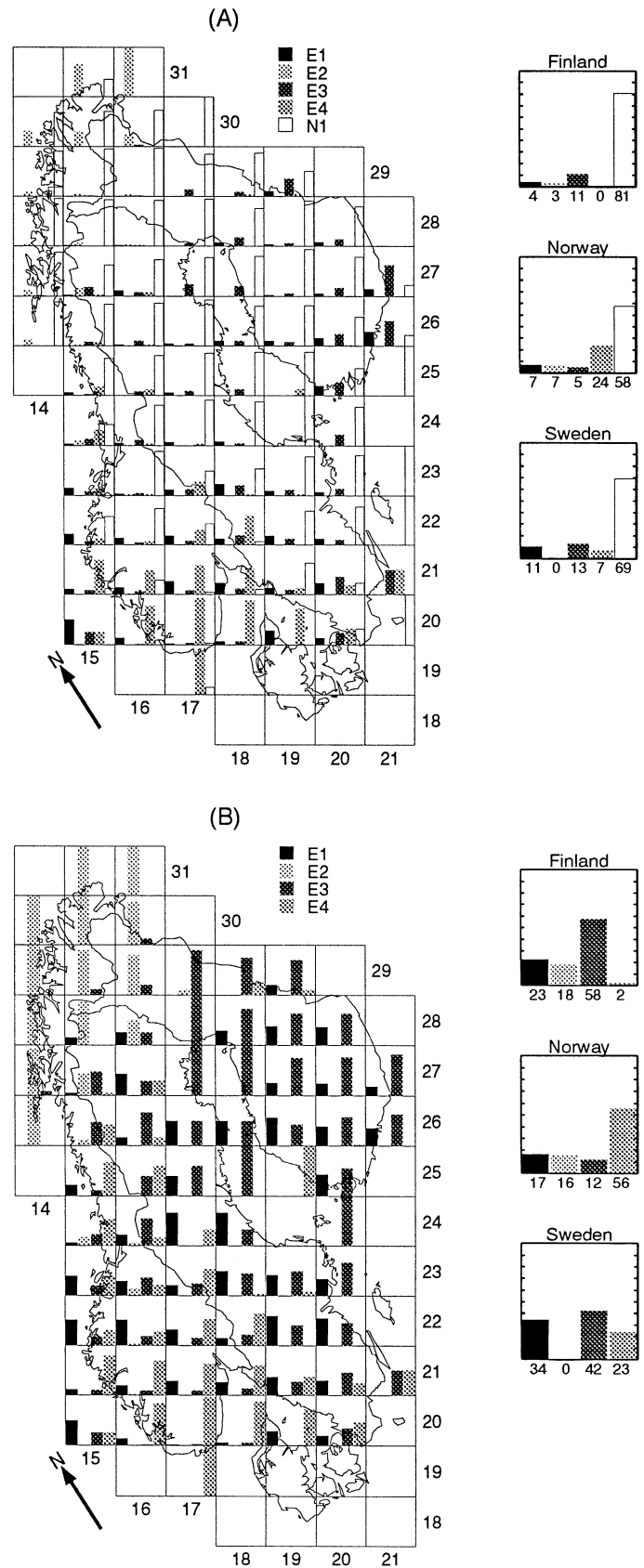
The amount by which S and N deposition must be reduced to achieve critical loads can be determined either by optimization, based on some predefined criteria (e.g., costs), or by fixing the deposition of one of the pollutants. For example, for computing reductions required in S deposition alone in order to achieve nonexceedance, a fixed scenario for N deposition has to be selected. Fixing  $N_{dep}$  in the exceedance function (equation 13) and calling the resulting S deposition the critical load of S at  $N_{dep}$ ,  $CL(S|N_{dep})$ , we obtain by setting  $Ex = 0$ :

$$CL(S|N_{dep}) = \begin{cases} L_{crit}/a_S & \text{for } N_{dep} \leq CL_{min}(N) \\ a_N(CL_{max}(N) - N_{dep})/a_S & \text{for } CL_{min}(N) < N_{dep} \\ & \leq CL_{max}(N) \\ -\infty & \text{for } N_{dep} > CL_{max}(N) \end{cases} \quad (17)$$

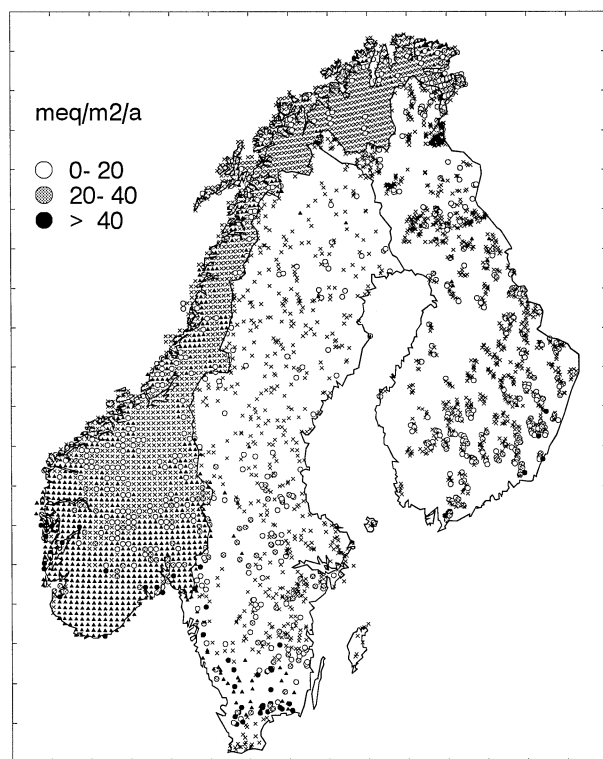
The  $-\infty$  in the above equation indicates that for  $N_{dep} > CL_{max}(N)$  even a 100% reduction in S deposition does not suffice to achieve nonexceedance. Analogous to equation 17, a critical load of N for a given S deposition,  $CL(N|S_{dep})$ , can be defined and, for example, used to investigate the necessary N deposition reductions after implementing the Second Sulfur Protocol.

For production of Figure 4, the N deposition was fixed at the present level, which roughly corresponds to the 1988 Sofia Protocol for the control of  $\text{NO}_x$  emissions. This way the overall reduction requirements, i.e., the difference between S deposition and  $CL(S|N_{dep})$ , before the Second Sulfur Protocol can be assessed. In Figure 4, which shows the locations of all lakes considered in this study, the potential for protecting a lake is plotted: lakes that are not exceeded are depicted as crosses; lakes that are exceeded, but S reductions are sufficient for protecting the lake (cases E1–E3 in Figure 1) are shown as circles, with the grey shade indicating





**Figure 3.** Bar charts depicting the percentage of lakes in each of the exceedance classes (N1 and E1–E5; see Figure 1) in every EMEP grid cell covering Finland, Norway, and Sweden: (A) Including all lakes (cases N1 and E1–E4; see Figure 1), and (B) only lakes exceeding the critical loads of S and N (cases E1–E4). Note that case E5 is not shown, since it rarely occurs (less than 1%).



**Figure 4.** Location of the Nordic lakes and their S reduction requirements, at present (1990) N deposition: crosses show nonexceeded lakes, shaded circles depict lakes that can be protected by S reductions alone (cases E1–E3 in Figure 1), and black triangles show lakes where (additional) N reductions are required (cases E4 and E5 in Figure 1).

the amount of S reductions required; and finally, those exceeded lakes that require (additional) reductions of N deposition (cases E4 and E5 in Figure 1) are displayed as black triangles.

In the optimization models employed in the negotiations on emission reductions in Europe, it is impossible to use the huge amount of individual critical load data, and therefore statistical descriptors, like the fifth percentile in a given grid, are used (see Downing and others 1993). On a national (regional) level, however, it is possible to make use of the full data sets, and in the following we give an example on how one can use the above methodology for gaining insight into the relative importance of S and N deposition reductions. The percentage of lakes that are protected for every possible percent deposition reduction of S and/or N is displayed in Figure 5 for the lakes in Finland, Norway, and Sweden. Figures 5A,C,E start from the present (1990) N and S deposition, whereas the origins in Figures 5B,D,F correspond to the present N deposition and the S deposition after implementation of the Second Sulfur Protocol (UN/ECE 1994). For the present situation,

Figure 5A shows that S reductions (in relative terms) are more effective in Finland than reductions in N deposition. For example, about 95% of the lakes can be protected by a 50% reduction in S deposition together with a 10% reduction in N deposition. In order to achieve the same protection level with a 39% S reduction, a N deposition reduction of about 60% is required. In Norway, however, S and N deposition reductions are equally effective (in relative terms), as can be inferred from the slope of the isolines. For Swedish lakes, S reductions are more effective than N reductions, but not in such a pronounced manner as for Finnish lakes.

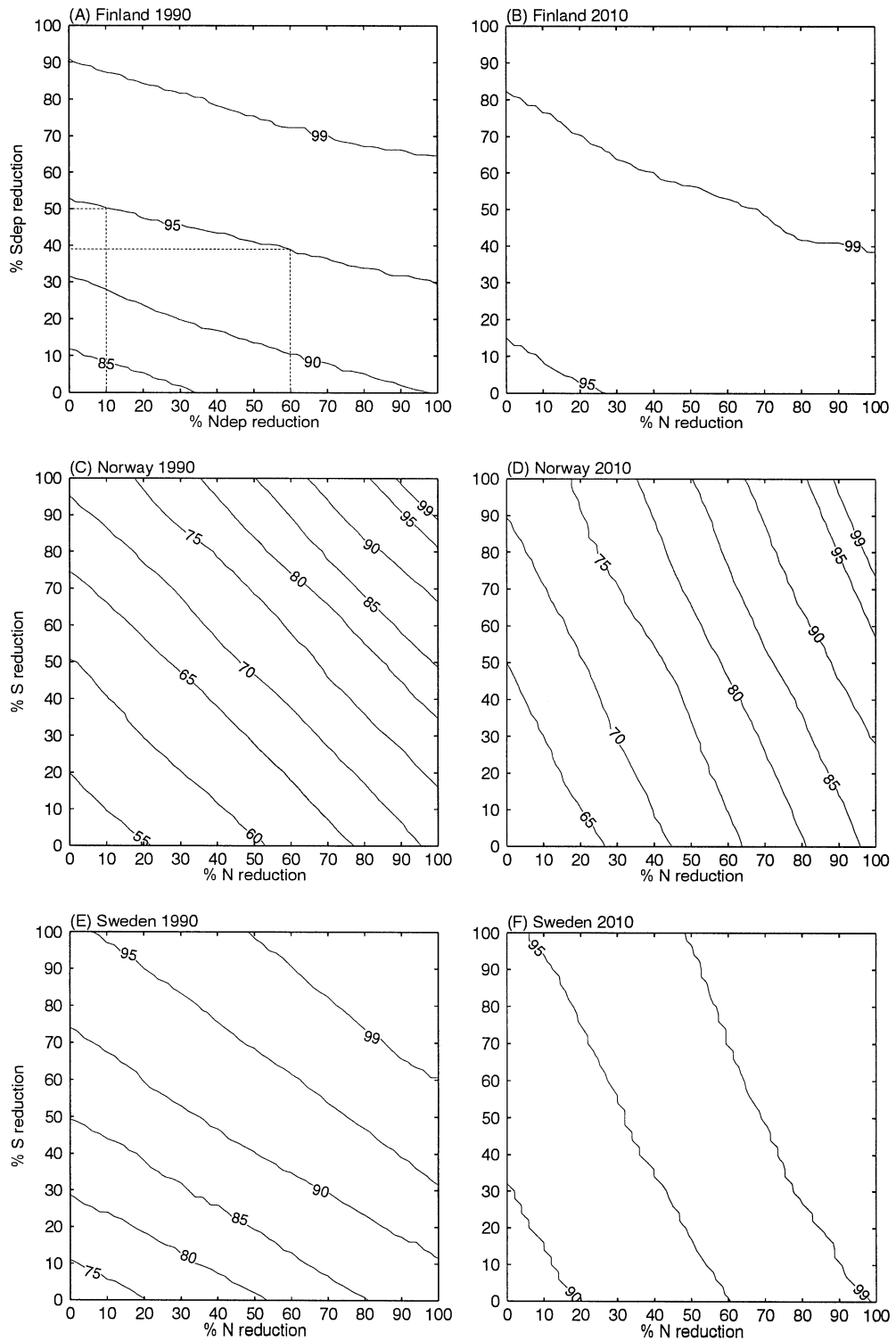
After the obligations of the Second Sulfur Protocol have been implemented, the number of affected lakes obviously decreases in each country, and the relative importance of S and N deposition changes considerably. In particular, for Norway and Sweden, N reductions become more effective for restoring lakes than S reductions. For Finnish lakes, S reductions still remain a more effective means for achieving nonexceedance. This type of graph offers the possibility for a quick assessment, both for comparing different scenarios (by replacing the present depositions with the scenario depositions) and for comparing different regions with respect to the relative importance of S and N deposition.

Knowledge on the level of uncertainty associated with the model results is an essential element when using the model's output for formulating policy options. In an earlier exercise, the uncertainty in the calculated critical loads for lakes in Finland was analyzed by Monte Carlo methods (Posch and others 1993, Kämäri and others 1993). That study showed that, despite the large uncertainty in some of the model parameters and input data, the resulting ranges of critical loads are in most cases narrow enough to draw meaningful conclusions on the sensitivity of the lake/catchment systems to acidifying deposition. Ideally, such an analysis should also be carried out for the critical load calculations presented in this paper, but the similarity of the situation in the other Nordic countries is a strong indication that the uncertainty ranges are of a similar magnitude.

## Conclusions

In this paper we have assessed the exceedances of the critical loads of acidifying N and S deposition. Both N and S deposition contribute to the acidification of surface waters, and thus the critical loads of N and S are interrelated, and they have to be treated together.

A tool is presented for analyzing the exceedance and the reductions needed for N and S depositions. The



**Figure 5.** Isolines of protection percentages of lakes in Finland, Norway, and Sweden for all possible uniform percentage reductions, starting from given deposition patterns: **(A, C, E)** present (1990) S and N deposition; **(B, D, F)** 2010 (Second Sulfur Protocol) S deposition and present (1990) N deposition. The dotted lines in Figure 5A compare two reduction scenarios (see text).

question of which pollutant has to be reduced in order to achieve critical loads is approached by comparing the present N and S deposition at each lake to the critical load function. The results show a clear regional pattern in the S and/or N exceedance. In practically the whole of Finland and the northern parts of Scandinavia, the acidification problem could be solved by reducing S deposition alone. In the southern parts of Sweden some reductions in N deposition are clearly needed in addition to those for S. In the southern parts of Norway even strong measures to reduce S deposition are not enough; N deposition also has to be reduced considerably.

The methodology described here allows the simultaneous analysis of N and S reduction requirements, e.g. by (cost) optimization models. Alternatively, the two pollutants can be assessed separately by fixing the level of one pollutant. This is demonstrated by analyzing the reductions required for S, given the present level of N deposition. In an iterative procedure, additional reductions requirements for N deposition may be investigated next, after an agreement for reducing S emissions on the basis of critical loads is finalized.

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