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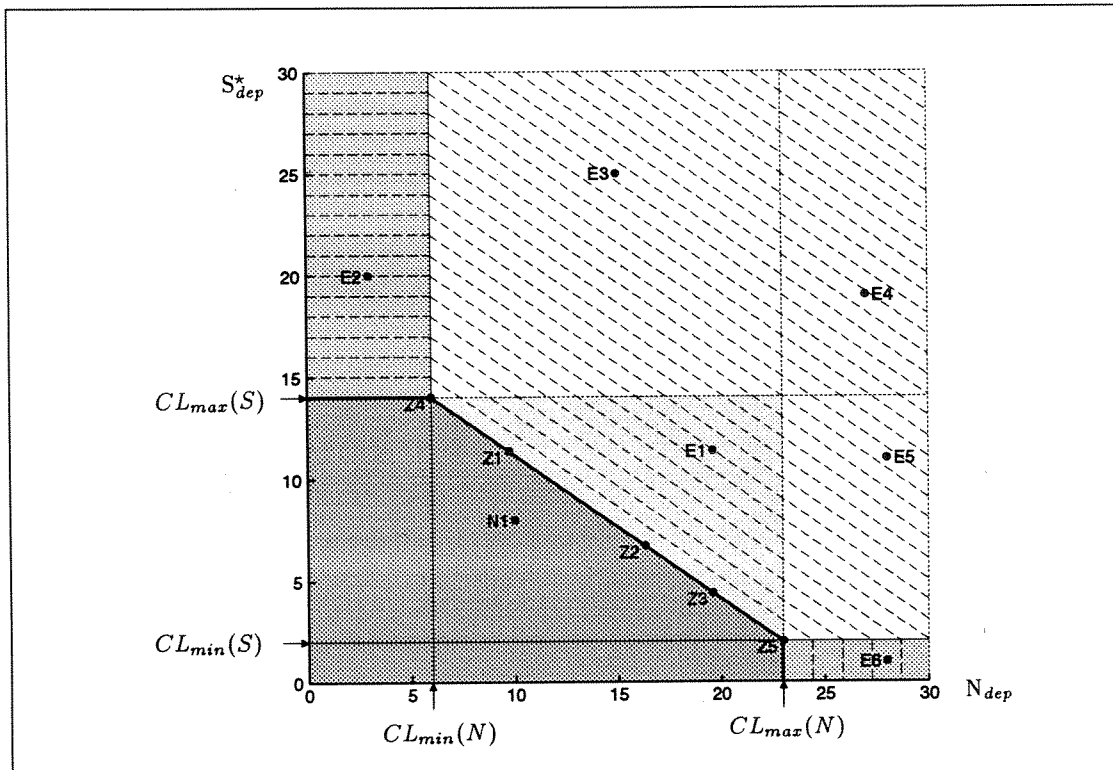
**Exeedance of Critical Loads
for Lakes in Finland, Norway
and Sweden:**

Reduction Requirements for
Nitrogen and Sulfur Deposition.

Norwegian Institute for Water Research NIVA

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Abstract:

The main goal of the present study was to evaluate the different ways of computing the exceedances and the reductions needed for N and S depositions. Two models for calculating the steady state critical loads for N and S acidity were applied to Finnish, Norwegian and Swedish lake data bases. The first model was used for calculating present exceedances from the critical loads of acidity and the second for assessing also the future situations. The critical loads of N and S deposition are interrelated, and therefore, one cannot be looked at without makong assumptions about the other. The question which pollutant has to be reduced in order to achieve the critical load was approached by investigating the present N and S deposition for each lake with respect to the critical load function. Secondly, the reduction required was assessed separately for S deposition by fixing N deposition to the present level. Only in southernmost Norway strong measures to reduce sulfur is not enough, strongest possible reductions of nitrogen deposition is also required.

4 keywords, Norwegian	4 keywords, English
1. Sur nedbør	1. Acid deposition
2. Innsjøer	2. Lakes
3. Tålegrenser	3. Critical loads
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Project leader

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For the Administration

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

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Preface

Critical loads have been widely accepted as a basis for control strategies for regional air pollution. In order to gain more insight in the magnitude and the spatial variation of critical loads, the UN ECE Executive Body on Long Range Transboundary Air Pollution has set up a Task Force on Mapping Critical Level/loads under the Working Group of Effects. The data from individual countries are collected, mapped and reported by the Coordination Center for Effects, located at the National Institute of Public Health and Environmental Protection (RIVM) in Bilthoven, the Netherlands. Since 1985 the Nordic Council of Ministers has supported projects dealing with critical loads of air pollutants. During the last four years the Nordic projects have had the objective to assist the Task Force on Mapping in establishing a scientific basis for mapping procedures.

The present report is the final report of a Nordic working group on *the basis for deriving critical and target loads for sulfur and nitrogen* with Arne Henriksen as the project leader. The data has been collected by the National Focal Centers of Finland, Norway and Sweden. The computations and the map displays have been prepared by Maximilian Posch. All authors have participated in the data analysis, derivation of inputs and reporting. It is the hope of the authors that the results of this document will highlight the potential and limitations of the critical load concept and help identifying further research needs. The Nordic Council of Ministers is gratefully acknowledged for financial support.

1. Introduction

The starting point for the scientific work on critical loads of sulfur and nitrogen was established at a workshop organized by the Nordic Council of Ministers in 1986 in Sundvollen, Norway. The workshop report (Nilsson 1986) provided for the first time agreed definitions for critical loads and first estimates for critical loads of sulfur and nitrogen for forest soils, groundwaters and surface waters. As a follow-up of the Sundvollen Workshop, the first workshop on critical loads held under the auspices of the UN/ECE Convention on the Long-Range Transboundary Air Pollution was sponsored and organized by the Nordic Council of Ministers at Skokloster, Sweden, in 1988 (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 U.S. 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 the UN/ECE workshop on mapping critical levels and loads held in 1989 in Bad Harzburg, Germany. The mapping methods were documented in a draft manual for mapping critical levels and loads (ECE 1990). The definitions and criteria for critical loads as well as methods for calculating them were largely based on the results of the Skokloster workshop. Furthermore, a workshop on critical loads for nitrogen was organized by the Nordic Council of Ministers in Lökeberg, Sweden, in April 1992 (Grennfelt and Thörnelöf 1992). Recommendations for deriving critical loads of nitrogen and their exceedances were elaborated in the workshop. The question of how much of the incoming N is leached and how much of the leaching can be permitted was approached in the workshop by empirical and process-oriented methods.

Since 1985 the Nordic Council of Ministers has supported projects dealing with critical loads of air pollutants. In 1990 a project on inter- and intra-regional variability of critical loads was carried out with funding from the Nordic Council. In this project, data for assessing the critical loads of sulfur acidity for lakes for the whole of Finland, Norway and Sweden were collected and harmonized. This variability study demonstrated the benefits of using simple methods to compute critical loads for surface waters. The results of the mapping exercise appeared to be consistent: the maps showed that the gradients, both in deposition and in critical loads, continue smoothly across country borders (Henriksen *et al.* 1990a, 1992). In 1992 the variability project was followed by the present project aiming at deriving critical loads of nitrogen and sulfur and providing the means for assessing target loads on the basis of these critical loads.

The main goal of the present study is to evaluate the different ways of computing the exceedances and the reductions needed for nitrogen and sulfur depositions. Quantitative estimates of the exceedances over the critical loads of nitrogen and sulfur provide the means for assessing the consequences of alternative target loads or deposition patterns resulting from alternative control strategies. Any proposed loading pattern can be evaluated as maps by comparing the deposition pattern with the respective critical load distributions.

As a first step, preliminary maps for critical loads of nitrogen for surface waters are produced relying on the outcome of the Lökeberg workshop. The working group for surface waters distinguished between two concepts: 1) the present exceedance and 2) the potential exceedance. In calculating the present exceedance of nitrogen and sulfur deposition over the critical load of acidity, nitrate leaching is used to indicate the proportion of nitrogen deposition which is not retained in the catchment. For the potential exceedance all relevant processes acting as net sinks for nitrogen and sulfur are considered and quantified.

Two models, based on the above two concepts, for calculating steady-state critical loads of sulfur and nitrogen acidity are applied to Finnish, Norwegian and Swedish lake data bases. The first model can be used for calculating the present exceedances from the critical loads and the second also for assessing future situations. In the latter approach, which considers the most important processes of nitrogen retention, the mechanisms are formulated as rate-limited processes, i.e. the magnitude depends on the input. The results on the critical loads are then used for calculating exceedances and reductions required for the depositions of nitrogen and sulfur.

2 Derivation of Models

2.1 Background

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 *et al.* 1985; De Vries *et al.* 1989) or steady-state models based on mass balance calculations (e.g. Sverdrup and Warfvinge 1990; Posch *et al.* 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. Therefore, dynamic processes such as cation exchange and adsorption/desorption of sulfate are neglected in the derivation of long-term critical loads.

In this section two steady-state approaches, one empirical and one process-oriented, used for calculating the critical loads of acidifying deposition to surface waters in the Nordic countries, are described. The first approach, the Steady-State Water Chemistry (SSWC) method, enables the calculation of critical loads of acidity and, based on that, present exceedances of incoming total acidity over the critical loads of acidity. While the SSWC method is restricted to the present situation, the second approach, the First-order Acidity Balance (FAB) model and its extension attempts to account for those processes considered to be relevant for the overall acidity balance of the catchments in a steady-state situation. The FAB model allows the evaluation of the effects of any scenario for S and N deposition.

Two assumptions and simplifications are made in the above approaches concerning the behavior of N in the terrestrial part of the catchment (Sverdrup *et al.* 1990; De Vries and Kros 1991): (1) The effect of nutrient cycling is ignored, and (2) the leaching of ammonium is negligible, implying a complete uptake and/or nitrification in the rootzone. The latter assumption is valid for practically all non-agricultural areas in the Nordic countries (see Kämäri *et al.* 1992). Furthermore, for S it is assumed that the immobilization and the reduction of sulfate in the catchment's soils are negligible. Although sulfate adsorption might occur in catchment soils (e.g. Johnson *et al.* 1986), this process should be neglected when considering a long-term critical load (De Vries 1992), because it is only a temporary phenomenon (several decades).

2.2 Steady State Water Chemistry (SSWC) method

2.2.1 Critical load of acidity

The Steady State Water Chemistry model described earlier (ECE 1990; Henriksen *et al.* 1990a,b) is used for calculating the critical load of S acidity for surface waters. A new version of this model, modified to consider both S and N acidity, was developed for the workshop on N critical loads in Lökeberg, Sweden (Kämäri *et al.* 1992). The equation for the critical load of acidity $CL(Ac)$ is given by

$$CL(Ac) = Q([BC]_0^\star - [ANC]_{limit}) - BC_{dep}^\star \tag{1}$$

where Q is the runoff, $[BC]_0^\star$ the original sea salt corrected base cation concentration, $BC_{dep}^\star$ the sea salt corrected atmospheric deposition of base cations and $[ANC]_{limit}$ is the selected critical ANC threshold.

It is assumed that the catchments were in steady-state relative to deposition inputs during the pre-industrial period. Hence, the change over time in the leaching of base cations (Ca, Mg, Na, K) can be related to long-term changes in inputs of strong acid anions by the F-factor (Henriksen 1984; Brakke *et al.* 1990):

$$[BC]_t^\star - [BC]_0^\star = F\left([SO_4^{2-}]_t^\star + [NO_3^-]_t - [SO_4^{2-}]_0^\star - [NO_3^-]_0\right) \tag{2}$$

where the subscripts 0 and t refer to the original and present concentrations, respectively. This formulation is based on the assumption that (a) the difference between uptake by vegetation and mineralization of base cations is constant over time, and (b) the base cation deposition has not changed over time.

2.2.2 Present exceedance of the critical load of acidity

In the present situation inorganic N leaching at the outlet of the catchment, N_{le}, is assumed to equal the N sources minus the N sinks, i.e.

$$N_{le} = N_{dep} - N_{den} - N_{upt} - N_{imm} - N_{ret} - N_{exp} \tag{3}$$

where N_{dep} is the total N deposition, N_{den} is N lost by denitrification, N_{upt} is the net growth uptake of N by the forests in the catchment, N_{imm} is N immobilized in the terrestrial catchment, N_{ret} is N retained in the lake, and N_{exp} is the organic N export out of the catchment.

Thus, the present exceedance of acidity can be expressed as

$$Ex(Ac) \;\; = \;\; S^\star_{dep} + N_{le} - BC^\star_{dep} - CL(Ac) \tag{4}$$

where $S^\star_{dep}$ is the total S deposition. That equation rests on the assumption that sulfate is a completely mobile anion and the S deposition itself can be used to indicate the acidifying effect of S.

2.3 First-order Acidity Balance (FAB) model

2.3.1 Acidity balance

The *present* exceedance of the critical load of acidity can be assessed without calculating the different (rate-limited) sink processes separately (Eqs.3,4). However, the prediction of future conditions requires the quantification of all rate-dependent N and S processes. The methodology for including such processes in the critical load and exceedance calculations for lakes has been presented by Posch *et al.* (1993) and later extended in Kämäri *et al.* (1992).

For the sum of S and N deposition the following acidity balance can be derived (Kämäri *et al.* 1992):

$$
\begin{aligned}
N_{dep} + S^\star_{dep} \;\; &= \; fN_{upt} + (1-r)(N_{imm} + N_{den}) + N_{exp} + rN_{ret} + fS_{upt} + rS_{ret} \\
&\;\; + BC^\star_{dep} + (1-r)BC_w - fBC_{upt} - Alk_{le}
\end{aligned}
\tag{5}
$$

where f is the fraction of forested land in the catchment area, r is the lake:catchment area ratio, S_{upt} is the net growth uptake of S in the terrestrial catchment, S_{ret} is the in-lake retention of sulfate, BC_w is the weathering of base cations, BC_{upt} is the net growth uptake of base cations, and Alk_{le} is the alkalinity leaching. Note that $(1-r)$ refers to the terrestrial catchment area fraction, which limits the influence of N_{imm}, N_{den} and BC_w to the terrestrial area only.

2.3.2 Description of rate-dependent processes

A simple description for the rate of denitrification is (De Vries *et al.* 1992):

$$
N_{den} \;\; = \; \begin{cases} f_{de}(N_{dep} - N_{imm} - N_{upt}) & \text{for forested land} \\ f_{de}(N_{dep} - N_{imm}) & \text{for open land} \end{cases}
\tag{6}
$$

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

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

$$r N_{ret} = \rho_N \big(N_{dep} - f N_{upt} - (1-r)(N_{imm} + N_{den}) \big) \tag{7}$$

where the factor ρ_N is modeled by a kinetic equation (Kelly et al. 1987):

$$\rho_N = \frac{s_N}{z/\tau + s_N} = \frac{s_N}{Q/r + s_N} \tag{8}$$

where z is the mean lake depth, τ is the lake's residence time, and s_N is the net mass transfer coefficient for N.

Inserting the expression for soil denitrification (6), in-lake N-retention (7) and an analogous equation for the in-lake S-retention ($\rho_S = s_S/(Q/r + s_S)$) in Eq.5 one obtains

$$
\begin{aligned}
a_N N_{dep} + a_S S_{dep}^\star &= b_1 N_{upt} + b_2 N_{imm} + N_{exp} + b_3 S_{upt} \\
&\quad + BC_{dep}^\star + (1-r)BC_w - f BC_{upt} - Alk_{le}
\end{aligned} \tag{9}
$$

where the dimensionless constants a_N, a_S, b_1, b_2 and b_3 are all smaller than one and depend on ecosystem properties only:

$$a_N = (1 - f_{de}(1-r))(1 - \rho_N) \tag{10a}$$

$$a_S = 1 - \rho_S \tag{10b}$$

$$b_1 = f(1 - f_{de})(1 - \rho_N) \tag{10c}$$

$$b_2 = (1-r)(1 - f_{de})(1 - \rho_N) \tag{10d}$$

$$b_3 = f(1 - \rho_S) \tag{10e}$$

Note that a_N and a_S are the fractions of N and S deposition, respectively, that contribute to acidification.

2.3.3 Critical base cation leaching

The amount of acidity that can be neutralized in the catchment, depends on the flux of base cations (ANC). When sufficient information is available, the regional distribution of weathering rates can be estimated with a model based on soil chemistry (Sverdrup 1990). An alternative approach is to estimate the net base cation leaching from water quality data by the steady-state model introduced by Henriksen (see above). In this formulation

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the quantity $BC^\star_{dep} + (1 - r)BC_w - fBC_{upt}$ can be approximated by the pre-acidification leaching of base cations ($Q[BC]^\star_0$) from the catchment area. Furthermore, replacing the critical alkalinity leaching, $Alk_{le,crit}$, by $Q[ANC]_{limit}$, where $[ANC]_{limit}$ is the tolerance criterion for the aquatic organisms considered (Henriksen $et\ al.$ 1990b), one obtains for the critical base cation leaching:

$$BC_{le,crit} = Q\left([BC]^\star_0 - [ANC]_{limit}\right) . \tag{11}$$

2.3.4 Critical load of N and S

Inserting a critical base cation leaching (Eq.11) into Eq.9, we get the following equation for the critical loads of N and S:

$$a_N CL(N) + a_S CL(S) = b_1 N_{upt} + b_2 N_{imm} + N_{exp} + b_3 S_{upt} + BC_{le,crit} \tag{12}$$

Note that in this formulation individual critical loads of N and S are not specified; each *pair* of depositions ($N_{dep}, S^\star_{dep}$) fulfilling Eq.12 is a valid (pair of) critical loads of N and S. The critical loads of N and S are limited by the following constraints, resp.:

$$CL(N) \leq \left(b_1 N_{upt} + b_2 N_{imm} + N_{exp} + BC_{le,crit}\right)/a_N =: CL_{max}(N) \tag{13a}$$

and

$$CL(S) \leq \left(b_3 S_{upt} + BC_{le,crit}\right)/a_S =: CL_{max}(S) \tag{13b}$$

Furthermore, if

$$N_{dep} \leq \left(b_1 N_{upt} + b_2 N_{imm} + N_{exp}\right)/a_N =: CL_{min}(N) \tag{14a}$$

or

$$S^\star_{dep} \leq b_3 S_{upt}/a_S =: CL_{min}(S) , \tag{14b}$$

all N or S is consumed by uptake (and immobilization) and the other of the two pollutants can be considered alone. The relationships between the depositions and critical loads, derived by the above formulations, is illustrated in Fig. 1. The thick lines in the figure indicate all possible pairs of critical loads of N and S acidity, and the slope of the tilted line is given by a_N/a_S.

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Figure 1. The relationships between the N and S depositions and critical loads. The case for the non-exceedance (N1) and the six cases for exceedance (E1-E6) as well as the five cases for critical load (Z1-Z5) are described in the text.

2.3.5 Exceedance of critical loads

Although we cannot sensibly define individual exceedances for S and N, we can define an **exceedance function** for a given S and N deposition:

$$Ex(S^\star_{dep}, N_{dep}) := \begin{cases} a_S S^\star_{dep} - R_S - BC_{le,crit} & \text{if } N_{dep} \leq CL_{min}(N) \\ a_S S^\star_{dep} + a_N N_{dep} - R & \text{if } S^\star_{dep} > CL_{min}(S) \ \& \ N_{dep} > CL_{min}(N) \\ a_N N_{dep} - R_N - BC_{le,crit} & \text{if } S^\star_{dep} \leq CL_{min}(S) \end{cases}$$

$$(15)$$

where we introduced the following abbreviations

$$R_S \quad := \quad b_3 S_{upt} \tag{16a}$$

$$R_N \quad := \quad b_1 N_{upt} + b_2 N_{imm} + N_{exp} \tag{16b}$$

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and

$$R \; := \; R_S + R_N + \text{BC}_{le,crit} \; = \; b_1 N_{upt} + b_2 N_{imm} + N_{exp} + b_3 S_{upt} + \text{BC}_{le,crit} \qquad (16c)$$

If $Ex > 0$, the deposition of N and S leads to an acidity leaching above the critical leaching (i.e. too low alkalinity leaching), and we say that the critical load of N and S is exceeded. In Fig. 1 isolines of constant exceedance are shown as dashed lines. If $Ex < 0$, we call this non-exceedance. This case is depicted as the dark shaded area in Fig. 1. If $Ex = 0$, the N and S deposition form a pair of critical loads (the thick lines in Fig. 1). Note, that for $N_{dep} \leq CL_{min}(N)$ or $S^\star_{dep} \leq CL_{min}(S)$ the deposition of N and S and their critical loads· can be considered alone.

In Fig. 1 example deposition scenarios for exceedance and non-exceedance are indicated by the black dots: N1 indicates a pair of S and N deposition causing no exceedance. The dots labeled Z1,...,Z5 all represent scenarios leading to $Ex = 0$. The dots E1-E6 illustrate different possibilities for exceedance: At E2 a reduction in S deposition below $CL_{max}(S)$ avoids exceedance, while at E6 the same is true for N. Also at E3 a minimum reduction of S deposition below $CL_{max}(S)$ is necessary, but further reductions in S and/or N are needed to get below the thick line; the same reasoning holds for E5 with respect to N. At E4 both minimum reductions in S and N are needed to get below $CL_{max}(S)$ and $CL_{max}(N)$, respectively. Finally, at E1 – or, more general, within the light grey shaded region – we have $Ex > 0$, but non-exceedance can be achieved by different combinations of S and N deposition reductions: Reducing only N leads to Z1, reducing S only brings the dot to Z3, while, e.g., Z2 is reached by reducing both S and N. In fact, non-exceedance can be also achieved by *increasing* the deposition of one of the pollutants, while at the same time heavily reducing the other one (e.g. to reach Z4 or Z5).

Note, that the exceedance calculated by Eq.15 is, in general, *not* the amount by which deposition (i.e. the sum of N and S deposition) has to be reduced in order to achieve non-exceedance. This can be seen as follows: Let $X < CL_{max}(S)$ and $Y < CL_{max}(N)$ be the S and N deposition causing a positive exceedance $Ex = a_S X + a_N Y - R > 0$ (light grey shaded region in Fig. 1); if we now reduce the depositions by this amount, i.e. we take $X' = X - \lambda Ex$ and $Y' = Y - (1 - \lambda)Ex$, where $0 \leq \lambda \leq 1$ is the fraction of Ex allocated to S-reduction, then the new exceedance is given by $Ex' = a_S X' + a_N Y' - R = $ ... $= \left(1 - a_S \lambda - a_N(1 - \lambda)\right) Ex > 0$, since $a_S < 1$ and $a_N < 1$.

However, let $S^\star_{dep}$ and N_{dep} be S and N depositions so that $Ex(S^\star_{dep}, N_{dep}) > 0$; then we have the following cases for the required *reductions* of S and N (called S_{red} and N_{red}, respectively) in order to achieve non-exceedance:

(1) $S^\star_{dep} > CL_{max}(S)$ and $N_{dep} < CL_{min}(N)$ (case E2 in Fig. 1):
In this case we have $S_{red} = S^\star_{dep} - CL_{max}(S)$ and $N_{red} = 0$ (actually, one could increase N_{dep} to $CL_{min}(N)$ without 'endangering' the ecosystem)

(2) $S^\star_{dep} < CL_{min}(S)$ and $N_{dep} > CL_{max}(N)$ (case E6 in Fig. 1):
In this case we have $S_{red} = 0$ (and an increase of $S^\star_{dep}$ to $CL_{min}(S)$ would be 'allowable') and $N_{red} = N_{dep} - CL_{max}(N)$.

(3) $S^\star_{dep} > CL_{min}(S)$ and $N_{dep} > CL_{min}(N)$:
In this case the exceedance is given by $Ex = a_S S^\star_{dep} + a_N N_{dep} - R > 0$. The reductions have to be chosen in such a way, that the exceedance function (Eq.15) becomes zero, i.e.

$$a_S(S^\star_{dep} - S_{red}) + a_N(N_{dep} - N_{red}) - R = 0$$

or

$$a_S S_{red} + a_N N_{red} = Ex \tag{17}$$

Eq.17 has to be fulfilled for every selected reduction scheme. In addition, the reductions have to obey the following obvious constraints:

$$CL_{min}(S) \leq S^\star_{dep} - S_{red} \leq CL_{max}(S) \tag{18a}$$

and

$$CL_{min}(N) \leq N_{dep} - N_{red} \leq CL_{max}(N) \tag{18b}$$

Selecting σ with $0 \leq \sigma \leq 1$ and

$$S_{red} = S^\star_{dep} - \sigma CL_{max}(S) - (1 - \sigma)CL_{min}(S) \tag{19a}$$

we cover the whole range of possible S reductions given by Eq.18a. For $\sigma = 0$ we get the maximum S reduction $S_{red} = S^\star_{dep} - CL_{min}(S)$ (reaching Z5 in Fig. 1), whereas for $\sigma = 1$ we get the minimum S reduction $S_{red} = S^\star_{dep} - CL_{max}(S)$ (reaching Z4 in Fig. 1).
Inserting Eq.19a into Eq.17 yields after some calculations

$$N_{red} = N_{dep} - (1 - \sigma)CL_{max}(N) - \sigma CL_{min}(N) \tag{19b}$$

covering the whole range of possible N reductions given by Eq.18b (maximum N reduction for $\sigma = 1$ and minimum N reduction for $\sigma = 0$).

The reductions in S and N deposition, calculated by Eq.19a and Eq.19b, resp., can be positive (i.e. real reductions) or negative (i.e. increases), depending on $S^\star_{dep}$ and N_{dep}.

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If $S_{dep}^{\star} > CL_{max}(S)$ (cases E3 and E4 in Fig. 1), we have $S_{red} > 0$ for every choice of σ; and for N we have $N_{red} > 0$ for all σ, if $N_{dep} > CL_{max}(N)$ (cases E4 and E5 in Fig. 1). If $S_{dep}^{\star} \leq CL_{max}(S)$ and $N_{dep} \leq CL_{max}(N)$ (case E1, light-grey shaded region in Fig. 1), one can chose σ in such a way that either $S_{red} < 0$ or $N_{red} < 0$.

As an example, let us look at case E1 in Fig. 1: With $\sigma = 0$ we reach Z5 with $S_{red} = S_{dep}^{\star} - CL_{min}(S) > 0$ and $N_{red} = N_{dep} - CL_{max}(N) < 0$. And with $\sigma = 1$ we reach Z4 with $S_{red} = S_{dep}^{\star} - CL_{max}(S) < 0$ and $N_{red} = N_{dep} - CL_{min}(N) > 0$. The points Z1 and Z3 represent the special case were either $S_{red} = 0$ or $N_{red} = 0$, resp. The respective σ-values can be easily computed from Eqs.19a,b. To reach Z1 ($S_{red} = 0$) one obtains $\sigma = (S_{dep}^{\star} - CL_{min}(S))/(CL_{max}(S) - CL_{min}(S))$ and $N_{red} > 0$ is obtained by inserting this σ into Eq.19b. Similarly, to reach Z3 ($N_{red} = 0$) one obtains $\sigma = (CL_{max}(N) - N_{dep})/(CL_{max}(N) - CL_{min}(N))$ and $S_{red} > 0$ is obtained by inserting this σ into Eq.19a. These two cases are of importance when one looks for the amount to be reduced for one pollutant, while the deposition of the other one is kept at a constant level.

Finally, the shortest distance to reach the line $Ex = 0$ is in this case obtained by moving to the point Z2. However, this is *not* the minimum overall reduction, i.e. the minimal $S_{red} + N_{red}$. Since $a_N < a_S$ for practically all ecosystems, it follows that $CL_{min}(S) + CL_{max}(N) > CL_{max}(S) + CL_{min}(N)$, i.e. the minimum reduction is always at Z5 (and the maximum at Z4). This is also intuitively clear, since this type of ecosystems can 'absorb' more N than S (due to the uptake and retention processes).

As mentioned above, Eq.17 holds for every reduction in S and N deposition. Since $a_S \leq 1$ and $a_N \leq 1$ it follows that Ex is a lower bound for the overall reduction $S_{red} + N_{red}$ (but not sufficient for achieving non-exceedance). This lower bound can be made sharper by observing that $a_N \leq a_S$: dividing both sides in Eq.17 by a_S we get

$$ S_{red} \; + \; N_{red} \;\; \geq \;\; S_{red} \; + \; \frac{a_N}{a_S} N_{red} \;\; = \;\; \frac{Ex}{a_S} \;\; =: \;\; Red_{min} \qquad (20) $$

This lower bound Red_{min} ($\geq Ex$) is the minimum reduction required to achieve non-exceedance. On the other hand, dividing by a_N we obtain

$$ S_{red} \; + \; N_{red} \;\; \leq \;\; \frac{a_S}{a_N} S_{red} \; + \; N_{red} \;\; = \;\; \frac{Ex}{a_N} \;\; =: \;\; Red_{max} \qquad (21) $$

showing that Red_{max} is the maximum reduction required to achieve non-exceedance. The actual value of $S_{red} + N_{red}$ will depend on which pollutant is reduced to which extent.

2.3.6 Critical load for eutrophication

In addition to acidification, an upper limit on the N deposition has to be set in order to avoid eutrophication. The mass balance for N reads

$$N_{dep} = fN_{upt} + (1-r)(N_{imm} + N_{den}) + N_{exp} + rN_{ret} + N_{le} \qquad (22)$$

where N_{le} stands for the N leaching. Adverse effects of inorganic N are considered to develop at concentrations exceeding $N_{le,crit}/Q$, where $N_{le,crit}$ is a critical leaching of N. Inserting the expressions for denitrification (Eq.6) and in-lake N retention (Eq.7) one obtains for the critical load of N related to eutrophication

$$CL_{eut}(N) = (b_1 N_{upt} + b_2 N_{imm} + N_{exp} + N_{le,crit})/a_N \qquad (23)$$

and the dimensionless factors b_1, b_2 and a_N are given by Eqs.10. Eq.23 sets an upper limit on the N deposition, however, in this report the critical N load to avoid eutrophication is not considered.

2.4 Separate critical loads of S and N and their exceedance

The international negotiations on emission reductions within the framework of the ECE have so far concentrated on producing separate protocols for S and N oxides. Therefore, much effort has been put on determining unique critical loads of these compounds. One way is to fix the σ-value in Eqs.19 above. In doing so we obtain for the critical load of S $(CL(S) = S^\star_{dep} - S_{red})$:

$$CL(S) := (b_3 S_{upt} + \sigma_f BC_{le,crit})/a_S \qquad (24)$$

where b_3 and a_S are given by Eqs.10; and for the critical load of N one obtains:

$$CL(N) := (b_1 N_{upt} + b_2 N_{imm} + N_{exp} + (1 - \sigma_f)BC_{le,crit})/a_N \qquad (25)$$

where the dimensionless factors b_1, b_2 and a_N are given by Eqs.10.

In the above critical load formulations the factor σ_f accounts for the fraction of base cations (ANC) balancing S input, and $1 - \sigma_f$ for the fraction balancing acidifying N input. Consequently, this factor is called the sulfur-factor, and its magnitude will depend on the fraction of S of the total acidic input (Hettelingh *et al.* 1991, Posch *et al.* 1993). Therefore – with S_{net} and N_{net} denoting the net fluxes of S and N to the lake – we define σ_f in the following way:

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$$\sigma_f \ = \ \frac{S_{net}}{S_{net} + N_{net}} \ . \tag{26}$$

The net fluxes of N and S are derived in the same way as the critical loads above; one obtains respectively

$$N_{net} \ = \ \big(1 - f_{de}(1-r)\big)N_{dep} \ - \ (1 - f_{de})\big(fN_{upt} + (1-r)N_{imm}\big) \tag{27}$$

and

$$S_{net} \ = \ S_{dep}^{\star} \ - \ fS_{upt} \ . \tag{28}$$

Obviously, negative values of N_{net} and S_{net} have to be set to zero, because in steady-state the system cannot take up more than there is supplied by deposition.

The disadvantage with this approach is that the critical loads of S and N become deposition-dependent, and this contradicts the notion of a critical load being solely a property of the ecosystem. As a consequence, the presentation of critical load values alone does not provide all of the information on the ecosystem's status. Only the comparison with the deposition (scenarios) of S and N gives a full picture of the present (and future) status of the ecosystem. This comparison is done by computing exceedances of these critical loads, defined as the differences between the deposition and the critical load of the respective pollutant (see Kämäri et al. 1992). It should be noted that exceedances computed in this way differ from the exceedance function defined in Eq.15.

3. Derivation of Inputs

3.1 Selection of sites

The data for calculating and mapping critical loads and their exceedances were largely obtained from national lake surveys conducted to assess the acidification status of surface waters, carried out in Finland (Forsius *et al.* 1990, Kämäri *et al.* 1991), Norway (Henriksen *et al.* 1988) and Sweden (Bernes 1991). Short descriptions of these surveys, and other data bases on which the critical load calculations were based, are given below.

3.1.1 Finland

For Finland the surveys of 1450 lakes conducted in 1987-1989 form the basis for assessing critical loads for Finnish surface waters. The spatial distribution of the lake data set reflects the actual lake density in different regions, and therefore no data were available for a few grids having very low number of lakes. In 1987, a statistical survey of 970 lakes was carried out (Kämäri *et al.* 1991). During the years 1987-1989 a total of 480 additional lakes were surveyed by the Lapland Water and Environment District.

The lakes for the 1987 lake survey were selected from two separate subregions, Subregion 1 (southern and central Finland) and Subregion 2 (northern Finland). The statistical sampling "frame" in each subregion consisted of a series of 1:50,000-scale topographic maps (20×30 km^2). Due to the uncertainty of the size distributions included in the lake frame population and the large variations in lake density in the different regions, a two-stage cluster sampling was used (Forsius *et al.* 1990). All basins considered as lakes with a surface area within the pre-defined size range were numbered, and 8 lakes were randomly sampled from each map. If the number of lakes on the map was 8 or less, all lakes were sampled. The lakes sampled represents ca. 2% of the total number of lakes in the size range $0.01 - 10$ km^2 in Subregion 1, and 5% of the lakes with surface areas $0.1 - 10$ km^2 in Subregion 2.

3.1.2 Norway

For Norway, a grid net of 0.5° latitude by 1° longitude subdivided into 16 subgrids was used as the basis for the selection of lakes and rivers. One lake or stream was selected to be representative for each grid from the 1:50,000 map series, based on subjective judgment. Three sources of data were used when selecting a site for each grid: First the lakes sampled within the 1000 Lake Survey conducted in 1986 were chosen whenever possible. Secondly additional lakes from the southern parts of Norway were sampled in fall of 1989 and 1990 for those grids not covered by the 1000 Lake Survey lakes. Thirdly, for the northern counties additional 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 thus 2305.

If there were wide variations in lake chemistry within the grid, a lake in the most sensitive area was selected to represent the subgrid, if it amounted to more than 25% of the grid's area. Sensitivity was evaluated on the basis of water chemistry, topography and bedrock geology. Thus, the Norwegian database for critical load calculations was based on the assumption that the water chemistry assigned to each subgrid is representative for the grid area. This approach is justified for Norway, because lakes are fairly evenly distributed throughout the country.

In the Norwegian 1000 Lake survey, water samples were collected during fall overturn 1986 from 1005 lakes throughout Norway (Henriksen *et al.* 1988). The lakes selected were located in areas underlain by types of bedrock expected to give runoff waters having low buffering capacities, such as granites, gneisses and migmatites. 305 of the lakes located in southern Norway had been sampled in the fall of 1974 and 1975, as part of the original survey of lakes conducted by the SNSF-project (Overrein *et al.* 1981). Most of the lakes sampled were larger than 0.2 km^2. The lakes had no appreciable direct human influence, were located in headwater areas and were not affected by local sources of pollution. Almost half of the lakes were selected from counties most affected by acidification.

3.1.3 Sweden

All Swedish lakes with an area greater than one hectare constituted the basis for selection for the 1990 lake survey (Bernes 1991). Some exceptional lake types were excluded: (a) overgrown or very shallow lakes (water depth less than 1 m), (b) certain dams used as reservoirs for industries or receiving bodies for sewage water, and (c) certain dams used for the generation of hydroelectric power. In order to obtain a reasonable geographic distribution the lake register was stratified based on county, and within each county the lakes were classified according to area into size classes $0.01 - 1$ km^2, $10 - 100$ km^2 and > 100 km^2. From each size class a pre-defined percentage of the number 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 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, the period representing winter situation. For the calculations presented in this paper a statistically representative subset of 760 lakes, which had been analyzed for N compounds and are not influenced by liming, was used.

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3.2 Concentrations of base cations and strong acid anions

The different methods for calculating critical loads of N and S require information on the present lake water concentrations of base cations, sulfate and N compounds. This information was available from the above survey data bases. Standard analytical methods have been used on all samples (Henriksen *et al.* 1988, Forsius *et al.* 1990, Bernes 1991)

3.3 Concentrations of organic N

For Finland, the organic N concentration was estimated for most lakes as the differences between the total N and the sum of inorganic N species. However, for 153 lakes in northern Finland the organic N was estimated by a linear regression from the chemical oxygen demand (COD) on the basis of data of 317 lakes in Lapland. For these lakes the equation for organic N (in μeq l^{-1}) as a function of COD (in mg l^{-1}) reads

$$\text{N}_{org} = 1.5COD + 9.0 \qquad (n = 317, r = 0.74) \tag{29}$$

For Norway organic N was estimated by linear regression from TOC, based on data from the Norwegian monitoring program

$$\text{N}_{org} = 2.0TOC + 4.2 \qquad (n = 453, r = 0.80) \tag{30}$$

For Sweden, the organic N was estimated as the differences between the total N and the sum of inorganic N species. The regression between organic N and TOC has fairly similar coefficients as in the Norwegian data base

$$\text{N}_{org} = 1.9TOC + 8.4 \qquad (n = 988, r = 0.57) \tag{31}$$

3.4 Background concentrations of sulfate

The background (pre-acidification) sulfate concentration (in μeq l^{-1}) was estimated from the relationship between $[SO_4^{2-}]_t^*$ and $[BC]_t^*$ from 251 Finnish, Norwegian and Swedish lakes located in northern regions, which receive the lowest acidic deposition in these countries:

$$[SO_4^{2-}]_0^* = 0.078[BC]_t^* + 18.75 \qquad (n = 251, r = 0.66) \tag{32}$$

This equation suggests that there is an atmospheric background contribution of sulfate and a geological contribution which is proportional to the concentration of base cations (see Fig. 2 and Henriksen *et al.* 1990a). However, although these lakes are located in remote

northern areas, they may still receive slightly elevated levels of acidic deposition. It was therefore decided to use as value for $[SO_4^{2-}]_0^*$ the mean value (Eq.32) minus one standard deviation (thick line in Fig. 2, see Posch *et al.* 1993). Only $[SO_4^{2-}]_0^*$-values less than the present-day sulfate concentrations $[SO_4^{2-}]_t^*$ were accepted.

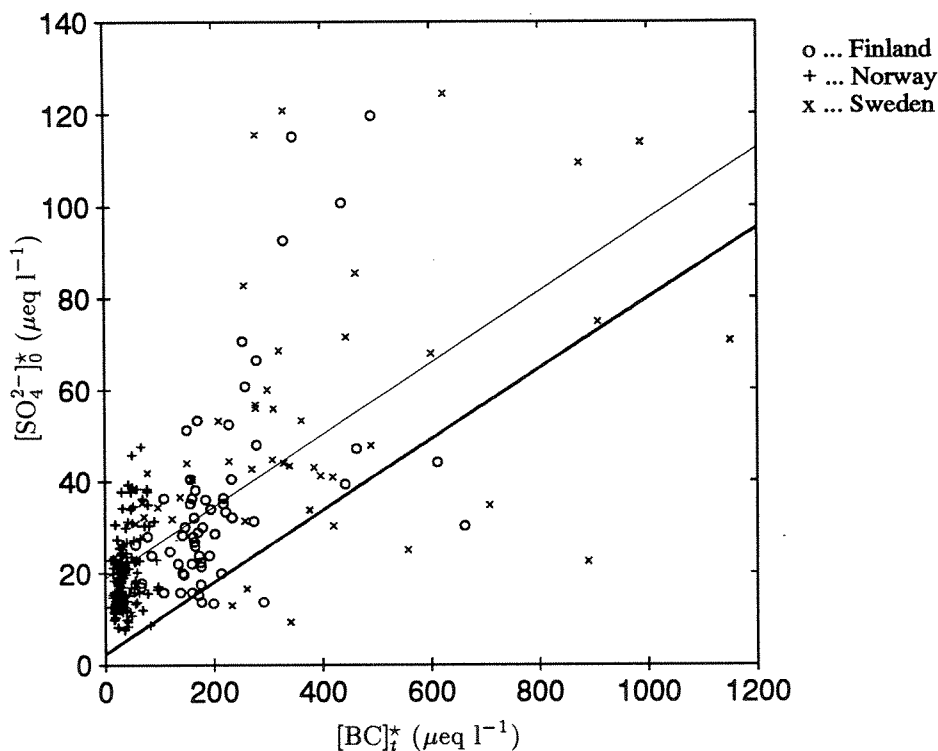*Figure 2. Background sulfate concentration as a function of base cation concentration. The thin line shows the regression equation (Eq.32) and the thick line the regression minus one standard deviation used for computing the background sulfate concentration.*

3.5 F-factor

The value of F (see Eq.2) 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 *et al.* 1990; Marmorek *et al.* 1990). In this study we have used the following non-linear relationship between F and the background base cation concentration

$$F = 1 - \exp(-[BC]_0^*/B) \tag{33}$$

where B is a scaling factor. Inserting this expression for F into Eq.2 gives a non-linear equation for $[BC]_0^*$ which is solved by an iterative procedure.

The parameter B requires the knowledge of the pre-acidification 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 (see Posch *et al.* 1993). The median value of this distribution, 131 μeq l^{-1}, was used for the calculations in this paper.

3.6 Chemical criterion

Acid Neutralizing Capacity (ANC), defined as the difference between non-marine base cations and strong acid anions, appears to be a suitable chemical criterion for sensitive indicator organisms in surface waters (Henriksen *et al.* 1990b; Lien *et al.* 1992):

$$[ANC] = [BC]^\star - [SO_4^{2-}]^\star - [NO_3^-] \tag{34}$$

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 *et al.* 1988, 1989), and these data have been used to calculate the probability for fish damage at different levels of ANC (Lien *et al.* 1992). According to these data an $[ANC]_{limit}$ of 20 μeq l^{-1} seems to be appropriate for calculating the critical load for freshwater fish, and this value was used also in the present study.

3.7 Mass transfer coefficients

For all three countries 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 m yr^{-1}, was used for the critical load calculations. The value for s_N, 5 m yr^{-1}, was obtained from a similar study of 12, mostly Canadian, lakes (Dillon and Molot 1990).

3.8 Lake to catchment area ratios

For Finnish lakes both lake and catchment areas were estimated from topographic maps (1:50 000). Similarly, for Norwegian 1000 Lake Survey lakes the lake and catchment areas were estimated from topographic maps (1:50 000); for the remaining Norwegian lakes the median ratio ($r = 0.072$) in the 1000 Lake Survey data base was used. For Sweden, the ratio was estimated from a regression between lake and catchment areas for 3343 limed lakes located in different parts of the country. The regression equations for the Finnish, Norwegian and Swedish data sets are, respectively

$$r = 0.0881 A_l^{0.1864} \qquad (n = 1441, r = 0.83) \tag{35}$$

$$r = 0.0728 A_l^{0.2281} \qquad (n = 1007, r = 0.59) \qquad (36)$$

$$r = 0.0772 A_l^{0.1829} \qquad (n = 3343, r = 0.75) \qquad (37)$$

3.9 Runoff

The annual runoff for Finnish lakes was obtained by interpolation from a national runoff map available at the Hydrological office of the Water and Environment Research Institute. For Norwegian lakes, the mean annual runoff values were read manually from the national runoff maps produced by the National Board for Water and Electricity. For Swedish lakes the runoff values were determined from a modified digitized runoff map available at the Swedish Meteorological and Hydrological Institute.

3.10 Nutrient uptake by forests

For Finland, the net uptake of N by forests (N_{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 the Norwegian critical loads data base one value for N_{upt} has been assigned to each grid. These values are based on inventory results provided by the Norwegian Forest Research Institute (NISK). The information on forest coverage was obtained from the 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, compiled by the Department of Forest Soils, Swedish Agricultural University (Rosén et al. 1992). The forest fraction f was obtained from a data base of Lövblad et al. (1992).

For all countries, the net uptake of S (S_{upt}) in the catchment area was considered negligible and thus set to zero.

3.11 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 considered to be low. However, high values may occur in areas with peatsoils (Klemedtsson and Svensson 1988, De Vries et al. 1992).

For Finland, information on land types was available for the lakes surveyed in 1987 for 978 catchments. For these catchments two different denitrification fractions were used (De Vries *et al.* 1992): 0.8 for peat soils and 0.1 for other soil types. The actual denitrification fraction was then computed by linear interpolation according to

$$f_{de} = 0.1 + 0.7 f_{peat} \tag{38}$$

where f_{peat} is the fraction of peatlands in the catchment area. For the remaining Finnish lakes, located mainly in Lapland, the peatland fraction was estimated from COD values (in mg l^{-1}) of Lappish lakes by linear regression

$$f_{peat} = 0.025 \, COD + 0.051 \quad (n = 189, r = 0.64) \tag{39}$$

For Norwegian and Swedish lakes no catchment-specific land-type information was available. Therefore, values for the peatland fraction were obtained from a non-linear relationship between TOC (in mg l^{-1}), f_{peat} and latitude (λ) in the Finnish data set (see Fig. 3)

$$TOC = a \, f_{peat}^{b} \left(\frac{60°}{\lambda}\right)^{c} \quad (n = 978) \tag{40}$$

with

$$a = 35.53 \text{mg } l^{-1} \quad b = 0.2939 \quad c = 7.856 \tag{41}$$

For the Norwegian and Swedish regions south of Finland the values for $\lambda = 60°$ were used.

Values for the long-term immobilization of N in forest soils (N_{imm}) have been estimated by Rosén by studying chronosequences (see Rosén *et al.* 1992). N_{imm} was estimated from the total N content in 16 Swedish forest soil profiles, and considering only stable C-N-compounds N_{imm} was in the range $0.2 - 0.5$ kgNha^{-1}a^{-1}. The latter value was used for the calculations in this study.

3.12 Deposition

Deposition estimates of S, reduced and oxidized N compounds in Finland were obtained from model calculations of the Finnish acidification model HAKOMA (Johansson *et al.* 1990) for the year 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. Base cation deposition estimates (bulk precipitation) were obtained from the monitoring network run by the Water and Environment Research Institute (Järvinen and Vänni 1990). Catchment-specific values were interpolated from the three nearest stations by bilinear interpolation.

For Norway and Sweden land-use weighed deposition of S, total N and base cations estimated by Lövblad *et al.* (1992) was used. Total deposition had been calculated as the sum of measured wet deposition and estimated dry deposition on 50 × 50 km^2 NILU-grid squares. Dry deposition of S and N was estimated by multiplying interpolated monitored air concentrations with annual mean deposition velocities in each grid. Deposition to spruce, pine and deciduous forests, open fields and lake surfaces were considered.
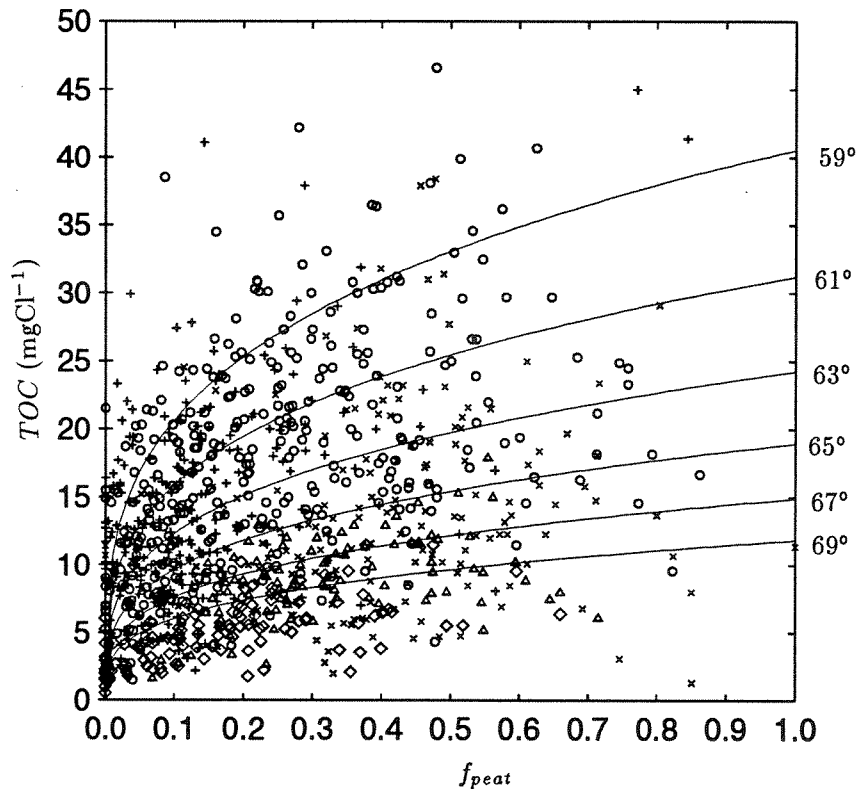

Figure 3. TOC versus peatland fraction for 978 Finnish lakes. The data for different latitudinal stripes are distinguished by the following symbols: plus-signs for $\lambda < 62°$, circles for $62° \leq \lambda < 64°$, ×-es for $64° \leq \lambda < 66°$, triangles for $66° \leq \lambda < 68°$, and diamonds for $\lambda \geq 68°$. The curves show the non-linear regression (Eq.40) between latitude $\lambda = 59°$ and $\lambda = 69°$ in steps of 2°.

4 Results and Discussion

4.1 Nitrate leaching

The most important question concerning the response of catchments to air-borne 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 a more or less open cycle, where the excess N is leached from the system, is often referred to as N saturation. The development of N saturation by increased N inputs involves a complex interaction of the processes in the N cycle. The knowledge on these processes is continuously increasing (see Grennfelt and Thörnelöf 1992).

The fate of NO_3^- is key in determining the effects of the N-cycle on catchment acidification. If nitrification rates or supply rates of inorganic N from external sources exceed the ecosystem's assimilation rate, then NO_3^- leaching occurs. Nitrate leaching results in a net cumulative production of H^+ that may be manifested as higher soil water concentrations of ionic Al, increased base cation export in runoff, declines in runoff alkalinity, etc., i.e. ecosystem acidification. While there have been reported cases of catchment leaching of NO_3^- caused by an imbalance between nitrification and assimilation rates (e.g. Van Miegroet and Cole 1984), most cases are attributed to excessive N supply from atmospheric sources (Van Breemen et al. 1982). Ecosystem leaching of NO_3^- signals that N contributes to the ecosystem acidification, usually in the form of increased base cation leaching. Nitrate leaching from the lake can therefore be used as an indicator of the acidifying role of N compounds for surface waters.

Low concentrations of both nitrate and ammonium were generally observed in the lakes of Finland, northern Norway and northern Sweden. In these areas the nitrate concentrations were mostly less than 2 μeq l^{-1}. The median of the data sets of the three countries was 1 μeq l^{-1}. In southern Norway and Sweden some 5% of the lakes had nitrate concentrations exceeding 10 μeq l^{-1}. Regarding nitrate leaching, there was a clear difference between southernmost parts and the rest of Fennoscandia. Only in southern Norway and southern Sweden the median of the leached nitrate was more than 10 meq m^{-2} yr^{-1} (Plate I). Elsewhere in Fennoscandia the leaching of nitrate was mostly at background levels. In 95% of the lakes the leaching of nitrate was less than 20% of that of sulfate, although the deposition of the compounds is of the same order of magnitude. Hence, according to these results, it is evident that N compounds *presently* contribute to long-term surface water acidification only in southern parts of Norway and Sweden.

4.2 Critical load of acidity and its present exceedance

The critical load of acidity for surface waters can be estimated using the traditional formula (Eq.1), originally given in the Mapping Manual (ECE, 1990). The results obtained by applying this formula show that the sensitive lakes in all Nordic countries can stand only very small additions of actual acidity (Plate IIa). In fact, some of the critical loads obtained are negative. However, as discussed in the Appendix, no negative critical load values exist in reality. In the SSWC model negative values of critical load of acidity are adjusted to zero by setting the $[ANC]_{limit}$ to $[BC]_0^*$, if $[BC]_0^* < 20$ μeq l^{-1}. The values for the 5th percentile critical load of acidity (Plate II) are generally lower than those given for S acidity in Henriksen et al. (1990a). This is due to the difference in the calculation method: In this report the base cation deposition is subtracted and the variable F is calculated in a slightly different way. In Henriksen et al. (1990a) the values were interpreted as S acidity and the base cation deposition was not subtracted.

When calculating the exceedance, the critical load values for the S acidity given in Henriksen et al. (1990a) were directly compared with the S deposition. The values in Plate IIa can, however, be related to the acidifying loads only by subtracting the base cation deposition also from the loading side. Moreover, when applying the concept of the present exceedance, the leaching of the acidifying elements are used to indicate their acidifying effect from the surface waters' point of view. In Plate IIb sulfate is assumed to be a mobile anion and thus S deposition is used together with nitrate leaching in the calculations of present exceedance (Eq.4).

4.3 Critical loads of S and N and their exceedances

As shown in the description of the FAB model, unique critical loads of N and S, that depend on ecosystem properties only, cannot be specified. Therefore, there are limitations in what can be displayed as critical loads and their exceedances. Displaying the constraints limiting the critical loads of N and S (and their respective exceedances) is one way of roughly illustrating the ecosystems' sensibility to acidifying deposition. Secondly, displaying the minimum and maximum amount of the combined S+N deposition reduction required to achieve non-exceedance illustrates the range within which one has the choice of allocating reductions of S and/or N deposition. Thirdly, the amount by which deposition has to be reduced in order to reach critical loads can be calculated for one pollutant by fixing the deposition level of the other. Fourthly, the question which pollutant *has to be* reduced and which one *can be* reduced in order to reach critical loads can investigated by classifying the present N and S deposition pair for each lake with respect to the exceedance function (cases N1 and E1-E6 in Section 2, see Fig. 1). Finally, if one insists on displaying individual critical loads of N or S it should be noted that these critical loads become deposition-dependent

and they are no longer unique ecosystem properties. In the sequel, examples of the above cases are discussed.

Individual critical loads of N and S that depend on ecosystem properties only, cannot specified; each *pair* of depositions (N_{dep}, S_{dep}^{*}) fulfilling Eq.12 is a valid (pair of) critical loads of N and S. However, in Eqs.13 and 14 the constraints limiting the critical loads of N and S are given. These limits depend on ecosystem properties only and thus give a rough indication of the ecosystems' sensibility to acidifying deposition. While for S it is assumed that S_{upt} is negligible, and thus $CL_{min}(S)$ is zero, $CL_{min}(N)$ is a function of N retention processes and its 5th percentile in each NILU-grid (not shown) lies below 20 meq m^{-2} yr^{-1} almost everywhere in the three countries. The maximum critical loads depend also on the available base cations (ANC) and their 5th percentiles in each NILU-grid are depicted in Plate III. The maps in Plate III indicate a large inter-regional variability ranging from values close to zero, mostly in Norway, to above 100 meq m^{-2} yr^{-1} in many locations in Finland and Sweden.

Not only is it impossible to specify individual critical loads of N and S in the above formulation, but it is in general also not possible to define a critical load of S+N, $CL(S+N)$. Only if $a_N = a_S$, i.e. when denitrification and in-lake N and S retention are neglected (or negligible), one can define a critical load of S+N: Putting $f_{de} = \rho_N = \rho_S = 0$ in Eq.12 (see also Eqs.10a-e) one obtains

$$CL(S+N) \;=\; CL(N) + CL(S) \;=\; fN_{upt} + (1-r)N_{imm} + N_{exp} + fS_{upt} + BC_{le,crit} \quad (42)$$

By subtracting the base cation deposition on both sides of the equation, one obtains the variable that has traditionally been referred to as the critical load of potential acidity (c.f. Hettelingh *et al.* 1991).

Since individual critical loads of N and S, solely dependent on ecosystem properties, cannot be defined, unique exceedances, i.e. unique reductions required, cannot be computed. However, it is possible to derive maximum and minimum reductions required for S+N in order to achieve non-exceedance (see Eqs.20-21). In Plate IV the 95th percentile for these minimum and maximum reductions are mapped for each NILU-grid for the present N and S deposition. These required reductions vary from negative values, i.e. non-exceedance, mainly in central and northern parts of the countries to values above 50 meq m^{-2} yr^{-1} mainly in southern parts of the three countries. From Eq.20-21 it is obvious that the regions of non-exceedance are identical in both maps. Moreover, it should be noted that the maps are fairly similar and thus overall amount of S+N to be reduced is quite narrowly constrained.

Since most lakes allow a certain degree of freedom in the selection of N and/or S to be reduced in order to achieve non-exceedance, unique S and N deposition reductions cannot be determined. The amount by which S and N deposition has to be reduced in order 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 non-exceedance, a fixed scenario for N deposition has to be selected. Fixing N_{dep} in the exceedance function (Eq.15) and calling the resulting S deposition *the critical load of S at* N_{dep}, $CL(S|N_{dep})$, we obtain by setting $Ex = 0$:

$$CL(S|N_{dep}) = \begin{cases} (R_S + BC_{le,crit})/a_S & \text{for } N_{dep} \leq CL_{min}(N) \\ (R - a_N N_{dep})/a_S & \text{for } CL_{min}(N) < N_{dep} \leq CL_{max}(N) \\ -\infty & \text{for } N_{dep} > CL_{max}(N) \end{cases}$$

The $-\infty$ in the above equation indicates that for $N_{dep} > CL_{max}(N)$ no reduction in S deposition suffices to achieve non-exceedance. The required S deposition reduction is simply given by subtracting the conditional critical load $CL(S|N_{dep})$ from $S_{dep}^\star$.

For producing Fig. 4 the N deposition was fixed at the present level which corresponds to the 1988 Sofia Protocol for the control of NO_x emissions. In Fig. 4 the required S deposition reduction is plotted for each country as inverse cumulative distribution function (cdf) both in deposition units and as a percentage of the present S deposition. The percentage of lakes that require a S deposition reduction can be read from the point where the cdf starts from the left vertical axis, varying from 18% of the lakes in Finland to 42% in Norway. The percentage of lakes which cannot be 'protected' by S reductions, and thus necessarily require a N reduction, can be seen from the intersection of the cdfs with the right vertical axes. In Finland only a small number of lakes remains exceeded after strong reductions of S deposition, whereas in Norway, even after a 100% reduction in S deposition, 22% of the lakes remain exceeded and require reductions of N deposition as well.
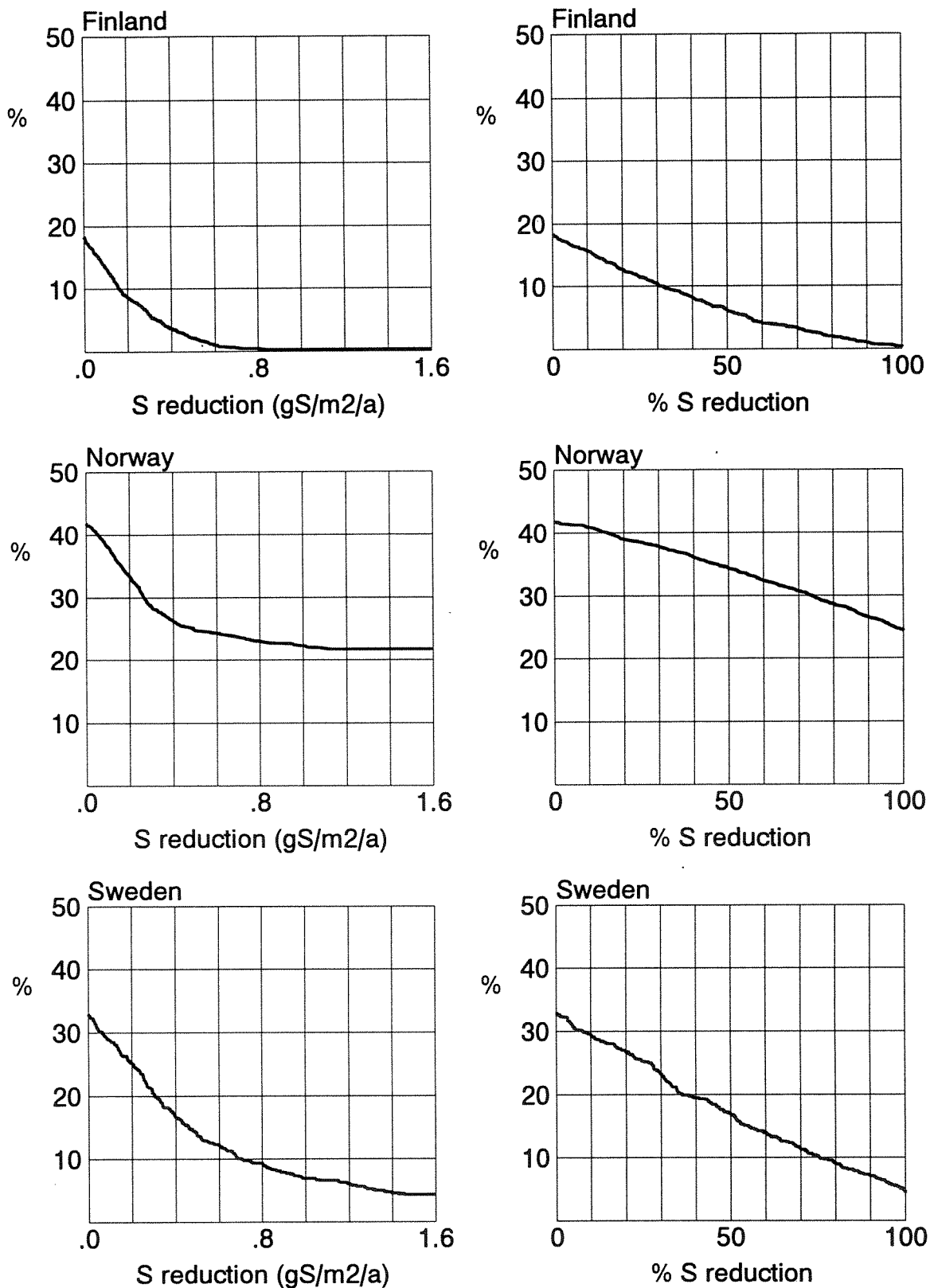
Figure 4. Inverse cumulative distribution functions for each country showing the reductions required for S deposition in absolute deposition units (left) and in percentages from present deposition (right), for the N deposition fixed at present level.

The question which pollutant it has to be reduced in order to reach critical loads can be approached by investigating the present S and N deposition for each lake with respect to the exceedance function. In other words, it can be evaluated on the basis of Fig. 1 which of the six cases, explained in detail on Section 2, the lake represents. In Fig. 5 the percentage of the different cases are plotted for each EMEP-grid. The results demonstrate a clear regional pattern in the exceedance. The critical loads are exceeded in about on third of the study lakes in Fennoscandia, with the greatest proportions in the southernmost parts of Norway and Sweden, where up to 100% of the lakes exceed the critical loads. 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 north-eastern 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%.

On the lower map of Fig. 5 the bars depict the percentages of lakes in each of the five exceedance cases, this time given 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 more 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 thus non-exceedance can in these regions be obtained by reducing S deposition alone (cases E1-E3). Mandatory reductions in N deposition are required only in a quite limited area in southern Sweden and in southern and central Norway. There the lakes 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 non-exceedance (case E4). In Finland only 2% and in Sweden 13% of the exceeded lakes 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 Fig. 5 (lower map) show that 44% of the exceeded lakes in Finland, 18% in Norway, and 6% in Sweden require a mandatory reduction in S deposition only (case E2). A significant proportion of the exceeded lakes in all three countries (Finland: 35%, Norway: 18%, Sweden: 65%) require a mandatory reduction in S deposition in addition to a reduction allocatable to either S or N (case E3). Furthermore, 12-19% of the exceeded lakes in the Nordic countries can be restored by any combination of reductions in S and N depositions (case E1). However, this case is fairly significant in southern Finland and Sweden (even above 50%), leaving a broad range for optimizing emission reductions between S and N.
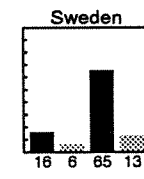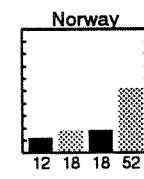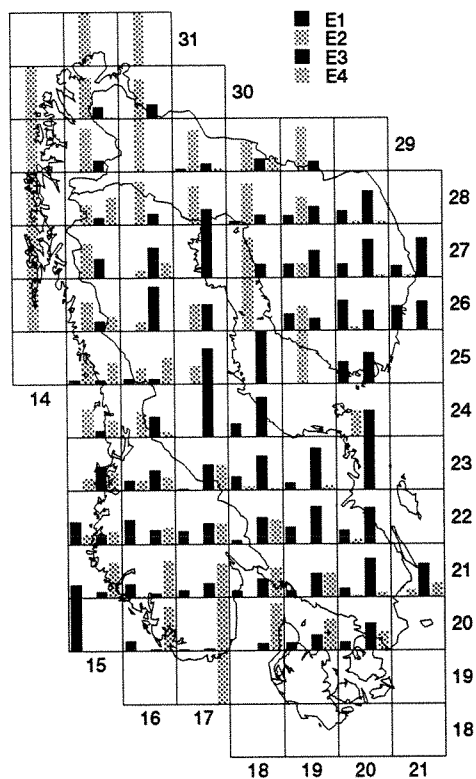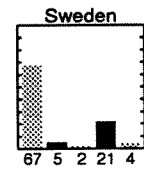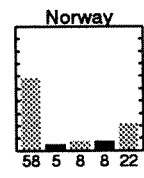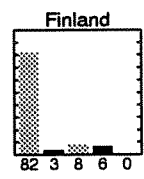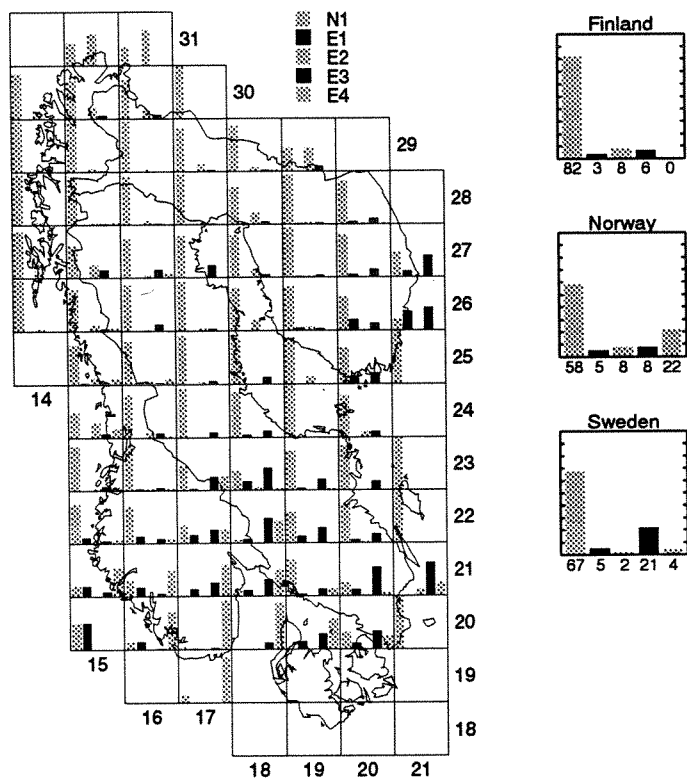
Figure 5. Barcharts depicting the share of lakes in each of the exceedance cases: Including the all lakes (top, cases N1 and E1-E5; see Fig. 1), and only lakes exceeding the critical loads of S and/or N (bottom, Ex(S, N) > 0, cases E1-E5).

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For producing separate critical loads of S and N the so-called sulfur-factor σ_f has been introduced (Eqs.24,25). As pointed out earlier, the disadvantage in doing this is that the critical loads become deposition-dependent, and they are no longer unique ecosystem properties. Obviously, the critical load values for S and N calculated according to Eqs.25,24 using σ_f, given by Eq.26, lie between the values for CL_{min} and CL_{max} for N and S, respectively. The individual critical loads of S and N calculated in this way should be referred to as "critical loads of S and N for a given deposition pair". (see Posch *et al.* 1993; Kämäri *et al.* 1993). Due to the deposition dependence of the sulfur-factor these critical loads have to be re-computed for every deposition scenario and therefore they are only of limited value for evaluating control strategies. The use of a single exceedance function, defined by Eq.15, involves less assumptions than the definition of individual exceedances and enables more straight-forward assessments of deposition scenarios.

5 Conclusions

In this report we have assessed the exceedances of the critical loads of acidifying N and S deposition. The critical loads of N and S are interrelated, and therefore they have to be treated together.

The critical load of actual acidity is less than 20 meq m^{-2} yr^{-1} practically throughout Fennoscandia, and therefore aquatic ecosystems in the Nordic countries can stand only very small loads of acidity. The present exceedance gives an indication of the lake areas currently most threatened by acidic deposition. The highest exceedances are computed for southernmost Norway and Sweden.

In this report a tool is provided for analyzing the exceedance and the reductions needed for N and S depositions. The question 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 an exceedance function. The results show a clear regional pattern in the S and/or N exceedance. Practically in the whole of Finland and in the northern parts of Fennoscandia, the acidification problem could be solved by reducing S deposition only. 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, also N deposition has to be reduced considerably. Reductions in N deposition are necessary in regions where also the present N leaching values are elevated.

The methodology described here allows the simultaneous analysis of N and S reduction requirements by optimization models, computing cost-effective ways to reduce N and/or S deposition. Alternatively, the two pollutants can be assessed separately by fixing the one pollutant to a level defined, e.g., in an earlier international Protocol. We demonstrated this procedure by analyzing the reductions required for S, given the level of N deposition. This iterative procedure additional reductions required 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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Appendix: Negative critical loads

Arne Henriksen

1. Background

When critical loads for surface water are calculated according to the ECE-Mapping manual (ECE, 1990) some of the calculations may turn out as "negative" critical loads values. This is not the case with the methods used for calculating critical loads for soil. Thus, the cause must lie in the method for calculating critical loads for surface waters.

2. The Steady-State Water Chemistry Method

The basis for the Steady State Water Chemistry method is as follows: Acid Neutralization Capacity (ANC) is used at the chemical criterion for sensitive indicator organisms in surface waters. ANC is defined as the difference between non-marine base cations (BC)* and strong acid anions (AN)*:

$$[ANC] = [BC]^* - [AN]^* = [HCO_3^-] + [A^-] - [H^+] - [Al^{n+}] \qquad (A1)$$

where $[HCO_3^-]$ is the hydrogencarbonate concentration, $[A^-]$ is the concentration of organic anions and $[Al^{n+}]$ is the sum of all positively-charged aluminium species.

For surface waters ANC has been selected as the *critical chemical value,* and is set using fish as a *biological indicator.* ANC_{limit} is thus the critical concentration for fish. The use of ANC represents a simplified approach in which numerous interacting factors affecting toxicity to fish, including pH, aluminium and TOC, are lumped into a single variable. Data for water chemistry and change in general fish status have been used for assessing the ANC_{limit} for fish in Norway (Lien *et al.* 1991). Few fish populations are damaged at ANC-concentrations above 20 µeq/l. Of the fish species studied, salmon, brown trout and roach were the most sensitive, and perch the least sensitive. Although ANC_{limit} will depend on the fish species considered, a value of 20 µeq/l seems to be appropriate for evaluation of critical load and critical load exceedance for fresh water fish, at least in Norway and Finland. Canada has set pH 6.0 (corresponds to an ANC range of 20 - 40 µeq/l for clearwater lakes) as a suitable chemical threshold for use in defining critical loads, disgarding those areas with historical pH-values less than 6.0 (Brydges and Summers 1989). This level is set to protect all aquatic biota. Also, Sweden consider pH > 6.0, or ANC = 50 µeq/l as a suitable national threshold value.

The critical load (CL) of acidity for a given indicator organism in the selected surface water is calculated by :

$$CL(Ac) = ([BC]^*_0 - [ANC_{limit}]) \cdot Q - BC^*_{dep} \qquad (A2)$$

where $[BC]^*_0$ is the original leaching of base cations within the catchment area, $[ANC_{limit}]$ is the critical ANC-concentration for the biological indicator considered, Q is the mean annual runoff and BC^*_{dep} is the non-marine base cation deposition.

To compute the critical load, values for BC^*_0 has to be estimated from present-day water chemistry data (see Henriksen et al. 1990).

As pointed out above the ANC_{limit} for a biological indicator has to be set in order to determine a critical load. The calculations then largely depend on the ANC_{limit} chosen. Figure

A1 shows the effect of choosing different values for ANC_{limit}. Here, the frequency distributions of the critical loads calculated for ANC_{limit} = 0, 20 and 50 µeq/l, respectively, are presented for Finland, Norway and Sweden. For Norway an ANC_{limit} of 50 µeq/l would lead to critical loads of zero and less ("negative" critical load) for more than 30% of the area (most of southern Norway). Even an ANC_{limit} of 20 µeq/l will result in zero critical load for nearly 10% of the area of Norway. For Finland and Sweden, however, the change in ANC_{limit} does not have the same effect as for Norway. The reason for this is largely that the runoff is on the average much higher in Norway than in the other countries, which, in areas with slowly weathering soils, leads to a higher number of lakes with a very low concentration of base cations The change in ANC_{limit} will result in a much greater effect on critical load for areas with high runoff than in areas with low runoff. Norway is thus most sensitive of the Nordic countries to the selection of the ANC_{limit}.
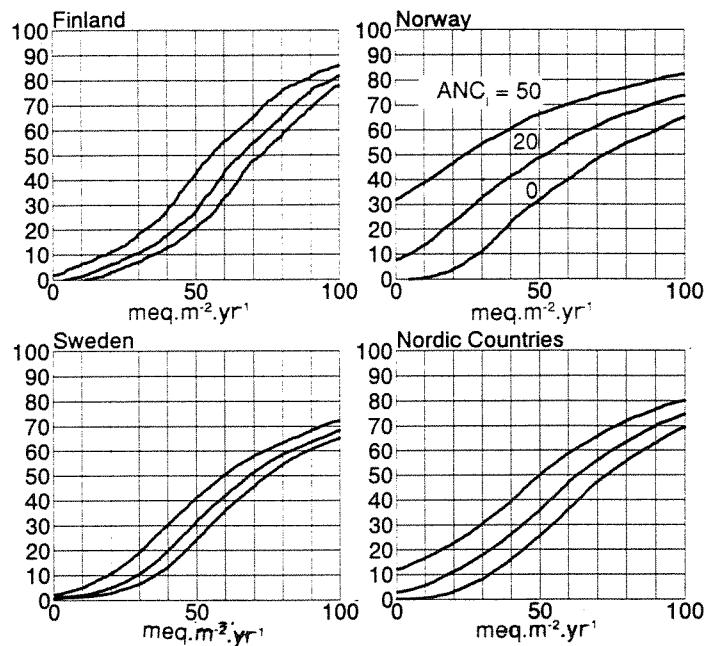

Figure A1. frequency distributions of critical loads of sulphur to lakes for three levels of ANC_{limit} (0,20 and 50 µeq/l) for Finland, Sweden and Norway.

It is evident from eq. A2 that lakes with $BC_0^* < ANC_{limit}$ or $([BC]^*_0 - [ANC_{limit}]) \cdot Q$ is less than BC^*_d will result in "negative" critical load values. When assessing the number and amount of critical load exceedance for a given deposition scenario one must bear in mind that for such lakes the critical load will always be exceeded, even assuming the strongest measures for air pollution control. These lakes have always had a chemical situation which does not allow many of the chosen indicator organisms to live. Alternative values for ANC_{limit} has to be chosen. It may thus be worthwhile to treat lakes for which the critical load is exceeded even in the absence of anthropogenic deposition separately. Canada has chosen to disregard such lakes in their scenarios. These lakes must however, be kept in mind when assessing the consequences of national target loads. In practice, a target load resulting from international negotiations will most likely be higher than the lowest critical load for a region. Thus, for a given target load there will be areas where lakes will not recover to acceptable conditions for fish even when the target load is reached. A political decision is needed on how to handle such naturally acidic lakes, should they be accepted to be permanently

acidified or should they for example be limed? Such consideration should be included in a national control strategy for air pollutants.

3. Lakes with negative critical loads in the Nordic Countries

Using the national lake surveys carried out in Norway (1986), Finland (1987) and Sweden (1990) we have calculated the critical loads according to the steadys state water chemistry method using $ANC_{limit} = 20$ and 50 µeq/l and separated those ending up with "negative" values. The numbers and the statistics of the chemistry of these lakes are given in tables A1 and A2.

For Norway 163 (16%) lakes out of the lakes included in the 1000-lake survey in 1986 have "negative" critical loads using ANC_{limit}.= 20 µeq/l. The corresponding figures for Sweden and Finland are 18 (0.4%) out of 4015 and 4 (0.3%) out of 1450, respectively. The reason for the much higher percentage of lakes with negative critical loads in Norway is due to the larger number of lakes with low base cation concentrations. The Norwegian lakes are all located in areas where the bedrock consist of granites and gneisses and the soil covers are thin. This is also the case for the Swedish and Finnish lakes. Also, the runoff of the "negative" critical load lakes in Norway are about five times higher than those in Sweden and Finland (table A1). The pH-values and the non-marine concentrations of the sum of calcium and magnesium and of sulfate are very similar, indicating similar water chemistry for "negative" critical load lakes in the three countries. The chloride concentrations reflects that the Norwegian lakes are more seasalt influenced than the Swedish and Finnish lakes. The Finnish lakes have the lowest nitrate concentrations. The concentrations of organic carbon (TOC) are lowest in the Norwegian lakes, mostly less than 2 mgC/l. The Finnish lakes are also low TOC-lakes for Finland (2-6 mgC/l), all located in Lapland. The Swedish lakes also show higher TOC-values. They may, however, be overestimated from the regression shown below. For 7 of the 18 lakes measured TOC were from 0.3 to 5.7 mgC/l, with an average of 2.7 mgC/l.

Table A1. Arithmetric means of the lake chemistry in Norway, Sweden and Finland with "negative critical loads for $ANC_{limit}=20$ µeq/l.

Component	Norway			Sweden			Finland		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
pH	5,08	4,44	6,14	5,09	4,64	5,66	5,10	4,68	5,41
Ca* + Mg, µeq/l	15	8	23	14	9	28	18	15	21
SO4*, µeq/l	33	10	75	30	70	48	34	22	47
TOC[1], mg/l	0,9	0,1	4,4	4,7	3,5	12,5	4,0	2,1	5,9
NO3, µg/l	108	<1	380	80	7	380	1717	0	59
CL-20, meq/m^2.yr	-11	-46	0	-2	-9	0	-1	-2	0
Runoff, l/km^2.sec	66	12	118	13	8	30	12	11	13
N. of lakes	128			18			4		
Percent of lakes	13			0,4			0,3		

Table A2. Arithmetric means of the lake chemistry in Norway, Sweden and Finland with "negative critical loads" for ANC_{limit}=50 μeq/l.

Component	Norway			Sweden			Finland		
	Mean	Min	Max	Mean	Min	Max	Mean	Min	Max
pH	5,23	4,32	5,58	5,16	3,94	6,32	5,24	4,14	6,75
Ca* + Mg, μeq/l	27	8	100	42	8	130	36	1	59
SO_4*, μeq/l	43	10	85	41	6	167	32	22	83
$TOC^{1)}$, mg/l	0,9	0,1	7,8	6,8	3,4	19,1	7,4	2,0	23,9
NO3, μg/l	98	<1	525	54	7	380	8	0	74
CL-50, meq/m^2.yr	-39	-44	0	-7	-37	0	-5	-14	0
Runoff, l/km^2.sec	53	11	120	15	8	40	12	9	16
N. of lakes	485			85			67		
Percent of lakes	48			2,1			4,6		

[1])*For the Swedish and the Finnish lake surveys TOC was not analyzed in all samples. All the Swedish lakes were, however, analyzed for Absorbance, and all the Finnish lakes were analyzed for COD (permanganate). The TOC-values for Sweden and Finland in the tables A1 and A2 were calculated from regression of TOC and Absorbance for Sweden (TOC = 3.43 + 38.8 * ABSORB, r = 0.87, N = 988), and from regression of TOC and COD for Finland (TOC = 2.0 + 0.68*COD, r = 0.96, N = 967), respectively.*

Using ANC_{limit}.= 50 μeq/l the number of "negative" critical load lakes increases significantly, to 485 (48%) for Norway, 85 (2.1%) for Sweden and 67 (4.6%) for Finland (Table A2). As expected the average values for pH and base cations increase, while the values for TOC and sulfate increase only slightly.

4. Conclusions

Negative critical loads occur only in lakes with low base cation concentrations and usually located in areas with bedrock consisting of granites and gneisses. This is valid for all three countries. Norway has the largest number of lakes with "negative" critical loads which can be ascribed to the thinner soil covers and higher runoff.. The "negative" critical load lakes are typical clearwater lakes in all three countries, but the TOC-levels are generally higher in Sweden and Finland than in Norway, most likely due to higher runoff in the latter country.

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Color plates and captions

Plate I. The median present nitrate leaching in each NILU-grid.

Plate II. The critical load of acidity (5th percentile; left), and its present exceedance (95th percentile; right) in each NILU-grid, calculated with present S deposition.

Plate III. The 5th percentile of the maximum critical load of N (left) and S (right) in each NILU-grid.

Plate IV. The 95th percentile of the minimum (left) and maximum (right) S+N reduction required to achieve non-exceedance in each NILU-grid.
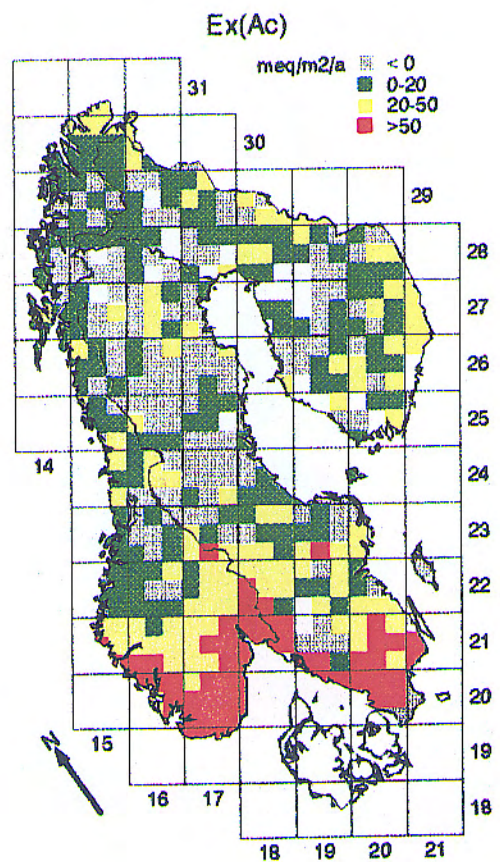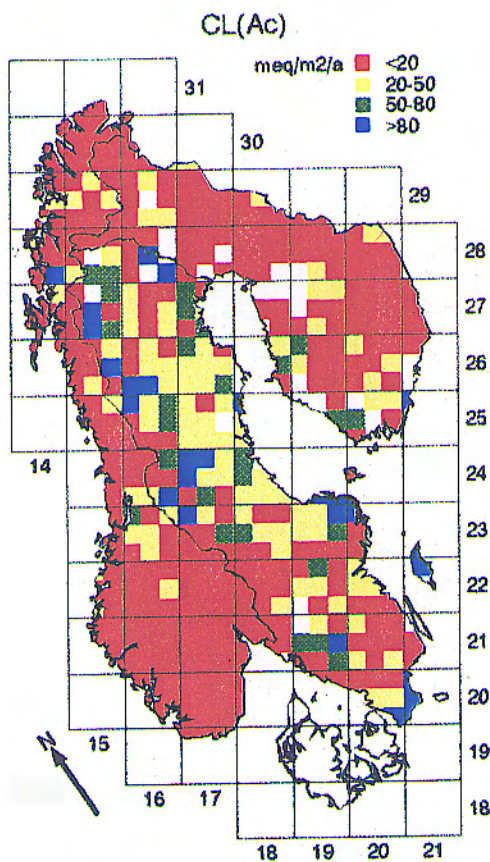
Plate I:

N leaching

meq/m2/a
- ■ < 1
- ■ 1- 5
- ■ 5-10
- ■ >10

Plate II:

CL(Ac)

meq/m2/a
- ■ <20
- ■ 20-50
- ■ 50-80
- ■ >80

Ex(Ac)

meq/m2/a
- < 0
- ■ 0-20
- ■ 20-50
- ■ >50

Plate III:

CLmax(N)

meq/m2/a	
🟥	<20
🟨	20-50
🟩	50-80
🟦	>80

CLmax(S)

meq/m2/a	
🟥	<20
🟨	20-50
🟩	50-80
🟦	>80

Plate IV:

Redmin

meq/m2/a	
▦	< 0
🟩	0-20
🟨	20-50
🟥	>50

Redmax

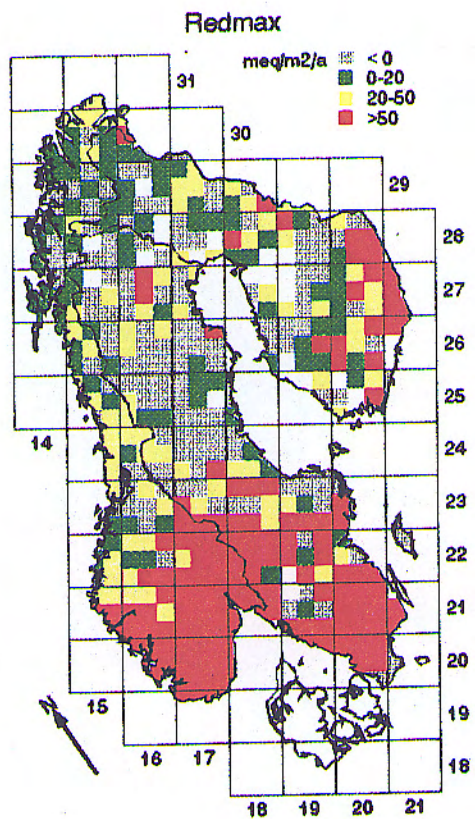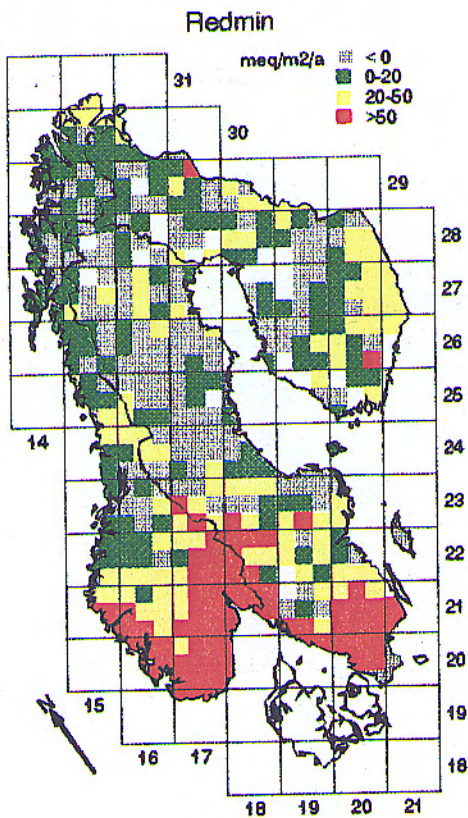| meq/m2/a	
▦	< 0
🟩	0-20
🟨	20-50
🟥	>50

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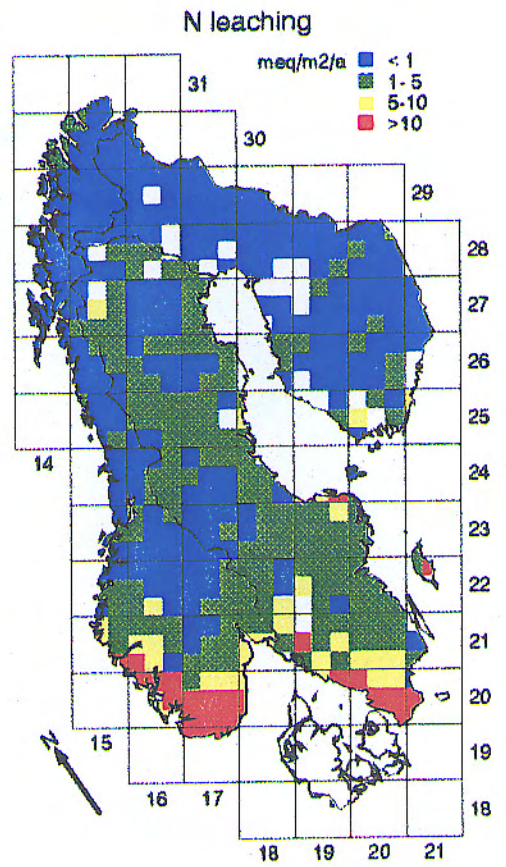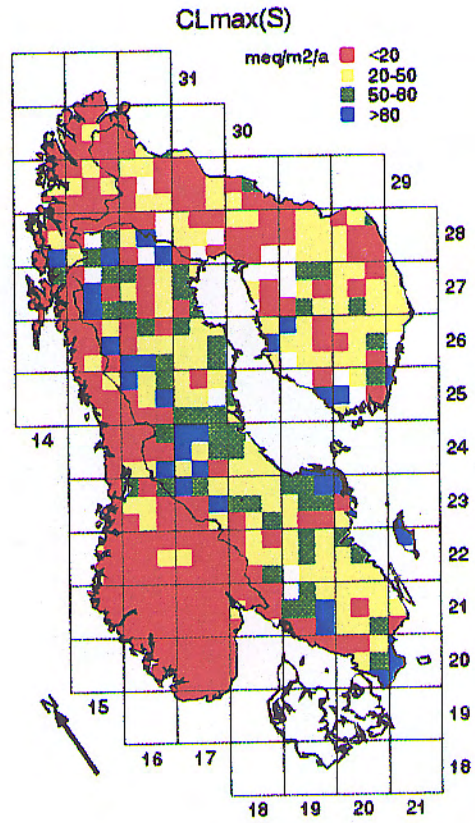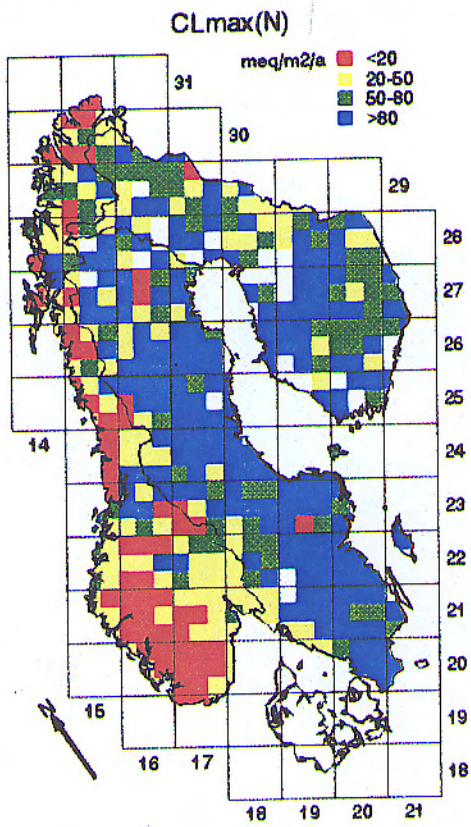
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