

# Present and potential nitrogen outputs from Norwegian soft water lakes – an assessment made by applying the steady-state First-order Acidity Balance (FAB) model

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## Abstract

The steady-state First-order Acidity Balance (FAB) model for calculating critical loads of sulphur (S) and nitrogen (N) is applied to 609 Norwegian soft-water lakes to assess the future nitrate ( $\text{NO}_3^-$ ) leaching potential under present (1992-96) S and N deposition. The lakes were separated into five groups receiving increasing levels of N deposition (<25, 25-49, 50-74, 75-99 and 100-125  $\text{meq m}^{-2} \text{yr}^{-1}$ ). Using long-term sustainable N sink rates presently recommended for FAB model applications, N immobilisation, net N uptake in forests, denitrification and in-lake N retention were estimated for each group of lakes. Altogether, the long-term N sinks constituted  $9.9 \pm 3.2$  to  $40.5 \pm 11.4$   $\text{meq m}^{-2} \text{yr}^{-1}$  in the lowest and highest N deposition categories, respectively. At most sites, the current N deposition exceeds the amount of N retained by long-term sustainable N sinks plus the  $\text{NO}_3^-$  loss via the lake outlets. This excess N, which is currently retained within the catchments may, according to the FAB model, leach as acidifying  $\text{NO}_3^-$  in the future. If these predictions are fulfilled,  $\text{NO}_3^-$  leaching at sites in the various N deposition categories will increase dramatically from present (1995) mean levels of 1-20  $\text{meq m}^{-2} \text{yr}^{-1}$ , to mean levels of 7-70  $\text{meq m}^{-2} \text{yr}^{-1}$  at future steady state. To illustrate the significance of such an increase in  $\text{NO}_3^-$  leaching, the mean Acid Neutralising Capacity (ANC) at sites in the highest N deposition category may decrease from  $-18 \pm 15$   $\mu\text{eq L}^{-1}$  at present, to  $-40 \pm 20$   $\mu\text{eq L}^{-1}$ . Under present S and N deposition levels, the FAB model predicts that 46% of the Norwegian lakes may experience exceedances of critical loads for acidifying deposition. In comparison, the Steady-State Water Chemistry model (SSWC), which considers only the present N leaching level, estimates critical load exceedances in 37% of the lakes under the same deposition level. Thus far, there are great uncertainties regarding both the time scales and the extent of future N leaching, and it is largely unknown whether the FAB model predictions will ever be fulfilled. Hence, long-term monitoring and further studies on N immobilisation processes under varying N deposition levels and ecosystem types seem necessary to make better predictions of future  $\text{NO}_3^-$  leaching.

**Key words:** Lakes, hydrochemistry, nitrogen, nitrate, sinks, leaching, acidification, critical loads, FAB model

## Introduction

Surveys of nearly 500 lakes in Norway conducted in both 1986 and 1995 have revealed widespread chemical recovery, mainly due to a 40% decrease in sulphate concentrations (Skjelkvåle *et al.*, 1998). During the same period, there have been no significant changes in  $\text{NO}_3^-$  concentrations in any part of the country, even though stores of N in catchments have increased due to several decades of elevated N deposition and retention. A crucial question is whether this N accumulation is sustainable over time, or is mobilisation of  $\text{NO}_3^-$  and resultant acidification of surface waters to be

expected in the future (Skeffington and Wilson, 1988; Aber *et al.*, 1989). This is one of the main issues faced in determining critical loads of acidity and the designing of future control strategies to reduce adverse effects of regional and transboundary air pollution (Posch *et al.*, 1999).

Two models – one empirical and one process-oriented – are commonly used to calculate critical loads of acidifying deposition (S and N) for surface waters (Henriksen and Posch, 2001). The empirical Steady-State Water Chemistry (SSWC) model allows the calculation of critical loads of acidity and their exceedances for any S deposition scenario,

given the *present* N leaching level (UBA 1996; Henriksen and Posch, 2001). The SSWC model does not consider any other N leaching scenarios since no equations on N sources and sinks are included. These are, however, included in the process-oriented First-order Acidity Balance (FAB) model, which calculates separate critical loads of S and N and their exceedances for any scenario of S and N deposition (Posch *et al.*, 1997; Henriksen and Posch, 2001). Both models use the same approaches for calculating the pre-acidification leaching of non-marine base cations and also the same  $ANC_{limit}$ , the lowest ANC flux that does not damage selected biota. During the negotiations for the new multi-pollutant, multi-effect protocol signed in Gothenburg in December 1999 (UN/ECE, 2000), the FAB model was used as a basis for calculating critical loads for surface waters.

In contrast to the SSWC model, which considers only the present  $NO_3^-$  leaching, the FAB model also takes into account anticipated long-term sustainable N sinks in the terrestrial catchment and in lakes. The model to a great extent proposes a “worst case” scenario by assuming that all N inputs not retained by long-term sustainable N sinks will be leached as  $NO_3^-$  sometime in the future. At present, the N leaching predicted by the FAB model is much higher than the actual leaching at Norwegian lake sites (Henriksen *et al.*, 1998). However, whether the present N accumulation is sustainable in the long term (Vitousek *et al.*, 1997) is largely unknown.

In Norway, two large databases are commonly used for whole-country or regional assessments of critical load exceedances in surface waters. One is the Norwegian critical load database, which divides the country into 2305 grid squares each covering about 12 x 12 km in southern Norway and a decreasing grid width at higher latitudes (Henriksen and Buan, 2000). Applying the SSWC and FAB models to this database, Henriksen and Buan (2000) found that between 1992 and 1994, the critical load of acidity in surface waters was exceeded in 20 and 29% of the land area, respectively. The other is a lake database, including 1006 statistically selected lakes covering the whole country, sampled during the autumn of 1995 as part of a joint Northern European lake survey (Henriksen *et al.*, 1998). Applying the SSWC model to these lakes showed that 29% of the lakes had exceedance of critical loads at 1990 S and N loading (Henriksen *et al.*, 1997).

In this paper, the Norwegian 1995 lake database has been used to:

- Apply the FAB model to assess the significance of anticipated long-term sustainable N sinks vs. present N deposition and N leaching level in Norwegian lakes and their catchments.

- Estimate potential N leaching in the future and evaluate its possible consequences for water chemistry and exceedances of critical loads for acidifying deposition.

## N sinks included in the FAB model

The FAB model is derived in Posch *et al.* (1997) and further refinements are given in Henriksen and Posch (2001). The charge balance (acidity balance) underlying the model can be written as:

$$N_{dep} + S_{dep} = N_i + N_u + N_{de} + N_{lake} + S_{lake} + BC_{le} - ANC_{le, crit} \quad (1)$$

where  $N_{dep}$  and  $S_{dep}$  are acidifying deposition,  $N_i$  — anticipated long-term sustainable nitrogen immobilisation in soils,  $N_u$  — nitrogen uptake by trees,  $N_{de}$  — soil denitrification,  $N_{lake}$  — in-lake nitrogen retention,  $S_{lake}$  — in-lake sulphur retention,  $BC_{le}$  — base cation leaching from catchment and  $ANC_{le, crit}$  — the lowest ANC flux that does not damage selected biota ( $ANC_{limit}$ ). The term  $BC_{le}$  minus  $ANC_{le, crit}$  is analogous to the critical load of acidity for surface waters,  $CL_A$ . In this paper,  $BC_{le}$  and  $ANC_{le, crit}$  are estimated from present water quality data as outlined in UBA (1996) and Henriksen and Posch (2001). Exceedance of critical loads,  $Ex_A$ , is calculated as the actual S and N leaching minus  $CL_A$ . Equations and recommended values for the N sink processes included in the model are listed in the sections below. In-lake S retention is estimated using the same equations as for in-lake N retention, but with a mass transfer coefficient which is an order of magnitude lower than for N (Dillon and Molot, 1990; UBA 1996).

*N immobilisation:* The long-term sustainable immobilisation of N ( $N_i$ ) denotes the continuous build-up of stable C and N compounds in forest soils since the last glacial period (Rosén *et al.*, 1992). This is assumed to be net N immobilisation, including N fixation. The equation included in the FAB model is written as:

$$N_i = x_i \left( 1 - \frac{A_o}{A_d} \right) \quad (2)$$

where  $x_i$  is an anticipated long-term sustainable immobilisation rate of N ( $meq\ m^{-2}\ yr^{-1}$ ) in the catchment soils and  $A_o:A_d$  the lake:catchment area ratio ( $km^2\ km^{-2}$ ).

*N uptake:* The FAB model considers only the net uptake in forests, i.e. the amount of N removed from the catchment by harvesting (UBA, 1996). N uptake is calculated as:

$$N_u = x_u f_{for} \quad (3)$$

where  $x_u$  is the mean net N uptake in forests ( $\text{meq m}^{-2} \text{ yr}^{-1}$ ), and  $f_{\text{for}}$  is the fraction of forest within the catchment ( $\text{km}^2 \text{ km}^{-2}$ ). N uptake in forests can be computed from the average amount of N in the harvested biomass, divided by the rotation period (in years).

**Denitrification:** The amount of N removed from the terrestrial catchment by denitrification ( $N_{\text{de}}$ ) ( $\text{meq m}^{-2} \text{ yr}^{-1}$ ) is calculated by multiplying the sum of available N (from atmospheric deposition) after N immobilisation and N uptake by a denitrification fraction ( $f_{\text{de}}$ ).

$$N_{\text{de}} = f_{\text{de}} (N_{\text{dep}} - N_i - N_u) \left(1 - \frac{A_o}{A_d}\right) \quad (4a)$$

This implies that  $N_i$  and  $N_u$  are considered to remove N faster than  $N_{\text{de}}$  (DeVries *et al.*, 1994). The denitrification fraction ( $f_{\text{de}}$ ) is assumed dependent on the soil type and its moisture status (DeVries *et al.*, 1994). A commonly used simplification in FAB applications has been to estimate  $f_{\text{de}}$  from the fraction of peatlands in the catchment ( $f_{\text{peat}}$ ) (Posch *et al.*, 1997):

$$f_{\text{de}} = 0.1 + 0.7 f_{\text{peat}} \quad (4b)$$

**In-lake N retention:** The FAB model considers the potential in-lake retention ( $N_{\text{lake}}$ ), assuming that all deposited N not exported to the lake is retained by long-term sustainable N sinks in the terrestrial part of the catchment.  $N_{\text{lake}}$  (in  $\text{meq m}^{-2} \text{ yr}^{-1}$ ) is estimated from the N inputs to the lakes times an estimated in-lake N removal coefficient ( $R_N$ ):

$$N_{\text{lake}} = R_N (N_{\text{dep}} - N_i - N_u - N_{\text{de}}) \quad (5a)$$

$R_N$  (the proportion of the N input retained in the lake) is modelled by a kinetic equation derived by Kelly *et al.* (1987):

$$R_N = \frac{S_N}{q_s + S_N} \quad (5b)$$

where  $S_N$  ( $\text{m yr}^{-1}$ ) is a net mass transfer coefficient (or settling velocity) for N in the lake and  $q_s$  the areal water load (per unit area of lake) ( $\text{m yr}^{-1}$ ). The  $S_N$  value used in FAB applications thus far, has been adopted from long-term studies of lakes in Canada and the eastern US (Kelly *et al.*, 1987; Dillon and Molot, 1990). The  $q_s$  variable can be calculated from the catchment runoff ( $\text{m yr}^{-1}$ ), divided by the lake:catchment area ratio ( $\text{km}^2 \text{ km}^{-2}$ ):

$$q_s = \frac{Q}{(A_o / A_d)} \quad (5c)$$

If either the actual  $\text{NO}_3^-$  input or output flux ( $\text{meq m}^{-2} \text{ yr}^{-1}$ ) from the lake is known, the present in-lake N retention ( $N_{\text{lake2}}$ ) can be estimated from

$$N_{\text{lake2}} = \text{NO}_{3(\text{in})} R_N \quad \text{or} \quad N_{\text{lake2}} = \frac{\text{NO}_{3(\text{out})}}{1 - R_N} R_N \quad (6)$$

## Input data and parameter values

### THE LAKE DATABASE

The material includes water chemistry data from a Norwegian survey in 1995 of 1006 statistically selected lakes covering the whole country (Henriksen *et al.*, 1998). All lakes were sampled after autumn overturn in October/November 1995. In addition to water chemistry data, the lake database includes catchment characteristics, point sources, hydrology and atmospheric deposition. Lake and catchment areas are measured from topographical maps, while the fractions of various land-cover types (forest, peat, lakes, arable) and atmospheric deposition have been obtained from digital maps and databases at the Norwegian Institute for Air Research (Tørseth and Semb, 1998). The mean catchment runoff (1961-90) at each site is obtained from a hydrological database operated by the Norwegian Water Resources and Energy Directorate (NVE).

A sorting procedure was performed to exclude lakes with (a) significant settlement or agricultural activities in the catchment, (b) catchments  $>100 \text{ km}^2$ , (c) upstream lakes with accumulated surface areas  $>100\%$  of that characterising the lake at the catchment outlet, (d) total phosphorus  $>10 \mu\text{g L}^{-1}$ , and (e) calcium  $>10 \text{ mg L}^{-1}$ . This reduced the lake population from 1006 to 609 sites. Subsequently, the sites were separated into five groups receiving increasing levels of N deposition ( $<25$ ,  $25-49$ ,  $50-74$ ,  $75-99$  and  $100-125 \text{ meq m}^{-2} \text{ yr}^{-1}$ ).

All water samples were analysed unfiltered at NIVA for several variables, of which nitrate ( $\text{NO}_3^-$ ), ammonium ( $\text{NH}_4^+$ ), total nitrogen (TN), pH, and total organic carbon (TOC) are presented in this paper.  $\text{NO}_3^-$ ,  $\text{NH}_4^+$  and TN (after oxidation with peroxodisulphate) were analysed by automated colorimetry. TOC was analysed by spectrophotometry after UV-oxidation.  $\text{NH}_4^+$  was analysed at only 145 lake sites where concentrations were expected to be measurable due to possible influences from settlement and agriculture. After the sorting procedure, the number of lakes with  $\text{NH}_4^+$  data was reduced to 38 (6% of the 609 remaining lakes). Organic N (TON) is defined as TN minus  $\text{NO}_3^-$  and  $\text{NH}_4^+$ . At sites with no  $\text{NH}_4^+$  data available, TON was estimated as TN minus  $\text{NO}_3^-$  and the median  $\text{NH}_4^+$  concentration within each N deposition category. Acid Neutralising Capacity (ANC) was calculated as the

equivalent sum of base cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ,  $\text{Na}^+$ ,  $\text{K}^+$ ) minus the equivalent sum of strong acid anions ( $\text{Cl}^-$ ,  $\text{SO}_4^{2-}$ ,  $\text{NO}_3^-$ ). Fluxes of chemical compounds were calculated from the 1995 water chemistry data and the 1961–90 mean catchment runoff ( $\text{m yr}^{-1}$ ) at each site.

Catchment characteristics aggregated to N deposition class are presented in Table 1. About 66% of the lakes are located in areas with relatively low N deposition ( $<50 \text{ meq m}^{-2} \text{ yr}^{-1}$ ), whereas 15% of the lakes are located in the most heavily affected areas in southernmost and southeastern Norway (N deposition  $\geq 75 \text{ meq m}^{-2} \text{ yr}^{-1}$ ) (Tørseth and Semb, 1998). The most acid sensitive lakes (lowest critical load) are mainly located in the highest N deposition areas (Table 1). In consequence, these lakes are most affected by acidification. The marked distribution of acid-sensitive lakes in the southernmost and southeastern Norway, where the acid deposition is highest, largely explains why Norway is massively affected by surface water acidification. TOC fluxes were highest at the high N deposition sites ( $>50 \text{ meq m}^{-2} \text{ yr}^{-1}$ ), even though there was no significant correlation between these two variables.

Uncertainties in the input data are associated mainly with the estimation of chemical fluxes and N deposition. Chemical fluxes incorporate uncertainties in water discharge estimates and chemical sampling strategy. Analytical uncertainties are believed to be of minor importance in this context. Since water discharge has not been measured at the study sites, chemical flux estimates are based on the 1961–90 mean catchment runoff at each site. This inevitably

introduces an error in the estimates of annual fluxes at each individual site, but should give fairly reliable estimates when considering aggregated lake data and steady-state conditions. All lakes were sampled shortly after autumn overturn, which gives a relatively good estimate of the annual mean concentrations of chemical solutes (Henriksen *et al.*, 1988; 1998). The atmospheric deposition data are interpolated from fixed sites to a regular grid by kriging interpolation, which is a statistical method that can be used to estimate unknown data from neighbouring measurements (Tørseth and Semb, 1998). The uncertainties associated with this operation and the application of grid-based deposition data on individual lakes in this paper are not quantified (Kjetil Tørseth, pers. comm.).

#### PARAMETER VALUES REFLECTING TERRESTRIAL AND AQUATIC N RETENTION

For the estimation of *long-term sustainable N immobilisation* in the soils ( $N_i$ ), a fixed value of  $3.6 \text{ meq m}^{-2} \text{ yr}^{-1}$  ( $0.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ ) was used. This value represents the lower end of the range ( $0.5$  to  $1.0 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) presently recommended for European critical load calculations (UBA, 1996). Values reflecting *N uptake* ( $N_u$ ) are based on inventory results in 560 plots in a  $9 \times 9 \text{ km}$  national grid, provided by the Norwegian Institute of Land Inventory (NIJOS) (Frogner *et al.*, 1994). According to this database,  $N_u$  values in forests may reach  $50 \text{ meq m}^{-2} \text{ yr}^{-1}$  in southern Norway and  $25 \text{ meq m}^{-2} \text{ yr}^{-1}$  in northern Norway. There is a significant

*Table 1.* Site characteristics (arithmetic mean  $\pm$  standard deviation) along an N deposition gradient (in  $\text{meq m}^{-2} \text{ yr}^{-1}$ ). Abbreviations: n – number of sites,  $A_d$  – catchment area,  $A_o$  – lake area,  $f_{\text{for}}$  – forest fraction,  $f_{\text{peat}}$  – peat fraction, Q – catchment runoff,  $N_{\text{dep}}$  – N deposition,  $S_{\text{dep}}$  – S deposition, and  $CL_A$  – critical loads of acidity. Deposition data cover the period 1992–1996 (Tørseth and Semb, 1998). Water chemistry data are from the Norwegian lake survey 1995 (Henriksen *et al.*, 1998). Critical loads of acidity are calculated by the SSWC model (UBA, 1996; Henriksen and Posch, 2001).

$N_{\text{dep}}$ ( $\text{meq m}^{-2} \text{ yr}^{-1}$ )	<25	25–49	50–74	75–99	100–125	All
n	229	170	116	42	52	609
$A_d$ ( $\text{m}^2$ )	5.4 $\pm$ 12.7	5.5 $\pm$ 11.1	4.2 $\pm$ 10.7	3.3 $\pm$ 8.6	3.1 $\pm$ 6.2	4.9 $\pm$ 11.2
$A_o:A_d$ ratio ( $\text{m}^2 \text{ m}^{-2}$ )	0.13 $\pm$ 0.08	0.12 $\pm$ 0.10	0.14 $\pm$ 0.08	0.15 $\pm$ 0.08	0.15 $\pm$ 0.08	0.13 $\pm$ 0.09
$f_{\text{for}}$ (.)	0.19 $\pm$ 0.29	0.18 $\pm$ 0.31	0.34 $\pm$ 0.38	0.30 $\pm$ 0.39	0.16 $\pm$ 0.28	0.22 $\pm$ 0.32
$f_{\text{peat}}$ (.)	0.04 $\pm$ 0.08	0.03 $\pm$ 0.07	0.02 $\pm$ 0.04	0.02 $\pm$ 0.03	0.02 $\pm$ 0.03	0.03 $\pm$ 0.07
Q ( $\text{m yr}^{-1}$ )	1.07 $\pm$ 0.55	1.42 $\pm$ 0.94	1.72 $\pm$ 0.99	2.03 $\pm$ 0.81	2.15 $\pm$ 0.57	1.45 $\pm$ 0.87
$S_{\text{dep}}$ ( $\text{meq m}^{-2} \text{ yr}^{-1}$ )	15.1 $\pm$ 6.3	21.1 $\pm$ 4.2	37.2 $\pm$ 4.8	53.5 $\pm$ 3.0	63.3 $\pm$ 2.4	27.7 $\pm$ 16.4
$N_{\text{dep}}$ (.)	16.7 $\pm$ 4.7	34.5 $\pm$ 7.0	62.1 $\pm$ 6.9	92.9 $\pm$ 7.3	112.0 $\pm$ 3.4	43.7 $\pm$ 31.0
$CL_A$ (.)	99.1 $\pm$ 112.7	64.9 $\pm$ 74.4	48.4 $\pm$ 46.1	50.2 $\pm$ 120.8	23.7 $\pm$ 19.0	70.1 $\pm$ 91.3
ANC (.)	100.0 $\pm$ 122.1	56.0 $\pm$ 94.5	21.0 $\pm$ 56.0	15.6 $\pm$ 124.7	-39.9 $\pm$ 32.5	54.9 $\pm$ 108.0
$\text{H}^+$ (.)	0.9 $\pm$ 1.7	3.3 $\pm$ 13.3	9.2 $\pm$ 10.7	14.4 $\pm$ 10.5	30.7 $\pm$ 14.4	6.6 $\pm$ 12.9
TOC ( $\text{mmol m}^{-2} \text{ yr}^{-1}$ )	179 $\pm$ 235	177 $\pm$ 192	280 $\pm$ 232	256 $\pm$ 245	379 $\pm$ 244	220 $\pm$ 233

( $p < 0.01$ ) relationship between  $N_u$  and latitude, and rough estimates of mean  $N_u$  in the lake database are made on the basis of this regression line. According to this, mean  $N_u$  values in forested areas are around  $15 \text{ meq m}^{-2} \text{ yr}^{-1}$  ( $2.1 \text{ kg ha}^{-1} \text{ yr}^{-1}$ ) in southernmost Norway and approximately  $8 \text{ meq m}^{-2} \text{ yr}^{-1}$  in northernmost Norway. These estimates are in the lower end of the range previously suggested for boreal forests in Europe (UBA, 1996).

*In-lake N retention* is estimated by applying a mean  $S_N$  value of  $5 \text{ m yr}^{-1}$ , which is obtained from long-term studies of lakes in Canada and the eastern US (Kelly *et al.*, 1987; Dillon and Molot, 1990). This is the same value as earlier recommended for FAB applications in Europe (UBA, 1996). For application of the FAB model to non-headwater lakes, Hindar *et al.* (2001) compared three different approaches to include in-lake N retention. These were (a) the “one-lake approach”, which only considers the lake at the catchment outlet, (b) the “big-lake” approach, which treats all lakes in the system as a single lake situated near the catchment outlet, and (c) the “lake system” approach, which treats each lake and sub-catchment separately. In a study performed in the Killarney Provincial Park, Canada, the “big-lake” and “lake system” methods increased the average in-lake N retention estimates by 47% and 73%, respectively, in comparison to the “one-lake” approach” (Hindar *et al.*, 2001). In Norway, where lake water residence times usually are shorter than in Canada (Ontario), the differences in model results using the three approaches are assumed to be smaller. In the present study, in-lake N retention is estimated by the “big-lake approach”. About 40% of the sites included in the present study are headwater lakes, while the remaining sites have upstream lakes with an accumulated surface area of maximum 100% of the primary lake at the catchment outlet. This limit was introduced to reduce errors in the  $N_{\text{lake}}$  estimates due to N retention in upstream lakes.

The parameter values reflecting long-term sustainable terrestrial and aquatic N sinks will have associated errors which can be reduced only if further empirical studies that

can improve the basis for specifying process rates on a spatial and temporal scale (Curtis *et al.*, 2000). In addition to the uncertainties associated with input data and parameter values, the FAB model also includes uncertainties due to model structure, process equations and the relationships between water chemistry and aquatic biota (critical limits). The various uncertainty types were evaluated previously in connection with FAB applications to Finnish lakes (Kämäri *et al.*, 1993; Posch *et al.*, 1993). Despite the large uncertainty in some of the model parameters and input data, the resulting ranges of critical loads were narrow enough, in most cases, to draw meaningful conclusions on lake/catchment sensitivity to acidifying deposition.

## Results

### N LEACHING ALONG AN N DEPOSITION GRADIENT

$\text{NO}_3^-$  concentrations and fluxes from the 609 lakes were positively correlated to N deposition,  $r^2=0.51$  and  $r^2=0.44$ , respectively ( $p < 0.01$ ). When arranging the sites into groups according to N deposition level, mean  $\text{NO}_3^-$  fluxes increased from nearly zero in the lowest deposition category to  $20.4 \pm 9.9 \text{ meq m}^{-2} \text{ yr}^{-1}$  at sites with the highest deposition (Table 2). In the most heavily affected category, surface water  $\text{NO}_3^-$  fluxes corresponded to over 30% of the non-marine sulphur ( $\text{SO}_4^*$ ) flux. This implies that  $\text{NO}_3^-$  under present conditions plays a significant role in the acidification of these lakes.

At lake sites where  $\text{NH}_4^+$  data are available,  $\text{NH}_4^+$  concentrations showed a weak positive correlation with N deposition ( $r^2=0.20$ ,  $p < 0.01$ ,  $n=38$ ), although the concentration level generally was very low ( $\leq 2 \text{ } \mu\text{eq L}^{-1}$  in 33 of 38 samples). The median  $\text{NH}_4^+$  flux ranged from  $0.4 \text{ meq m}^{-2} \text{ yr}^{-1}$  in the lowest N deposition category to  $4.6 \text{ meq m}^{-2} \text{ yr}^{-1}$  at high deposition sites.  $\text{NH}_4^+$  concentrations generally were much lower than  $\text{NO}_3^-$  concentrations, corresponding to 14-40% of  $\text{NO}_3^-$  in the various N deposition

Table 2. N fluxes from the lake outlets ( $\text{meq m}^{-2} \text{ yr}^{-1}$ ) along the N deposition gradient. Arithmetic mean values  $\pm$  standard deviation.  $\text{NH}_4^+$  fluxes are estimated from a limited number of samples (3-20 sites within each N deposition category). Chemical data are from Henriksen *et al.* (1998).

$N_{\text{dep}}$ ( $\text{meq m}^{-2} \text{ yr}^{-1}$ )	<25	25-49	50-74	75-99	100-125	All
$\text{NO}_3^-$ flux (..)	$1.0 \pm 1.5$	$4.4 \pm 7.6$	$9.7 \pm 8.2$	$13.8 \pm 9.5$	$20.4 \pm 9.9$	$6.1 \pm 8.9$
$\text{NH}_4^+$ flux (..)	0.4	0.6	2.0	2.2	4.6	1.2
TON flux ( $\text{mmol m}^{-2} \text{ yr}^{-1}$ )	$6.2 \pm 5.7$	$7.0 \pm 6.8$	$9.2 \pm 7.2$	$8.7 \pm 7.6$	$12.3 \pm 8.6$	$7.7 \pm 6.9$
TN flux (..)	$7.6 \pm 5.7$	$11.9 \pm 10.3$	$20.9 \pm 9.6$	$24.7 \pm 10.1$	$37.3 \pm 11.0$	$15.1 \pm 12.5$

categories. The highest relative importance of  $\text{NH}_4^+$  was found at low deposition sites where  $\text{NO}_3^-$  concentrations were low.

At many sites, TON constituted a considerable fraction of TN in the lake outlets, varying from  $78 \pm 23\%$  in the lowest N deposition category to  $33 \pm 20\%$  in the highest N deposition category. In contrast, the relative contribution of  $\text{NO}_3^-$  to TN increased from  $14 \pm 21\%$  in the lowest N deposition category, to  $54 \pm 21\%$  in the two highest N deposition categories. Hence,  $\text{NO}_3^-$  was the dominant N species when N deposition exceeded  $50\text{--}75 \text{ meq m}^{-2} \text{ yr}^{-1}$ . There was no significant relation between TON export and N deposition ( $p > 0.1$ ). TON concentrations were, however, positively correlated to the concentration of TOC in the 609 study lakes ( $r^2 = 0.78$ ,  $p < 0.01$ ). Hence, the TON flux, to a large extent, appears to be uncoupled from the short-term input-output N balance, but is closely related to TOC status in the catchments.

#### QUANTIFICATION OF TERRESTRIAL AND AQUATIC N SINKS

Whereas the long-term N immobilisation in soils ( $N_i$ ) and the N uptake by forests ( $N_u$ ) is assumed to be independent of N deposition, both denitrification ( $N_{de}$ ) and in-lake N retention ( $N_{lake}$ ) are assumed to be functions of N deposition.

The only factor leading to variation in the  $N_i$  estimates is the terrestrial fraction of the catchments (Eqn. 2), whereas latitude and the forest fraction in each catchment solely determine the  $N_u$  estimates (Eqn. 3). This implies that the relative importance of  $N_{de}$  and  $N_{lake}$  as long-term N sinks increases with N loading.

At the low N deposition sites ( $< 25 \text{ meq m}^{-2} \text{ yr}^{-1}$ ), all quantified N sink processes were of comparable magnitude,  $1.2 \pm 0.9$  to  $3.5 \pm 2.0 \text{ meq m}^{-2} \text{ yr}^{-1}$  (Table 3). At sites receiving the highest N loading ( $100\text{--}125 \text{ meq m}^{-2} \text{ yr}^{-1}$ ),  $N_{lake}$  appeared to be the most important N sink, reaching  $25.0 \pm 10.5 \text{ meq m}^{-2} \text{ yr}^{-1}$ . This corresponded to over 60% of all quantified long-term N sinks in this category, whereas  $N_i$ ,  $N_u$  and  $N_{de}$ , accounted for 7, 6 and 25%, respectively.

At most sites, the current N deposition exceeds the amount of N retained by long-term sustainable N sinks plus the  $\text{NO}_3^-$  loss via the lake outlets (Tables 2 and 3). This excess N retention was positively correlated to N deposition ( $r^2 = 0.78$ ,  $p < 0.01$ ,  $n = 609$ ) and increased from  $5.9 \pm 3.7 \text{ meq m}^{-2} \text{ yr}^{-1}$  in the lowest N deposition category to  $51.1 \pm 12.8 \text{ meq m}^{-2} \text{ yr}^{-1}$  at sites receiving the highest N deposition. If this fraction of N deposition starts to leach as  $\text{NO}_3^-$  in the future,  $\text{NO}_3^-$  fluxes in the most heavily affected areas may increase to levels around  $70 \text{ meq m}^{-2} \text{ yr}^{-1}$  (Fig. 1). This is 3.5 times the present  $\text{NO}_3^-$  leaching at these sites.

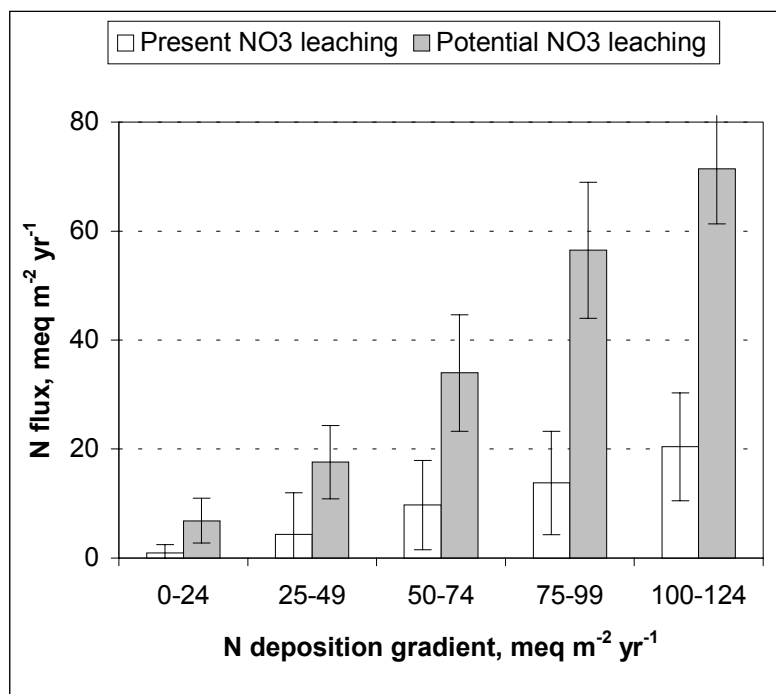


Fig. 1. Present (1995)  $\text{NO}_3^-$  leaching at 609 Norwegian lake sites and FAB model prediction of potential  $\text{NO}_3^-$  leaching (steady-state) under present (1992-96) N deposition levels. Standard deviation is indicated by vertical bars.

Table 3. Long-term sustainable N sinks estimated from 1995 water chemistry (Henriksen *et al.*, 1998), 1992-1996 deposition data (Tørseth and Semb, 1998) and default values presently recommended for FAB model applications.

$N_{dep}$ (meq m <sup>-2</sup> yr <sup>-1</sup> )	<25	25-49	50-74	75-99	100-125	All
$N_i$ (.)	3.1±0.3	3.1±0.4	3.1±0.3	3.0±0.3	3.0±0.3	3.1±0.3
$N_u$ (.)	2.0±3.0	2.3±4.1	4.7±5.3	4.3±5.5	2.3±4.1	2.8±4.2
$N_{de}$ (.)	1.2±0.9	3.1±1.4	5.2±1.3	8.1±1.6	10.2±2.4	3.7±3.1
$N_{lake}$ (.)	3.5±2.0	8.4±5.3	15.2±7.1	21.0±9.1	25.0±10.5	10.1±9.1
$\Sigma$ N sinks (.)	9.8±3.2	16.9±7.2	28.2±9.3	36.4±10.3	40.5±11.4	19.7±12.7

## Discussion

In the FAB model, the acidifying potential of N is expressed as N deposition minus the sum of N sinks presumed to be sustainable in the long term. Some of this N is already leaching as NO<sub>3</sub><sup>-</sup> and causes acidification today (Table 2), whereas the remainder presently is accumulated within the catchments. The long-term stability of this N retention fraction is largely unknown (Vitousek *et al.*, 1997; Wright, 1998). Thus, a crucial question is whether the present N accumulation rate can proceed into the future, or if the ecosystems gradually or suddenly will become N saturated (Skeffington and Wilson, 1988; Aber *et al.*, 1989).

### PRESENT N RETENTION CAPACITY *v.* LONG-TERM SUSTAINABLE N SINKS

Most ecosystems — especially those located in high-deposition areas — presently retain more N than reflected in the long-term N sinks recommended for FAB model applications (Rosén *et al.*, 1992; Curtis *et al.*, 2000). According to combined input-output and process studies in forested catchments, a significant fraction of this excess N accumulation can be ascribed to N immobilisation in the soil organic pool (Gundersen *et al.*, 1998a; Kjønnaas *et al.*, 1998). At two forested catchments in SW Norway receiving N deposition levels of 115-175 meq m<sup>-2</sup> yr<sup>-1</sup>, Mulder *et al.* (1997) ascribed 51-81% of the net N retention to  $N_i$ . This is substantially higher than the long-term  $N_i$  values of approximately 3.0 meq m<sup>-2</sup> yr<sup>-1</sup> that were estimated for the various N deposition categories in the present study (Table 3). Hence, the FAB model suggests that a considerable fraction of the present N accumulation must be regarded as non-sustainable in the long term.

Denitrification rates ( $N_{de}$ ) generally are low in well-drained soils, such as those often characteristic for forest and heathland sites. Data from several forested sites within the NITREX project (“Nitrogen saturation experiments”)

indicated  $N_{de}$  fluxes in the range 0.5-3.6 meq m<sup>-2</sup> yr<sup>-1</sup> (Gundersen *et al.*, 1998a). Bogs and wetlands are often characterised by much higher  $N_{de}$  values, in some cases >150 meq m<sup>-2</sup> yr<sup>-1</sup> (UBA, 1996). However, such ecosystem types usually occupy only small areas within a catchment (on average 3% in the present data material; Table 1). The  $N_{de}$  estimates generated from this study were about an order of magnitude higher than values usually measured in forests, but well within the range reported earlier for mixed ecosystems in boreal and temperate regions (UBA, 1996). Hence, Eqn. 4 probably gives quite reasonable  $N_{de}$  estimates for the Norwegian sites at the ambient N deposition level. In areas experiencing higher N loading (as e.g. in central Europe),  $N_{de}$  estimates calculated by Eqn. 4 might, however, be much higher than the current values measured in soils (Curtis *et al.*, 1998, 2000). A possible reason for the low  $N_{de}$  values measured under present conditions may be that current high  $N_i$  rates reduce the supply of N for denitrification. However, in recent FAB applications to UK sites Curtis *et al.* (1998, 2000) decided to use fixed  $N_{de}$  values for certain soil types, independent of N deposition, to obtain better conformity between estimated *v.* currently observed  $N_{de}$  values.

In-lake N retention ( $N_{lake}$ ) values estimated by the FAB model are based on the assumptions that all N not retained by long-term sustainable terrestrial N sinks will be exported to the lakes as NO<sub>3</sub><sup>-</sup>, and that in-lake N removal rates are first-order reactions (proportional to the input concentrations). The first assumption is the main principle behind the FAB model, whereas the second is supported by a number of Canadian lake studies, which demonstrate that NO<sub>3</sub><sup>-</sup> reduction rates increase with input concentrations (Kelly *et al.*, 1987). Thus, the  $N_{lake}$  value can be regarded as potential in-lake N retention given the increases in NO<sub>3</sub><sup>-</sup> leaching predicted by the FAB model. When estimating  $N_{lake}$ , the FAB model applies a mean mass transfer coefficient for NO<sub>3</sub><sup>-</sup> ( $S_{NO_3}$ ), obtained from long term-studies of lakes in

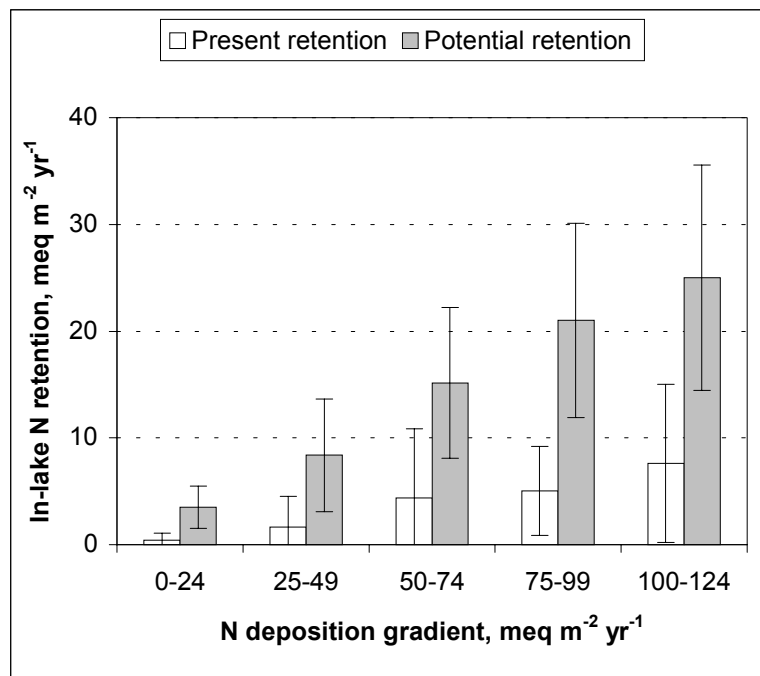


Fig. 2. Present (1995) and potential in-lake N retention at the Norwegian lake sites estimated by Eqns. 5 and 6 under present (1992–96) N deposition levels. The FAB model considers the potential in-lake N retention when calculating critical load exceedances. Standard error bars are indicated.

Canada and the US (Kelly *et al.*, 1987; Dillon and Molot, 1990). According to new data from Norwegian lakes and prolonged time series from Canadian lakes, the mean  $S_{NO_3}$  value adopted by the FAB model ( $5 \text{ m yr}^{-1}$ ) seems representative for lakes spanning a wide range in N inputs and hydrological conditions.

At the Norwegian lake sites, the  $N_{lake}$  values estimated by the FAB model generally were much higher than the current in-lake retention ( $N_{lake2}$ ) estimated from observed  $NO_3^-$  leaching and Eqn. 6 (Fig. 2). In fact, the predicted mean  $N_{lake}$  value at sites receiving the highest N loading was more than three times higher than the mean  $N_{lake2}$  value. Hence, it seems evident that lakes have a large potential in reducing negative effects from possible increases in terrestrial N leaching from catchments in high-deposition areas. According to the theoretical future steady state predicted by FAB, the potential in-lake retention at sites receiving N deposition higher than  $50 \text{ meq m}^{-2} \text{ yr}^{-1}$  may exceed the sum of all long-term terrestrial N sinks (Table 3). When taking into account that lakes within these N deposition categories cover only 13–15% of the total catchment area, FAB suggests that the aquatic ecosystems at these sites may become several times more efficient in retaining N (per unit of surface area) at future steady state than the terrestrial parts.

#### THE ROLE OF $NH_4^+$ AND TON

The FAB model considers only the leaching of  $NO_3^-$  and not  $NH_4^+$  and TON. It is assumed that the deposited  $NH_4^+$  is taken up (preferentially) by vegetation, and — what is not taken up — is nitrified (Posch *et al.*, 1997). In general, this is confirmed at most experimental and monitoring sites by the fact that  $NH_4^+$  is found rarely in appreciable amounts in soil leachate or in the runoff.

In contrast to  $NH_4^+$ , TON constitutes a significant fraction of TN outputs at the study sites (Table 2). However, TON is not an ion and therefore does not enter the charge balance, from which the FAB model is derived. If TON is to be included in the FAB model, it has to be considered a sink of N — such as immobilisation, for example — irrespective of whether it is exported with the runoff or stays in the catchment. This would result in a corresponding decrease in the present estimates on critical load exceedances. It is, however, important to avoid double counting: The terms in the FAB model which account for the amount of N that is immobilised in the catchment or retained in lake sediments might already contain the N which is leaving the catchment as TON.

On the basis of mainly Norwegian data from small, calibrated catchments, national lake surveys and large-scale



N addition experiments, Wright *et al.* (1997) found no significant relationship between N deposition and TON runoff. Also in the present study, which partly overlaps the lake surveys included by Wright *et al.* (1997), TON fluxes showed no correlation with N deposition, while about 80% of the variance could be explained by the TOC concentrations. Thus, it does not seem reasonable to regard TON as a part of the short-term inorganic N cycle, since a large fraction of TON leaching today may be due to ancient N deposition, N fixation or weathering. On the other hand, it seems likely that the TON leached is derived from previously immobilised N in the soils and is released at a fixed rate over time. This suggests that the long-term immobilisation of inorganic N in soils ( $N_i$ ) might be underestimated and — even if derived from ancient sources — TON may still represent a “missing sink” in the FAB model.

#### CONSEQUENCES FOR LAKE WATER QUALITY IF THE FAB PREDICTIONS ARE FULFILLED

If the future rate of N immobilisation decreases according to the FAB model predictions, it may involve dramatic consequences for aquatic ecosystems. Under present (1992–

96) N deposition,  $\text{NO}_3^-$  leaching at sites in the highest N deposition category ( $100\text{--}125 \text{ meq m}^{-2} \text{ yr}^{-1}$ ) may increase by a factor of 3.5, from about  $20 \text{ meq m}^{-2} \text{ yr}^{-1}$  at present to more than  $70 \text{ meq m}^{-2} \text{ yr}^{-1}$  at future steady state (Fig. 1). In these already heavily acidified catchments,  $\text{NO}_3^-$  leaching from the terrestrial ecosystem will be accompanied by  $\text{H}^+$ ,  $\text{Al}^{3+}$  and base cations. The ratio of change in non-marine base cation concentrations due to changes in strong acid anion concentration can be simulated by the empirical F-factor (Henriksen and Posch, 2001). Under present (1992–96) S and N deposition, the predicted  $\text{NO}_3^-$  increase within the highest N deposition category may involve reductions in average ANC values from  $-18 \pm 15 \text{ } \mu\text{eq L}^{-1}$  at present, to  $-40 \pm 20 \text{ } \mu\text{eq L}^{-1}$  at future steady state (Fig. 3). Within the groups receiving intermediate amounts of N deposition ( $50\text{--}74$  and  $75\text{--}100 \text{ meq m}^{-2} \text{ yr}^{-1}$ ), ANC would decrease from mean levels of 32 and  $10 \text{ } \mu\text{eq L}^{-1}$  to 19 and  $-10 \text{ } \mu\text{eq L}^{-1}$ . This probably would lead to extensive reductions of brown trout populations, which have critical ANC limits around  $0\text{--}20 \text{ } \mu\text{eq L}^{-1}$  (Lien *et al.*, 1996). In addition to increased acid stress/damages to freshwater organisms, the predicted increase in surface water  $\text{NO}_3^-$  fluxes would cause a significant increase in the N loading on coastal marine areas (Hessen *et al.*, 1997).

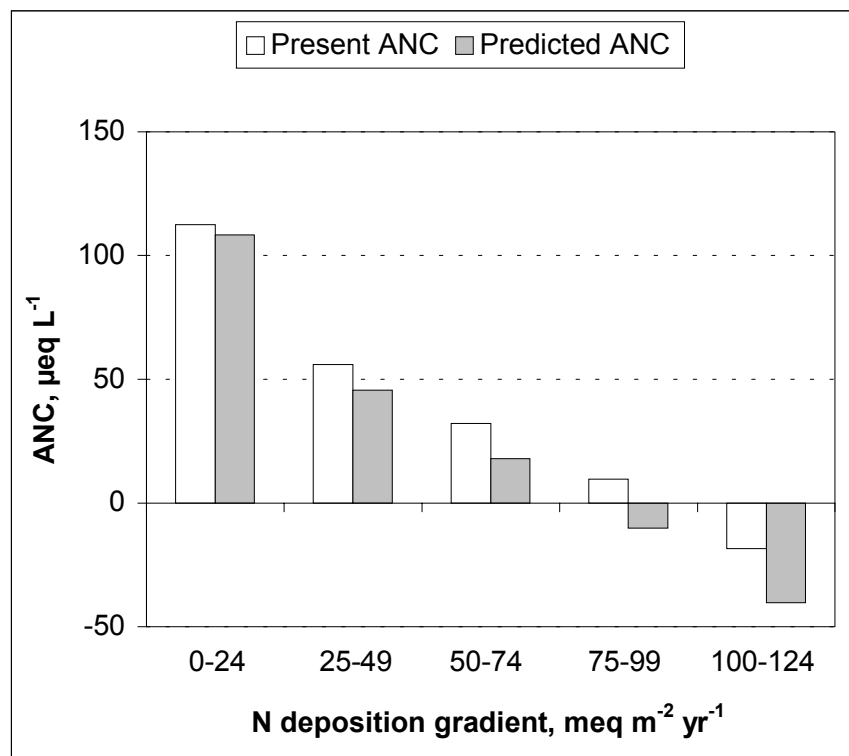


Fig. 3. Present (1995) ANC values at the Norwegian lake sites and FAB model prediction of future (steady-state) ANC under present (1992-96) S and N deposition.

In a FAB application to 13 catchments in the UK, Curtis *et al.* (1998) predicted a mean increase in  $\text{NO}_3^-$  losses by  $10.5 \text{ kg ha}^{-1} \text{ yr}^{-1}$  ( $75 \text{ meq m}^{-2} \text{ yr}^{-1}$ ) from the ambient level of  $5.4 \text{ kg ha}^{-1} \text{ yr}^{-1}$  ( $39 \text{ meq m}^{-2} \text{ yr}^{-1}$ ). The most significant effects of increased  $\text{NO}_3^-$  concentrations were found at sites where current ANC values were less than  $100 \mu\text{eq L}^{-1}$ . Here, the predicted increase in  $\text{NO}_3^-$  concentrations would reduce ANC to around zero. The UK sites investigated by Curtis *et al.* (1998) leach, on average, nearly twice as much  $\text{NO}_3^-$  as the Norwegian high-deposition sites today. In comparison, the ambient N deposition at the UK sites (mean  $146 \text{ meq m}^{-2} \text{ yr}^{-1}$ ) is about 30% higher than at the Norwegian high-deposition sites. However, the estimated long-term N sinks at the UK sites and the Norwegian high-deposition sites were of comparable magnitude ( $33 \text{ v. } 41 \text{ meq m}^{-2} \text{ yr}^{-1}$ ), such that the relative difference in the  $\text{NO}_3^-$  leaching potential seem largely to be reflected by the N deposition level.

Under present (1992–96) S + N deposition and N leaching level, the SSWC model estimates that 37% of the Norwegian lakes may experience exceedances of critical loads for acidifying deposition at future steady state (Fig. 4). About 10% of these may exhibit exceedances higher than  $50 \text{ meq m}^{-2} \text{ yr}^{-1}$ . With increased  $\text{NO}_3^-$  leaching as predicted by the FAB model, the number of exceeded lakes might increase to 46% at future steady state (Fig. 4). Of these, 23% may experience exceedances higher than  $50 \text{ meq m}^{-2} \text{ yr}^{-1}$ . This scenario will surely be modified before 2010, due to reduced

S emissions following the second sulphur protocol (UN/ECE, 1994) and also the implementation of the multi-pollutant, multi-effect protocol (UN/ECE, 2000), which for the first time entails reductions of N emissions in Europe.

WILL THE FAB PREDICTIONS EVER BE FULFILLED?

At present, it seems difficult to decide whether long-term terrestrial N sinks included in the FAB model are underestimated, or if the present N accumulation level may be just a temporary and non-sustainable phenomenon. There is a possibility that some permanent N retention processes may have been omitted in the FAB model (e.g. TON leaching derived from N previously immobilised in the soils). The greatest uncertainty, however, seems to be associated with the sustainability of present N accumulation rates in terrestrial ecosystems.

Even though the current N retention seems to increase with increasing N deposition (Table 3), there was also a clear positive relationship between N deposition level and  $\text{NO}_3^-$  outputs in the present material (Table 2). Several studies have demonstrated a close relationship between N deposition level, soil N status and  $\text{NO}_3^-$  leaching (Stoddard, 1994; Dise and Wright, 1995; Tietema *et al.*, 1998). In high-deposition areas, there are even examples where N outputs exceed the input (e.g. Bredemeier *et al.*, 1998). Decades with elevated atmospheric N inputs have led to decreased

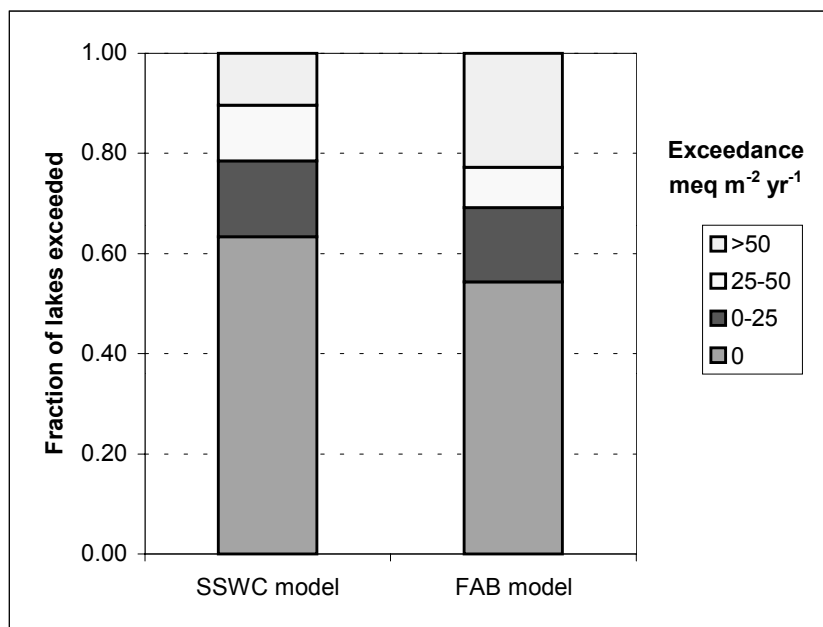


Fig. 4. Critical load exceedances at steady state under 1992-96 S and N deposition levels; calculated from the SSWC model (considering present  $\text{NO}_3^-$  leaching) and the FAB-model (considering potential  $\text{NO}_3^-$  leaching).

soil C:N ratios in several forested sites in Europe, and empirical data demonstrate a negative correlation between  $\text{NO}_3^-$  leaching and C:N ratios in the forest floor (Gundersen *et al.*, 1998b).

Unfortunately, there are presently few sites with simultaneous long-term data on C:N ratios and  $\text{NO}_3^-$  leaching, such that the time perspective for shifts in  $\text{NO}_3^-$  leaching level (and possibly fulfilment of the FAB predictions) is unclear. However, an overall lack of significant trends in  $\text{NO}_3^-$  concentrations in acid-sensitive streams in Europe experiencing elevated levels of N deposition may indicate that N saturation is a slow process requiring several decades (Wright *et al.*, 2001). On the other hand, reductions in N deposition (e.g. by roof experiments) usually have resulted in an immediate reduction in  $\text{NO}_3^-$  output concentrations (e.g., Tietema *et al.*, 1998). Hence, the future  $\text{NO}_3^-$  leaching level may be determined by two opposing factors, i.e. gradually increasing N saturation and reduced N deposition (Wright *et al.*, 2001). In addition, increased air and soil temperatures caused by a possible climate change may accelerate  $\text{NO}_3^-$  leaching by increasing mineralisation of soil organic matter. This was demonstrated by the CLIMEX project (Climate Change Experiment), where a three-year increase in  $\text{CO}_2$  and air temperature tripled the  $\text{NO}_3^-$  leaching in an enclosed forested catchment in southernmost Norway (Wright, 1998).

Due to the large uncertainties associated with future  $\text{NO}_3^-$  leaching as a response to decreasing C:N ratios, altered N deposition levels and possible changes in the global climate, long-term monitoring and further studies of N retention processes in various ecosystem types under varying N deposition levels and climatic conditions seem necessary. In the meantime, it seems reasonable to base the FAB model on the precautionary principle, assuming that all N not taken up by presumed long-term sinks may leach as acidifying  $\text{NO}_3^-$  some time in the future.

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