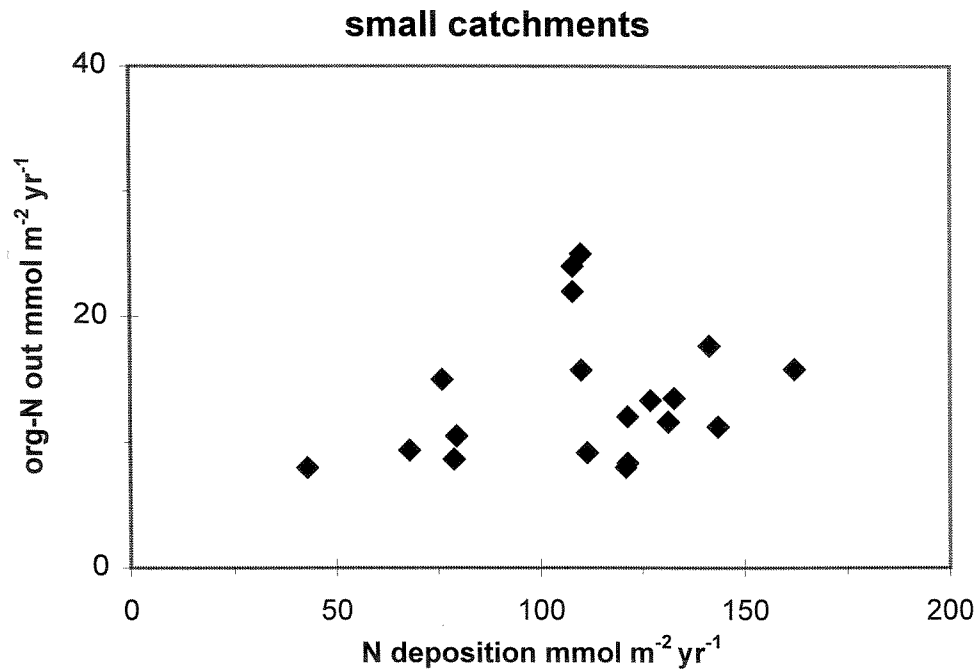


# Atmospheric deposition of nitrogen, runoff of organic nitrogen, and critical loads for soils and waters



Miljøverndepartementet  
Fagrapport nr. 83



## Naturens Tålegrenser

Programmet Naturens Tålegrenser ble satt igang i 1989 i regi av Miljøverndepartementet. Programmet skal blant annet gi innspill til arbeidet med Nordisk Handlingsplan mot Luftforurensninger og til pågående aktiviteter under Konvensjonen for Langtransporterte Grensoverskridende Luftforurensninger (Genevekonvensjonen). I arbeidet under Genevekonvensjonen er det vedtatt at kritiske belastningsgrenser skal legges til grunn ved utarbeidelse av nye avtaler om utslippsbegrensning av svovel, nitrogen og hydrokarboner.

En styringsgruppe i Miljøverndepartementet har det overordnede ansvar for programmet, mens ansvaret for den faglige oppfølgingen er overlatt en arbeidsgruppe bestående av representanter fra Direktoratet for naturforvaltning (DN), Norsk polarinstitutt (NP) og Statens forurensningstilsyn (SFT).

Arbeidsgruppen har for tiden følgende sammensetning:

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Else Løbersli - DN  
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Styringsgruppen i Miljøverndepartementet består av representanter fra avdelingen for naturvern og kulturminner, avdelingen for vannmiljø, industri- og avfallssaker og avdelingen for internasjonalt samarbeid, luftmiljø og polarsaker.

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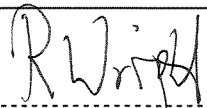
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**Abstract**

Here we test the hypothesis that increased deposition of inorganic N compounds leads to increased leaching and runoff of organic N and thus a higher critical load. We use mainly Norwegian data from input-output fluxes at small catchments, national lake surveys, and large-scale experiments with N deposition to whole catchments. Concentrations of organic-N are not significantly related to N deposition. Much of the variance in organic-N levels are explained by TOC concentrations. For the small catchments, there is a significant relationship between C/N ratio in dissolved organic matter and the N deposition. The sites with high N deposition have low C/N ratio. Chronically high N deposition and long-term accumulation of N in soils and biomass may have led to organic matter more enriched in N relative to pristine sites. Time trend data from manipulated catchments do not show changes in organic-N leaching over 4-10 years. Although organic N levels may have increased as a result of N deposition, the resultant effect on estimate of critical load for nitrogen for freshwaters is minor. For practical purposes organic N outputs can be neglected in estimating and mapping critical loads for nitrogen in Norway.

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1. tålegrenser	1. critical loads
2. nitrogen	2. nitrogen
3. vann	3. water
4. jord	4. soil

  
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# **Atmospheric deposition of nitrogen, runoff of organic nitrogen, and critical loads for soils and waters**

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## **Preface**

This report is prepared as part of work carried out under the programme “Naturens tålegreiser” under contract L-85/96 NATÅL from DN (Directorate for Protection of Nature) and contract 96-6-712-00 from MD (Ministry of Environment). The project was conducted by NIVA, NISK, and Department of Soil and Water Sciences, NLH. We thank Ann-Kristin Buan for technical assistance.

Oslo, November 1996

*Richard F. Wright, project leader*

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## Summary

Standard methods for calculation of critical loads of nitrogen for soils and surface waters consider only deposition and runoff of inorganic nitrogen compounds. In theory it is possible that increased deposition of inorganic nitrogen compounds ( $\text{NO}_x$  and  $\text{NH}_x$ ) could lead to increased leaching of organic forms of nitrogen. Increased leaching of organic N thus implies a higher critical load.

Here we test the hypothesis that increased deposition of inorganic N compounds leads to increased leaching and runoff of organic N from terrestrial ecosystems. We use mainly Norwegian data from input-output fluxes at small catchments, national lake surveys, and large-scale experiments with N deposition to whole catchments.

In Norwegian lakes surveyed in 1995 concentrations of organic-N are not significantly related to N deposition. About 80% of the variance in organic-N levels are explained by TOC concentrations.

In runoff from small catchments nitrate fluxes are correlated to N deposition, but there is no relationship between N deposition and organic-N flux in runoff. About 80% of the variance in organic-N levels are explained by TOC concentrations. For the small catchments, there is a significant relationship between C/N ratio in dissolved organic matter and the N deposition. The sites with high N deposition have low C/N ratio. This may indicate that chronically high N deposition and long-term accumulation of N in soils and biomass has led to organic matter more enriched in N relative to pristine sites. Time trend data from manipulated catchments do not show changes in organic-N leaching over 4-10 years.

Increased N-deposition may lead to increased decomposition of organic matter in soil due to increased microbiological activity, which in turn may lead to increased total organic carbon (TOC) and organic-N concentrations in soil solution. The data do not suggest that increased N deposition to forest ecosystems leads to changes in production and transport of organic-N in soil solution.

We conclude that while there is evidence from small catchment studies that organic N levels may have increased as a result of N deposition, the resultant effect on estimate of critical load for nitrogen for freshwaters is minor. For practical purposes organic N outputs can be neglected in estimating and mapping critical loads for nitrogen in Norway.

# 1. Introduction

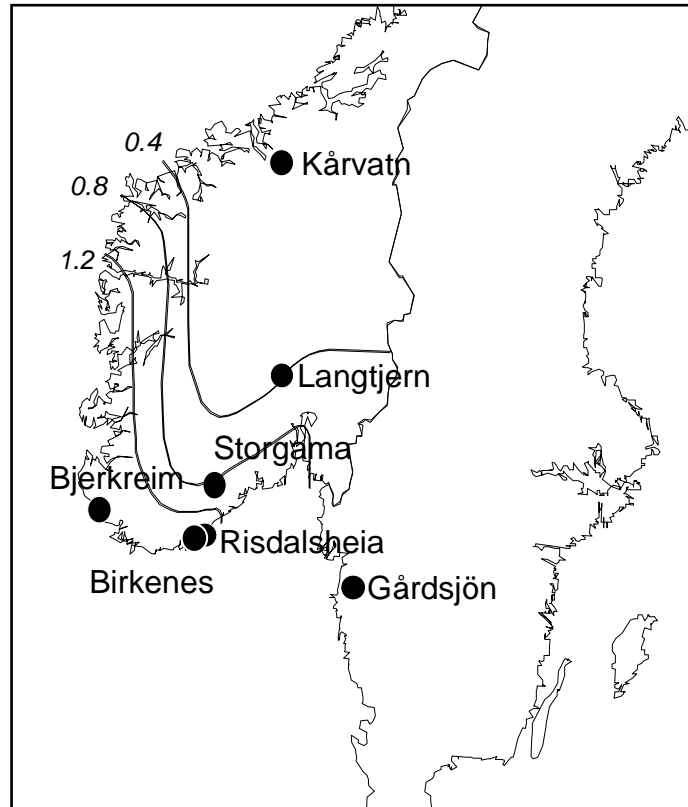
Standard methods for calculation of critical loads of nitrogen for soils and surface waters consider only deposition and runoff of inorganic nitrogen compounds (Hornung et al. 1994, Posch et al. 1995). The equations include processes involving inorganic N species such as uptake and mineralisation, but do not consider transformation of inorganic to organic N forms. In theory it is possible that increased deposition of inorganic nitrogen compounds ( $\text{NO}_x$  and  $\text{NH}_x$ ) could lead to increased leaching of organic forms of nitrogen. Organic nitrogen is commonly defined as the concentration of total dissolved nitrogen less the concentrations of nitrate and ammonium. Organic nitrogen is electrochemically neutral and thus by itself does not contribute to the acidification of soils or waters. If deposition of inorganic N leads to increased leaching of organic N, the system tolerates a higher N loading before leaching of inorganic N exceeds the set criterion; this thus implies a higher critical load.

Here we test the hypothesis that increased deposition of inorganic N compounds leads to increased leaching and runoff of organic N from terrestrial ecosystems. This hypothesis can be tested in two ways:

- (1) by examining empirical relationships between nitrogen deposition and runoff at catchments and lakes located over a geographical area covering a range of N deposition. This is an indirect test in that it relies on the “time-space” analogy, by which present-day geographic patterns are assumed to reflect changes in time at any given site.
- (2) by examining results from whole-ecosystem experiments at which the N deposition is experimentally changed and the effects on runoff are measured. Although this is a direct test, it has the disadvantage that the manipulated ecosystems have only experienced changed N deposition for a small number of years (4-10). These ecosystems probably have not yet fully adjusted to the changed deposition.

We use mainly Norwegian data from input-output fluxes at small catchments, national lake surveys, and large-scale experiments with N deposition to whole catchments (Figure 1). These data include measurements of major ions, Al-species, total organic carbon (TOC), and total dissolved nitrogen (TON). Together they provide a basis for assessing geographical patterns of organic-N leaching across a gradient in N deposition, and changes in organic-N leaching over time under changing N deposition.





**Figure 1.** Map of Norway and adjacent area of Sweden showing location of experimental sites and monitored catchments. Isolines indicate ambient deposition of inorganic N ( $\text{g N m}^{-2} \text{yr}^{-1}$ ) (deposition data from Skjelkvåle 1996).

## 2. Data sources

### 2.1 National lake surveys

In autumn 1995 a survey of 1500 Norwegian lakes was conducted to document the current status of acidification and changes in acidification since earlier surveys in 1974 and 1986 (Skjelkvåle *et al.*, in preparation) (Figure 2). About 1000 of the lakes were selected to statistically represent the entire population of Norwegian lakes, while the other 500 were lakes previously sampled in 1986. The latter are mostly small, acid-sensitive lakes unaffected by local pollution sources such as agricultural or municipal effluents, and as such are more suited for our analysis of the role of organic N. We use these 500 lakes. The 1995 survey included measurement of total dissolved N, and for the first time provides data for geographical patterns of organic N in Norwegian freshwaters. We excluded 47 of the 498 lakes from the analysis because they had concentrations of TOC below the detection limit of  $0.2 \text{ mgC L}^{-1}$  ( $16 \text{ mmol L}^{-1}$ ) and/or concentrations of organic-N below  $5 \text{ } \mu\text{mol L}^{-1}$ .



**Figure 2.** Map of Norway showing locations of the 500 lakes in the 1995 national survey. These lakes are small and acid-sensitive, and were originally sampled in the 1974-75 and/or 1986 surveys (from Skjelkvåle et al., in preparation).

## 2.2 Catchment studies

### 2.2.1 Norwegian monitoring program

Atmospheric deposition and runoff of water and dissolved chemical compounds are measured regularly at 6 small catchments as part of a national program to monitor deposition and effects of long-range transported air pollutants (Skjelkvåle 1996). Four of these catchments have complete data for several years. The catchments Birkenes, Storgama, Langtjern, and Kårvatn span a gradient in ambient N deposition (1994 values) from 120 mmol m<sup>-2</sup> yr<sup>-1</sup> (17 kg ha<sup>-1</sup> yr<sup>-1</sup>) at Birkenes in the south to 25 mmol m<sup>-2</sup> yr<sup>-1</sup> (3.6 kg ha<sup>-1</sup> yr<sup>-1</sup>) at Kårvatn on the west coast (Skjelkvåle 1995) (Figure 1). Two of the catchments (Birkenes and Langtjern) are in coniferous forests, while the other 2 (Storgama and Kårvatn) have mostly alpine heath and peatlands. These catchments have been monitored since as early as 1972 (Birkenes), but complete yearly data for all 3 nitrogen species (NH<sub>4</sub>, NO<sub>3</sub> and organic-N) are available only beginning in 1994.

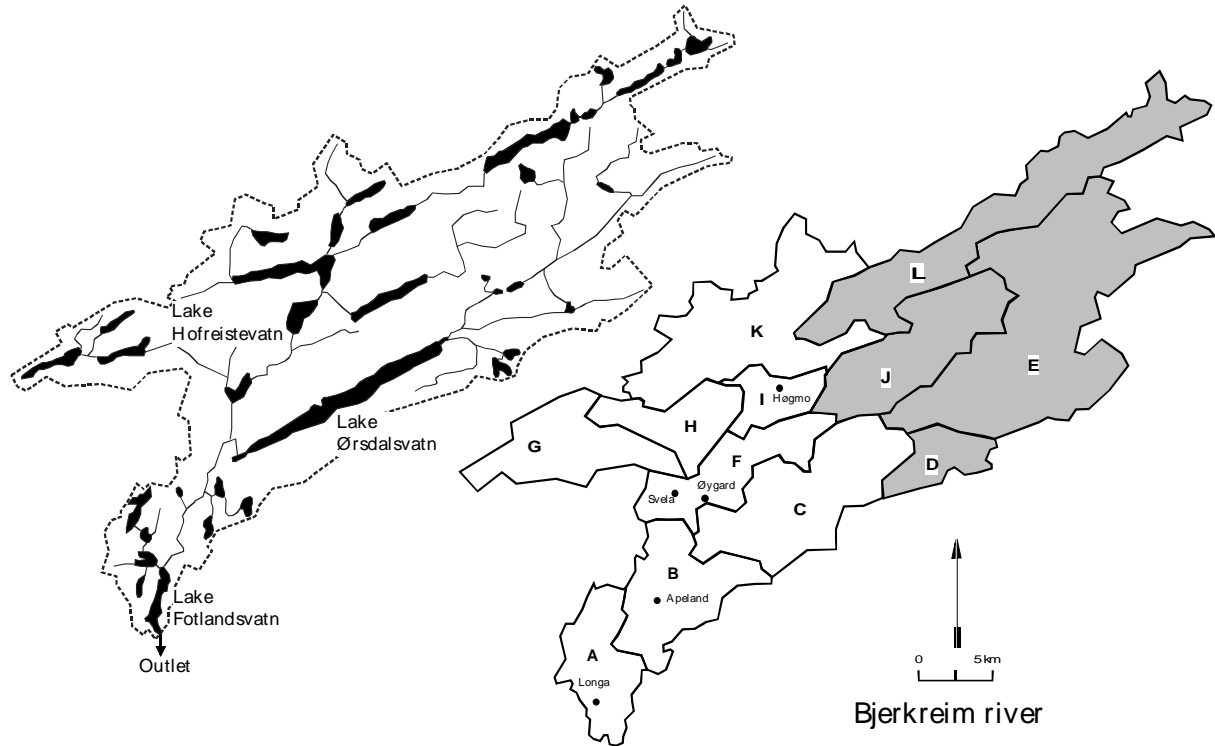
### 2.2.2 River Bjerkreim

During the period 1993-1995 River Bjerkreim, south-western Norway, was intensively studied as a part of the project “Nitrogen from mountains to fjords”, which includes studies of atmospheric deposition, N fluxes through agricultural and forest soils, and N fluxes to surface waters (Hessen and Henriksen 1994).

River Bjerkreim is situated in south-west Norway, in an area which is characterised by relative high N concentrations in precipitation (up to 70 mmol L<sup>-1</sup>) and high precipitation amounts (1.5-3.5 m yr<sup>-1</sup>). Total N deposition (wet + dry) was 11-27 kg N ha<sup>-1</sup> yr<sup>-1</sup> in different subcatchments during the project period. The river drains 685 km<sup>2</sup> and has an average specific runoff of 77 L s<sup>-1</sup> km<sup>-2</sup>.

N budgets were calculated for 19 subcatchments within the river system (Kaste *et al.*, in preparation), of which 9 are used here for evaluation of the role of organic-N (Figure 3). The subcatchments differ by their dominant land cover characteristics. Catchments affected by agriculture were not included here.

Forested catchments : SVELA, HØGMO, APELAND 1  
Heathland catchments: LONGA, ØYGARD  
Mountainous catchments: D, E, J, L



**Figure 3.** Map of the River Bjerkreim basin, south-western Norway, showing locations of small subcatchments and lakes at which input-output budgets were measured (from Kaste *et al.*, In preparation).

Chemical data are available from the outlet streams of all subcatchments. At the sites D, E, J, and L point samples are collected biweekly during the period 1992-1995, except at sites J and L where sampling frequency in 1994 was reduced to monthly, due to low chemical variation (the sites are located downstream large lakes). In five small catchments, which were picked out for more detailed studies (HØGMO, LONGA, ØYGARD, SVELA, APELAND 1), volume proportional samples are collected.

Soil solution data are available from the forest subcatchment Svela (Mulder *et al.*; in preparation). About 60% of the catchment is planted with forest. SVELA is mainly afforested with conifers in the lower areas, primarily Norway spruce. A large stand is dominated by mountain pine and downy birch. A recent clear-cut has been planted with larch. The parent material at SVELA is sandy till, containing large amounts of stones and boulders. Soil water is analysed at 10, 30 and 70 cm depth at two plots: S1 is a middle-aged spruce stand, and S2 is a young larch tree plantation.

In the Bjerkreim catchment input-output data are available from three lakes during the period 1993-1995 (Berge *et al.*; in preparation). Lake Ørsdalsvatn and Lake Hofreistevatn are deep and oligotrophic with little biological activity, whereas Lake Fotlandsvatn is dominated by shallow areas with substantial growth of macrophytes, mainly *Juncus bulbosus* and *Sphagnum* sp. Lake Fotlandsvatn are characterised by a very short residence time (~ one day).

## 2.3 Large-scale experiments

### 2.3.1 RAIN project, Risdalsheia, Norway

As part of the RAIN project (Reversing Acidification In Norway) a large-scale whole catchment manipulation experiment was conducted at Risdalsheia, near Grimstad, southernmost Norway (Wright *et al.* 1993). At the 860-m<sup>2</sup> KIM catchment ambient precipitation inputs of acid, sulphur and nitrogen compounds were removed by means of a transparent roof above the canopy and clean rain with natural levels of seasalts added beneath the roof by a sprinkling system. Nitrogen deposition was reduced from ambient levels of about 80 mmol m<sup>-2</sup> yr<sup>-1</sup> (12 kg ha<sup>-1</sup> yr<sup>-1</sup>) to about 18 mmol m<sup>-2</sup> yr<sup>-1</sup> (2.5 kg ha<sup>-1</sup> yr<sup>-1</sup>). Runoff discharge and concentrations of NH<sub>4</sub> and NO<sub>3</sub> have been measured since 1983, and total N since June 1991. The experiment started in 1984 and continued uninterrupted through May 1994 when it was succeeded by the CLIMEX project (Climate change experiment) (Jenkins *et al.* 1993).

### 2.3.2 NITREX project, Gårdsjön, Sweden

The NITREX project (Nitrogen saturation experiments) entails large-scale experiments with manipulation of nitrogen deposition (Wright and van Breemen 1995). One of these experiments is underway at Gårdsjön near Gothenburg on the Swedish west coast (Moldan and Wright, *in press*). Here nitrogen deposition has been experimentally increased by weekly additions of NH<sub>4</sub>NO<sub>3</sub> to a 0.56 ha catchment with mature spruce forest. The experiment increased nitrogen deposition from ambient 75 mmol/m<sup>2</sup>/yr (11 kg ha<sup>-1</sup> yr<sup>-1</sup>) to 350 mmol m<sup>-2</sup> yr<sup>-1</sup> (49 kg ha<sup>-1</sup> yr<sup>-1</sup>). Runoff discharge and concentrations of NH<sub>4</sub>, NO<sub>3</sub>, and total N have been measured since April 1989. Additions began April 1991.

At Gårdsjön studies of soil and soil solution are part of the investigations (Raastad and Mulder, *in press*; Stuanes and Kjønnaas, *in press*), and data from these studies include concentrations of N species in soil solution before and following the N additions.

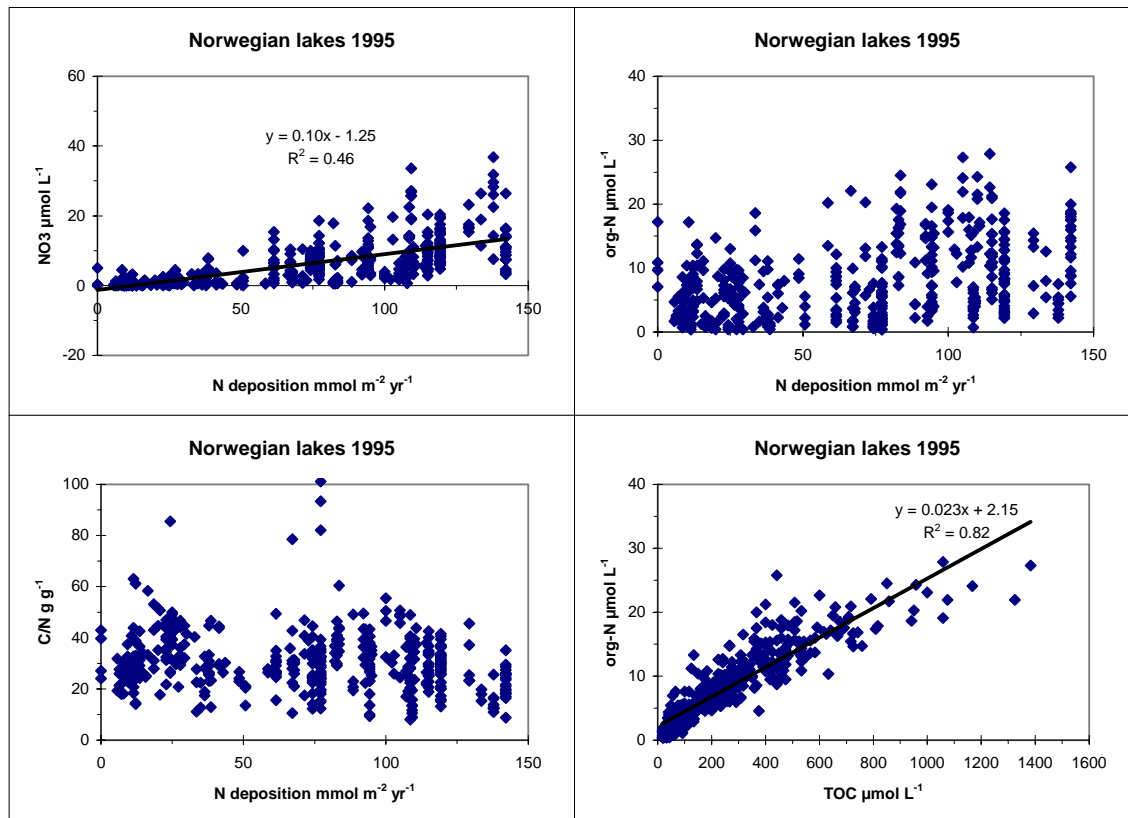
## 3. RESULTS

### 3.1 Geographic patterns

#### 3.1.1 National lake survey

The lake survey data show the familiar relationship of elevated concentrations of NO<sub>3</sub> in areas receiving N deposition greater than about 50 mmol m<sup>-2</sup> yr<sup>-1</sup> (Figure 4). At high (for Norway) levels of N deposition, some lakes have elevated NO<sub>3</sub> concentrations while others have very low concentrations. N deposition appears to be a necessary but not a sufficient factor in accounting for elevated NO<sub>3</sub> concentrations in Norwegian lakes. Only about 46% of the variance is explained by N deposition, and clearly there are other factors which govern the NO<sub>3</sub> concentrations in lakes.

Concentrations of organic-N, on the other hand, are not significantly related to N deposition (Figure 4). Organic-N levels are clearly related to TOC concentrations; TOC explains about 80% of the variance. The C/N ratio of dissolved organic matter in Norwegian lakes is not related to N deposition; the average C/N ratio is about 31.



**Figure 4.** Relationships between  $\text{NO}_3^-$ , organic-N, and C/N ratio in dissolved organic matter and TOC and N-deposition in 451 Norwegian lakes sampled in autumn 1995 (data from Skjelkvåle *et al.*, in preparation). Statistically significant linear regressions are shown.

### 3.1.2 Small catchments

At the 4 small catchments in the Norwegian monitoring programme runoff of inorganic nitrogen compounds (mostly  $\text{NO}_3^-$ ) is moderate at Birkenes and Storgama, but very low at Langtjern and Kårvatn (Table 1). Runoff of organic N (and TOC) is moderate at Birkenes, Storgama and Langtjern, and very low at Kårvatn (Skjelkvåle 1995).

Surface water in the Bjerkreim River catchment generally has low TOC concentrations (Table 1). In some catchments  $\text{NO}_3^-$  contributes up to 40% of the surface water acidification (Henriksen *et al.* in prep.). Yearly average  $\text{NO}_3^-$  concentrations were closely related to land use and land cover characteristics of the catchments: The lowest  $\text{NO}_3^-$  concentrations (6-11  $\mu\text{eq L}^{-1}$ ) and fluxes were recorded in outlet streams of the forested catchments HØGMO, APELAND-1, and SVELA. In mountainous areas (D, E, J, L)  $\text{NO}_3^-$  levels were a bit higher (15-17  $\mu\text{eq L}^{-1}$ ), while the heathland sites (ØYGARD, LONGA) were intermediate.

Organic N concentrations were remarkably uniform (5-12  $\mu\text{mol L}^{-1}$ ) compared to  $\text{NO}_3^-$  (6-45  $\mu\text{eq L}^{-1}$ ) in the subcatchments. As  $\text{NO}_3^-$  varied by a factor of 7 among the catchments, the corresponding organic N ratio was only 2. Further, there was no distinct coupling between organic N concentrations and land use or land cover characteristics, as were recorded for  $\text{NO}_3^-$ . On the other hand, there seemed to be a rather tight coupling between organic N and total organic C concentrations in streamwater (Fig. 4). Average molar organic C/N-ratios were rather uniform in forests, heathlands, mountainous areas (11-20).

**Table 1.** Volume-weighted fluxes of inorganic nitrogen inputs and nitrogen and carbon fluxes in runoff at small catchments in Norway and Gårdsjön in Sweden. Units:  $\text{mmol m}^{-2} \text{yr}^{-1}$ . Data sources: Risdalsheia (Wright and Lükewille, in press); Gårdsjön (Moldan and Wright, in press); monitored catchments (Skjelkvåle 1995); Bjerkreim (Kaste et al., in preparation).

**Monitored, control and reference catchments**

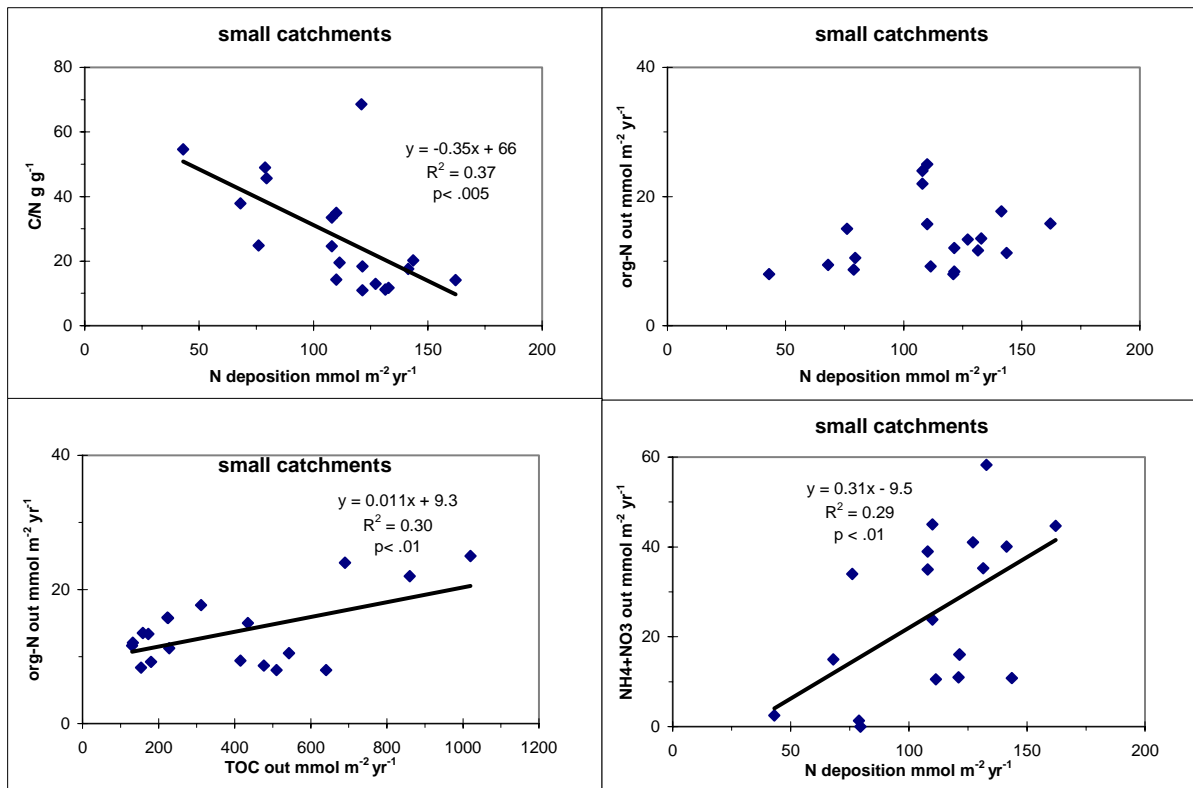
	period	site	deposition			runoff					
			NH <sub>4</sub>	NO <sub>3</sub>	sum	NH <sub>4</sub>	NO <sub>3</sub>	NH <sub>4</sub> + NO <sub>3</sub>	org-N	TOC	C/N g/g
Risdalsheia	93-94	EGIL	35	41	76	6	28	34	15	435	25
	93-94	ROLF	51	59	110	16	29	45	25	1020	35
	93-94	CEC	50	58	108	7	28	35	24	690	25
	93-94	MET	50	58	108	10	29	39	22	860	34
Gårdsjön	89-91	G2	27	53	80	0	0	0	11	543	46
	89-95	F1	27	52	79	1	1	1	9	477	49
monitored catchments	1994	Birkenes	63	58	121	1	10	11	8	640	69
	1994	Storgama	29	39	68	1	14	15	9	415	38
	1994	Langtjern	23	20	43	1	2	3	8	510	55
	1994	Kårvatn	12	13	25	0	2	2	1	140	92
Bjerkreim	1993-95	HØGMO			144	1	10	11	11	228	20
	1993-95	LONGA			110	1	23	24	16	225	14
	1993-95	ØYGARD			121	1	15	16	12	132	11
	1993-95	SVELA			121	1	15	16	8	154	18
	1993-95	APELAND 1			111	1	10	11	9	180	20
	1993-95	D			127	1	40	41	13	173	13
	1993-95	E			141	1	39	40	18	311	18
	1993-95	J			162	1	44	45	16	223	14
	1993-95	L			131	1	34	35	12	131	11
	1993-95	Main outlet			133	1	57	58	14	159	12
<b>Manipulated catchments</b>											
Risdalsheia	93-94	KIM	5	9	14	1	1	2	20	860	37
Gårdsjön	92-95	G2	173	192	365	2	11	13	12	629	47

Runoff from the untreated reference catchments at Risdalsheia (RAIN project) has high concentrations of TOC and organic-N as well as high levels of NO<sub>3</sub>. Also NH<sub>4</sub> concentrations are high relative to the other catchments included here (Table 1). The data clearly indicate that the site is saturated.

At the untreated control catchment F1 at Gårdsjön (NITREX) runoff has moderate concentrations of TOC and organic N and very low levels of inorganic N species, despite the relatively high ambient N deposition (Table 1).

Together the data for 20 of these small catchments show many of the same patterns as the lake data (Figure 5). Data from Kårvatn were omitted in the analysis because of the very low TOC and organic-N concentrations at Kårvatn. Nitrate fluxes in runoff are again correlated to N deposition, but there is no relationship between N deposition and organic-N flux in runoff. Organic-N flux is related to TOC flux, but only 30% of the variation in organic-N flux is explained by TOC flux. For the small catchments, there is a significant relationship between C/N ratio in dissolved organic matter and the N

deposition. The sites with high N deposition have low C/N ratio. This may indicate that chronically high N deposition and long-term accumulation of N in soils and biomass has led to organic matter more enriched in N relative to pristine sites.



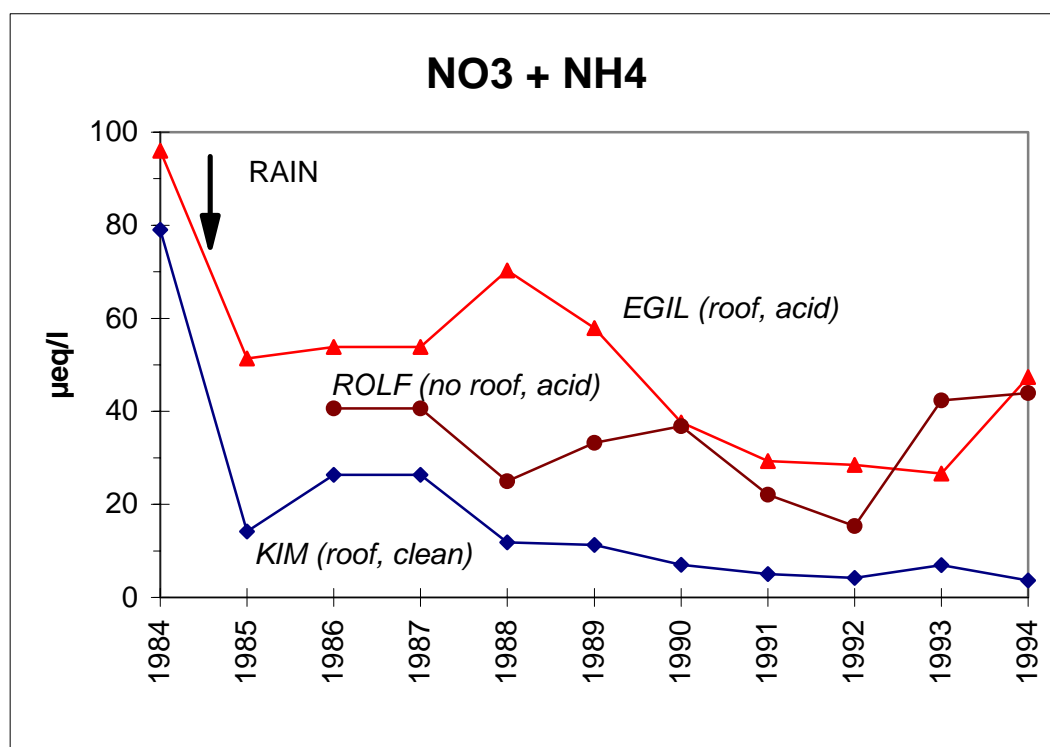
**Figure 5.** Relationships between fluxes in runoff of NO<sub>3</sub> and organic-N and C/N ratio in dissolved organic matter vs. TOC flux and N-deposition in 20 small catchments (see Table 1 for data sources). Statistically-significant linear regressions are shown.

## 3.2 Time trends

### 3.2.1 Risdalsheia (RAIN project)

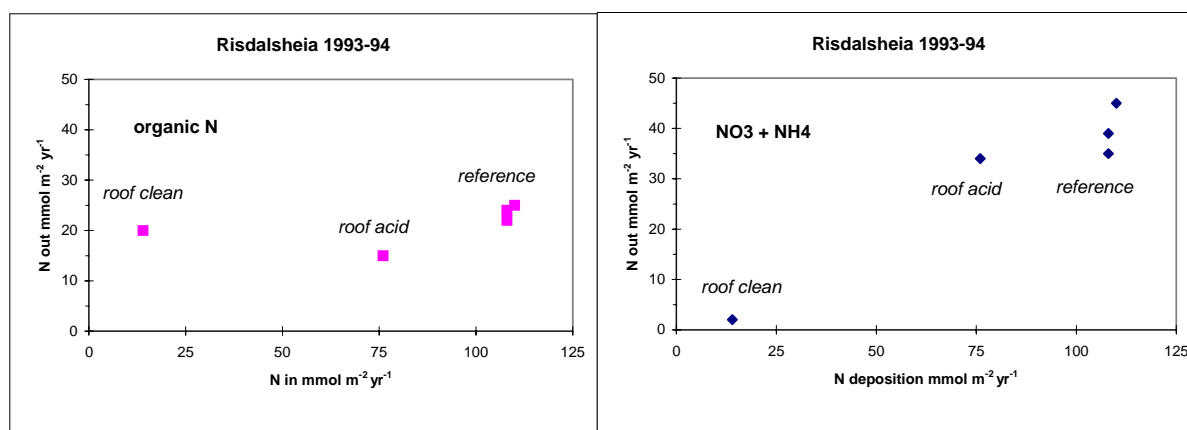
At Risdalsheia the experimentally-reduced N deposition resulted in immediate and large decreases in concentrations and fluxes of inorganic N species in runoff (Figure 6). The 10-year record from KIM catchment (roof, clean rain) shows permanent decreases in both NH<sub>4</sub> and NO<sub>3</sub> from the first year of treatment, while levels remained high at both EGIL catchment (roof, acid rain) and ROLF (no roof, acid rain).





**Figure 6.** Time trends of volume-weighted mean concentrations of inorganic N ( $\text{NH}_4 + \text{NO}_3$ ) in runoff at the 3 RAIN project catchments at Risdalsheia over the 10-year period 1984-1994.

Data for organic-N at these catchments and at 2 additional untreated reference catchments (CEC and MET) indicate no major difference among the catchments related to N deposition (Figure 7). Measurements of total N (and thus organic N) began only in June 1991 when the experiment was already in its seventh year of treatment, and thus there are no direct data from Risdalsheia to show the effect of reduced N deposition on organic N in runoff. Nevertheless the fact that the organic N outputs at these 5 catchments are quite similar and show no relation to N deposition, strongly indicates that organic N outputs at KIM catchment have not changed as a result of treatment.

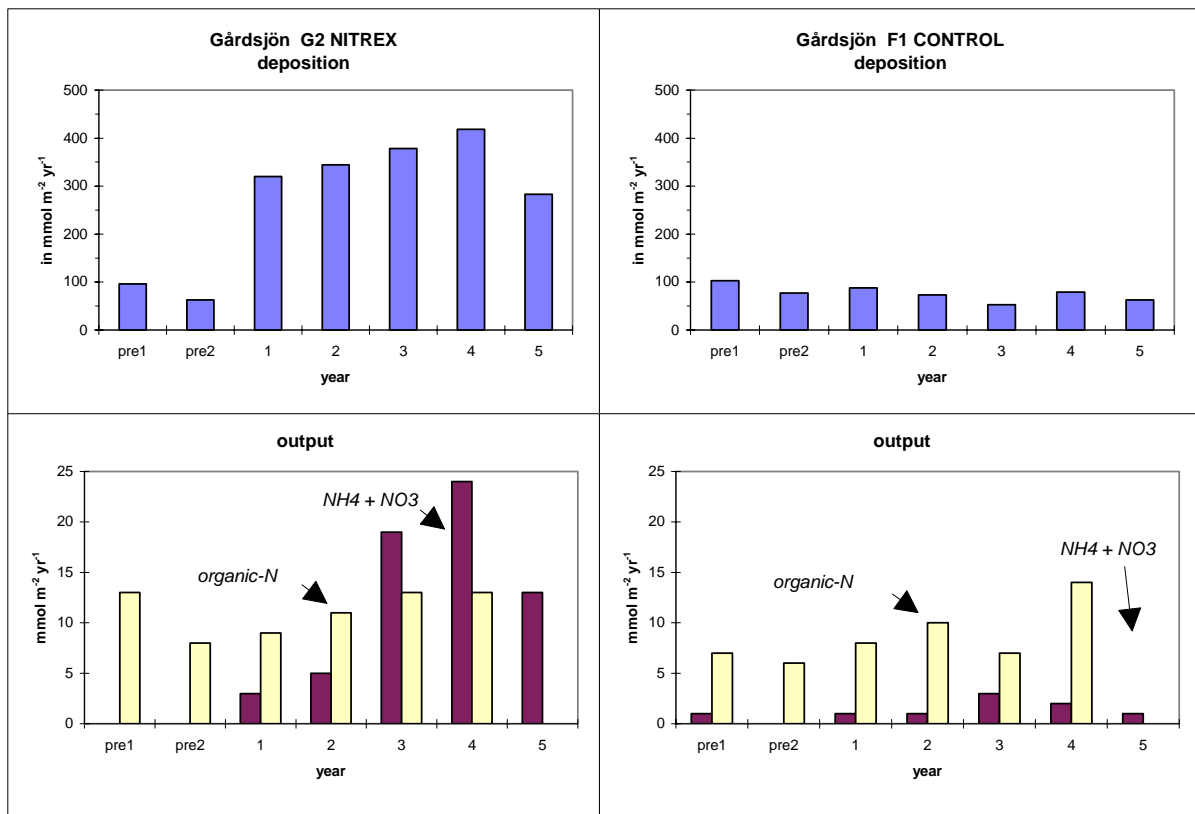


**Figure 7.** Fluxes of inorganic-N and organic-N in runoff vs. N-deposition at 5 catchments at Risdalsheia for the year June 1993-June 1994, the last year of measurements prior to the onset of CLIMEX treatments.

Concentrations of organic N in runoff, on the other hand, are highly correlated to concentrations of total organic carbon (TOC). The C/N ratio ( $\text{g g}^{-1}$ ) of dissolved organic matter in runoff at Risdalsheia ranges from 25-45, apparently unrelated to N deposition (Table 1).

### 3.2.2 Gårdsjön (NITREX project)

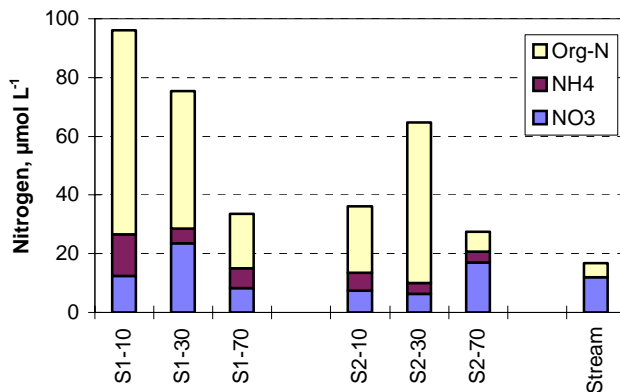
At Gårdsjön the experimentally-increased nitrogen deposition at catchment G2 NITREX resulted in increased runoff of inorganic N compounds, but no change in runoff of organic-N (Figure 8). Flux of organic-N from catchment G2 NITREX is about  $10 \text{ mmol m}^{-2} \text{ yr}^{-1}$ , somewhat higher than the flux at the untreated catchment F1 CONTROL. This difference can be explained by the similar differences in fluxes of TOC; the average over 6 years at G2 is  $640 \text{ mmol m}^{-2} \text{ yr}^{-1}$  and at F1 is  $510 \text{ mmol m}^{-2} \text{ yr}^{-1}$ . The average C/N ratio ( $\text{g g}^{-1}$ ) of dissolved organic matter in runoff is about 47 at both catchments.



**Figure 8.** Time trends of deposition and runoff of nitrogen compounds at G2 NITREX catchment and F1 CONTROL catchment at Gårdsjön, Sweden, over the 6-year period 1989-1995 (data from Moldan and Wright, in press).

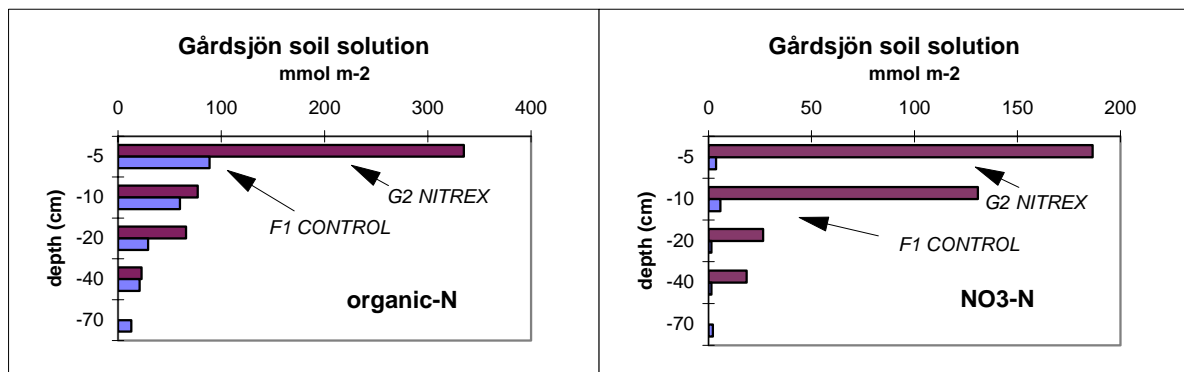
### 3.3 Soil solution

In a comparison of soil water and stream water chemistry in the forested catchment SVELA, streamwater concentrations of total N and organic N were lower than those in soil water from 10, 30, or 70 cm depths (Figure 9). Higher inorganic N/organic N - ratios in streamwater suggests that mineralisation of organic N in ground water or in the stream could have taken place. On the other hand, due to the great spatial variability in soil water chemistry and uncertainties attributed to water pathways, it is difficult to make direct comparisons to stream water chemistry at the catchment scale.



**Figure 9.** Middle N concentrations in soil water and stream water at SVELA 1994-1995. Soil water data from Mulder *et al.* (In prep.).

Similarly data from soil solution at the Gårdsjön catchments do not indicate changes in mobilisation of organic N as a result of the nitrogen additions of the NITREX experiment (Raastad and Mulder, in press). Concentrations of nitrate increased in soil solution, especially in the surface layers, but there was no apparent change in organic N concentrations (Figure 10) (Stuanes and Kjønås, in press). Again there is a close correlation between TOC and TON both from plot-to-plot within each catchment, and over time during the growing season (Raastad and Mulder, in press).



**Figure 10.** Volume-weighted average concentrations of nitrogen compounds in 1994 in soil solution at catchment G2 NITREX and F1 CONTROL at Gårdsjön, Sweden. Concentrations of DON were calculated as the difference between total dissolved N and dissolved inorganic N ( $\text{NH}_4^+\text{-N}$  and  $\text{NO}_3^-\text{-N}$ ). If measured  $\text{NH}_4^+$  and  $\text{NO}_3^-$  were below the detection limit, concentrations are set to 50 % of the detection limit (data from Stuanes and Kjønås, in press).

A study of dissolved organic matter (DOM) in Gårdsjön showed that there were large variations in the median C/N ratios in soil solution at different plots and at different depths (Raastad and Mulder, in press) (Table 2), but no changes in the C/N ratio was found in soil solution due to manipulation (N addition and reversal of acidification). This was mainly because of large spatial variation within catchments.

**Table 2.** Median C/N weight-ratios in DOM in soil solution (podzol) at Gårdsjön (from Raastad and Mulder, (in press).

Depth [cm]	NITREX (G2Ly2)	Roof (G1 IVL Ly1)	Control (FILy1)
5	28	18	35
10	18	20	31
20	53	28	38
40	43	35	34

### 3.4 In-lake processes

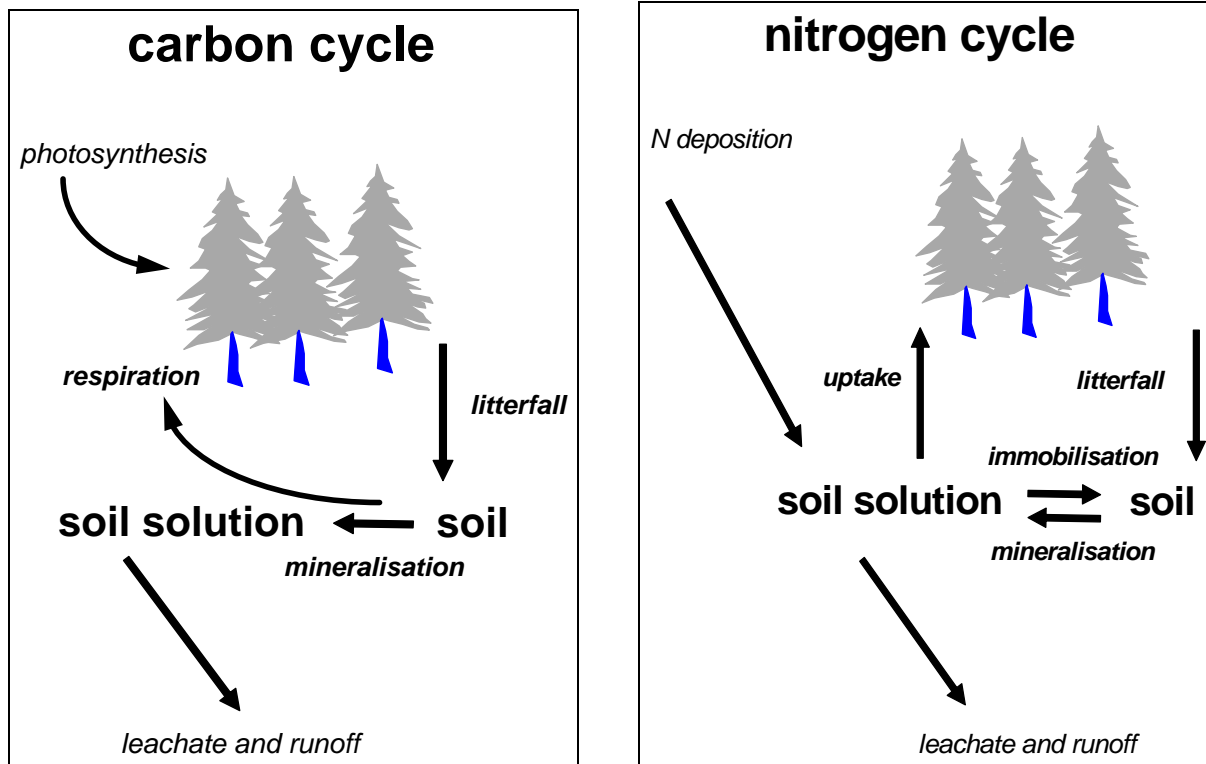
Input-output fluxes from the lakes in the Bjerkreim River catchment indicate a net retention of N in Lake Ørsdalsvatn and Lake Fotlandsvatn (Table 3) (Berge *et al.*; in preparation). Expressed per surface unit, Lake Fotlandsvatn had the highest N retention, about 50 kg ha<sup>-1</sup> yr<sup>-1</sup>. The seeming lack of retention in Lake Hofreistevatn is probably due to an underestimation of N inputs from the local catchment. In the two most oligotrophic lakes, Lake Ørsdalsvatn and Lake Hofreistevatn, a transformation of incoming organic N to NO<sub>3</sub><sup>-</sup> seems to occur. In Lake Fotlandsvatn, on the other hand, relatively more NO<sub>3</sub><sup>-</sup> was held back, while there seemed to be a net export of organic N.

**Table 3.** Nitrogen retention in lakes, calculated from chemical input-output data 1993-1995. Negative values indicate N-retention.

		Tot-N	NO <sub>3</sub> -N tonnes yr <sup>-1</sup>	Org-N	Tot-N kg ha <sup>-1</sup> yr <sup>-1</sup>	NO <sub>3</sub> -N kg ha <sup>-1</sup> yr <sup>-1</sup>	Org-N lake surface
Ørsdalsvatn	93/95	-11.2	5.1	-16.3	-9.1	4.2	-13.2
Hofreistevatn	93/95	2.2	5.4	-3.3	7.7	19.4	-11.7
Fotlandsvatn	93/95	-4.5	-6.5	2.0	-50.1	-71.9	21.8

## 4. Discussion

Interpretation of the relationships between inorganic nitrogen, organic nitrogen and total organic carbon in leachate and runoff necessitates consideration of the nitrogen and carbon cycles in terrestrial ecosystems (Figure 11). Most of the inorganic N is deposited on vegetation and soils. Typically only a small fraction of incoming precipitation falls directly on stream and lake surfaces. A fraction of the inorganic nitrogen (mostly nitrate) in surface waters may be “hydrologic nitrate”, a term used to denote nitrate that is leached when the contact time between precipitation and soil is too short to allow full opportunity for removal of nitrogen compounds by inorganic or organic processes.



**Figure 11.** Schematic view of the nitrogen and carbon cycles in terrestrial ecosystems.

Nitrogen deposition enters the nitrogen cycle either by processes such as microbial immobilisation and ion exchange in soil and uptake by vegetation. Most terrestrial catchments in areas of Norway not receiving significant nitrogen deposition are nitrogen limited, and typically over 90% of the incoming inorganic nitrogen is retained in the catchment. These ecosystems have very tight nitrogen cycles. The little nitrogen lost is generally lost in the form of organic N.

The nitrogen in dissolved organic matter derives from incomplete decomposition of litter and soil organic matter. The link between increased deposition of inorganic N and a possible increase in runoff of organic N thus goes through the entire N cycle. But because the organic N is a component of the dissolved organic matter, the key to understanding organic-N lies in understanding factors controlling TOC in runoff. Catchments may export more organic-N either by exporting more dissolved organic matter, or by exporting organic matter more enriched in nitrogen (i.e. lower C/N ratio).

Increased nitrogen deposition leads to increased storage of N in vegetation and soils. In forests chronic deposition of nitrogen may cause increased growth. But also the C/N ratio in both vegetation and soils increases, as has been shown by both fertilisation experiments (Tamm 1992) and the whole-ecosystem manipulations of the NITREX project (Boxmann et al., in press).

The influence of nitrogen deposition on the C/N ratios of biomass and soil organic matter depends on the amount of deposition, the length of time, and the capacity of the system to retain nitrogen. The capacity is governed by such factors as soil depth, amount of biomass, and percent carbon in the soil. But not all soil organic matter actively participates in the N cycle; in soils of cold, temperate regions probably a major fraction of the soil organic matter is essentially “inert”, in that it has accumulated over the thousands of years since deglaciation and the easily-decomposable fractions have been lost long ago.

Catchment data indicate that the forested sites have higher C/N ratios in dissolved organic matter, and also lower NO<sub>3</sub> concentrations in runoff (Table 1). This suggests that forests store more N in active biomass and soil organic matter relative to heathlands. This is certainly the case at Bjerkreim -- dissolved organic matter in runoff from forested catchments have C/N 18-20, whereas the non-forested catchments (with 1 exception) have C/N 11-14.

Quantitative estimates of the amount of active biomass and soil organic matter can be derived from applications of nitrogen-process models to manipulated catchments. One such model, MERLIN (Model of Ecosystem Retention and Loss of Inorganic Nitrogen; Cosby et al., in press), has been applied to both Risdalsheia (Beier and Wright, in preparation) and Gårdsjön G2 NITREX (Kjønaas et al. in preparation). A rough estimate indicates that at Risdalsheia about 30 years and Gårdsjön 60 years of present-day N deposition would be necessary to change the C/N ratios of active vegetation and soil organic matter by 10 (Table 4). This length of time is of the same order of magnitude to the actual number of years of historical N deposition at present-day levels.

**Table 4.** Carbon and nitrogen pools and fluxes for untreated catchments at Risdalsheia and Gårdsjön. SOM: soil organic matter. Also shown are estimates for number of years of present-day deposition required to change the C/N ratios of the active pools assuming all incoming N is retained and there is no change in carbon pools. In the case of Risdalsheia the calculation is made from the past to the present, while at Gårdsjön the calculation is for the future. (Data from Beier et al., in preparation; Kjønaas et al., in preparation).

	Risdalsheia			Gårdsjön		
	C	N	C/N	C	N	C/N
<b>pools mol m<sup>-2</sup></b>						
active vegetation	44	1.05	42	99	1.58	62
woody vegetation	192	1.64	117	546	1.63	335
active SOM	149	7	21	397	11.3	35
passive SOM	594	28	21	1226	38	32
<b>fluxes mmol m<sup>-2</sup> yr<sup>-1</sup></b>						
deposition		83			84	
runoff						
inorg N		51			0	
org N		20			11	
TOC		810			350	
C/N		40			32	
<b>to change from</b>	<b>C/N</b>	<b>ΔN mol m<sup>-2</sup></b>		<b>C/N</b>	<b>ΔN mol m<sup>-2</sup></b>	
active vegetation	52 to 42	0.2		62 to 52	0.3	
active SOM	31 to 21	2.2		35 to 25	4.6	
sum		2.4			4.9	
years of deposition		30			60	

If the long-term accumulation of N deposition results in a change in the C/N ratio by about 10 of dissolved organic matter in runoff, the flux of N out of the system would increase by 7 mmol m<sup>-2</sup> yr<sup>-1</sup> at Risdalsheia and 5 mmol m<sup>-2</sup> yr<sup>-1</sup> at Gårdsjön. These increases are small relative to present-day N

deposition of about  $80 \text{ mmol m}^{-2} \text{ yr}^{-1}$  at both sites. Thus there would be little change in estimate of critical load for nitrogen at these sites if organic N were taken into consideration.

Runoff at both Risdalsheia and Gårdsjön contains relatively high concentrations of TOC (and organic N). Thus the potential change in critical load due to consideration of organic N would be even smaller for the majority of Norwegian freshwaters.

After entering surface waters N-compounds can undergo transformations along river stretches and in lakes. In the two deep oligotrophic lakes in the Bjerkreim river basin a transformation of incoming organic N to  $\text{NO}_3^-$  seems to occur (Berge *et al.*, in prep.). This may be ascribed to loss by sedimentation, light degradation and mineralisation by bacteria. In the shallow lake, however, relatively more  $\text{NO}_3^-$  was retained, while there seemed to be a net export of organic N. In this case biological uptake of inorganic N by macrophytes, planktonic algae, and bacteria may be responsible. The elevated organic N concentrations in the lake outlet can be due to very short residence time with extensive drift of pelagic organisms and plant fragments. Expressed per surface unit this lake also had the highest N retention of the 3 lakes. This was probably due to higher biological N uptake and immobilisation relative to the 2 highly oligotrophic lakes.

The lake survey data do not indicate that the C/N ratio in lakewater is related to the N deposition. Lake data, however, do not give a direct picture of catchment runoff, as in-lake processes act on the dissolved organic matter and thus change the C/N ratio, as well as the TOC concentration. Nevertheless, with respect to critical loads for surface waters, the lake data would thus indicate that organic-N can be neglected in determining critical loads.

Dissolved organic matter (DOM) is produced during decomposition of organic matter in forest soil. Much of the DOM leached from the forest floor originates from initially soluble substances in litterfall and throughfall, but the structure of these components are changed during humification in the forest floor (Qualls *et al.* 1991). Production, decomposition and transport of DOM is influenced by factors such as type of vegetation, hydrology, temperature and soil moisture content. In addition management and atmospheric deposition of acidity and of nitrogen compounds may affect concentrations and dynamics of DOM. Increased N-deposition may lead to increased decomposition of organic matter in soil due to increased microbiological activity, which in turn may lead to increased DOC and DON concentrations in soil solution.

Studies of DON and possible effects of N addition in forest soil solution is complicated by large spatial variations both within and between catchments, in addition to differences as soil type, vegetation and hydrology which further complicates the interpretation of data. The data do not suggest that increased N deposition to forest ecosystems leads to changes in production and transport of DON in soil solution. At the NITREX experiment at Gårdsjön increased N deposition causes significant changes and increased leaching of  $\text{NO}_3^-$ , but no effect on organic-N (Stuanes and Kjønås, in press.). Other factors such as temperature may be more important to the decomposition of Soil organic matter and thereby concentrations of DOC and DON than N addition and acidification.

We conclude that while there is evidence from small catchment studies that organic N levels may have increased as a result of N deposition, the resultant effect on estimate of critical load for nitrogen for freshwaters is minor. For practical purposes organic N outputs can be neglected in estimating and mapping critical loads for nitrogen in Norway.

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