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**Assessment of Trends  
and Leaching of Nitrogen  
at ICP Waters Sites  
(Europe and North America)**

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**Abstract**

The ICP Waters database currently holds data for 204 sites in Europe and North America. These data were evaluated with respect to nitrate concentrations. About 50% of the sites currently have nitrate concentrations indicative of nitrogen saturation, that is, elevated level of nitrate above that expected in undisturbed systems not receiving significant amounts of N deposition. The relative importance of nitrate as an acid anion has increased at the ICP sites since the early 1990s, mostly due to the decrease in sulphate. The data indicate no major change in N saturation at the ICP sites during the 1990s, indicating that progression to increased N saturation is a slow process with a time scale of decades. There is no consistent pattern of trends in nitrate in the ICP waters sites. The overall lack of significant trends may be the result of 2 opposing factors (Wright et al. 2001); continued high deposition of nitrogen should tend to increase N saturation and give increased nitrate in runoff, whereas the decline in N deposition over the past 5-10 years in large parts of Europe should give decreased nitrate in runoff. Short and long-term variations in climate affect nitrate in streamwater, and thus contribute "noise" which masks long-term trends.

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CONVENTION ON LONG-RANGE  
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON  
ASSESSMENT AND MONITORING OF ACIDIFICATION  
OF RIVERS AND LAKES

**Assessment of Trends and Leaching of Nitrogen at  
ICP Waters Sites (Europe and North America)**

Prepared by the Programme Centre  
Norwegian Institute for Water Research  
Oslo, August 2001

## Preface

The International Cooperative Programme on Assessment and Monitoring of Rivers and Lakes (ICP Waters) was established under the Executive Body of the Convention on Long-Range Transboundary Air Pollution at its third session in Helsinki in July 1985. The Executive Body also accepted Norway's offer to provide facilities for the Programme Centre, which has been established at the Norwegian Institute for Water Research, NIVA. Berit Kvæven, Norwegian State Pollution Control Authority, has led the ICP Waters programme.

This report is a continuation of previous work within the ICP Waters programme about assessment of nitrogen in surface water. The report has been made in cooperation with European Monitoring and Assessment Programme (EMEP) Meteorological Synthesising Centre - West (MSC-W) through Leonor Tarrason and Chemical Coordination centre for Chemistry (CCC) through Kjetil Tørseth. The idea is to compare status and trends in nitrogen concentrations in surface waters with those in deposition.

Many individuals and institutions have assisted in the preparation of this report. We thank all national Focal Centres for submitting the necessary data to the ICP Waters Programme Centre, and for giving valuable comments to the report. At NIVA we thank Liv Bente Skancke and Torulv Tjomsland for technical assistance, and Ann Kristin Buan for database management.

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Oslo, August 2001

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## Summary and conclusions

The ICP Waters database currently holds data for 204 sites in Europe and North America. These data were evaluated with respect to status and trends in nitrate ( $\text{NO}_3^-$ ) concentrations.

### Status

- About 1/3 of the ICP sites have mean annual concentrations of  $\text{NO}_3^- < 5 \mu\text{eq L}^{-1}$ . More than half of the sites have  $\text{NO}_3^-$  concentrations  $> 10 \mu\text{eq L}^{-1}$ . In total, the results suggest that a large fraction of the ICP Waters sites have  $\text{NO}_3^-$  concentrations above those expected from catchments with no impact from nitrogen (N) deposition.
- In about 5% of the sites,  $\text{NO}_3^-$  makes a greater contribution to acidification than sulphate ( $\text{SO}_4^{2-}$ ). This means that for the majority of ICP Waters sites, on annual mean basis,  $\text{SO}_4^{2-}$  is still the dominant acidifying ion.
- $\text{SO}_4^{2-}$  is the most important acidifying anion, but  $\text{NO}_3^-$  constitutes more than 10% of the non-marine acid anions at 50% of the ICP Waters sites.
- The ICP Waters data support the general picture that increased N leakage occurs in catchments receiving N deposition above a threshold of 5-10  $\text{kg N ha}^{-1} \text{yr}^{-1}$ .
- About half of the 108 European ICP Waters sites exhibit a high degree of N saturation (Stage 2 or 3) in 1998. Such sites are found predominantly in Central Europe. Most of the sites in the Scandinavian countries are classified in Stage 0 or 1. In Eastern US 26% of the sites show high degree of N saturation (Stage 2) and no sites are classified in Stage 3.
- There is a clear connection between N deposition from air and N Stage at European ICP Waters sites. N-deposition levels are all below 10  $\text{kg N ha}^{-1} \text{year}^{-1}$  at Stages 0 and 1, while at Stages 2 and 3 no sites had air deposition below 10  $\text{kg N ha}^{-1} \text{year}^{-1}$ .

### Trend

- Deposition of both oxidised and reduced airborne N in Europe increased during the 1900's and levelled out in the 1970's and 1980's. Trend analyses on the EMEP data conducted by Barrett et al. 2000a and Barrett et al. 2000b indicate a decline in deposition of N of approximately 20% in Central Germany and Southern Scandinavia during the period 1989-1998. This is interpreted as a response to declining emissions of N compounds during the 1990s.
- Of 16 EMEP sites situated in the same region as the ICP Waters sites only three sites show significant changes in N deposition in the 10-year period 1989-1998. Measured wet deposition of N compounds at EMEP sites shows relatively large year-to-year variations, probably mainly due to natural variations in meteorological conditions such as amount of precipitation. This "noise" in the record thus means that trends in N deposition, therefore, must be relatively large before they become statistically significant. This probably explains why there are no significant trends in N deposition during the 1990s at many of the EMEP sites.
- The majority of the 98 ICP Waters sites with sufficient data for trend analysis showed no significant trends in  $\text{NO}_3^-$  for the time period 1989-1998. Only 8 sites showed significant increase and 11 showed significant decrease. Previous results from the ICP Waters programme (Stoddard et al. 1999, Lükewille et al. 1997) showed that in the 1980s for the ICP Waters program as a whole, more sites exhibited upward trends of  $\text{NO}_3^-$  concentrations (33%) than downward (8%). The data for the 1990s now indicate that this increasing trend has stopped.
- The frequency distribution of  $\text{NO}_3^-$  concentrations is approximately the same for 150 sites with data from 1990/92 and 1999. A division of data between US and Europe reveals that most of the increase in  $\text{NO}_3^-$  from 1990/92 to 1999 has occurred in the US sites, while in the European sites there is a tendency towards a decrease in  $\text{NO}_3^-$ .
- $\text{NO}_3^-$  plays a larger role in acidification in 1999 relative to 1990/92. This is mostly due to the decline in  $\text{SO}_4^{2-}$  concentration over this period, rather than an increase in  $\text{NO}_3^-$  concentrations.

- Of 36 ICP Waters sites in Europe classified relative to N-saturation stages in 1990/92 and 1998 in 1990/92, 24 had no change in N saturation stage, while 8 went down and 4 went up.
  - The results indicate that progression to increased N-saturation stages is a slow process with a time scale of decades.
  - The results show that there is no consistent pattern of trends in  $\text{NO}_3^-$  in the ICP waters sites. The overall lack of significant trends in  $\text{NO}_3^-$  concentrations in surface waters may be the result of 2 opposing factors (Wright et al. 2001); Continued high deposition of nitrogen (above the  $10 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  threshold) should tend to increase N saturation and give increased nitrate concentrations in runoff, whereas the decline in N deposition over the past 5-10 years in large parts of Europe should give decreased nitrate concentrations in runoff. Short and long-term variations in climate affect nitrate concentrations in streamwater, and thus contribute “noise” which masks long-term trends.
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# 1. Background

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) was established under the Executive Body of the Convention on Long-Range Transboundary Air Pollution (LRTAP) at its third session in Helsinki in July 1985 (EB AIR/7, Annex/V). It is one of six ICPs (Waters, Forests, Materials, Crops, Integrated Monitoring, Mapping and Modelling). The LRTAP convention went into effect in 1983 and was the first step to enforce emission reduction measures in the international sphere.

ICP Waters is designed to assess the degree and geographical extent of acidification of surface waters, evaluate dose/response relationships and long-term trends in aquatic chemistry and biota.

During the 13 years of operation, the programme has produced yearly reports and an in-depth report every three years; 3-year report (Wathne 1991), 6-year report (Skjelkvåle et al. 1994), 9-year report (Lükewille et al. 1997), and the latest 12-year report (Skjelkvåle et al. 2000). The major findings of the ICP Waters programme are listed in Appendix B. Reports and publications from the programme are listed in Appendix C. An assessment of N leaching in the ICP Waters sites based on data through 1992 is reported in Traaen and Stoddard 1995) and Stoddard and Traaen 1995) and through 1995 in Lükewille *et al.* (1997). Here we use ICP Waters data through 1998 and 1999 to update this assessment.

## 2. Introduction

During the 1900's large regions of Europe received elevated deposition of N compounds. Emissions of oxidised N species from combustion of fossil fuels and emissions of reduced N compounds from agriculture increased dramatically in Europe during the 1900's to reach peak levels about 1980. (Which reference?) European emissions of nitrogen to the atmosphere have levelled off during the 1980s and a slight decrease of approx. 15 % has been reported between 1990 and 1998. The European averaged decrease in N deposition from air during the 1990s is somewhat smaller than the reported emission decrease although there are significant differences from country to country (Tarrason and Schaug 2000).

Excess N deposition has long been viewed as a threat to the nutrient balance and health of forest and semi-natural terrestrial ecosystems. N is usually the growth-limiting nutrient in these ecosystems. Chronic excess N deposition can lead to N saturation, defined by Aber et al. 1989) as "the availability of ammonium and  $\text{NO}_3^-$  in excess of total combined plant and microbial nutritional demand". By this definition N saturation is manifest by increased leaching of inorganic N (generally  $\text{NO}_3^-$ ) below the routing zone. Inasmuch as  $\text{NO}_3^-$  is a strong acid anion, increased leaching of  $\text{NO}_3^-$  enhances acidification of soils and surface waters. Increased concentrations of inorganic N in runoff (streamwater) thus indicate N saturation of catchment ecosystems, under the conditions, of course, that there are no significant sources of N in the catchment (such as fertilisers, municipal and industrial wastewater).

The importance of  $\text{NO}_3^-$  in the acidification of surface waters has received much-deserved attention, especially as the importance of  $\text{SO}_4^{2-}$  declines. Results from the ICP Waters programme have shown that significant decreases in  $\text{SO}_4^{2-}$  concentrations at European and North American ICP Waters sites can be explained by a decline in sulphur (S) deposition due to successful emission reduction measures (Stoddard et al. 1999, Skjelkvåle et al. 2000). At many catchments, increasing  $\text{NO}_3^-$  concentrations



during the 1980s and decreasing values during the 1990s can be observed (Lükewille et al. 1997). The pattern cannot be explained by patterns in air deposition or other changes in the catchments, such as point sources, agriculture, clearcutting and forest fire. Hence, dose/response relationships are much more complicated for N than for S. Changes in N deposition may not always be directly correlated with changes in NO<sub>3</sub><sup>-</sup> leaching. The “nitrogen status” of an ecosystem, or changes in N-status, seems to be an important indicator of N-saturation.

Reports of increased NO<sub>3</sub><sup>-</sup> concentrations in runoff first appeared in the 1980s. The Norwegian 1000 lake survey in 1986 revealed that NO<sub>3</sub><sup>-</sup> concentrations in some areas in Southern Norway had almost doubled since investigations in 1974-75 (Henriksen and Brakke 1988). Increasing trends in NO<sub>3</sub><sup>-</sup> concentrations were also revealed in the U.S. EPA's Long-term Monitoring Project. Nine of 15 drainage lakes monitored in the Adirondack area showed increases in NO<sub>3</sub><sup>-</sup> concentrations, ranging from 0.5 to 2.0 µeq L<sup>-1</sup> yr<sup>-1</sup> in the period 1982-1990 (Driscoll and van Dreason 1993). Further, 5 of 8 streams in the Catskill area exhibited significant upward trends in NO<sub>3</sub><sup>-</sup>, ranging from 1.3 to almost 3 µeq L<sup>-1</sup> yr<sup>-1</sup> in the period 1983 - 1989 (Murdoch and Stoddard 1993). These trends have largely disappeared in the 1990s (Stoddard et al. 1998), a phenomenon that can also be observed at many ICP Waters sites. Even if NO<sub>3</sub><sup>-</sup> concentrations are relatively unchanging, however, leaching of NO<sub>3</sub><sup>-</sup> from forested catchments will have the effect of removing base cations from soil and mobilising aluminium and H<sup>+</sup> ions, contributing to catchment acidification.

The aim of this report is to:

- give an updated report (since the evaluation of Lükewille et al. 1997) on status of NO<sub>3</sub><sup>-</sup> in surface waters at ICP Waters sites in Europe and North America and
- give a more detailed assessment on trends in N in surface waters in subregions as a follow up from the 12-year report (Skjelkvåle et al. 2000), and relate these to trends in N deposition from air.

### 3. Methods and Site Selection

The surface water data used in this report are from the ICP Waters Programme, and the deposition data are from EMEP (European Monitoring and Assessment Programme).

The ICP Waters data comprise 204 sites in Europe (Austria, Belarus, Czech Republic, Estonia, Finland, Germany, Hungary, Italy, Ireland, Latvia, Norway, Poland, Russia, Spain, Sweden, Switzerland and UK) and North America (Canada, United States) (**Figure 1**). All sites were selected according to the Programme Manual (Norwegian Institute for Water Research 1996) to increase the likelihood of their being representative of acid-sensitive terrain in each country. In accordance with these criteria, the sites are lakes and streams draining headwater catchments and are free from local disturbance. Sampling frequencies vary, but all sites included here have as a minimum both summer/fall baseflow and spring runoff samples.

Nitrate concentrations reported in ICP Waters are measured using a variety of analytical methods. Annual cross-comparisons of methods using audit samples of known concentration, conducted by ICP Waters (Hovind, 2000) show excellent agreement among all of the national labs participating. Data used for assessment of N-status are from 1998 and 1999; trends data are from 1989-1998.

The data in the ICP Waters database illustrates a problem with large international databases, based on voluntary contributions. Even if the programme has a manual and the data are checked carefully before they are stored in the database at the ICP programme centre, there are many sites that does not

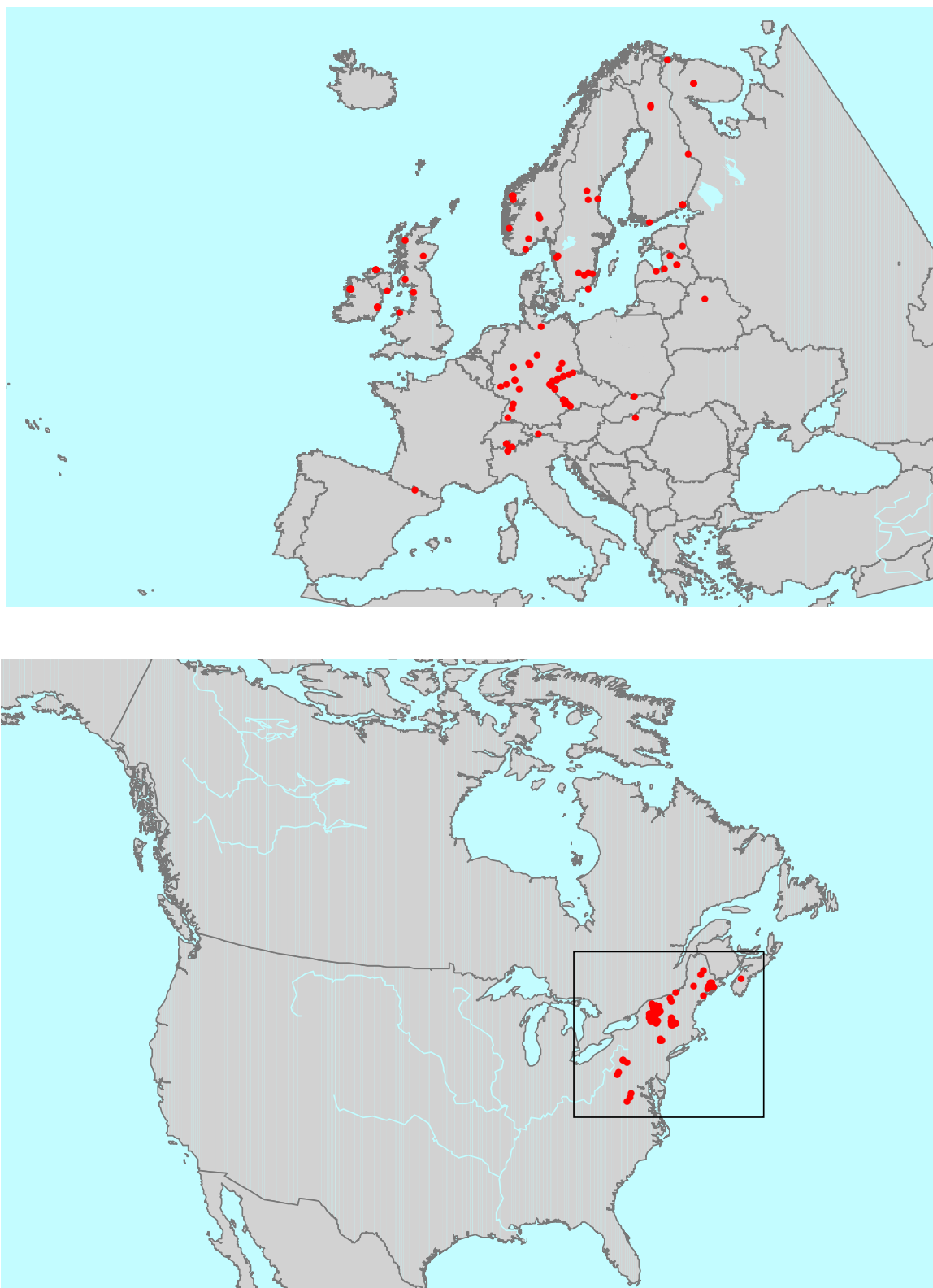
have sufficient data for all types of analysis. In the figures in the text it is always presented how many sites that are used in the particular analysis. For overview, this information is also given in **Table 1**. Name, location and most of the results presented in this report is listed in Appendix A.

Criteria for selection of sites used for assessment of N-saturation stages were the same as used by Stoddard and Traaen 1995), which is minimum 3 samples for the year 1998 and these are spread among seasons. Criteria for selection of sites used for assessment of trends were the same as used in the 12-year report (Skjelkvåle et al. 2000), which is minimum 2 samples per year, and that there are data for at least 7 of the 10 years 1989-98.

All deposition data used here are provided from European Monitoring and Assessment Programme (EMEP) both from Meteorological Synthesising Centre - West (MSC-W) and Coordination centre for Chemistry (CCC). Deposition from EMEP is only for European sites. Results reported here are measured wet deposition at EMEP stations located in the same region as the streamwater sites (Barrett et al. 2000a), and modelled total (wet + dry) deposition by country (Tarrason and Schaug 2000). The data are modelled deposition in 50x50 km grid squares in Europe. The model uses estimated annual emissions and actual meteorology for each year to calculate wet and dry deposition of S, oxidised N and reduced N compounds.

**Table 1.** Number of ICP Waters sites used in the different type of assessments

Total number of ICP Waters sites with data in 1999	204
Number of sites with NO <sub>3</sub> <sup>-</sup> data both from 1990/92 and 1999	150
Number of sites with nitrogen data both for deposition and runoff in 1999 (only European sites)	99
Number of sites with sufficient NO <sub>3</sub> <sup>-</sup> data to estimate N-saturation stages	198
Number of sites with sufficient NO <sub>3</sub> <sup>-</sup> data to estimate N-saturation stages both for 1990/92 and 1998	36
Number of sites with deposition and sufficient data for estimation of N-saturation stages	82
Number of sites with sufficient data for trend analysis	96



**Figure 1.** Location of 204 ICP Waters sites as of October 2000 and used in the evaluation of  $\text{NO}_3^-$  concentrations. The box for North America indicates the area shown in the following figures.

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## 4. Results

### 4.1 Trends in Nitrate Concentrations in Surface Waters

Much of the current concern about N saturation has resulted from evidence of increasing  $\text{NO}_3^-$  losses from catchments in high deposition areas of Europe and North America in the 1980s (e.g., Murdoch and Stoddard 1993, Driscoll and van Dreason 1993, Henriksen and Brakke 1988). More recent examinations of trends in the Northeastern U.S. have suggested that these trends have not continued into the 1990s (Mitchell et al. 1996).

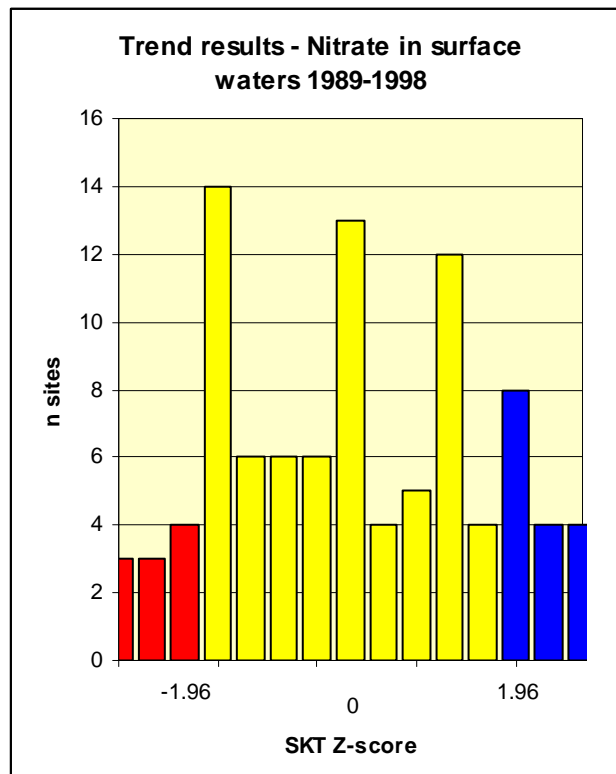
The ICP Waters data have provided the basis for evaluation of  $\text{NO}_3^-$  trends in surface waters in the 1990s over larger regions (Skjelkvåle et al. 2000). Sites with data of sufficient frequency (minimum 3 samples per year) and duration (at least 7 years in the 10-year period 1989-1998) were used (96 sites). Significance of trends over time were tested by the Seasonal Kendall Tau (SKT) statistic (details of statistical methods are given in Skjelkvåle et al. (2000) and Stoddard et al. (1999).

Of the 96 ICP-Waters sites with sufficient data for trend analysis 8 sites showed significant upward trends in the period from 1989-1998, 10 showed significant downward trends, while the remaining 78 sites showed no significant trends (**Table 2**). Four of the sites with significant upwards trends are located in Canada.

There is no consistent pattern of trends in  $\text{NO}_3^-$ . The overall lack of significant trends in nitrate concentrations in streams in Europe may be the result of 2 opposing factors (Wright et al. 2001). Continued high deposition of nitrogen (above the  $10 \text{ kgN ha}^{-1} \text{ yr}^{-1}$  threshold) should tend to increase N saturation and give increased nitrate concentrations in runoff, whereas the decline in N deposition over the past 5-10 years in large parts of Europe should give decreased nitrate concentrations in runoff. Short and long-term variations in climate affect nitrate concentrations in streamwater, and thus contribute "noise" which masks long-term trends. More detailed discussions about trends in  $\text{NO}_3^-$  for single countries and regions are presented in Vesely et al. (2001 in prep), Kopáček et al. (1998) (Czech Republic), Jeffries et al. 2001 in press (Canada), Monteith et al. 2000 (UK), Mitchell et al. 1996(US), Europe (Wright et al. 2001).

The sites were grouped by chemical criteria and catchment characteristics (Skjelkvåle et al. 2000). The only group that exhibits significant change is the low ANC group, where the data indicate significant increase in  $\text{NO}_3^-$  concentrations during the 1990s. Neither the high  $\text{NO}_3^-$  or low  $\text{NO}_3^-$  groups of sites exhibited significant trends in  $\text{NO}_3^-$  concentrations (both groups showed considerable trend variability).

Previous results from the ICP Waters programme (Stoddard et al. 1999, Lükewille et al. 1997) showed that in the 1980s for the ICP Waters program as a whole, more sites exhibited upward trends of  $\text{NO}_3^-$  concentrations (33%) than downward (8%). The data for the 1990s now indicate that this increasing trend has stopped.



**Figure 2.** Trend results showing SKT Z-scores for  $\text{NO}_3^-$ , for all the analysed sites (n=98). Negative scores indicate decreasing trends, while positive scores indicate increasing trends. Scores  $> 1.96$  or  $< -1.96$  are significantly different from 0 at  $p < 0.05$ . The positive slopes are blue and the negative are red.

**Table 2.** Trends in NO<sub>3</sub> concentration at ICP Waters sites in each country 1989-1998. Z-score > 1.96 or < -1.96 are significant at p<0.05. Statistically significant trends are indicated in bold. Unit of slope is µeq L<sup>-1</sup>. The results are taken from Skjelkvåle et al. 2000).

Site	Z-score	Slope	Site	Z-score	Slope	Site	Z-score	Slope	Site	Z-score	Slope
CA01	0.84	0.1	DE06	<b>-1.98</b>	<b>-6.5</b>	IT01	-1.19	-0.6	UK01	1.10	0.0
CA02	1.13	0.3	DE07	<b>-2.56</b>	<b>-7.1</b>	IT03	0.08	0.0	UK04	<b>2.02</b>	1.3
CA03	1.95	0.6	DE08	-0.73	-0.2	IT05	1.89	1.6	UK07	<b>2.62</b>	0.8
CA04	<b>2.38</b>	<b>0.4</b>	DE10	0.43	0.6	IT06	1.72	1.5	UK10	0.33	0.3
CA05	0.77	0.2	DE11	-1.80	-1.0	NO01	<b>-2.29</b>	-0.3	UK15	0.76	0.3
CA06	0.89	0.0	DE12	<b>-2.63</b>	<b>-1.6</b>	NO03	1.19	0.0	UK21	1.07	1.0
CA07	1.64	0.2	DE15	-1.50	-1.3	NO04	1.86	0.0	US07	0.00	0.0
CA08	-1.81	-0.2	DE17	-0.76	-0.8	NO05	<b>1.98</b>	0.1	US08	0.19	0.0
CA09	<b>2.49</b>	<b>0.3</b>	DE18	-0.36	0.0	NO06	-0.35	-0.1	US09	0.00	0.0
CA11	1.27	0.0	DE20	0.95	0.7	NO07	0.84	0.1	US10	-0.16	0.0
CA13	<b>2.48</b>	<b>0.1</b>	DE21	-1.93	-7.1	NO08	-0.26	0.0	US11	-1.79	-0.4
CA14	<b>2.33</b>	<b>0.1</b>	DE22	-1.56	-3.8	NO09	<b>2.00</b>	0.2	US12	-0.35	-0.3
CA16	0.00	0.0	DE23	<b>-2.87</b>	<b>-1.8</b>	NO10	-0.44	-0.1	US13	-1.37	-1.0
CA17	1.18	0.0	DE26	-1.69	-1.7	PL01	<b>-2.65</b>	-2.8	US14	-1.07	-0.4
CZ01	-1.64	-1.3	DE27	-1.63	-1.3	PL02	-1.90	-1.5	US15	-1.80	-1.0
CZ02	-1.67	-1.4	DE31	-1.46	-11.4	SE01	-0.97	-0.1	US16	-1.66	-0.9
CZ03	-0.74	-0.9	DE32	-0.51	-0.3	SE02	0.75	0.1	US17	-1.34	-0.6
CZ04	0.98	0.7	DE33	<b>-3.03</b>	<b>-5.4</b>	SE03	-0.97	-0.3	US19	-0.46	0.0
CZ05	0.20	0.3	FI01	1.28	0.2	SE05	-0.25	0.0	US20	0.40	0.0
CZ06	-0.08	0.0	FI02	0.92	0.1	SE06	-1.10	0.0	US21	-0.10	0.0
DE01	1.66	0.5	FI03	-0.31	0.0	SE08	0.91	0.3	US22	-1.94	-5.6
DE02	1.71	0.6	FI05	-0.85	0.0	SE09	<b>-2.25</b>	-0.8	US23	<b>-2.10</b>	-2.5
DE03	1.67	1.2	FI06	-1.15	0.0	SE10	1.14	0.1	US24	-1.82	-1.9
DE05	-0.35	0.0	FI08	0.91	0.1	SE11	-1.68	-0.1			
						SE12	<b>-2.90</b>	-0.6			

**Table 3.** Number of sites with significant increasing or decreasing (p< 0.05) NO<sub>3</sub><sup>-</sup> trends from 1989-1998 in the different countries.

Country	Total Number of Sites	Number of Sites with:	
		Upward Trends	Downward Trends
Canada	14	4	
Czech republic	6		
Finland	6		
Germany	22		5
Italy	4		
Norway	9	2	1
Poland	2		1
Sweden	10		2
U.K.	6	2	
U.S.	17		1
<b>Total</b>	<b>96</b>	<b>8</b>	<b>10</b>

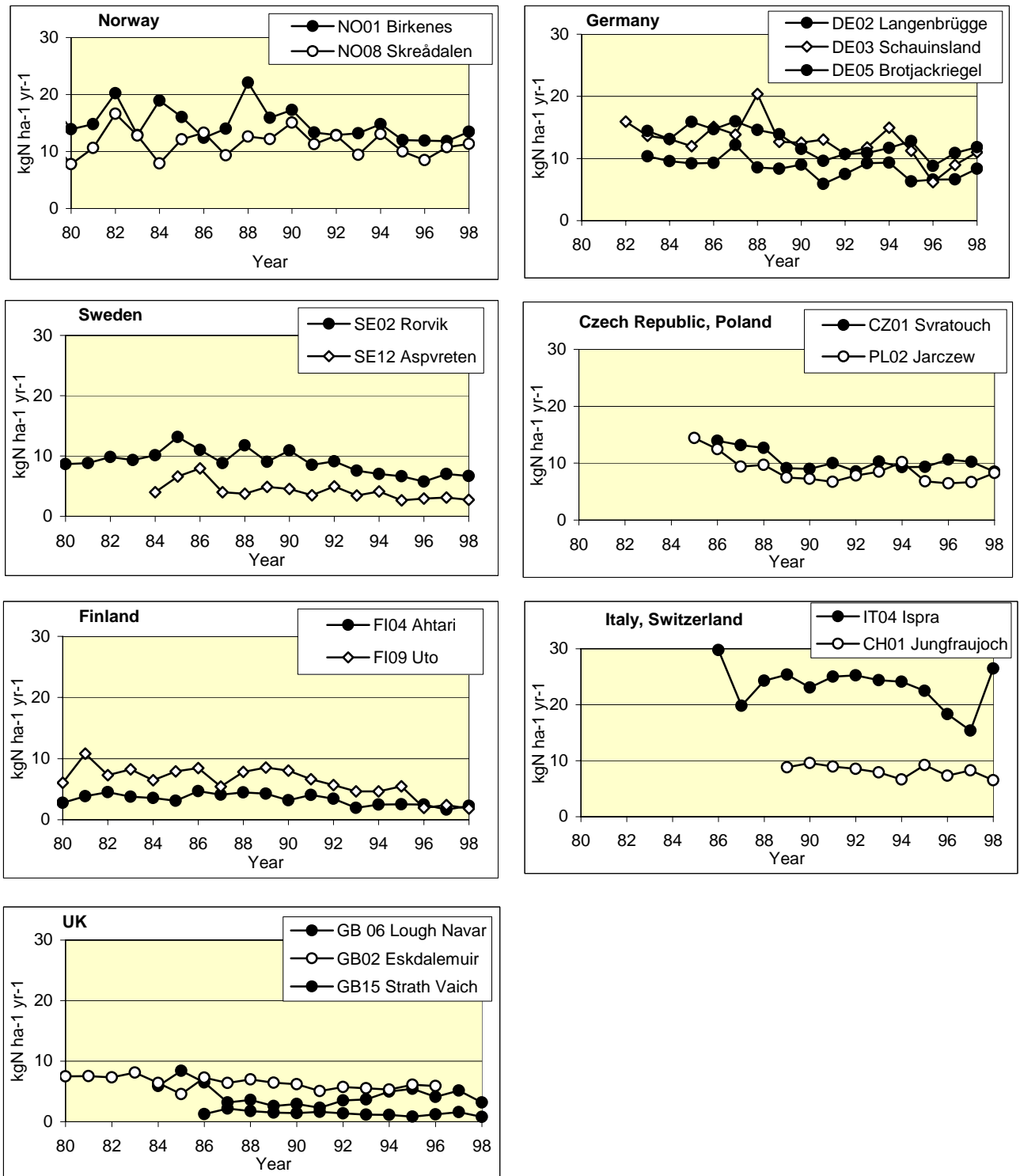
## 4.2 Trends in deposition

Emissions and following deposition of oxidized and reduced N ( $\text{NO}_3+\text{NH}_4$ ) have increased substantially the last 50-100 years, and reached their maximum in the late 1980ies and early 1990ies. Levels have decreased somewhat during the late 1990s in Central and Northern Europe, in response to modest declines in emissions (Tarrason and Schaug 2000). Trend analyses on the EMEP data conducted by Barrett et al. 2000a) and Barrett et al. 2000b) indicate a decline in deposition of N of approximately 20% in Central Germany and Southern Scandinavia during the period 1989-1998. This is interpreted as a response to declining emissions of N compounds during the 1990s.

For a selection of EMEP sites situated in the same region as the ICP Waters sites (**Table 4** and **Figure 3**) we have calculated trends in  $\text{NO}_3+\text{NH}_4$  deposition for the time period 1989-1998. Only the Finnish and the Swedish sites show significant changes in N deposition in this period, and in Sweden one site shows an increasing and the other a decreasing trend. Measured wet deposition of N compounds at EMEP sites shows relatively large year-to-year variations, probably mainly due to natural variations in meteorological conditions such as advections patterns and amount of precipitation. This “noise” in the record thus means that trends in N deposition, therefore, must be relatively large before they become statistically significant. This probably explains why there are no significant trends in N deposition during the 1990s at many of the EMEP sites.

**Table 4.** Linear regression of annual  $\text{NO}_3+\text{NH}_4$  deposition 1989-98 at 16 EMEP sites in 9 countries in Europe. Significant trends indicated in bold. NS: not significant. Units:  $\text{kg N ha}^{-1} \text{ yr}^{-1}$ .

Country	EMEP-site	slope	$r^2$	p
Czech Republic	CZ01 Svatouch	+0.05	0.04	NS
Germany	DE02 Langenbrügge	-0.10	0.05	NS
	DE03 Schauinland	-0.42	0.28	NS
	DE05 Brotjackriegel	-0.12	0.07	NS
Finland	FI04 Ahtari	<b>-0.24</b>	0.66	<0.05
	FI09 Uto	<b>-0.08</b>	0.90	<0.05
UK	GB 06 Lough Navar	-0.11	0.30	NS
	GB02 Eskdalemuir	+0.22	0.37	NS
	GB15 Strath Vaich	-0.05	0.29	NS
Italy	IT04 Ispra	-0.52	0.20	NS
Norway	NO01 Birkenes	-0.42	0.48	NS
	NO08 Skreådalen	-0.34	0.29	NS
Poland	PL02 Jarczew	0	0	NS
Sweden	SE02 Rorvik	<b>-0.43</b>	0.70	<0.05
	SE12 Aspvreten	<b>+0.23</b>	0.64	<0.05
Switzerland	CH01 Jungfrauoch	-0.23	0.42	NS



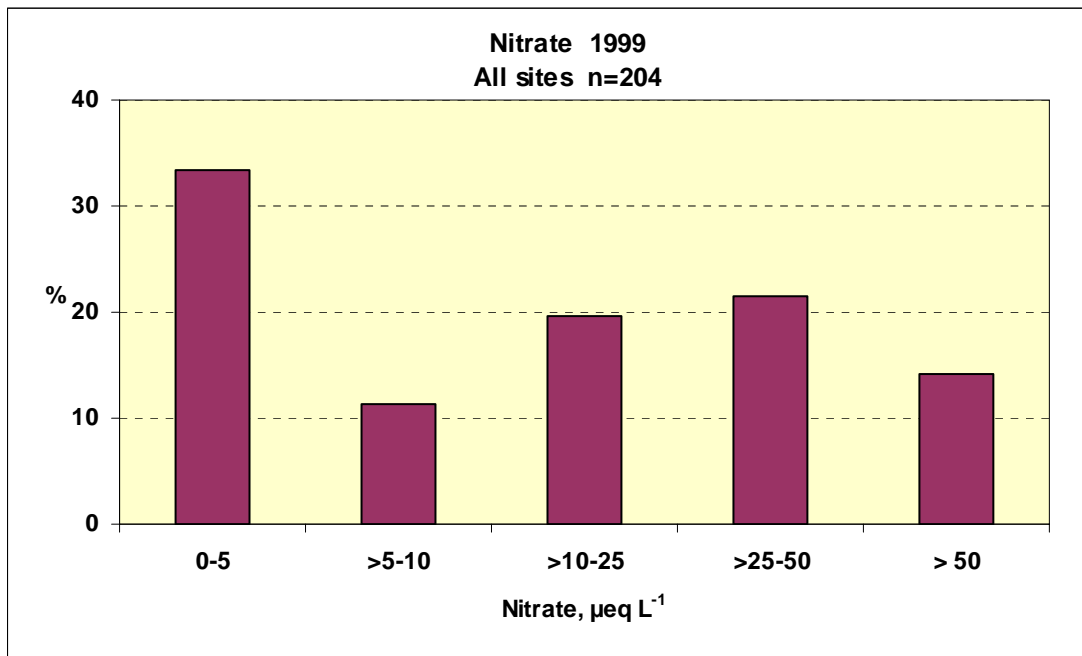
**Figure 3.** Measured wet deposition of nitrogen ( $\text{NO}_3 + \text{NH}_4$ ) at several EMEP stations in Europe (data from EMEP Chemical Coordinating Centre <http://www.nilu.no/projects/ccc>, Barrett et al. 2000a).



### 4.3 Nitrate in Runoff

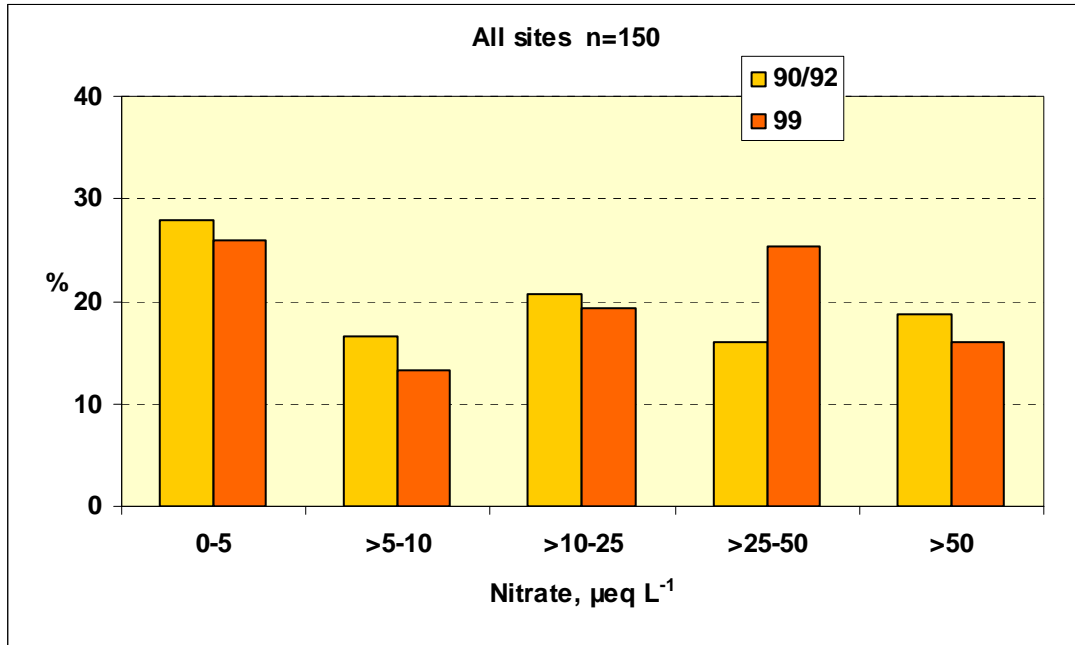
About 1/3 of the ICP sites (located primarily in the Nordic countries, Ireland, Canada and the U.S.) have mean annual concentrations of  $\text{NO}_3^-$  below  $5 \mu\text{eq L}^{-1}$  (**Figure 4**). More than half of the sites have  $\text{NO}_3^-$  concentrations above  $10 \mu\text{eq L}^{-1}$ . Due to the episodic pattern of  $\text{NO}_3^-$  at many sites, the significance of  $\text{NO}_3^-$  in acidification is often greater than shown by average figures. Mean concentrations greater than  $50 \mu\text{eq L}^{-1}$  are found at 14% of the sites, mainly in Germany, Latvia, Estonia, Italy, Czech Republic, Belarus and Hungary. In total, the results suggest that a large fraction of the ICP Waters sites have  $\text{NO}_3^-$  concentrations above those expected from catchments with no impact from N deposition (Stoddard 1994). Although all sites are among the least disturbed in each country, agricultural influences may affect some of the highest  $\text{NO}_3^-$  concentrations.

Unweighted mean values, as used here, may underestimate actual annual means, particularly in catchments with heavy snowmelt in spring. The results for the ICP Waters sites should therefore be considered conservative estimates of mean  $\text{NO}_3^-$  concentrations.



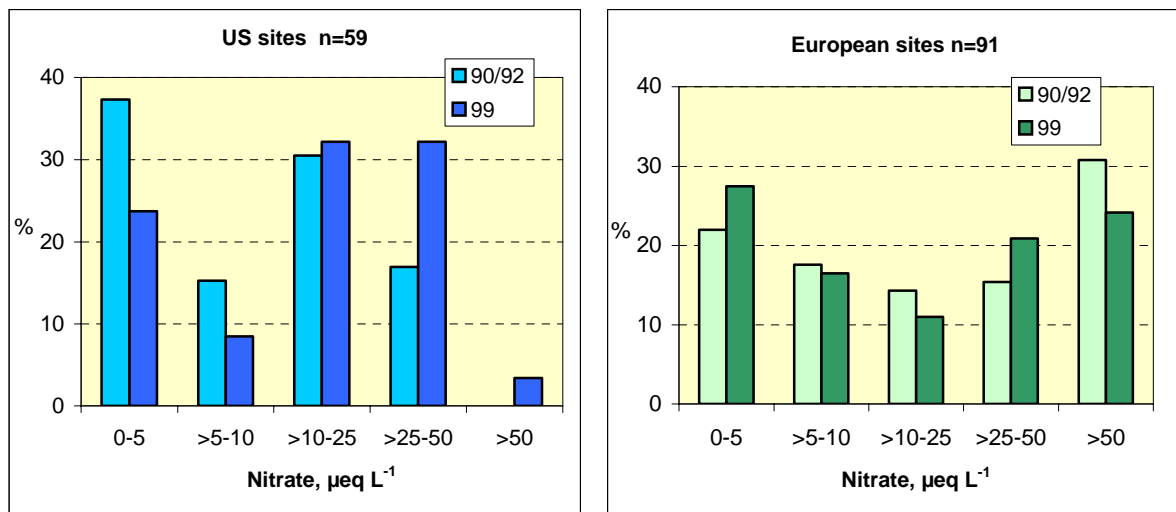
**Figure 4.** Frequency distribution of mean annual  $\text{NO}_3^-$  concentrations for the 204 ICP Waters sites with data from 1999 (a few sites 1998; see Appendix A).

The frequency distribution of  $\text{NO}_3^-$  concentrations is approximately the same for 150 sites with data from 1990/92 and 1999 (**Figure 5**). There has been some increase in sites with  $\text{NO}_3^-$  in the range from 25-50  $\mu\text{eq L}^{-1}$ , and decrease in all the other groups.

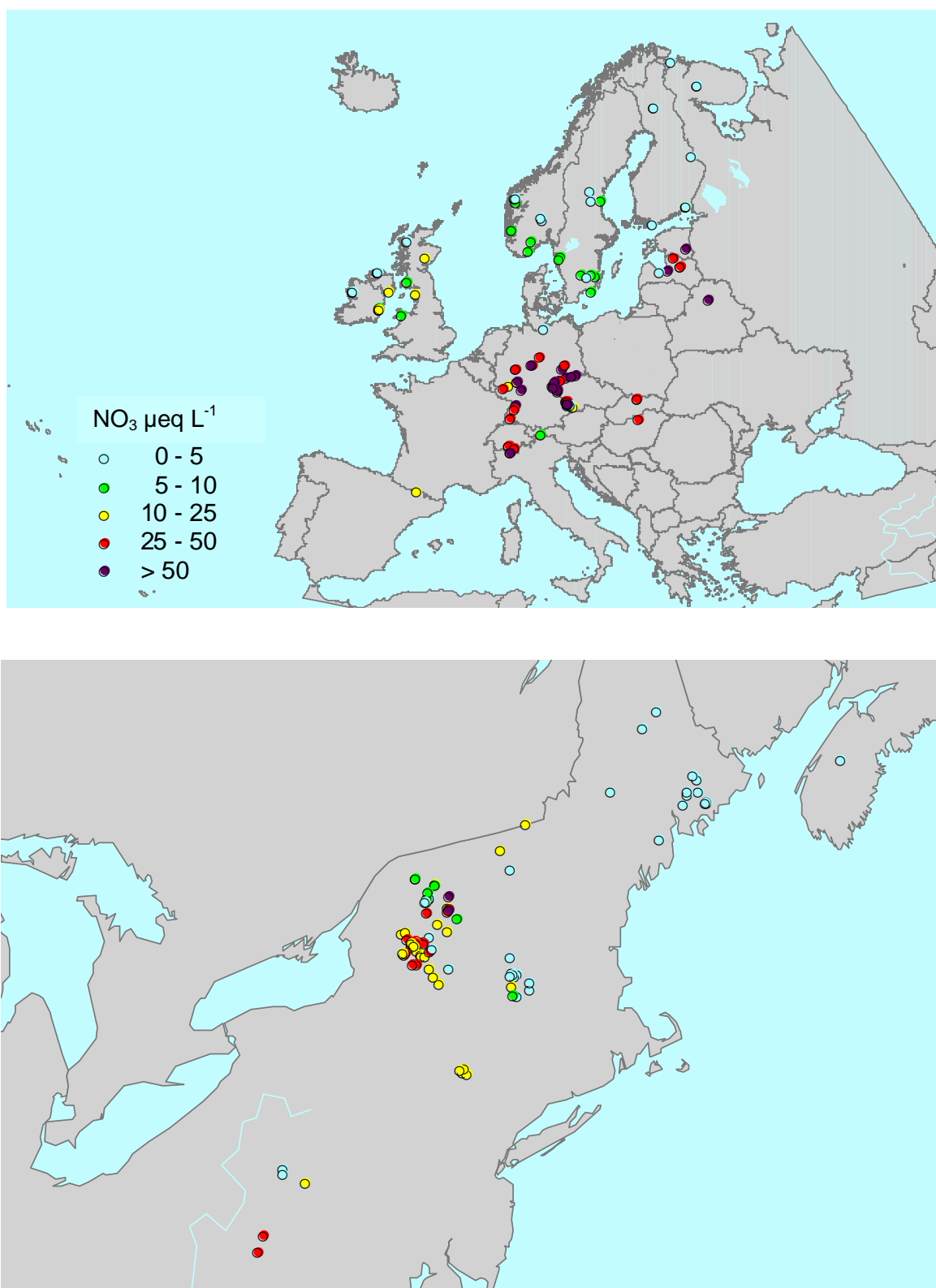


**Figure 5.** Frequency distribution of mean annual  $\text{NO}_3^-$  concentrations at 150 ICP Waters sites with data from 1999 and the period 1990-92.

A division of data between Europe and US reveals that most of the increase in  $\text{NO}_3^-$  from 1990/92 to 1999 has occurred in the US sites, while the European sites show tendency towards decrease in  $\text{NO}_3^-$  (**Figure 6**).



**Figure 6.** The same data shown in **Figure 5** but divided between North America and Europe.

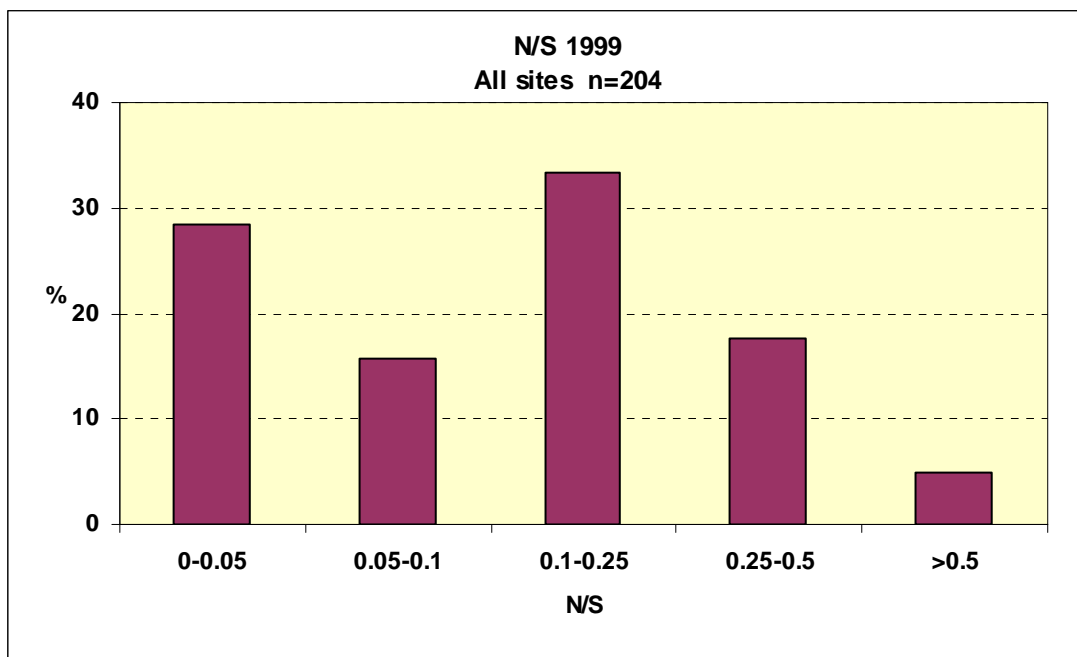


**Figure 7.** Map showing mean annual  $\text{NO}_3^-$  concentrations in ICP Waters sites in 1999 (unit  $\mu\text{eq L}^{-1}$ ) in Europe and Eastern US.

#### 4.4 Relative importance of Nitrate and Sulphate

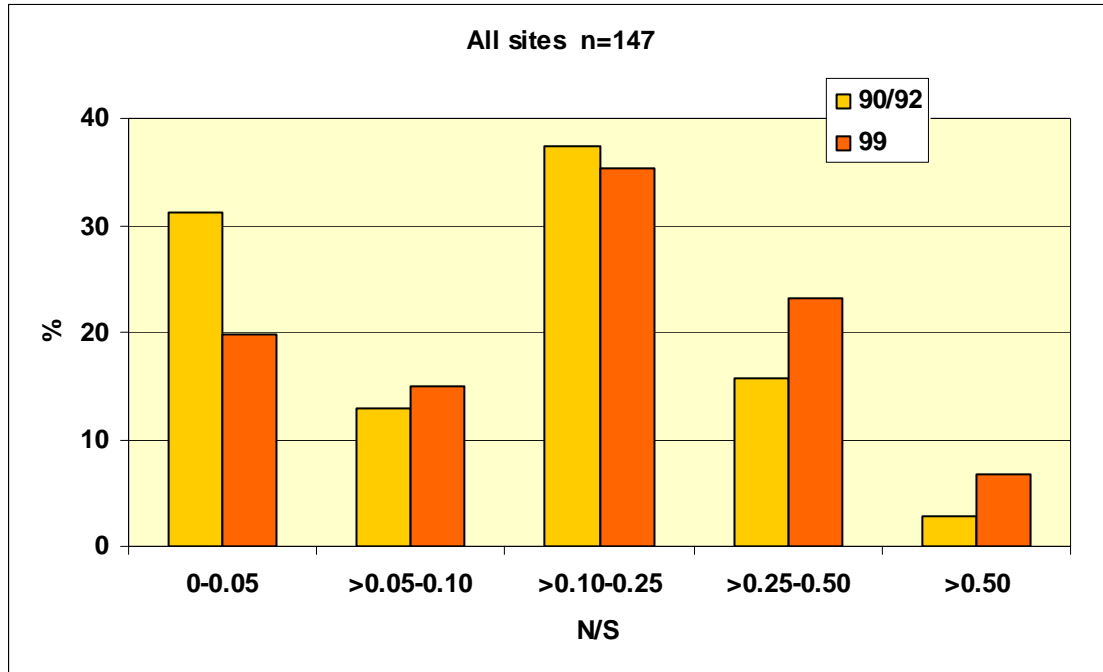
The importance of  $\text{NO}_3^-$  in acidification relative to  $\text{SO}_4^{2-}$  can be quantified by ratio N/S defined as the concentration of  $\text{NO}_3^-$  divided by the sum of non-marine  $\text{SO}_4^{2-}$  (denoted by asterisk) and  $\text{NO}_3^-$  ( $\mu\text{eq L}^{-1}$ ):  $\text{N/S} = \text{NO}_3^- / (\text{SO}_4^* + \text{NO}_3^-)$ .

N/S is  $> 0.1$  at about 55% of the 204 ICP-sites in 1999 (**Figure 8**), but only 5% have N/S values  $> 0.5$ . At these sites  $\text{NO}_3^-$  makes a greater contribution to acidification than  $\text{SO}_4^{2-}$ . These sites are situated in Germany and Italy (**Figure 11**). This means that for the majority of ICP Waters sites, on annual mean basis,  $\text{SO}_4^{2-}$  is still the dominant acidifying ion.



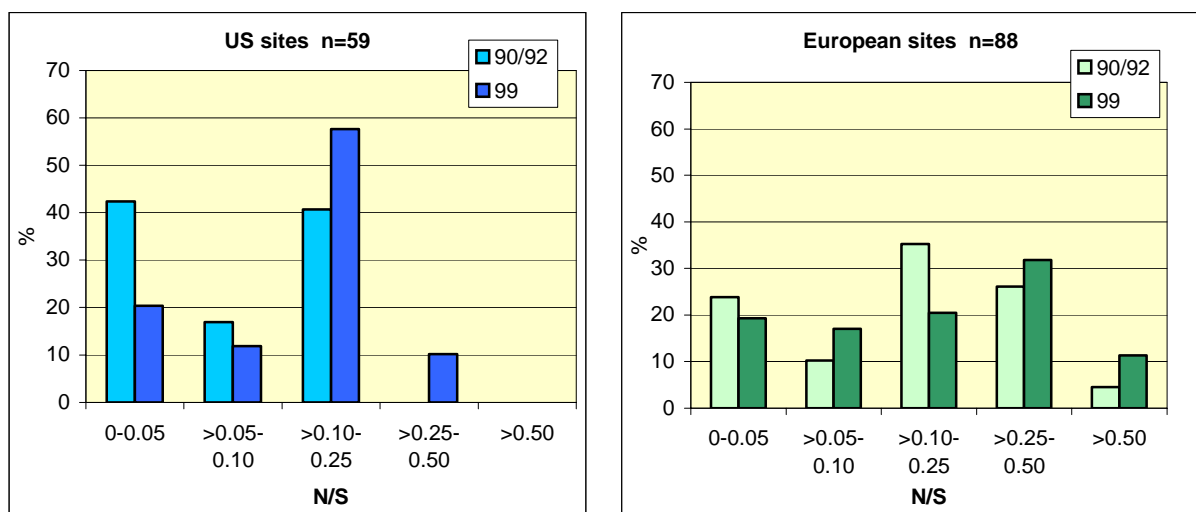
**Figure 8.** Relative importance of  $\text{NO}_3^-$  in acidification in 204 sites with data from 1999 (a few sites from 1998 (see Appendix A. ).

Comparison with the frequency distribution of N/S in the 1990/92 data from 147 ICP sites indicates that there has been a systematic shift to larger values (**Figure 9**).  $\text{NO}_3^-$  plays a larger role in acidification in 1999 relative to 1990/92. This is most certainly due to the decline in  $\text{SO}_4^{2-}$  concentration over this period, rather than an increase in  $\text{NO}_3^-$  concentrations.

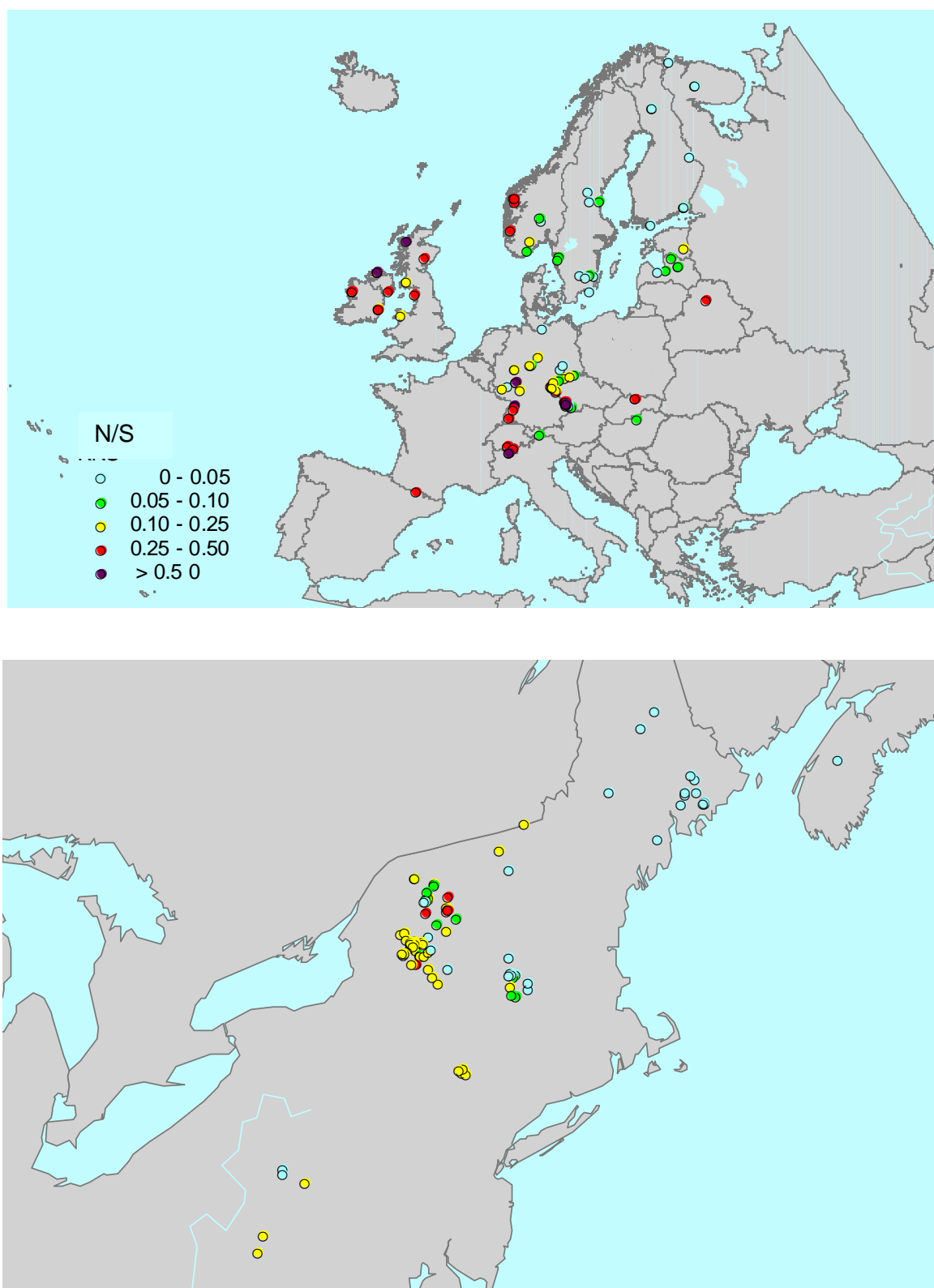


**Figure 9.** Relative importance of  $\text{NO}_3^-$  in acidification at 147 ICP Waters sites in 1999 compared to the period 1990/92 for the same sites.

A division of data between US and Europe (**Figure 10**) show that for the US sites, there has been a shift towards higher N/S from a median value of 0.07 in 1990/92 to 0.16 in 1999, while for the European sites there has been a shift from N/S at 0.16 to 0.20 from 1990/92 to 1999.



**Figure 10.** The same data as shown in **Figure 9** but divided between North America and Europe.



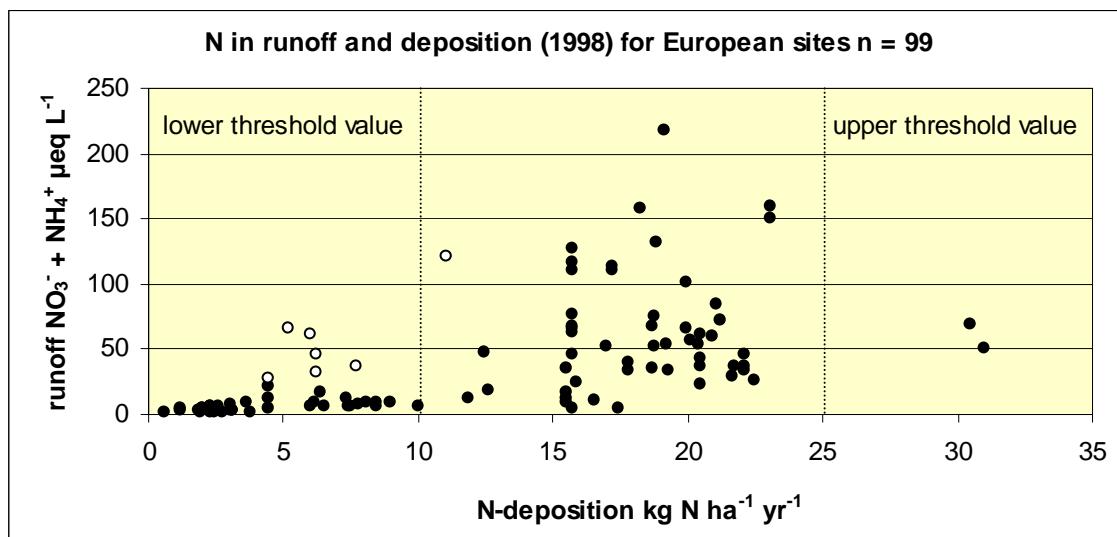
**Figure 11.** Map showing N/S (ICP Waters sites in 1999 Europe and Eastern US.  $N/S = \text{NO}_3^- / (\text{SO}_4^{2-} + \text{NO}_3^-)$  in  $\mu\text{eq L}^{-1}$ ).

## 4.5 Nitrogen Deposition and Inorganic Nitrogen in Runoff

Empirical data from forested ecosystems in Europe show a relationship between N deposition and N loss (Dise and Wright 1995, Gundersen et al. 1998). These data indicate very little  $\text{NO}_3^-$  leaching occurs at N deposition below  $9\text{--}10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , leaching can occur at intermediate deposition between  $9$  and  $25 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , and significant leaching occurs at most all sites receiving deposition greater than  $25 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . These thresholds are based on data from monitoring plots across a gradient of current N deposition in Europe and from several experimental sites.

Input/output data for  $\text{NO}_3^-$  are not available for most of the ICP Waters sites. However, a general view of N leakage is obtained by comparing the concentrations in runoff ( $\text{NO}_3^- + \text{NH}_4^+$ ) and estimated total (wet + dry) N deposition fluxes (**Figure 12**). Deposition fluxes are values for the  $50 \times 50 \text{ km}$  EMEP grids in which the respective ICP Waters sites are situated. The North American sites are not included in this analysis.

The results support the general picture that increased N leakage occurs in catchments receiving N deposition above some threshold. Some catchments have a pronounced  $\text{NO}_3^-$  leakage at a total N deposition as low as  $5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . In the case of the Latvian, Estonian and the Belarussian sites (marked with open circles in the figure) influence of agriculture (e.g., pasture land) on water quality cannot be excluded. For North American sites (Stoddard 1994) observed substantial N-leakage at wet deposition as low as  $2.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , corresponding to a wet + dry deposition of approximately  $5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . The possible difference in threshold values between the European and North American continents is an interesting phenomenon whose explanation is not yet available.



**Figure 12.** Nitrogen ( $\text{NO}_3^- + \text{NH}_4^+$ ) concentration in runoff and total N deposition fluxes (wet + dry) for 99 European ICP Waters sites in 1999. The deposition data are 1998 values for the EMEP  $50 \times 50$  grids in which the respective sites are situated (data from Tarrason and Schaug 2000). Threshold values of  $10$  and  $25 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  are from Dise and Wright 1995).

Absolute threshold values for ICP Waters sites are uncertain for several reasons: the deposition data are not site specific; the sampling frequency is low at many sites; the amount of N leaked is influenced

by both deposition and the duration of the deposition; N concentrations in runoff (instead of N fluxes) were compared with modelled N deposition fluxes; the site selection does not necessarily include the most sensitive sites with respect to N leakage; the threshold may be different for non-forested ecosystems; for lakes there may be a significant amount of retention of N in the lake itself. Despite these uncertainties, the data in **Figure 12** are in reasonably good agreement with the data from Dise and Wright 1995) and Gundersen et al. 1998).

## 4.6 Classification of Nitrogen saturation

Nitrogen is the growth-limiting nutrient in many terrestrial ecosystems. Long-term atmospheric N deposition may lead to a situation where the availability of inorganic N is in excess of the total amounts required for the growth of plants and soil micro-organisms. N saturation is defined here as persistent losses of  $\text{NO}_3^-$  and/or  $\text{NH}_4^+$  in streamflow or groundwater discharge, which may be accompanied by increases in N mineralisation and nitrification in soil (Stoddard 1994).

Stoddard 1994) has elaborated the term “persistent” by addressing short-term temporal variations in  $\text{NO}_3^-$  leaching from catchments thus considering the seasonality of biological N turnover processes. He names different stages in a hypothetical time course through which ecosystems can pass if atmospheric N deposition remains high or increases.

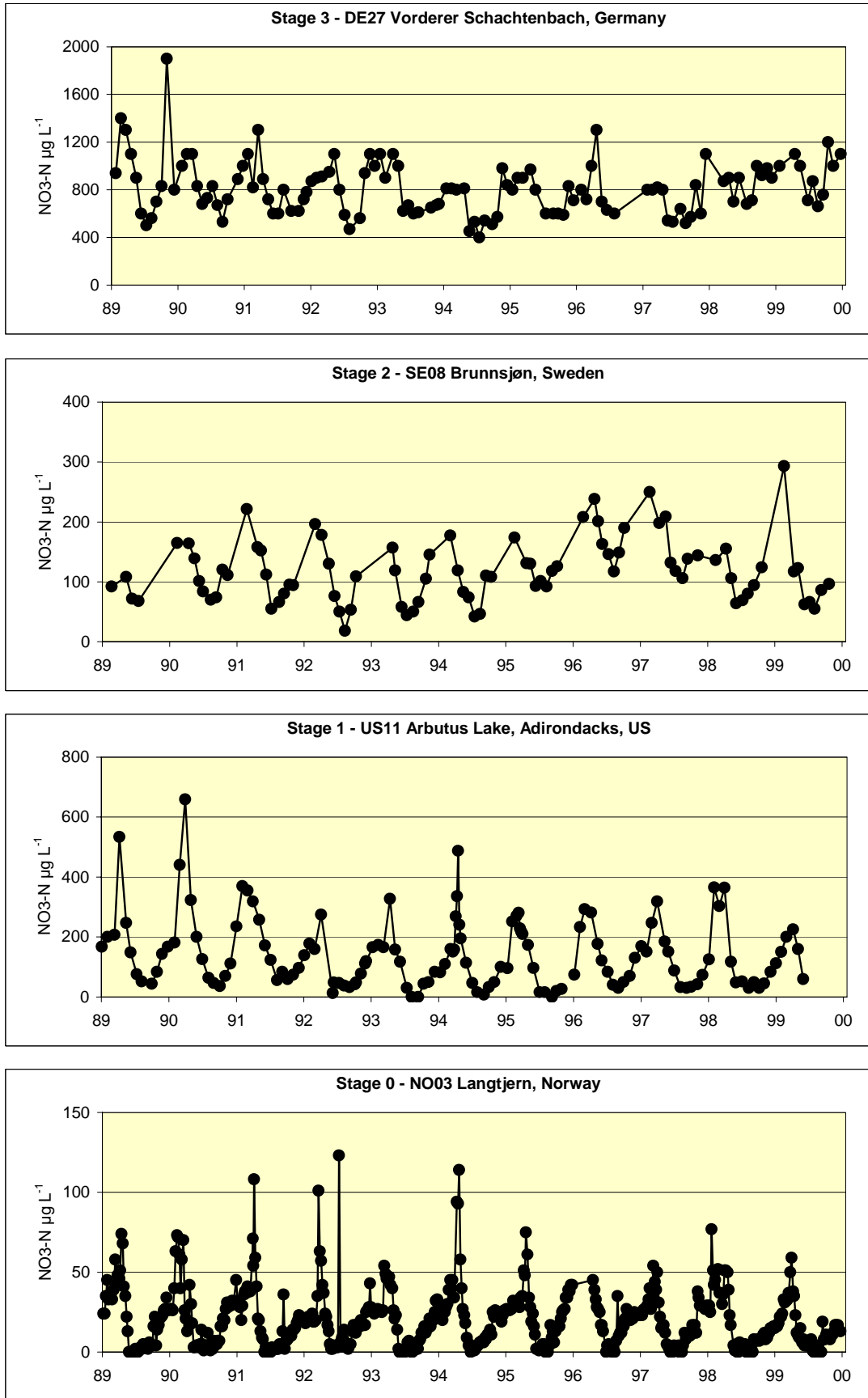
The N cycle at **Stage 0** is dominated by forest and microbial uptake governing the seasonal  $\text{NO}_3^-$  pattern of runoff water. Nitrate concentrations are very low during most of the year, and measurable concentrations may only be found during snowmelt or major hydrologic events. At **Stage 1**, this pattern is amplified: the switch from physical to nutrient limitation in spring is postponed. Substantial  $\text{NO}_3^-$  may leave the catchment during extreme hydrological events, resulting in peaks of episodically high  $\text{NO}_3^-$  concentrations. In **Stage 2**, the seasonal onset of N limitation is even further delayed so that biological demand no longer controls  $\text{NO}_3^-$  concentrations in winter and spring. The period of N limitation during the growing season is much reduced. Additional nitrification, stimulated by a build-up of  $\text{NH}_4$  in soils, results in an increase in  $\text{NO}_3^-$  baseflow concentrations to levels as high as those found in deposition. **Stage 3** is characterised by a lack of any coherent seasonal pattern in  $\text{NO}_3^-$  output. Mineralisation of stored N can add substantially to  $\text{NO}_3^-$  output in surface waters, which may, together with gaseous emissions ( $\text{N}_2\text{O}$ ), exceed inputs from N deposition alone.

The original criteria of Stoddard 1994) were adapted for the range of sampling frequencies that are used to collect the ICP Waters data (for details see Stoddard and Traaen 1995; Traaen and Stoddard 1995). Separate criteria for sites with frequent samples and infrequent samples were developed in order to characterise the majority of available ICP Waters; the inclusion of sites with relatively infrequent samples (<4 times per year) made it difficult to separate Stages 0 and 1, and Stages 2 and 3, and resulted in two classifications in addition to Stoddard's Stages 0 through 3 (Stage 0/1 and Stage 2/3). Typical seasonal  $\text{NO}_3^-$  patterns for sites in each country in various N saturation stages are illustrated in **Figure 13**.

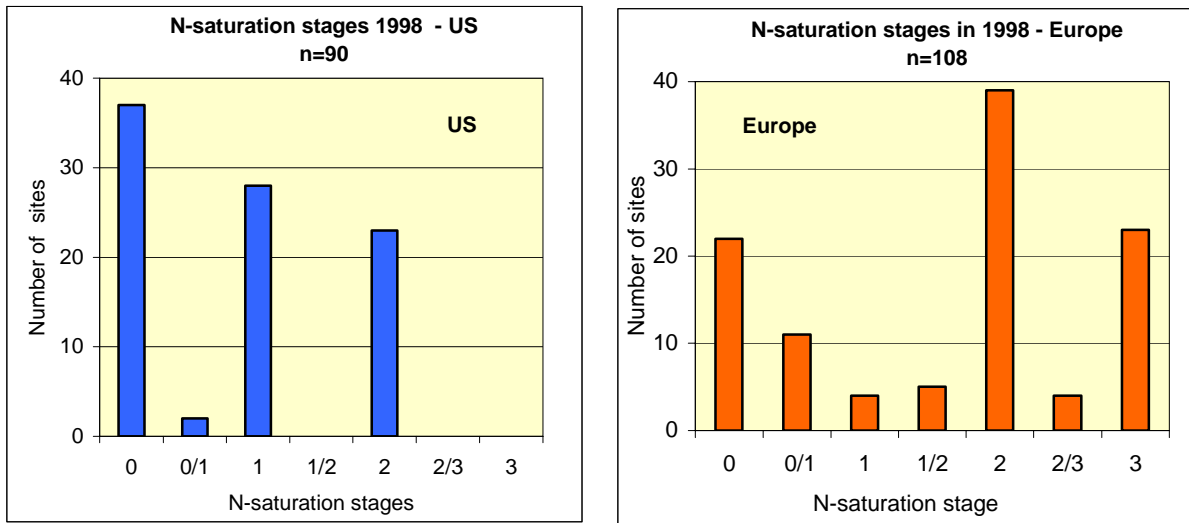
The distribution of N saturation stages in 1998 among all of the sites in ICP Waters is shown in **Figure 14** and **Figure 16**. About half of the 108 ICP sites exhibit a high degree of N saturation (Stage 2 or 3) in 1998. Such sites are found predominantly in Central Europe, although 2 of the Swedish sites are also classed as Stage 2. Most of the sites in the Scandinavian countries are classed Stage 0 or 1 (**Figure 16**).

Thirty-six of the sites were also classified in 1990/92. Of these 24 had no change in N-saturation stage from 1990/92 to 1998, while 8 went down and 4 went up (**Figure 15**).

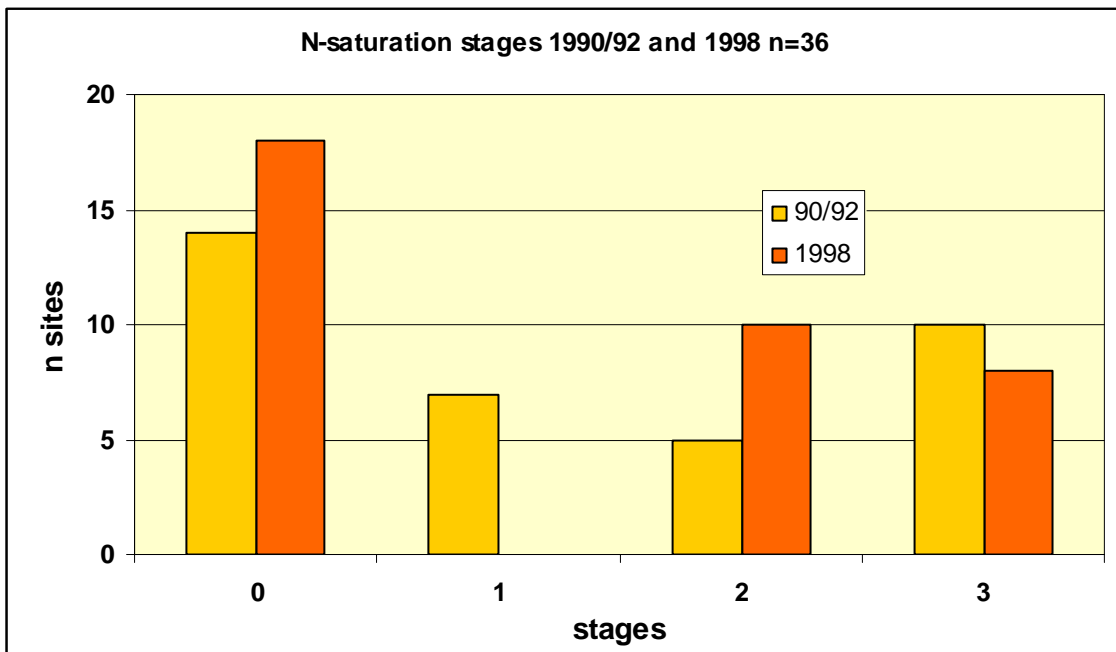




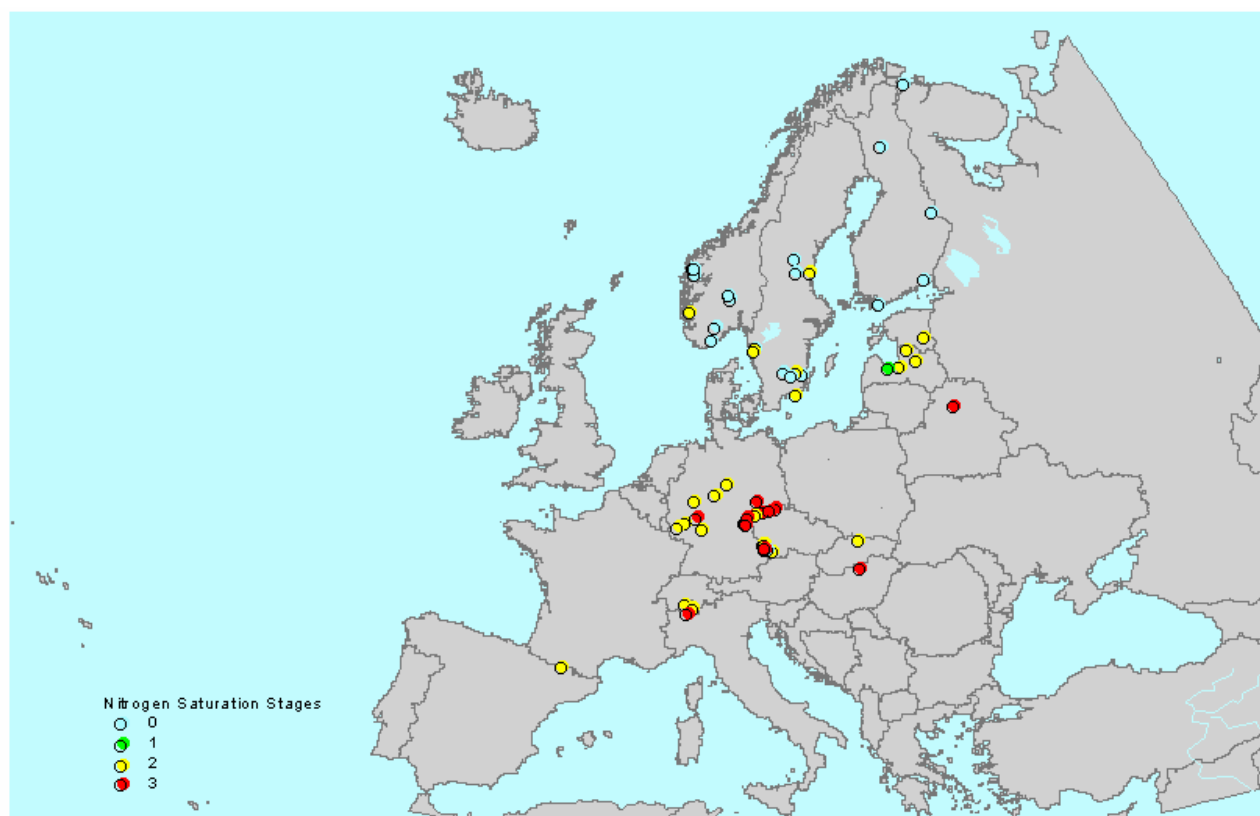
**Figure 13.** Seasonal  $\text{NO}_3^-$  patterns for typical sites illustrating the various N saturation stages.



**Figure 14.** Number of ICP Waters sites in various stages of N saturation in 1998 (stages modified from Stoddard (1994); criteria for assigning sites to stages explained in Traaen and Stoddard (1995)) for US and European and ICP Waters sites.



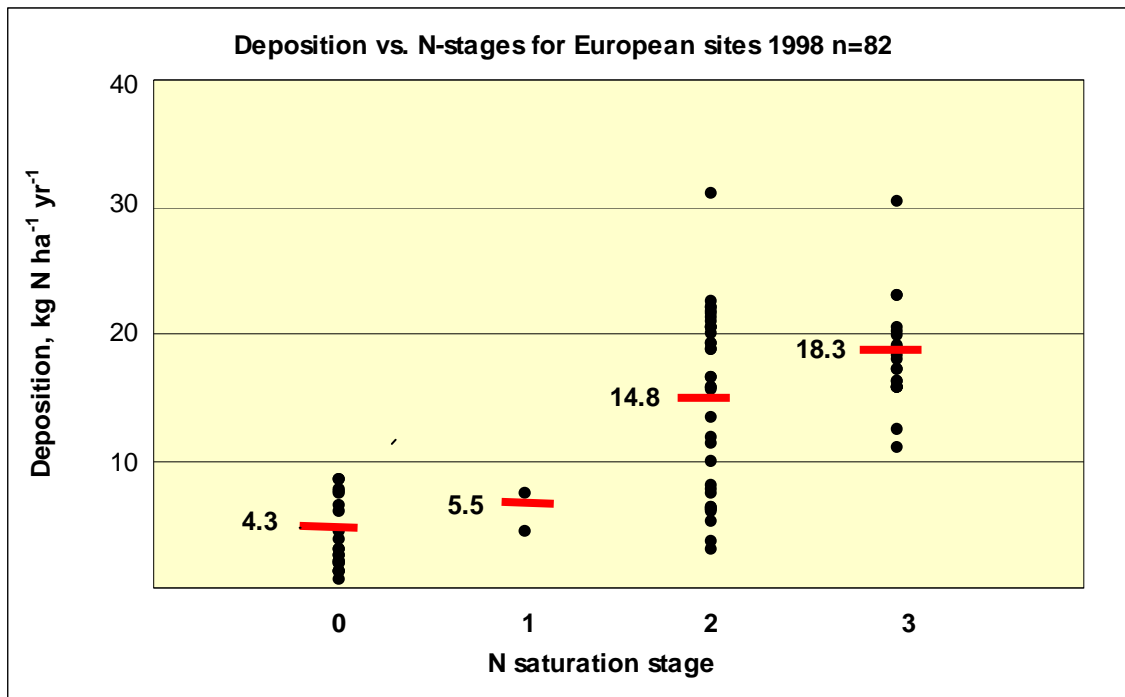
**Figure 15.** Comparison of N-saturation stages between 1998 and 1990/92 for 36 ICP sites.



**Figure 16.** Map showing N-saturation stages in ICP Waters sites in Europe. N saturation stages modified from Stoddard (1994); criteria for assigning sites to stages explained in Traaen and Stoddard (1995).

## 4.7 Relationship between N deposition and N saturation Stage

There is a clear connection between N deposition and Stage classification at European ICP Waters sites (**Figure 17**). N deposition levels are all below  $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  at Stages 0 and 1, while at Stages 3 no sites had deposition below  $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ . This is the same general picture as found in the evaluation of 1990/92 data (Traaen and Stoddard 1995). The picture confirms that there is a correlation between N-saturation stages and the general deposition level.



**Figure 17.** Relationship between N deposition (wet + dry, estimated from EMEP grid data) and stages of N saturation in 1998 for 82 European ICP Waters sites (North America not included). Average values are indicated by horizontal bars, and the numbers indicate average deposition value in  $\text{kg N ha}^{-1} \text{ yr}^{-1}$ .

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## Appendix A. Data

Results from single sites are listed in this table.

- Location, EMEP 50\*50 grid cell (EMEP grid cell do not exist for US and Canda)
- Deposition 1998 for the EMEP grid cell
- Average concentration in 1998/1999 (\* indicates data only from 1998, \*\* data only from 1997)
- N/S is  $(\text{NO}_3^- / (\text{NO}_3^- + \text{SO}_4^{2-}))$ .
- SKT Z-scores and Theil trend slope for  $\text{NO}_3^-$  for the period 1989-1998. Negative scores indicate decreasing trends, while positive scores indicate increasing trends. Scores  $> 1.96$  or  $< -1.96$  are significantly different from 0 at the  $p < 0.05$  level.
- Estimated N-stage, based on 1998 data.

Country	ICP Site	Site Name	EMEP grid square 50*50				Deposition 1998 ox + red kg N ha <sup>-1</sup> yr <sup>-1</sup>	Average concentration					Trends 1989-1998 Z-score slope µeq L <sup>-1</sup> yr <sup>-1</sup>		N-Stage 1998
			I	J	Lat.	Long.		NH <sub>4</sub> µeq L <sup>-1</sup>	NO <sub>3</sub> µeq L <sup>-1</sup>	TOT-N µg L <sup>-1</sup>	SO <sub>4</sub> µeq L <sup>-1</sup>	N/S	Z-score	slope	
Austria	AU03	Tirol, Schwarzsee ob Sölden	107	54	46.96	10.94	19.6	0.6	7.5	105.2	0.07				
Belarus	BY01	Berezinsky Biosphere Reserve	108	84	54.73	28.35	11.1	28.9	47.8	77.6	7.5	0.24		3	
Canada	CA01	Ontario, Algoma Region, Batchawana Lake			47.06	-84.39		4.7	19.9	40.6	95.1	0.17	0.84	0.08	
	CA02	Ontario, Algoma Region, Wishart Lake			47.04	-84.40		2.4	36.2	41.9	8.5	0.27	1.13	0.33	
	CA03	Ontario, Algoma Region, Little Turkey Lake			47.04	-84.41		3.3	33.2	39.9	100.7	0.25	1.95	0.61	
	CA04	Ontario, Algoma Region, Turkey Lake			47.05	-84.41		2.8	30.1	36.0	9.5	0.22	2.38	0.38	
	CA11	Nova Scotia, Mountain Lake			44.32	-65.26			1.4	5.9	34.9	0.04	1.27	0.00	
	CA13	Nova Scotia, Kejimikujik Lake			50.58	-60.38			1.4	7.7	38.1	0.04	2.48	0.10	
Switzerland	CH05	Laghetto (Lago Inferiore), Ticino						1.4	19.9	39.5					
Czech Republic	CZ01	Bohemian Forest, erné	106	59	49.18	13.18	20.5	2.1	69.2	16.5	0.42	-1.64	-1.30	3	
	CZ02	Bohemian Forest, ertovo	106	59	49.17	13.20	20.5	5.4	47.3	112.7	0.30	-1.67	-1.38	2	
	CZ03	Bohemian Forest, Plešné	107	60	48.78	13.87	16.5	2.1	32.0	17.5	0.20	-0.74	-0.90	2	
	CZ04	Bohemian Forest, Prášílské	106	59	49.08	13.40	20.5	1.9	39.6	66.1	0.37	0.98	0.68	2	
	CZ05	Bohemian Forest, Laka	106	59	49.12	13.33	20.5	2.1	42.6	18.5	0.46	0.20	0.33	2	
	CZ06	Bohemian Forest, Zd'árské	106	60	48.93	13.65	15.7	1.4	6.6	136.2	0.05	-0.08	0.00	(2)	
Germany	DE01	Schwarzwald, Dürreychbach	101	54	48.75	8.44	21.1	1.6	88.9	19.5	0.54	1.66	0.51		
	DE02	Fichtelgebirge, Eger	103	59	50.09	11.82	20.1		56.4	100.0	0.36	1.71	0.59	3	
	DE03	Rothaargebirge, Elberndorfer Bach	97	58	50.99	8.20	30.4	3.2	78.9	20.5	0.22	1.67	1.22	3	
	DE04	Sächsische Tieflandsbucht, Ettelsbach	102	62	51.12	12.76	18.2	6.6	163.9	3591.7	0.04			3	
	DE05	Schwarzwald, Goldersbach	101	52	47.87	8.06	19.3	1.9	41.6	21.5	0.37	-0.35	0.00		
	DE06	Hunsrück, Gräfenbach	98	56	49.92	7.62	22.4	1.5	40.6	522.3	0.07	-1.98	-6.46	2	
	DE07	Erzgebirge, Grosse Pyra	103	61	50.41	12.53	18.7	14.3	80.1	22.5	0.15	-2.56	-7.10	3	
	DE08	Bayerischer Wald, Grosse Ohe	106	60	48.93	13.41	15.7	2.4	64.9	68.2	0.49	-0.73	-0.23	3	
	DE09	Sächsische Tieflandsbucht, Heidelberg	102	63	51.43	12.92	17.0	23.5	87.5	23.5	0.02				
	DE10	Bayerischer Wald, Hinterer Schachtenbach	106	60	48.94	13.41	15.7		92.6	64.9	0.59	0.43	0.60	3	



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			I	J	Lat.	Long.		NH <sub>4</sub> µeq L <sup>-1</sup>	NO <sub>3</sub> µeq L <sup>-1</sup>	TOT-N µg L <sup>-1</sup>	SO <sub>4</sub> µeq L <sup>-1</sup>	N/S			
	DE11	Schwarzwald, Kleine Kinzig	101	53	48.42	8.36	19.2	1.4	55.5	24.5	0.44	-1.80	-1.01		
	DE12	Harz, Lange Bramke	98	62	51.86	10.42	21.7	5.4	35.0	245.6	0.12	-2.63	-1.58	2	
	DE14	Kaufunger Wald, Nieste 3	99	60	51.29	9.74	20.4	6.2	36.7	465.1	0.07				
	DE15	Kaufunger Wald, Nieste 5	99	59	51.30	9.62	21.2	5.9	59.7	26.5	0.09	-1.50	-1.32	2	
	DE16	Lauenburgische Seenplatte, Pinnsee	96	65	53.63	10.74	17.5	2.6	3.6	106.9	0.03				
	DE17	Bayerischer Wald, Rachelsee	106	60	48.97	13.40	15.7	3.6	48.4	27.5	0.36	-0.76	-0.83		
	DE18	Fichtelgebirge, Röslau	104	59	50.04	11.90	18.7		38.6	239.2	0.14	-0.36	0.00	2	
	DE19	Taunus, Rombach 2	99	56	50.21	8.44	23.1	5.3	205.9	28.5	0.38			3	
	DE20	Taunus, Rombach 3	99	56	50.20	8.44	23.1	5.2	136.9	226.3	0.38	0.95	0.70	3	
	DE21	Erzgebirge, Rote Pockau	103	62	50.62	13.19	18.9	275.9	104.9	29.5	0.12	-1.93	-7.14	3	
	DE22	Odenwald, Schmerbach 3	100	56	49.66	8.89	20.9	6.1	56.6	347.6	0.14	-1.56	-3.84	2	
	DE23	Bayerischer Wald, Seebach	106	60	48.94	13.41	15.7		54.6	30.5	0.47	-2.87	-1.79	2	
	DE24	Erzgebirge, Talsperre Sosa	103	61	50.49	12.64	18.7	3.0	51.8	484.0	0.10			2	
	DE25	Elbsandsteingebirge, Taubenbach	103	63	50.84	14.13	17.2	1.4	116.1	31.5	0.09			3	
	DE26	Hunsrück, Traunbach 1	97	54	49.72	7.11	21.6	1.0	34.0	169.7	0.17	-1.69	-1.72	2	
	DE27	Bayerischer Wald, Vorderer Schachtenbach	106	60	48.94	13.41	15.7		56.4	32.5	0.42	-1.63	-1.30	3	
	DE28	Oberpfälzer Wald, Waldnaab 2	103	60	49.77	12.42	20.0	4.4	103.5	148.9	0.41				
	DE29	Oberpfälzer Wald, Waldnaab 8	103	60	49.79	12.42	20.0	3.3	66.0	33.5	0.16				
	DE30	Erzgebirge, Wilde Weisseritz	103	63	50.72	13.71	17.2	4.5	112.4	606.5	0.16			3	
	DE31	Erzgebirge, Wolfsbach	102	60	50.32	12.14	19.1	11.0	260.1	34.5	0.26	-1.46	-11.37	3	
	DE32	Rothaargebirge, Zinse	97	57	51.00	8.20	31.0	3.1	57.3	243.2	0.19	-0.51	-0.25	2	
	DE33	Fichtelgebirge, Zinnbach	104	59	50.01	11.90	18.7		72.1	35.5	0.13	-3.03	-5.35	3	
Estonia	EE01	River Ahja, Kiidjärve	101	87	58.15	26.98	5.2	2.9	68.6	101.6	0.19			2	
Spain	ES01	Lake Redó	100	35	42.64	0.77	11.9	2.1	10.7	18.2	0.29			2	
Finland	FI01	Hirvilampi	96	91	60.70	27.92	4.4	2.6	3.2	18.4	0.02	1.28	0.16		
	FI02	Vuorilampi	96	91	60.73	27.92	4.4	7.5	5.0	33.5	0.03	0.92	0.05	0	
	FI03	Mäkilampi	96	91	60.74	27.88	4.4	3.0	2.6	23.8	0.02	-0.31	0.00	0	
	FI05	Lapland, Suopalampi	84	96	67.06	26.10	1.2	0.9	2.1	20.7	0.07	-0.85	0.00	0	
	FI06	Lapland, Vasikkajärvi	84	96	67.11	26.09	1.2	0.8	2.7	10.7	0.06	-1.15	0.00	0	
	FI07	Vusimaa, Vitsjön	94	86	59.96	23.32	6.5	1.6	2.7	23.1	0.02			0	
	FI08	N-Karelia, Kakkisenlampi	92	96	63.65	29.94	1.8	1.5	1.2	12.1	0.03	0.91	0.09	0	
Hungary	HU01	Matra Mountains, Csórrét Reservoir	114	66	47.93	19.96	12.5	6.4	146.9	544.3	0.21			3	
Ireland	IE01	Wicklow, Glendalough, Lake Upper, Mid L.	77	49	53.00	-6.35	15.5	1.6	12.1	19.5	0.14				
	IE02	Wicklow, Glendalough, Lake Upper, Inflow 1	77	49	53.00	-6.38	15.5	0.9	11.9	19.6	0.11				
	IE03	Wicklow, Glendalough, Lake Upper, Inflow 2	77	49	53.00	-6.35	15.5	0.7	23.9	38.4	0.16				
	IE04	Wicklow, Glendalough, Lake Upper, Inflow 3	77	49	53.00	-6.35	15.5	0.8	14.4	23.7	0.16				
	IE05	Galway, Lough Maumwee, Mid Lake	72	49	53.47	-9.55	3.2	1.0	6.7	7.1	0.09				
	IE06	Galway, Lough Maumwee, Inflow 1	72	49	53.47	-9.53	3.2	0.9	3.6	4.2	0.05				
	IE07	Galway, Lough Maumwee, Inflow 2	72	49	53.48	-9.53	3.2	0.8	4.2	6.7	0.06				
	IE08	Donegal, Lough Veagh, Mid Lake	73	53	55.13	-7.70	2.3	1.7	5.7	5.4	0.09				

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			I	J	Lat.	Long.		NH <sub>4</sub> µeq L <sup>-1</sup>	NO <sub>3</sub> µeq L <sup>-1</sup>	TOT-N µg L <sup>-1</sup>	SO <sub>4</sub> µeq L <sup>-1</sup>	N/S			
	IE09	Donegal, Lough Veagh, Inflow 1	73	53	55.13	-7.70	2.3	0.9	2.8	2.3	46.5	0.05			
	IE10	Donegal, Lough Veagh, Inflow 2	73	53	55.13	-7.70	2.3	0.9	1.6	1.8	48.7	0.03			
	IE11	Donegal, Lough Veagh, Inflow 3	73	53	55.13	-7.70	2.3	0.7	2.4	3.3	47.5	0.04			
	IE12	Donegal, Lough Veagh, Inflow 4	73	53	55.13	-7.70	2.3	0.6	5.2	6.4	63.2	0.08			
Italy	IT01	Piemonte, Lake Paione Inferiore	105	48	46.17	8.19	22.1	0.5	25.9		48.5	0.37	-1.19	-0.63	2
	IT02	Piemonte, Lake di Mergozzo	105	49	45.95	8.46	22.1	0.3	46.0	56.5	188.0	0.20			
	IT03	Piemonte, Lake Paione Superiore	105	48	46.17	8.19	22.1	2.4	25.1		49.5	0.41	0.08	0.04	2
	IT04	Piemonte, River Cannobino	106	49	46.07	8.69	15.7	0.5	47.5	56.0	115.0	0.29			2
	IT05	Piemonte, River Pellino	105	49	45.80	8.39	15.7	0.6	111.4	130.5	50.5	0.53	1.89	1.58	3
	IT06	Piemonte, River Pellesino	105	49	45.79	8.38	15.7	2.4	110.1	131.3	69.5	0.61	1.72	1.49	3
Latvia	LV01	Burtnieku Lake, hydrosite	100	85	57.72	25.30	7.7	5.3	73.1	91.6	51.5	0.11			2
	LV02	Barta, Dukupji	103	84	57.13	25.92	6.2	8.5	76.0	149.6	663.3	0.10			2
	LV03	Liela Jugla, Zaki	101	83	56.97	24.48	6.0	5.9	96.4	178.3	52.5	0.08			2
	LV04	Tulija, Zoseni	103	84	57.13	25.92	6.2	14.1	45.9	127.5	591.8	0.07			2
	LV05	Zvirbuli stream, hydrosite	100	82	56.92	23.47	4.4	21.9	6.8		53.5	0.04			1
Norway	NO01	Aust-Agder, Birkenes	86	70	58.38	8.25	7.5		6.9	20.9	94.2	0.07	-2.29	-0.25	0
	NO03	Buskerud, Langtjern	85	75	60.37	9.72	3.8		1.4	20.6	54.5	0.03	1.19	0.02	0
	NO04	Finnmark, Dalelv	80	102	69.68	30.38	0.6		1.6	9.3	101.3	0.02	1.86	0.03	0
	NO05	Oppland, Aurdøla	84	75	60.50	9.50	3.0		3.1	16.5	55.5	0.06	1.98	0.07	0
	NO06	Rogaland, Vikedalselva	83	71	59.53	5.97	3.6		11.5	16.4	40.6	0.22	-0.35	-0.05	2
	NO07	Sogn og Fjordane, Gaula	80	74	61.33	6.12	2.6		6.4	12.3	56.5	0.23	0.84	0.13	0
	NO08	Sogn og Fjordane, Nausta	80	75	61.57	5.88	2.0		5.0	11.1	21.8	0.19	-0.26	-0.03	0
	NO09	Sogn og Fjordane, Trodøla	80	75	61.57	5.93	2.0		5.4	10.2	57.5	0.19	2.00	0.19	0
	NO10	Telemark, Storgama	86	71	59.02	8.53	6.0		5.7	22.9	43.5	0.12	-0.44	-0.05	0
Poland	PL01	Tatra Mountains, Dlugi Staw Gasienicowy	112	67	49.22	20.01	17.8	2.6	48.3	53.1	58.5	0.43	-2.65	-2.76	2
	PL02	Tatra Mountains, Zielony Staw Gasienicowy	112	67	49.22	20.00	17.8	4.3	30.5	42.9	64.1	0.32	-1.90	-1.50	2
Russia	RU07	Chuna, Kola peninsula			51.60	104.00		1.4	2.8	7.1	62.5				
	RU08	Svetloe, Kola peninsula			51.60	104.00		1.7	0.1	15.6	29.1				
	RU09	Arkashino, Kola peninsula			67.82	33.68		1.6	0.1	26.1	63.5				
	RU10	Glubokoe, Kola peninsula			67.82	33.68		0.3	0.1	6.8	54.1				
Sweden	SE01	Delångersån Iggesund	88	82	61.64	17.09	3.0	0.5	7.8	34.6	64.5	0.07	-0.97	-0.14	2
	SE02	Alsterån Getebro	96	74	57.01	16.16	7.8	1.4	7.9	57.1	232.5	0.03	0.75	0.09	0
	SE03	Alsterån Strömsborg	95	74	57.09	15.66	7.3	1.8	12.4	67.6	65.5	0.04	-0.97	-0.25	2
	SE05	Tvåringen	86	82	62.24	15.67	1.9	0.6	1.3	26.1	49.3	0.03	-0.25	-0.02	0
	SE06	Stensjön	87	82	61.64	15.90	2.5	0.7	1.2	26.4	66.5	0.02	-1.10	-0.02	0
	SE08	Brunnsjön	97	73	56.01	15.73	6.2	1.9	10.5	55.6	225.7	0.04	0.91	0.34	2
	SE09	Fiolen	94	73	57.09	14.53	8.5	0.9	3.2	36.1	67.5	0.01	-2.25	-0.83	0
	SE10	Storasjö	94	74	56.94	15.27	7.4	2.4	2.6	42.4	91.3	0.03	1.14	0.06	0
	SE11	Fräcksjön	90	73	58.14	12.18	8.4	1.0	5.2	40.6	68.5	0.03	-1.68	-0.12	0
	SE12	Härsvatten	90	72	58.02	12.03	8.1	4.0	8.9	28.8	136.9	0.06	-2.90	-0.56	2

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United Kingdom	UK01	Scotland, Loch Coire nan Arr	73	59	57.42	-5.65	2.7	3.1	11.1	69.5	0.07	1.10	0.00		
	UK04	Scotland, Lochnagar	76	60	56.76	-3.22	6.4	22.1	25.4	55.3	0.29	2.02	1.25		
	UK07	Scotland, Round Loch of Glenhead	77	55	55.09	-4.42	10.0	12.5	22.3	70.5	0.17	2.62	0.75		
	UK10	England, Scoat Tarn	79	55	54.48	-3.29	12.6	25.9	30.7	59.9	0.30	0.33	0.34		
	UK15	Wales, Llyn Llagi	80	51	53.01	-4.01	9.0	12.6	20.4	71.5	0.18	0.76	0.27		
	UK21	N.Ireland, Blue Lough	76	52	54.15	-5.97	15.9	37.2	51.9	91.1	0.29	1.07	1.00		
United States	US05	Maine, Little Long Pond			44.64	-68.08		1.6	0.3	66.6	0.00				
	US06	Maine, Tilden Pond			44.63	-68.07		1.6	0.3	54.1	0.01				
	US11	New York, Adirondack Mnt., Arbutus			43.99	-74.24		0.9	22.8	114.4	0.17	-1.79	-0.44		
	US12	New York, Adirondack Mnt., Constable			43.83	-74.80		0.3	18.2	111.5	0.14	-0.35	-0.33		
	US13	New York, Adirondack Mnt., Dart Lake			43.80	-74.86		2.3	14.6	99.2	0.13	-1.37	-0.99		
	US14	New York, Adirondack Mnt., Heart Lake			44.18	-73.97		2.9	14.2	92.1	0.13	-1.07	-0.35		
	US15	New York, Adirondack Mnt., Lake Rondaxe			43.76	-74.91		1.6	13.1	99.8	0.12	-1.80	-1.01		
	US17	New York, Adirondack Mnt., Otter Lake			43.79	-74.85		2.4	13.4	111.3	0.11	-1.34	-0.60		
	US23	New York, Catskill Mnt., E. Br. Neversink, Headw.			43.19	-74.50		0.5	26.1	100.7	0.21	-2.10	-2.54		
	US24	New York, Catskill Mnt., Rondout Creek			41.96	-74.45		2.6	4.1	90.5	0.04	-1.82	-1.94		
	US34*	Little Clear Pond. Adirondacks			41.93	-74.38		3.4	9.9	93.2	0.10				
	US35	Loon Hollow Pond. Adirondacks			42.01	-74.41		2.9	24.1	96.5	0.20				
	US36	Willys Lake. Adirondacks			41.99	-74.50		0.1	25.3	81.3	0.24				
	US37	Woods Lake. Adirondacks			44.52	-74.13		0.4	31.6	82.4	0.28				
	US38	Middle Settlement Lake. Adirondacks			44.51	-74.13		2.6	11.8	50.1	0.19				
	US39	Grass Pond. Adirondacks			44.31	-74.37		0.4	34.4	52.0	0.40				
	US40	Middle Branch Lake. Adirondacks			44.34	-74.30		1.4	11.8	38.1	0.24				
	US41	Limekiln Lake. Adirondacks			44.35	-74.29		0.6	20.6	84.4	0.20				
	US42	Squaw Lake. Adirondacks			44.16	-73.95		0.3	10.6	35.4	0.23				
	US43	Indian Lake. Adirondacks			44.66	-74.50		0.6	20.5	37.4	0.35				
	US44	Brook Trout Lake. Adirondacks			44.66	-74.50		0.8	11.4	83.6	0.12				
	US45	Lost Pond. Adirondacks			43.96	-75.05		0.9	27.8	105.0	0.21				
	US46	South Lake. Adirondacks			43.97	-74.96		0.5	34.8	98.0	0.26				
	US47	North Lake. Adirondacks			43.87	-74.97		0.1	33.3	84.7	0.28				
	US48	Willis Lake. Adirondacks			43.68	-75.10		1.1	4.9	103.0	0.05				
	US49	Long Pond. Adirondacks			43.69	-75.06		0.8	3.6	91.5	0.04				
	US50	Carry Pond. Adirondacks			43.70	-75.10		1.6	2.0	96.1	0.02				
	US51	Lake Colden. Adirondacks			43.71	-74.81		0.6	45.7	91.9	0.33				
	US52	Avalanche Lake. Adirondacks			43.64	-74.74		1.1	59.5	95.7	0.38				
	US53	Little Simon Pond. Adirondacks			43.62	-74.76		0.6	42.7	89.2	0.32				
	US54	Raquette Lake Reservoir. Adirondacks			43.60	-74.66		0.2	28.6	90.9	0.24				
	US55	G Lake. Adirondacks			43.65	-74.56		0.4	19.9	86.1	0.19				
US56	Middle Pond. Adirondacks			43.51	-74.88		3.4	7.1	98.2	0.07					
US57	Sagamore Lake. Adirondacks			43.52	-74.95		0.3	27.1	92.6	0.23					
US58	Black Pond Outlet. Adirondacks			43.37	-74.25		0.1	6.7	79.0	0.08					

Country	ICP Site	Site Name	EMEP grid square 50*50				Deposition 1998 ox + red kg N ha <sup>-1</sup> yr <sup>-1</sup>	Average concentration					Trends 1989-1998 Z-score slope µeq L <sup>-1</sup> yr <sup>-1</sup>	N-Stage 1998
			I	J	Lat.	Long.		NH <sub>4</sub> µeq L <sup>-1</sup>	NO <sub>3</sub> µeq L <sup>-1</sup>	TOT-N µg L <sup>-1</sup>	SO <sub>4</sub> µeq L <sup>-1</sup>	N/S		
	US59	Windfall Pond Outle. Adirondacks			43.84	-74.48		0.7	28.5		67.6	0.30		
	US60	Queer Lake. Adirondacks			43.68	-74.49		1.1	19.5		90.9	0.18		
	US61	Big Moose Lake. Adirondacks			44.12	-73.98		1.0	25.6		89.4	0.22		
	US62	Cascade Lake Outlet. Adirondacks			44.13	-73.97		0.4	26.4		110.0	0.19		
	US63	Little Echo Pond. Adirondacks			44.16	-74.44		0.5	0.6		125.0	0.01		
	US64	Squash Pond Outlet. Adirondacks			43.80	-74.65		1.1	30.5		88.8	0.26		
	US65	West Pond Outlet. Adirondacks			43.42	-74.64		1.2	12.6		106.5	0.11		
	US66	Bubb Lake Outlet. Adirondacks			44.34	-74.37		0.6	17.8		117.5	0.13		
	US67	Owen Pond. Adirondacks			43.77	-74.63		0.7	54.2		107.3	0.34		
	US68	Jockeybush Lake. Adirondacks			44.44	-74.29		1.1	25.1		113.6	0.18		
	US69	Clear Pond. Adirondacks			43.81	-74.85		0.4	8.5		101.9	0.08		
	US70	Nate Pond. Adirondacks			43.81	-74.81		0.5	15.5		101.3	0.13		
	US71	Bean Pond. Maine			43.83	-74.85		1.3	2.4		107.1	0.02		
	US72	Bracey Pond. Maine			43.79	-74.80		1.6	0.6		37.9	0.02		
	US73	Anderson Pond. Maine			44.31	-74.40		0.0	0.5		90.9	0.01		
	US74	Mud Pond. Maine			43.83	-74.89		1.5	0.4		82.2	0.01		
	US75	Salmon Pond. Maine			43.81	-74.88		1.4	0.9		95.7	0.01		
	US76	Wiley Pond. Maine			43.77	-74.85		0.9	0.9		125.2	0.01		
	US77	Second Pond. Maine			44.32	-73.90		0.6	1.1		99.0	0.01		
	US78	Abol Pond. Maine			43.30	-74.59		0.7	0.9		100.7	0.01		
	US79	Duck Pond. Maine			44.00	-73.82		1.5	0.7		118.1	0.01		
	US80	Jellison HI Pd. Maine			43.86	-74.09		1.1	0.9		61.2	0.01		
	US81	Crystal Pond. Maine			45.13	-69.98		1.0	0.5		44.3	0.01		
	US82	Newbert Pond. Maine			44.96	-68.12		2.4	1.1		49.9	0.02		
	US83	Partridge Pond. Maine			44.65	-68.06		1.1	0.9		86.5	0.01		
	US84	Benner Run. Mid-Apps			44.63	-65.09		0.9	15.9		49.7	0.24		
	US85	Linn Run. Mid-Apps			44.63	-68.09		0.2	39.5		52.0	0.43		
	US86	Roberts Run. Mid-Apps			46.00	-68.54		0.6	3.1		62.0	0.05		
	US87	Stone Run. Mid-Apps			44.70	-68.58		0.6	1.4		58.4	0.02		
	US88	Baldwin Creek. Mid-Apps			45.84	-68.93		0.1	41.6		40.4	0.51		
	US89*	Bourn. Vermont			44.80	-68.19			1.6		72.8	0.02		
	US90*	Grout. Vermont			44.81	-68.43			5.3		34.3	0.13		
	US91*	Hardwood. Vermont			45.03	-68.19			2.9		70.7	0.04		
	US92*	Little - Woodford. Vermont			44.34	-69.27			16.5		55.7	0.23		
	US93*	Stamford. Vermont			44.84	-68.40			6.2		131.2	0.05		
	US94*	Stratton. Vermont			40.93	-78.02			0.0		203.6	0.00		
	US95*	Sunset. Vermont			40.14	-79.21			1.7		194.3	0.01		
	US96*	Big Mud. Vermont			41.17	-78.41			2.6		207.0	0.01		
	US97*	Branch. Vermont			41.10	-78.45			1.4		202.4	0.01		
	US98*	Beaver Pond. Vermont			40.35	-79.05			11.6		56.2	0.17		
	US99*	Big Muddy. Vermont			43.11	-73.00			11.1		56.6	0.16		

Country	ICP Site	Site Name	EMEP grid square 50*50				Deposition 1998 ox + red kg N ha <sup>-1</sup> yr <sup>-1</sup>	Average concentration					Trends 1989-1998 Z-score slope µeq L <sup>-1</sup> yr <sup>-1</sup>	N-Stage 1998
			I	J	Lat.	Long.		NH <sub>4</sub> µeq L <sup>-1</sup>	NO <sub>3</sub> µeq L <sup>-1</sup>	TOT-N µg L <sup>-1</sup>	SO <sub>4</sub> µeq L <sup>-1</sup>	N/S		
	US100*	Howe. Vermont			43.05	-72.95			4.3		65.9	0.06		
	US101*	South - Marlboro. Vermont			44.47	-72.50			0.0		79.9	0.00		
	US102*	Forester. Vermont			42.93	-73.07			0.0		76.1	0.00		
	US103	Paine Run. Virginia			42.82	-73.07		0.2	4.0		63.4	0.06		
	US104	Piney River. Virginia			43.10	-72.97		0.4	11.7		71.1	0.14		
	US105	Staunton River. Virginia			42.92	-72.68		2.4	2.5		59.1	0.04		
	US97*	Branch. Vermont			43.31	-72.93			1.4		62.2	0.02		
	US98*	Beaver Pond. Vermont			43.08	-73.02			11.6		72.6	0.14		
	US99*	Big Muddy. Vermont			45.01	-71.94			11.1		55.5	0.17		
	US100*	Howe. Vermont			44.76	-72.60			4.3		63.0	0.06		
	US101*	South - Marlboro. Vermont			42.79	-72.99			0.0		82.8	0.00		
	US102*	Forester. Vermont			42.84	-72.71			0.0		75.1	0.00		
	US103	Paine Run. Virginia			43.08	-72.87		0.2	4.0		116.3	0.03		
	US104	Piney River. Virginia			38.20	-78.79		0.4	11.7		70.1	0.14		
	US105	Staunton River. Virginia			38.70	-78.27		2.4	2.5		46.0	0.05		
					38.44	-78.37								

## Appendix B. Major Programme Findings

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) is designed to assess the degree and geographical extent of acidification of surface waters, evaluate dose/response relationships and long-term trends in aquatic chemistry and biota. The major programme findings are listed here.

### The 3-year period 1996-98

#### *The ICP Waters database*

The ICP Waters database has been renovated into a new working database with sites and data selected on the basis of three criteria. (1) Only active sites are included in the new database. These are currently monitored and for which data in the period 1996-98 have been reported to the programme centre. (2) Only the required or optional parameters are included. In addition we have included total organic carbon (TOC), reactive and non-labile aluminium and colorimetric analysed  $\text{SO}_4$ , where available. (3) Only the upper depth is included for lakes. The ICP Waters database now includes 142 sites with chemical data and 123 sites with biological data. Quality control of data is performed annually, and most laboratories have participated in intercalibration exercises in the period.

#### *Representativeness of the database*

Many of the sites included in the ICP Waters programme are especially sensitive to acidification. They need not be representative of all surface waters in a region, but rather represent the acid-sensitive surface waters. Most of the sites appear to be well suited to monitor changes in acidification in response to changes in acid deposition. The sites are generally representative of the lower ANC, lower critical load levels of the distributions for all the waters surveyed in the region.

The ICP Waters sites cover most of the acid-sensitive areas in Europe that receive significant acid deposition. There are no ICP sites, however, in several regions that have been or are potentially affected. Furthermore there are insufficient data from many regions in Eastern Europe to adequately assess both the risk of acidification and the sensitivity of surface waters. For North America the ICP sites cover several, but not all of the acid-sensitive regions that receive significant acid deposition, and that can be expected to show changes in response to changing levels of deposition in the future.

#### *Trends in water chemistry*

Data from 98 ICP Waters sites with sufficient data were tested for trends in concentrations of major chemical components for the 10-year period 1989-1998 using the non-parametric Seasonal Kendall test (SKT). The sites were grouped into regions by means of meta-analysis.

All of the regions had highly significant downward trends in sulphate ( $\text{SO}_4$ ). The majority of the single sites (67 out of 98) showed a significant decrease in  $\text{SO}_4$ . Nitrate ( $\text{NO}_3^-$ ), on the other hand, showed no regional patterns of change. Central Europe did show some significant decreasing  $\text{NO}_3^-$  trends, but the heterogeneity within the region was too large to identify a regional pattern. Decreasing trends in lake water  $\text{NO}_3^-$  occurred in the so-called Black Triangle, while southwest Germany and Italy (Southern Alps) exhibited no or increasing  $\text{NO}_3^-$  trends.

Recovery in acidification reflected by an increase in surface water ANC and pH is significant in the Nordic countries/UK region. Lack of recovery during the last decade at the single UK sites has been attributed to (a) the absence of significant S-deposition reductions in western areas, and (b) the impact of natural climatic variations on water chemistry (seasalt episodes). In Central Europe, there was a regional tendency toward increasing ANC, but significant heterogeneity; 10 out of 28 sites in this region showed no significant change in ANC. Two important regions failed to show significant recovery. In Eastern North America there was no regional pattern for ANC or pH, while northern Nordic countries showed increase in pH but no regional pattern for ANC. Concentrations of base cations declined in most regions. All of the regions showed tendencies towards increasing concentrations of dissolved organic carbon (DOC).

The sites were also grouped according to several characteristics. Sites with low acid neutralising capacity (ANC) showed the largest rates of recovery. Neither the high  $\text{NO}_3^-$  or low  $\text{NO}_3^-$  groups of sites exhibited significant trends in  $\text{NO}_3^-$  concentrations. Non-forested sites showed clear and consistent signals of recovery in ANC and pH, and appropriate (relative to  $\text{SO}_4$  trends) rates of base cation declines. Hence, the recovery observed, in fact, associated with declining  $\text{SO}_4$ .

### ***Biology***

Effects of acidification are described for Ireland, UK, Scandinavia and Central Europe including lowland as well as mountain areas. Critical limits of ANC are suggested for the different regions. For Ireland, UK and Norway, ANC of  $20 \mu\text{eq L}^{-1}$  are proposed. For Sweden, Germany and the Vosges Mountains of France the limit is set to  $50 \mu\text{eq L}^{-1}$ . In the high Alps and Pyrenees the present information indicates a limit of about  $30 \mu\text{eq L}^{-1}$ . For UK and most sites in Germany no statistically significant trend in acidification was recorded, but positive signals of improvements in the invertebrate fauna were observed. A clear positive trend was found for the Norwegian sites and for most of the Swedish sites. In the most acidic sites in Central Europe, improvements in water quality have not yet reached a level where stable effects on biology can be detected. Biological recovery of such sites requires considerable and stable improvements in water quality with respect to acidification.

### ***Heavy metals***

The ICP Waters database contains a number of sites with heavy metal data, and these sites are located in relatively few countries, and heavy metals are reported from fewer sites in each of these countries than major solutes. To be able to give a good picture of the general level of heavy metals in surface waters throughout Europe and North America, heavy metal data for more sites with a larger geographical cover are needed. Some Focal Centres reported that there exist data on heavy metals from additional rivers. These sites are not likely to be suitable for monitoring the effects of air-transported heavy metals to surface waters.

Few sites have long time series on heavy metals. The analytical methods have changed and the detection limit has generally decreased through the monitoring period for the sites with long-term trends.

Heavy metal data in the ICP Waters database has been analysed with different analytical methods and analytical precision. All the laboratories reporting data to ICP Waters participate in national and international intercalibrations. To be able to compare the data within the programme it is of great importance to conduct intercalibration of analysis of heavy metals for improvements of results.

### ***Future perspectives***

The ICP Waters programme is well positioned to monitor changes over the next years as the new Protocol on reductions in emissions of S and N compounds comes into effect. The trends already documented in water chemistry and biology in the ICP Waters data are thus likely to continue to reflect the reduced emissions of S and N.

The results from the ICP Waters Programme, as reported here in the 12-year report, clearly show that surface waters respond to changes in atmospheric deposition. Indeed waters are much more responsive than either soils or terrestrial vegetation to changes in long-range transported acid pollutants. Lakes and rivers also have the advantage that they reflect responses integrated over the entire catchment area. The ICP site network is geographically extensive and includes long-term data series (> 15 years) for many sites. The network is thus well poised to document changes that result from implementation of the protocols.

The ICP Waters programme is best developed with respect to major-ion chemistry and chemical parameters associated with acidification. Addition of new biological parameters and expansion of sampling programmes at individual sites to include existing biological parameters could substantially strengthen the network. As reported here, the invertebrate fauna responds to changes in water chemistry, and is a suitable group of organisms for monitoring purposes. As the acidification of surface waters continues to diminish in the future, the recovery of damaged groups of organisms should increase.

The ICP Waters programme presently includes only a minor amount of data for heavy metal concentrations. An analysis of these data presented here indicates that ecological effects of long-range transported heavy metals are probably minor. Intercalibration of heavy metal analysis methods should be a part of the yearly intercalibration exercise. In the event that the Working Group on Effects (WGE) deems it prudent to include heavy metals in its

activities, the ICP Waters sampling programme and database provides a ready vehicle for sampling, quality control and assessment of heavy metals in surface waters.

### **The 3-year period 1993-95**

#### ***Trends in Surface Water Chemistry***

Trends in water chemistry indicate that  $\text{SO}_4$  concentrations are decreasing at almost all ICP Waters sites, and in almost all cases the decreases in the 1990s are larger than in the 1980s.  $\text{NO}_3^-$  is of increasing importance as a strong-acid anion. In the Nordic countries (Finland, Sweden, and Norway) alkalinity decreased in the 1980s (acidification), but then increased in the 1990s (recovery). At many European sites (Italy, Germany, Netherlands, Denmark) alkalinity also increased in the 1980s, and the rate accelerated in the 1990s. Other regions (e.g. Adirondacks/Quebec, Midwestern North America, UK) show either no recovery or further acidification.

Regions with declining  $\text{SO}_4$  that fail to show recovery in alkalinity in the 1990s (Adirondacks/Quebec, Midwestern North America) are characterised by strongly declining concentrations of sum base cations ( $\text{SBC} = \Sigma \text{Ca, Mg, K, Na}$ ). SBC concentrations are no longer declining in the Nordic Countries. All of the hydrogen ion ( $\text{H}^+$ ) trends detectable at a regional level are consistent with the alkalinity trends observed within each region.

The 1980s were characterised by increases in  $\text{NO}_3^-$  in almost all regions. These increases have generally not continued in the 1990s. Regional-scale phenomena other than N deposition may be responsible for the development in the 1990s (e.g., changes in climate or climate extremes).

#### ***Nitrogen Leaching from ICP Waters Sites***

$\text{SO}_4$  is the most important acidifying anion at ICP Waters Sites, but  $\text{NO}_3^-$  constitutes more than 10% of the non-marine acid anions at 63% of the sites. Exceptions are sites in Canada, Finland, Sweden and Russia. More than 50% of the sites have yearly-average  $\text{NO}_3^-$  concentrations  $> 10 \mu\text{eq L}^{-1}$ , and 27% are  $> 50 \mu\text{eq L}^{-1}$ . Dutch sites have ammonium values  $> 100 \mu\text{eq L}^{-1}$ .

More than 50% of the ICP Waters sites show high degree of N saturation (stages 2 or 3 according to Stoddard's classification system). There is a clear relationship between high N saturation stage and high N deposition load. Leaching of inorganic N occurs at sites with very high annual precipitation (Norway, Ireland, Italy, and UK) even though deposition is below the empirical threshold for leaching of  $10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ .

Compared with sulphur (S), nitrogen (N) is much more involved in biological processes within ecosystems. Hence changes in N deposition may not always directly correlate with changes in inorganic N leaching in runoff. Besides N-deposition, the overall N-status of ecosystems, changes in climate or climate extremes and hydrology can strongly influence leaching of excess  $\text{NO}_3^-$  (and ammonium) from a catchment.

#### ***Effects of Acidification on Aquatic Fauna***

Water acidification (including acidic episodes) changes the composition of invertebrate assemblages in surface waters. Long-term monitoring of both water chemistry and biology is necessary to assess dose/response relationships.

In areas with originally high pH (6.0 - 8.0) and high Ca-concentrations (e.g., Southern Sweden, Germany) a critical alkalinity limit of  $50 \mu\text{eq L}^{-1}$  is proposed. In areas where fauna is adapted to water with low conductivity, low pH (5.5 - 6.5) and low calcium concentrations (e.g., Finland, Norway), alkalinity values should be  $\geq 20 \mu\text{eq L}^{-1}$  to protect invertebrates and fish. These critical levels are the basis for setting critical loads of acidity for surface waters. By comparing invertebrate samples taken before and after 1990, improvements at many Norwegian and some German sites can be observed. This is confirmed by correlation analyses between time and acidity index applied to Norwegian long-term data series. Detrended canonical correspondence analysis (Norwegian datasets) shows a high correlation of invertebrate assemblages with pH and total aluminium (Al), and also a significant correlation with calcium. Trend analyses indicate recovery of the invertebrate fauna at many sites.

#### ***Intercalibration Exercises***

The programme conducts yearly chemical and biological intercalibrations. The number of participating laboratories have increased from 9 in 1987 to 57 laboratories in 24 countries in the 13<sup>th</sup> chemical intercomparison in 1999. The investigated variables are pH,  $\text{K}_{25}$  (conductivity),  $\text{HCO}_3^-$  (alkalinity),  $\text{NO}_3^- + \text{NO}_2^-$ ,



Cl<sup>-</sup>, SO<sub>4</sub><sup>2-</sup>, Ca<sup>2+</sup>, Mg<sup>2+</sup>, Na<sup>+</sup>, K<sup>+</sup>, Al, reactive Al, non-labile Al, DOC and COD-Mn. The intercomparison confirms that the results are generally compatible between laboratories.

In 1998, 6 laboratories participated in the 4<sup>th</sup> intercalibration of invertebrates. The results from this intercalibration are the best of all tests performed so far. The biological intercalibration proved that the methods suggested in the Programme Manual are reliable and suitable to assess the effects of acidification on aquatic fauna.

#### ***Implications for the Assessment of Critical Loads***

An assessment of the status for critical load and critical load exceedences, using the methods described in the mapping manual (UN/ECE 1996) showed that at 46% of the European sites the critical loads of acidity is less than 50 meq/m<sup>2</sup>/yr, and 68% of the sites have critical loads less than 100 meq/m<sup>2</sup>/yr. The corresponding figures for the North American sites are 40% and 75%, respectively. The results indicated that many of the selected ICP Waters sites are sensitive to acidification. This raises the question whether the sensitivity of the surface waters has been underestimated in large parts of Europe, and that in several of the EMEP-grid cells the low percentiles would be even lower if critical loads for surface waters had been included.

### **The 3-year period 1990-93**

#### ***Trends in Water Chemistry***

Significantly decreasing SO<sub>4</sub> concentrations were a major feature of the ICP Waters sites. A decline in Ca<sup>2+</sup> was found at many European locations. Several surface waters were characterised by upward trends in NO<sub>3</sub><sup>-</sup> concentrations, and constant or increasing N deposition seemed to be the driving force. A combined decrease in Ca<sup>2+</sup> and increase in NO<sub>3</sub><sup>-</sup> may account for the lack of recovery from acidification at sites with decreasing SO<sub>4</sub>.

#### ***Correlation between Surface Water Trends and Deposition***

Statistically significant links between observed surface water trends and *changes* in deposition chemistry were not discovered. This was certainly due to the kind of deposition data used both in space and time. Information on atmospheric inputs was not available for the single sites. Deposition from the nearest existing monitoring station, up to 160 km away, was paired with the surface waters, respectively. The development of S deposition at EMEP sites shows a steady decrease since the 1970s. The ICP on Waters Programme includes mainly data from 1980 onwards. Response to decreases in SO<sub>4</sub> deposition at certain sites might have been influenced by earlier changes in deposition.

#### ***Dose / Response Relationships***

In general, sensitive invertebrate species were associated with high ANC and conductivity and tolerant species with high Al, low pH and ANC. The chemical parameter most strongly related to fauna composition was pH. It was possible to predict pH from invertebrate assemblages at single sites. The effect of acidification on invertebrates varied from region to region, depending on Ca<sup>2+</sup> concentrations, humic substances and ionic strength. In areas with originally oligotrophic waters, the fauna was adapted to low conductivity, pH values of 5.5 - 6.5 and an ANC of 10 - 30 µeq L<sup>-1</sup>. In Central Europe, the fauna was much more sensitive to acidification. It was adapted to waters with high ionic strength, rich in Ca<sup>2+</sup>, with pH values of 6.0 - 8.0 and an ANC of 50 - 200 µeq L<sup>-1</sup>.

#### ***Intercalibration Exercises***

Chemical intercalibration exercises showed that the results were generally compatible between laboratories. Biological intercalibration in 1992 proved that the methods suggested in the Programme Manual are reliable and suitable to assess the effects of acidification on aquatic fauna. The tests included, however, only a selection of the species present at the various sites.

#### ***Implications for the Assessment of Critical Loads***

The results of the ICP Waters monitoring activities from 1989 to 1991 stress the importance of considering multiple pollutants (i.e., S and N) which together affect sensitive organisms in freshwater ecosystems. Thus, critical load assessments should focus on deposition of both S and N compounds.

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## Appendix C. Reports and publications from the programme

1. Manual for Chemical and Biological Monitoring. Programme Manual. Prepared by the Programme Centre, Norwegian Institute for Water Research. NIVA, Oslo 1987.
2. Norwegian Institute for Water Research, 1987. Intercalibration 8701. pH, Ks, SO<sub>4</sub>, Ca. Programme Centre, NIVA, Oslo.
3. Norwegian Institute for Water Research, 1988. Data Report 1987 and available Data from Previous Years. Programme Centre, NIVA, Oslo.
4. Norwegian Institute for Water Research, 1988. Intercalibration 8802. pH, K<sub>25</sub>, HCO<sub>3</sub>, NO<sub>3</sub>, SO, Cl, Ca, Mg, Na, K. Programme Centre, NIVA, Oslo.
5. Proceedings of the Workshop on Assessment and Monitoring of Acidification in Rivers and Lakes, Espoo, Finland, 3rd to 5th October 1988. Prepared by the Finnish Acidification Research Project, HAPRO, Ministry of Environment, October 1988.
6. Norwegian Institute for Water Research, 1989. Intercalibration 8903: Dissolved organic carbon and aluminium fractions. Programme Centre, NIVA, Oslo. NIVA-Report SNO 2238-89. ISBN 82-577-1534-4.
7. Note: Some reflections about the determination of pH and alkalinity. Prepared by the Programme Centre, Norwegian Institute for Water Research. Håvard Hovind, NIVA, Oslo October 1989.
8. Hovind, H. 1990. Intercalibration 9004: pH and alkalinity. Programme Centre, NIVA, Oslo. NIVA-Report SNO 2465-90. ISBN 82-577-1776-2.
- Skjelkvåle, B.L. and Wright, R.F. 1990. Overview of areas sensitive to acidification: Europe. Programme Centre, NIVA, Oslo. Acid Rain Research Report 20/1990. NIVA-Report 2405-90. ISBN 82-577-1706-1.
9. Johannessen, M. 1990. Intercalibration in the framework of an international monitoring programme. Proceedings of the third annual Ecological Quality Assurance Workshop, Canada Centre for Inland Waters, Burlington Ontario. Programme Centre, NIVA, Oslo.
10. Norwegian Institute for Water Research, 1990. Data Report 1988. Programme Centre, NIVA, Oslo.
11. Norwegian Institute for Water Research, 1990. Data Report 1989. Programme Centre, NIVA, Oslo.
12. Proceedings for the 5th Meeting of the Programme Task Force Freiburg, Germany, October 17 -19, 1989. Prepared by the Umweltbundesamt, Berlin July 1990.
13. Hovind, H. 1991. Intercalibration 9105: pH, K<sub>25</sub>, HCO<sub>3</sub>, NO<sub>3</sub> + NO<sub>2</sub>, Cl, SO<sub>4</sub>, Ca, Mg, Na, K and TOC. Programme Centre, NIVA, Oslo. NIVA-Report 2591-91. ISBN 82-577-1931-5.
14. Norwegian Institute for Water Research, 1991. The Three Year Report. Summary and results 1987 – 1989: Results from the International Co-operative Programme on Assessment and Monitoring of Acidification in Rivers and Lakes. Programme Centre, NIVA, Oslo.
15. Norwegian Institute for Water Research, 1991. Summary of The Three Year Report 1987 – 1989. Programme Centre, NIVA, Oslo.
16. Scientific papers presented at the Sixth Task Force meeting in Sweden 23 - 24 October 1990. Swedish Environmental Protection Agency, Sweden, September 1991.
17. Seventh Task Force meeting of international Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes. Galway, Ireland. September 30 - October 3 1991. Proceedings.
18. Johannessen, M., Skjelkvåle, B.L. and Jeffries, D. 1992. International cooperative Programme on Assessment and Monitoring of Rivers and Lakes. In: Conference Abstracts, Intern. Conference on Acidic Deposition, Glasgow 16-21, sept. 1992, p. 449. Kluwer Academic Press.
19. Hovind, H. 1992. Intercalibration 9206: pH, K<sub>25</sub>, HCO<sub>3</sub>, NO<sub>3</sub> + NO<sub>2</sub>, Cl, SO<sub>4</sub>, Ca, Mg, Na, K, Al and DOC. Programme Centre, NIVA, Oslo. NIVA-Report 2784-92. ISBN 82-577-2164-6.
20. Norwegian Institute for Water Research, 1992. Data Report 1990. Programme Centre, NIVA, Oslo.

21. Norwegian Institute for Water Research, 1992. Evaluation of the International Co-operative Programme on Assessment and Monitoring of Acidification in Rivers and Lakes. Programme Centre, NIVA, Oslo.
22. Hovind, H. 1993. Intercalibration 9307: pH,  $k_{25}$ ,  $\text{HCO}_3$ ,  $\text{NO}_3 + \text{NO}_2$ , Cl,  $\text{SO}_4$ , Ca, Mg, Na, K, total aluminium, reactive and non-labile aluminium, TOC and COD-Mn. Programme Centre, NIVA, Oslo. NIVA-Report 2948-93. ISBN 82-577-2370-3.
23. Raddum, G.G. 1993. Intercalibration of Invertebrate Fauna 9301. Programme Centre, NIVA, Oslo. NIVA-Report SNO 2952-93. ISBN 82-577-2376-2.
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