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ASSESSMENT OF TRENDS AND LEACHING OF NITROGEN AT ICP WATERS SITES
(EUROPE AND NORTH AMERICA)

Technical report by the Programme Centre of the International Cooperative Programme on
Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters)

Introduction

1. During the twentieth century large regions of Europe received elevated deposition of nitrogen compounds. Emissions of oxidized nitrogen (N) species from combustion of fossil fuels and emissions of reduced N compounds from agriculture increased dramatically in Europe during the 1900s to reach peak levels around 1980. Since then emissions have levelled off and then decreased slightly in most countries (Tarrason and Schaug, 2000).
2. 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 NO₃ in excess of total combined plant and microbial nutritional

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demand". By this definition N saturation is manifest by increased leaching of inorganic N (generally NO_3^-) below the rooting zone. Since NO_3^- is a strong acid anion, increased leaching of NO_3^- further acidifies soils and surface waters. Increased concentrations of inorganic N in run-off (stream-water) thus indicate N saturation of catchment ecosystems, under the assumption, of course, that there are no significant sources of N in the catchment (such as fertilizers, municipal and industrial waste-water).

3. The role of NO_3^- in the acidification of surface waters has received much-deserved attention of late, especially as the importance of SO_4^{2-} declines. Results from ICP Waters have shown that significant decreases in SO_4^{2-} concentrations at its European and North American sites can be explained by a decline in sulphur deposition due to successful emission reduction measures (Stoddard *et al.*, 1999; Skjelkvåle *et al.*, 2000). At many catchments, increasing 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 deposition or other changes in the catchments, such as point sources, agriculture, clear-cutting and forest fire. Hence, dose/response relationships are much more complicated for N than for sulphur (S). Changes in N deposition may not always be directly correlated with changes in NO_3^- leaching. The "nitrogen status" of an ecosystem, or changes in N status, seems to be an important indicator of N saturation.

4. Reports of increased NO_3^- concentrations in run-off first appeared in the 1980s. The Norwegian 1000-lake survey in 1986 revealed that NO_3^- concentrations in some areas in southern Norway had almost doubled since investigations in 1974-75 (Henriksen and Brakke, 1988). Increasing trends in NO_3^- concentrations were also revealed in the United States Environmental Protection Agency's (EPA) long-term monitoring project. Nine of the fifteen drainage lakes monitored in the Adirondack area showed increases in NO_3^- concentrations, ranging from 0.5 to 2.0 $\mu\text{eq l}^{-1} \text{ year}^{-1}$ in the period 1982-1990 (Driscoll and van Dreason, 1993). Furthermore, five of the eight streams in the Catskill area exhibited significant upward trends in NO_3^- , ranging from 1.3 to almost 3 $\mu\text{eq l}^{-1} \text{ year}^{-1}$ in the period 1983 - 1989 (Murdoch and Stoddard, 1993). These trends largely disappeared in the 1990s (Stoddard *et al.*, 1998), a phenomenon that can also be observed at many ICP Waters sites. Even if NO_3^- concentrations are relatively stable, leaching of NO_3^- from forested catchments will have the effect of removing base cations from soil and mobilizing aluminium and H^+ ions, contributing to catchment acidification.

5. The aim of this report is to provide an updated account (since the evaluation of Lükewille *et al.*, 1997) on the status of NO_3^- in surface waters at ICP Waters sites in Europe and North America, to give a more detailed assessment of trends in N in surface waters in subregions as a follow-up to the ICP Waters 12-year report (Skjelkvåle *et al.*, 2000), and relate these results to trends in N deposition.

I. METHODS AND SITE SELECTION

6. The surface water data used in this report are from ICP Waters, and the deposition data are from the Cooperative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe (EMEP).

7. The ICP Waters data are from 204 sites in Europe (Austria, Belarus, Czech Republic, Estonia, Finland, France, Germany, Hungary, Italy, Ireland, Latvia, Norway, Poland, Romania, Russian Federation, Spain, Sweden, Switzerland and United Kingdom) and North America (Canada, United States). 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/autumn base-flow and spring run-off samples.

8. NO₃ concentrations reported to ICP Waters are measured using a variety of analytical methods. Annual cross-comparisons of methods conducted by ICP Waters (Hovind, 2000), using audit samples of known concentration, show excellent agreement among all of the participating national laboratories. Data used for assessing the N status are from 1998 and 1999; trends data are from 1989-1998.

9. The data in the ICP Waters database illustrate 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 Waters Programme Centre, there are many sites that do not have sufficient data for all types of analyses. The text of the figures presented always shows the number of sites that were used in each particular analysis.

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

11. All EMEP deposition data used here were provided by the Meteorological Synthesizing Centre - West (MSC-W) and the Chemical Coordination Center (CCC). EMEP deposition data cover only European sites. The data represent (i) deposition measured at EMEP stations located in the same region as the stream-water sites (Barrett *et al.*, 2000a); and (ii) deposition modelled on a country basis (Tarrason and Schaug, 2000). The modelled deposition data were calculated for 50 km x 50 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, oxidized N and reduced N compounds.

II. RESULTS

A. Nitrate in run-off

12. About one third of the ICP Waters sites (located primarily in the Nordic countries, Ireland, Canada and the United States) have mean annual concentrations of NO₃⁻ below 5 µeq l⁻¹. More than half the sites have NO₃ concentrations above 10 µeq l⁻¹. Due to the episodic pattern of NO₃ at

many sites, the significance of 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 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 NO_3 concentrations.

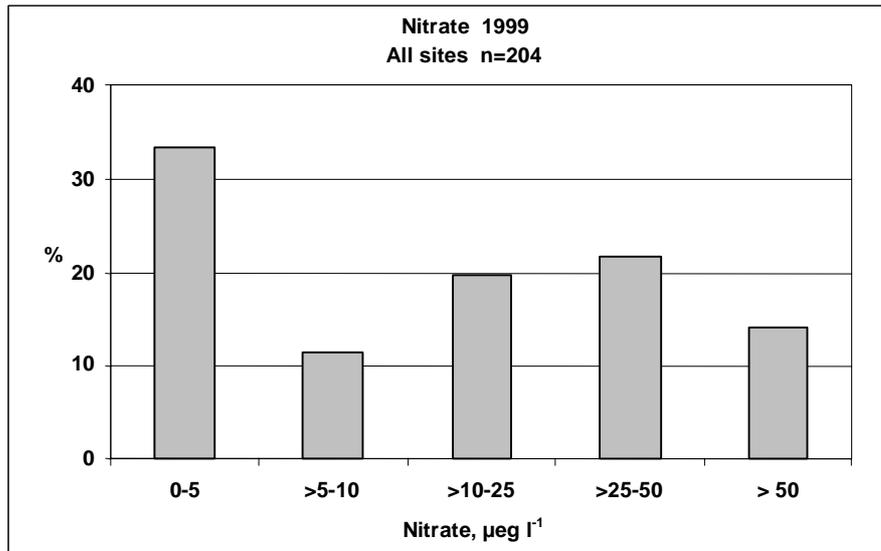


Figure I. Frequency distribution of mean annual NO_3 concentrations for the 204 ICP Waters sites with data from 1999

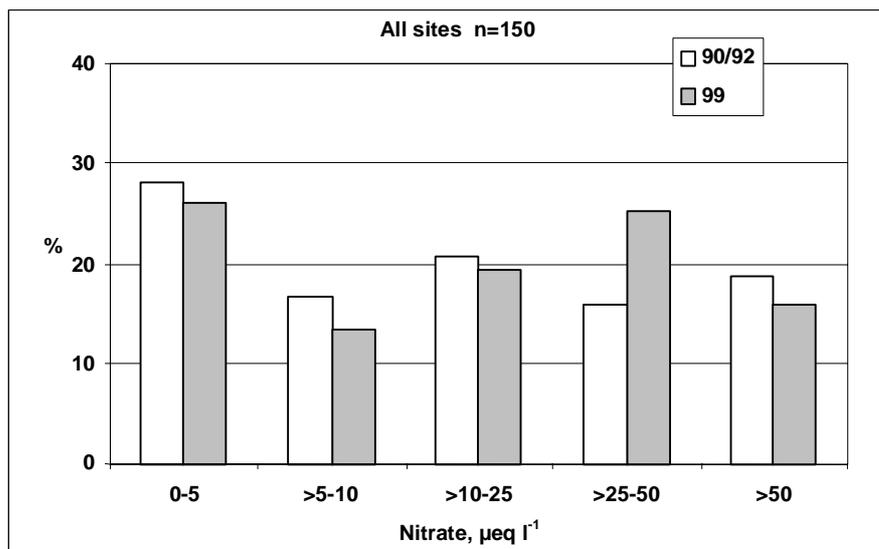


Figure II. Frequency distribution of mean annual NO_3^- concentrations at 150 ICP Waters sites with data from 1999 and the period 1990-1992

13. 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 NO_3 concentrations.

14. The frequency distribution of NO_3 concentrations is approximately the same for 150 sites with data from 1990/92 and 204 sites with 1999 data (figs. I and II). There has been some increase in sites with NO_3 in the range $25\text{-}50 \mu\text{eq l}^{-1}$, and decreases in all the other groups. A division of data between Europe and the United States reveals that most of the increase in NO_3 from 1990/92 to 1999 occurred in the United States sites, while the European sites show a tendency towards lower NO_3 .

B. Relative importance of nitrate and sulphate

15. The importance of NO_3 in acidification relative to SO_4 can be quantified by the ratio N/S defined as the concentration of NO_3^- divided by the sum of non-marine SO_4 (denoted by asterisk) and 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 sites in 1999, but only 5% have N/S values > 0.5 . At these sites NO_3 makes a greater contribution to acidification than SO_4 . These sites are situated in Germany and Italy. This means that for the majority of ICP Waters sites, on an annual mean basis, SO_4 is still the dominant acidifying ion.

16. Comparison of these results 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 (fig. III). NO_3 plays a larger role in acidification in 1999 than in 1990/92. This is most certainly due to the decline in SO_4 concentration over this period, rather than to an increase in NO_3 concentrations.

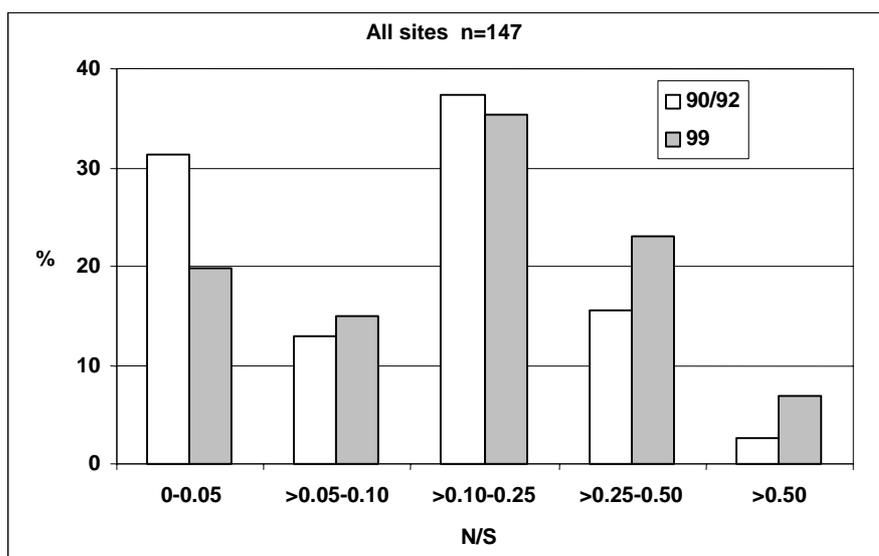


Figure III. Relative importance of NO_3^- in acidification at 147 ICP Waters sites in 1999 compared to the period 1990-92 for the same sites

17. A division of data between the United States and Europe shows that for the United States 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 in N/S from 0.16 to 0.20 between 1990/92 and 1999.

C. Nitrogen deposition and inorganic nitrogen in run-off

18. 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 that very little NO_3^- leaching occurs at N deposition below $9\text{--}10\text{ kg N ha}^{-1}\text{ year}^{-1}$, leaching can occur at intermediate deposition between 9 and $25\text{ kg N ha}^{-1}\text{ year}^{-1}$, and significant leaching occurs at almost all sites receiving deposition greater than $25\text{ kg N ha}^{-1}\text{ year}^{-1}$. These thresholds are based on data from monitoring plots across a gradient of current N deposition in Europe and from several experimental sites.

19. Input/output data for 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 (fig. IV). Deposition fluxes are values for the $50\text{ km} \times 50\text{ km}$ EMEP grids in which the respective ICP Waters sites are situated. The North American sites are not included in this analysis.

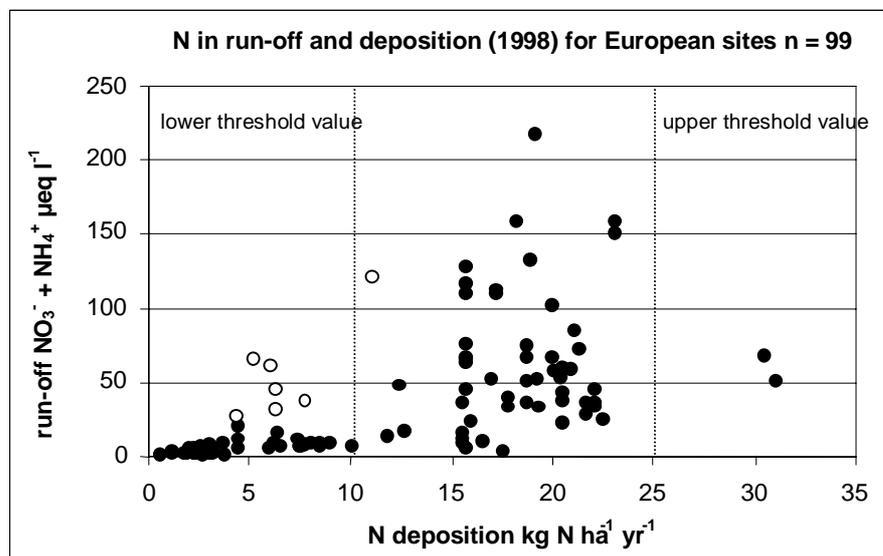


Figure IV. Nitrogen ($\text{NO}_3^- + \text{NH}_4^+$) concentration in run-off and total N deposition fluxes (wet + dry) for 99 European ICP Waters sites in 1999. The deposition data are 1998 values for the EMEP $50\text{ km} \times 50\text{ km}$ grids in which the respective sites are situated (data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

20. The results support the general picture that increased N leakage occurs in catchments receiving N deposition above some threshold. Some catchments have a pronounced NO_3 leakage at a total N deposition as low as $5 \text{ kg N ha}^{-1} \text{ year}^{-1}$. In the case of the Latvian, Estonian and the Belarusian sites (marked with open circles in the figure), influence of agriculture (e.g. pasture land) on water quality cannot be excluded. For the North American sites, Stoddard (1994) observed substantial N leakage at wet deposition as low as $2.5 \text{ kg N ha}^{-1} \text{ year}^{-1}$, corresponding to a wet + dry deposition of approximately $5 \text{ kg N ha}^{-1} \text{ year}^{-1}$. The possible difference in threshold values between the European and North American continents is an interesting phenomenon for which an explanation is not yet available.

21. 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 the deposition and its duration; N concentrations in run-off (instead of N fluxes) were compared with 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 presented in figure IV are in reasonably good agreement with the data from Dise and Wright (1995) and Gundersen *et al.* (1998).

D. Classification of nitrogen saturation

22. Nitrogen is the growth-limiting nutrient in many terrestrial ecosystems. Long-term atmospheric N deposition may lead to a situation where the available inorganic N exceeds the total amounts required for the growth of plants and soil micro-organisms. N saturation is defined here as persistent losses of NO_3^- and/or NH_4^+ in stream-flow or groundwater discharge, which may be accompanied by increases in N mineralization and nitrification in soil (Stoddard, 1994). Stoddard (1994) has elaborated the term "persistent" by addressing short-term temporal variations in 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.

23. 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 characterize the majority of available ICP Waters sites; 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).

24. The distribution of N saturation stages in 1998 among all ICP Waters sites is shown in figure V. About half of the 108 sites exhibited a high degree of N saturation (stage 2 or 3) in 1998. Such sites are found predominantly in central Europe, although two of the Swedish sites are also classed as stage 2. Most of the sites in the Scandinavian countries are classed stage 0 or 1. Thirty-six of these 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.

E. Relationship between N deposition and N saturation stage

25. There is a clear connection between N deposition and stage classification at European ICP Waters sites (fig. VI). N deposition levels are all below $10 \text{ kg N ha}^{-1} \text{ year}^{-1}$ at stages 0 and 1, while at stage 3 no sites had deposition below $10 \text{ kg N ha}^{-1} \text{ year}^{-1}$. This is the same general picture as found in the evaluation of 1990/92 data (Traaen and Stoddard, 1995).

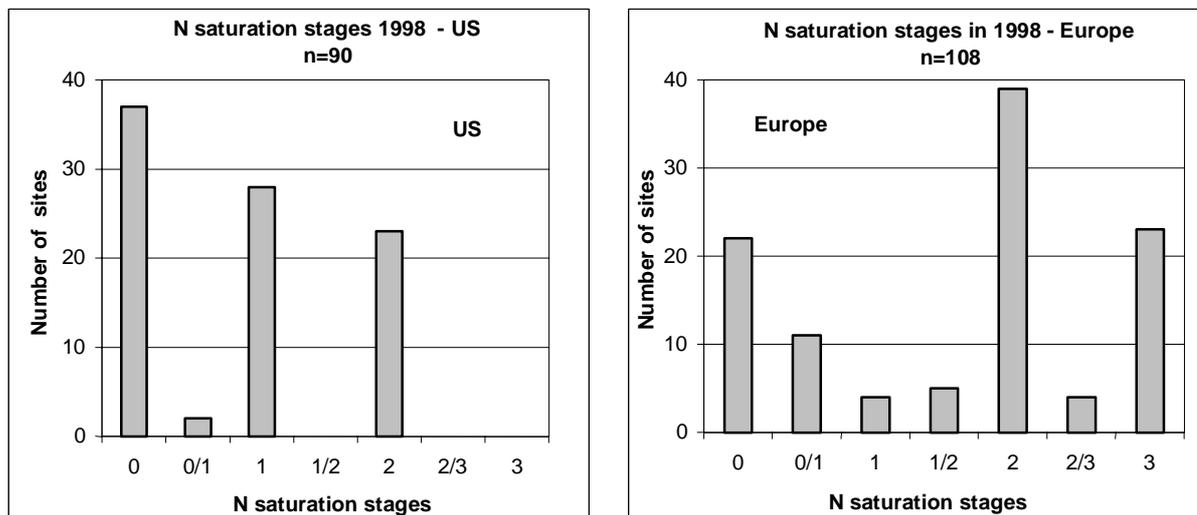


Figure V. Number of ICP Waters sites in various stages of N saturation in 1998 (stages modified from Stoddard, 1994; criteria for assigning sites to stages explained in Traaen and Stoddard, 1995) for the United States (US) and European ICP Waters sites

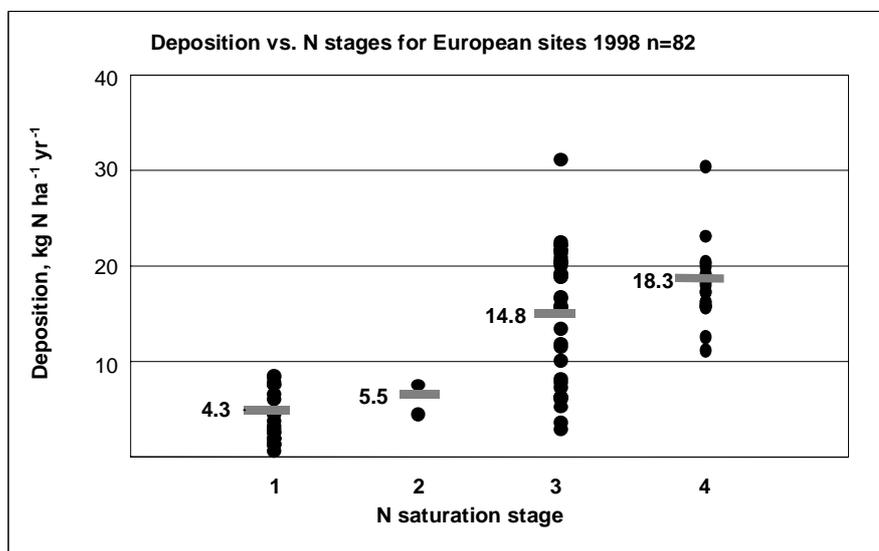


Figure VI. Relationship between N deposition (wet + dry, estimated from EMEP grid data) and stages of N saturation in 1998 for 82 European ICP Waters sites (North America not included). Average values are indicated by horizontal bars, and the numbers indicate average deposition value in $\text{kg N ha}^{-1}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

F. Trends in nitrate concentrations in surface waters

26. Much of the current concern about N saturation has resulted from evidence of increasing 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 United States have suggested that these trends have not continued into the 1990s (Mitchell *et al.*, 1996).

27. The ICP Waters data have provided the basis for the evaluation of NO_3^- trends in surface waters in the 1990s over larger regions (Skjelkvåle *et al.*, 2000). Sites with data of sufficient frequency (minimum three samples per year) and duration (at least 7 years in the 10-year period 1989-1998) were used (98 sites). The significance of trends over time was tested by the Seasonal Kendall Tau (SKT) statistic (details of statistical methods are given in Skjelkvåle *et al.*, 2000; and Stoddard *et al.*, 1999).

28. Of the 96 ICP Waters sites with sufficient data for trend analysis, 8 sites showed significant upward trends in the period from 1989 to 1999, 11 showed significant downward trends, while the remaining 77 sites showed no significant trends. It is, however, interesting to note that 5 of the 6 sites in the Czech Republic show decreasing trends in NO_3 (table 1).

29. The sites were grouped by chemical criteria and catchment characteristics (Skjelkvåle *et al.*, 2000). The only group that exhibits a significant change is the one with low acid neutralizing capacity (ANC), where the data indicate a significant increase in NO_3 concentrations during the 1990s. Neither the high NO_3 nor the low NO_3 groups of sites exhibited significant trends in NO_3 concentrations (both groups showed considerable trend variability).

30. Previous ICP Waters results (Stoddard *et al.*, 1999; Lükewille *et al.*, 1997) showed that in the 1980s for ICP Waters as a whole, more sites exhibited upward trends in NO_3 concentrations (33%) than downward trends (8%). The data for the 1990s now indicate that the trends showing increases have ceased to do so.

Table 1. Number of sites with significant increasing or decreasing ($p < 0.05$) NO_3 trends from 1989 to 1998

Country	Total number of sites	Number of sites with:	
		Upward trends	Downward trends
Canada	14	4	
Czech republic	6		5
Finland	22		
Germany	6		
Italy	4		
Norway	9	2	1
Poland	2		
Sweden	10		2
United Kingdom	6	2	
United States	17		1

Total	96	8	10
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G. Trends in deposition

31. Deposition of both oxidized and reduced N in Europe increased during the 1900s and levelled out in the 1970s and 1980s. The values somewhat decreased during the late 1990s in central and northern Europe, in response to modest declines in emissions (Tarrason and Schaug, 2000). Trend analyses of 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.

32. For a selection of EMEP sites situated in the same region as the ICP Waters sites (table 2), trends in NO₃+NH₄ deposition were calculated 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 amount of precipitation. This “noise” in the record thus means that changes 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 2. Linear regression of annual NO₃+NH₄ deposition for the period 1989-1998 at 16 EMEP sites in 9 countries in Europe. Significant trends are indicated in bold. NS: not significant. Units: kg N ha⁻¹ yr⁻¹

Country	EMEP-site	slope	r ²	p
Czech Republic	CZ01 Svartouch	+0.05	0.04	NS
Germany	DE02 Waldhof	-0.10	0.05	NS
	DE03 Schauinland	-0.42	0.28	NS
	DE05 Brotjackriegel	-0.12	0.07	NS
Finland	FI04 Ahtari	-0.24	0.66	<0.05
	FI09 Uto	-0.08	0.90	<0.05
United Kingdom	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	-0.43	0.70	<0.05
	SE12 Aspvreten	+0.23	0.64	<0.05
Switzerland	CH01 Jungfrauoch	-0.23	0.42	NS

III. SUMMARY AND CONCLUSIONS

33. The ICP Waters database currently holds data from 204 sites in Europe and North America. These data were evaluated with respect to status and trends in nitrate (NO_3) concentrations.

34. Status:

(a) About one third of the ICP sites have mean annual concentrations of $\text{NO}_3 < 5 \mu\text{eq l}^{-1}$. More than half the sites have NO_3 concentrations $> 10 \mu\text{eq l}^{-1}$. In total, the results suggest that a large fraction of the ICP Waters sites have NO_3 concentrations above those expected from catchments with no impact from nitrogen deposition;

(b) In about 5% of the sites, NO_3 makes a greater contribution to acidification than sulphate (SO_4). This means that for the majority of ICP Waters sites, on an annual mean basis, SO_4 is still the dominant acidifying ion;

(c) SO_4 is the most important acidifying anion, but NO_3 constitutes more than 10% of the non-marine acid anions at 50% of the ICP Waters sites;

(d) The ICP Waters data support the general picture that increased N leakage occurs in catchments receiving N deposition above a threshold of $5\text{-}10 \text{ kg N ha}^{-1} \text{ yr}^{-1}$;

(e) About half of the 108 European ICP Waters sites exhibited 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 the eastern United States 26% of the sites show a high degree of N saturation (stage 2) and no sites are classified in stage 3;

(f) There is a clear connection between N deposition 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 stage 3 no sites had deposition below $10 \text{ kg N ha}^{-1} \text{ year}^{-1}$.

35. Trend:

(a) The frequency distribution of NO_3 concentrations is approximately the same for 150 sites with data for the period 1990/1992 and for 1999. A division of data between the United States and Europe reveals that most of the increase in NO_3 from 1990/1992 to 1999 occurred in the United States sites, while in the European sites there is a tendency towards a decrease in NO_3 ;

(b) NO_3 played a larger role in acidification in 1999 than in 1990/1992. This is mostly due to the decline in SO_4 concentration over this period, rather than an increase in NO_3 concentrations;

(c) Of the 70 sites included in 1998, 36 were also classified in 1990/1992. Of these 24 had no change in N saturation stage, while 8 went down and 4 went up;

(d) The majority of the ICP Waters sites with sufficient data for trend analysis in 1998 showed no significant trends in NO_3 for the time period 1989-1998. Only 8 sites showed a significant increase and 11 showed a significant decrease. Previous results from ICP Waters (Stoddard *et al.*, 1999; Lükewille *et al.*, 1997) showed that in the 1980s, for ICP Waters as a whole, more sites exhibited upward trends in NO_3 concentrations (33%) than downward trends (8%). The data for the 1990s now indicate that this increasing trend has stopped;

(e) Deposition of both oxidized and reduced N in Europe increased during the 1900s and levelled out in the 1970s and 1980s. Trend analyses of 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;

(f) Of the 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 changes in N deposition, therefore, must be relatively large before they become statistically significant. This probably explains why there are no significant changes in N deposition trends during the 1990s at many of the EMEP sites;

(g) Overall, the data indicate that progression to increased N saturation is a slow process with a timescale of decades.

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