# CONVENTION ON LONG-RANGE TRANSBOUNDARY AIR POLLUTION

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

# The Nine Year Report:
## Acidification of Surface Water in Europe and North America - Long-term Developments (1980s and 1990s)

Prepared by the Programme Centre, Norwegian Institute for Water Research

**NIVA**

# Norwegian Institute for Water Research

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Abstract

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. During the last ten years international emission reduction measures in Europe and North America have resulted in a decrease in atmospheric sulphur deposition of up to 50%. Nitrogen deposition has stayed almost constant. Trends in surface water chemistry at the ICP Waters sites were calculated by applying the Nonparametric Seasonal Kendall Test and, on a regional scale, by Trend Meta-Analyses. Empirical relationships between nitrogen deposition and stages of nitrogen saturation were used to assess the importance of nitrogen leaching at the sites. To show effects of acidification on aquatic fauna (geographic extent and long-term trends) presence/absence studies, acidification indexes and correlation analyses were used. Trends in water chemistry indicate that sulphate 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. This is partly also reflected in a recovery of the invertebrate fauna at many sites. Decreasing sulphate concentrations emphasize the importance of nitrate as the second important acidifying anion. Besides nitrogen deposition, the overall nitrogen status of ecosystems, changes in climate or climate extremes and hydrology can have strong influences on leaching of excess nitrate (and ammonium) from a watershed.

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# 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 1995. The Executive Body has also accepted Norway's offer to provide facilities for the Programme Centre which has been established at the Norwegian Institute for Water Research, NIVA. The ICP-Waters programme has been lead by Berit Kvaeven, Norwegian State Pollution Control Authority.

The work plan of the Programme includes in-depth evaluations every third year. This nine year report summarises the results achieved so far, along the lines of the programme objectives. Focus has been on trends and regional trends in surface water chemistry, assessment of nitrogen leaching and effects of acidification on biota. Project leader for the nine year report has been Anke Lükewille, NIVA. Dean Jeffries, Canada, and John Stoddard, USA, are responsible for the trends work, Tor Traaen, NIVA, for the nitrogen evaluations and Gunnar Raddum, University of Bergen, for the biological section.

The programme was evaluated by external experts in 1992. As reported to Working Group on Effects "the Programme had been sucsessfull in establishing the metodologies and monitoring network". However, it is also concluded that "only time can create the necessary data base". This nine year report brings the ICP-Waters Programme a long step forward in that respect.

*Oslo, 17.3.1997*

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# Acknowledgements

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# Summary

The International Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) is designed to assess, on a regional basis, the degree and geographical extent of acidification of surface waters. The data collected should provide information on dose/response relationships under different conditions and correlate changes in acidic deposition with the physical, chemical and biological status of lakes and streams.

During the last ten years international emission reduction measures in Europe and North America have resulted in a decrease in atmospheric sulphur deposition of up to 50%. Nitrogen deposition has stayed almost constant. To relate these developments to changes in surface water chemistry and biology, the ICP Waters data base was used to:

- Compile a subset of selected and quality controlled water chemistry and site description data ( > 120 sites) that can be used, e.g., for statistical analyses.

- Calculate trends in surface water chemistry separately on data from the 1980s and 1990s for the single ICP Waters sites and for geographic regions (clustering based on similarities in water chemistry).

  *(Methods: Nonparametric Seasonal Kendall Test; Regional Trend Meta-Analyses)*

- Assess the importance of nitrogen leaching at ICP Waters sites.

  *(Methods: empirical relationships between nitrogen deposition, stages of nitrogen saturation and inorganic nitrogen and sulphate in runoff)*

- Show effects of acidification on aquatic fauna (geographic extent, dose/response relationships, long-term trends)

  *(Methods: presence/absence studies, acidification index, correlation analysis, detrended canonical correspondence analysis)*

## Trends in Surface Water Chemistry

Regular quarterly sampling (spring, summer, autumn, winter) is the minimum criteria for data sets chosen for statistical analyses. Results of more than 40 ICP Waters sites show that curvilinear trends are common, particularly for sulphate ($SO_4^{2-}$), acid alkalinity (ALK) and strong base cations (SBC = $\Sigma$ Ca, Mg, K, Na). 1990 can be seen as the inflection point. Important results of the regional trend analyses are:

⇨ $SO_4^{2-}$ concentrations are decreasing at almost all sites, and in almost all cases the decreases in the 1990s are larger than in the 1980s.

⇨ In the Nordic Countries (Finland, Sweden, Norway) ALK decreased in the 1980s (acidification), but increased in the 1990s (recovery). At many European sites (Italy, Germany, Netherlands, Denmark) ALK also increased in the 1980s, but the rate accelerated in the 1990s. The remaining regions (Adirondacks and Quebec, midwestern North America, U.K.) show either no recovery or further acidification.

⇨ Regions with declining $SO_4^{2-}$ that fail to show recovery in ALK in the 1990s (Adirondacks/Quebec, Midwestern North America) are characterized by strongly declining SBC concentrations. SBC concentrations are no longer declining in the Nordic Countries (regional recovery in ALK in the 1990s).

⇨ All of the hydrogen ion ($H^+$) trends detectable at a regional level are consistent with the ALK trends observed within each region.

⇨ The 1980s were characterized by increases in nitrate ($NO_3^-$) in almost all regions. These increases have disappeared in the 1990s. It seems likely that regional-scale phenomena other than nitrogen deposition are responsible for the development in the 1990s (e.g., changes in climate or climate extremes).

## Nitrogen Leaching from ICP Waters Sites

Concerning the leaching of nitrogen important observations for the ICP Waters sites are:

⇨ More than 50% of all sites have yearly average $NO_3^-$ concentrations > 10 μeq l[-1]. $NO_3^-$ values > 50 μeq l[-1] are found at 27% of the sites. All Dutch sites have ammonium values > 100 μeq l[-1].

⇨ $SO_4^{2-}$ is the most important acidifying anion, but $NO_3^-$ constitutes more than 10% of the non-marine acid anions at 63% of the ICP Waters sites. All Canadian, Finnish, Swedish and Russian sites are exceptions.

⇨ There is evidence that inorganic N leaching below the empirically found deposition limit of 10 kg N ha[-1] yr[-1] occurs especially at sites with very high annual precipitation (Norway, Ireland, Italy, U.K.).

⇨ More than 50% of the analyzed ICP Waters sites show a high degree of nitrogen saturation (stages 2 or 3 according to Stoddard's classification system). There is a clear coherence between high N saturation stages and high N deposition loads.

Compared with sulphur, 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.

A watershed's "nitrogen status" has a significant influence on the timing and extent of response to changes in N deposition. Even at high deposition, changes in N status may occur in certain compartments of the ecosystem (e.g., in current needles, forest floor). C/N ratios in such compartments seem to be reliable indicators of N saturation. Detailed information on this is available from long-term (manipulation) experiments (e.g., NITREX) and intensively studied sites within the ICP Integrated Monitoring Programme. In combination with regional monitoring data (ICP Waters, ICP Forests) results from these studies can be useful tools to link process level data with regional-scale questions, and to reassess critical loads of nitrogen.

## Effects of Acidification on Aquatic Fauna

Water acidification (including acidic episodes) changes the composition of invertebrate assemblages in surface waters. Both long-term monitoring of water chemistry and biology is necessary to assess dose/reponse relationships. Important findings within the ICP Waters Programme are:

⇨ From different invertebrate assemblages, pH can be predicted with an accuracy of 0.3 pH units.

⇨ In areas with originally high pH (6.0 - 8.0) and high calcium concentrations (e.g., southern Sweden, Germany) a critical ALK limit of 50 $\mu$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), ALK values should be $\geq 20$ $\mu$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 German sites can be observed. This is confirmed by correlation analyses between time and acidification index applied to Norwegian long-term data series (periods chosen: 1981 - 1988 and 1989 - 1994).

⇨ Detrended canonical correspondence analysis (Norwegian data sets) shows a high correlation of invertebrate assemblages with pH and total aluminium (Al), and also a significant correlation with calcium. Trend analyses indicate considerable improvements from 1991 to 1995 (increase in pH, decrease in total Al).

# Conclusions

*Trend analyses (1980s and 1990s) indicate that the reduction in sulphur deposition has lead to an improvement in water chemistry and partly also to a recovery of the invertebrate fauna at many ICP Waters sites. Decreasing sulphate concentrations emphasize the importance of nitrate as the second important acidifying anion. Besides nitrogen deposition, the overall nitrogen status of ecosystems, changes in climate or climate extremes and hydrology can have strong influences on leaching of excess nitrate (and ammonium) from a watershed.*

# 1. Introduction

## 1.1 Programme Reason and Background

Over the past 25 years acid atmospheric deposition, "acid rain", has received considerable attention as an international environmental problem in Europe and North America. Polluted air masses containing sulphur and nitrogen compounds travel long distances across national boundaries and are affecting surface waters, groundwater and forest soils in other countries. Rethinking of air pollution control strategies was necessary, including long-term monitoring of effected receptors such as rivers and lakes.

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 five ICPs (Waters, Forests, Materials, Crops, Integrated Monitoring). The LRTAP convention went into effect in 1983 and was the first step to enforce emission reduction measures in the international sphere.

Canada was appointed as lead country for the first phase of the ICP Waters. The delegation from the Union of the Soviet Socialist Republics offered to support activities in the leading Programme. The Executive Body also accepted Norway's offer to provide facilities for the Programme Centre.

The monitoring programme was designed to assess, on a regional basis, the degree and geographical extent of acidification of surface waters. The data collected should provide information on dose/response relationships under different conditions and correlate changes in acidic deposition with the physical, chemical and biological status of lakes and streams (EB.AIR/W6.1./R21).

The Programme Task Force has the mandate to plan and supervise activities undertaken within this Programme. At its first meeting, held in Grafenau (Germany) on 27 April 1986, the Programme Task Force discussed the scope, objectives and general organisation of the programme and formulated a monitoring manual to be recommended to the Working Group on Effects.

As reported to the fourth session of the Executive Body, the International Co-operative Programme is based on existing programmes in participating countries, implemented by voluntary contributions. The operational part of the programme using standardised methodologies started in1987 and has proceeded over a nine year period.

At its second meeting in Oslo (14 October 1986) the Programme Task Force adopted the Programme Manual compiled by the Programme Centre in Norway and based on a draft outline prepared by Canada. After finalising the Programme manual, Canada entrusted Norway to take the role as lead country in the subsequent implementation phase of the programme. This was confirmed by the Working Group on Effects at its sixth session and reported to the Executive Body on its fifth session (EB.AIR/16 - Annex II) in December 1987.

## 1.2 Programme Aims and Objectives

The ICP Waters Programme has been designed to establish the degree and geographic extent of surface water acidification in UN ECE countries. By means of long-term monitoring, trends and variations in aquatic chemistry and biota are analysed. The results are related to changes in (acidic) atmospheric deposition. Another aim is to evaluate dose/response relationships between water chemistry and biota.

The Programme objective is to establish an international network of surface water monitoring sites and promote international harmonisation of monitoring practices by:

- Compiling a Programme Manual on methods and operations.
  Sample collection at the ICP Waters sites and analytical methodologies used by the paticipating countries need to be standardized. The manual is also an overall guide to activities of, and priorities set, by the Programme Centre.

- Conducting inter-laboratory quality assurance tests.
  The bias between analyses carried out by the individual participants of the Programme have to be clearly identified and controlled. Bias may arise through the use of different analytical methods, errors in the laboratory calibration solutions, or through inadequate within-laboratory control.

- Compiling a centralised database with data quality control and assessment capabilities.
  The purpose of the ICP Waters' database management is to assure accuracy of data, to accumulate and archive data, and to retrieve and summarise the data in response to user requirements. The management may include facilities for extensive data manipulation for interpretative purposes.

Another objective of the Programme is to develop and/or recommend chemical and biological methods for monitoring purposes. The ICP Waters also conducts workshops on topics of central interest to the Programme Task Force and the aquatic effects research community.

Reporting of the ICP's findings cover the following aspects:

- Geographic extent of acidification using terrain sensitivity and current information sources.

- Long-term trends in water chemistry by using sites with data of more than 5 years.

- Dose/response relationships by:

    a.  determining changes in systems with similar buffering capacity and hydrology but different acid inputs;

    b.  comparing chemical and biological responses of systems with different buffering capacities but similar deposition levels.

These topics are addressed by 3-year summary reports: 1987-89 (WATHNE et al. 1991), 1989-1992 (SKJELKVÅLE et al. 1994), 1993-1995 (this report).

## 1.3  Programme Status

21 Countries are participating in the ICP Waters Programme (**Table 1**). Data of 124 sites have been used in this report (**Figure 1** and **Figure 2**).

**Table 1.** Programme status as of November 1996.

| Country | Participating in Task Force meetings | Data reported until | Participating in last intercalibration |
|---|---|---|---|
| Austria | √ | 1995 | √ |
| Belgium | √ | --- | --- |
| Bulgaria | √ | --- | --- |
| Canada | √ | 1994 | √ |
| Czech Republic | √ | 1995 | √ |
| Denmark | √ | 1995 | --- |
| Estonia | √ | --- | √ |
| Finland | √ | 1995 | √ |
| France | √ | 1995 | √ |
| Germany | √ | 1995 | √ |
| Hungary | √ | 1995 | --- |
| Italy | √ | 1995 | √ |
| Ireland | √ | 1995 | √ |
| Latvia | √ | 1995 | √ |
| Lithuania | √ | --- | √ |
| The Netherlands | √ | 1995 | --- |
| Norway | √ | 1995 | √ |
| Poland | √ | 1995 | √ |
| Romania | √ | --- | √ |
| Russia | √ | 1994 | √ |
| Spain | √ | --- | √ |
| Sweden | √ | 1995 | √ |
| Switzerland | √ | --- | √ |
| United Kingdom | √ | 1995 | √ |
| USA | √ | 1994 | √ |
| **Sum** | **25** | **18** | **20** |

For about half of the ICP Waters sites both chemical and biological data have been reported (**Annex A**).
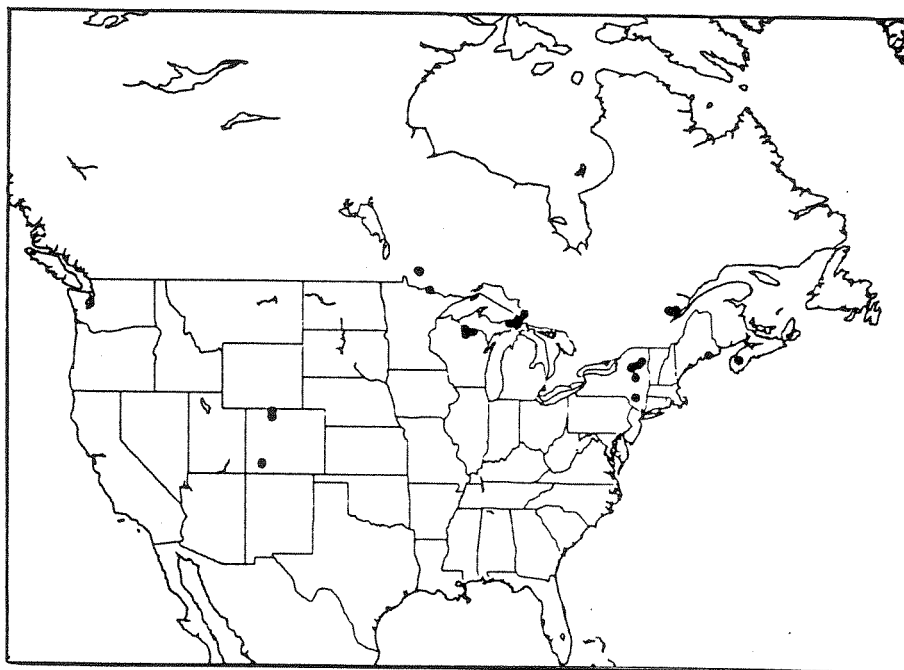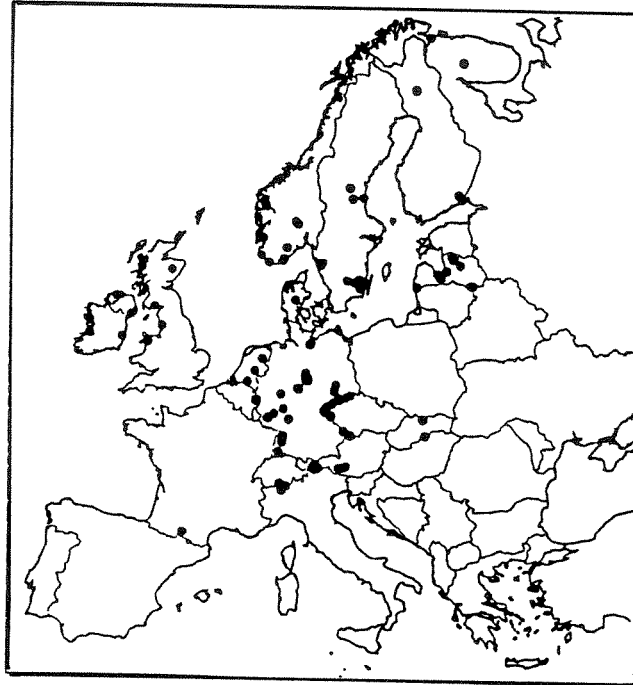




**Figure 1.** and **Figure 2.** Location of monitoring sites in Europe and North America.

# 1.4 Summary of Programme Findings 1989 - 1992

## 1.4.1 Trends in Water Chemistry

Significantly decreasing sulphate concentrations were a major feature of the ICP Waters sites. A decline in $Ca^{2+}$ was found at many European locations. Several surface waters were characterised by upwards trends in nitrate concentrations, and constant or increasing nitrogen deposition seemed to be the driving force. A combined decrease in $Ca^{2+}$ and increase in $NO_3^-$ may account for the lack of recovery from acidification at sites with decreasing $SO_4^{2-}$.

## 1.4.2 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 singles sites. Deposition from the nearest existing monitoring station, up to 160 km away, was paired with the surface waters, respectively. The development of sulphur 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 sulphate deposition at a certain site might have been influenced by earlier changes in deposition.

## 1.4.3 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 strongest 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 $Ca^{2+}$ 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. In Central Europe, the fauna was much more sensitive to acidification. It was adapted to waters with high ionic strength, rich in $Ca^{2+}$, with pH values of 6.0 - 8.0 and an ANC of 50 - 200 µeq/L

## 1.4.4 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.

## 1.4.5 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., sulphur *and* nitrogen) which together effect sensitive organisms

in freshwater ecosystems. Thus, critical load assessments should focus on both deposition of sulphur and nitrogen compounds.

## 1.5 Aim of the 9 Year Report

The aim of the 9 Year Report is to provide an overall synthesis and assessment of information on water chemistry and biology accumulated in the ICP Waters Programme within the last 3-4 years.

This report covers three topics:

1.  **Trends in water chemistry** at the single ICP sites and on regional scales (chapter 3);
2.  Update of the **assessment of nitrogen leaching** at ICP Waters sites (chapter 4);
3.  **Dose/response relationships** between water quality and aquatic fauna and their long-term trends (chapter 5).

# 2. Quality Control and New Data Base

## 2.1 General Aspects of Quality Control and Methods Used

Standardisation of sample collection and analytical methodologies are addressed in the latest version of the ICP Waters Programme Manual (1996). Aspects of site selection, water chemistry/biological monitoring and data handling are also described in detail within the manual.

Three levels of quality control of water chemistry data can be distinguished: in-laboratory controls in the single countries, between-laboratory controls (intercalibrations; see 2.2) and quality control of data reported to the National Focal Points and to the Programme Centre at NIVA. The last step does not focus on the physical-chemical analysis of single parameters in the laboratory, but is a more technical procedure including:

- looking for outliers,
- looking for continuity in time series,
- calculating ionic balances (see 2.4.1),
- comparing measured and calculated conductivities (see 2.4.2).

Methods used for trend analyses (chapter 3), assessment of nitrogen leaching (chapter 4) and dose/response relationships (chapter 5) are briefly described in the respective chapters.

## 2.2 Chemical and Intercalibration Exercise 1996

The latest between-laboratory quality control of chemical data was carried out in 1996 at NIVA (HOVIND 1996). 36 laboratories from 20 different countries participated. Two sample sets, one for the major ions and one for organic matter and aluminium fractions, were used. According to the general target accuracy of 20%, 70% of the results were acceptable. The best results were obtained for conductivity, nitrate, calcium, sodium and dissolved organic carbon. Rather poor comparability was observed for pH, alkalinity, chloride and aluminium species. To improve intercalibration results for these parameters, further harmonisation of the analytical methodologies is necessary.

## 2.3 New Site Description Data

By March 1996 most countries had reported a new, shortened list of site information data (description of catchment and biological sampling sites) to the Programme Centre at NIVA (**Annex A**). This updated information could be used when sites for trend analyses were chosen (see also chapter 3.1).

## 2.4 Quality Control of Reported Data

### 2.4.1 Ion Balances

Based on the principle of electroneutrality, ion balances are a useful tool for checking the accuracy of chemical analyses. An important criterium, though, is that concentrations of the most important cations and anions have been measured. Depending on the data available, two different procedures to calculate ion balances are used by the Programme Centre. Concentrations $\{\ \}$ are given in $\mu eq\ l^{-1}$ ($\cong$ mol charge $(mmol_c)\ m^{-3}$).

**I.**

| | | |
|---|---|---|
| **Sum anions** | **: SAN =** | $\{Cl^-\} + \{NO_3^-\} + \{SO_4^{2-}\} + \{ALK\}$ |
| **Sum cations** | **: SCAT =** | $\{Ca^{2+}\} + \{Mg^{2+}\} + \{Na^+\} + \{K^+\} +$ |
| | | $\{H^+\}$ |
| **Difference cations - anions** | **: DIFF =** | **SCAT - SAN** |
| **Difference in percent** | **: D-PRO =** | **DIFF in % of SCAT (DIFF*100/SCAT)** |

If measurements of Al, $NH_4^+$ and TOC are available, an extended set of equations is used:

**II.**

| | | |
|---|---|---|
| **Sum of anions** | **: SAN2 =** | **SAN + OAN⁻** |
| **Sum of cations** | **: SCAT2 =** | **SCAT + $\{LAL^{(*)}\}$ + $\{NH_4\}$** |
| **Difference cations - anions** | **: DIFF2 =** | **SCAT2 - SAN2** |
| **Difference in percent** | **: D-PRO2 =** | **DIFF2 in % of SCAT2** |
| | | **(DIFF2 * 100/SCAT2)** |

[*] "labile" aluminium $= Al^{3+}, Al(OH)^{2+}, Al(OH)_2^+$

OAN⁻ (organic anions in $\mu eq\ l^{-1}$) are calculated by using TOC concentrations, based on the following empirical equation derived from Norwegian water chemistry data:

$$OAN^- = 4.7 - 6.87 * exp(-0.322 * TOC)$$

The difference between sum cations (SCAT or SCAT2) and sum anions (SAN or SAN2) should be less than 10%.

## 2.4.2 Measured and Calculated Conductivity

Another means of quality control is the comparison of measured (CM) and calculated (CE) conductivity. CE can be obtained by multiplying the individual ion concentration or activity ($c_i$) with the respective molar, or in case of multivalent ions equivalent, ionic conductance ($\lambda_i$):

$$CE = c_i \, \lambda_i \quad (\text{unit: S cm}^2 \text{ eq}^{-1})$$

The difference in percent (CD) is given by the ratio:

$$CD = 100* \, |(CE\text{-}CM)| \, / \, CM$$

Values for $\lambda_i$ at standard conditions (20 or 25 °C) may be taken from tables in reference books. Based on individual ion concentrations ($c_i$), ionic strength (Ic) can be calculated as follows:

$$Ic = 0.5 \, \Sigma \, c_i \, z_i^2 \, / \, w_i \quad (\text{unit} = \text{mol}_c \, \text{m}^{-3})$$

$c_i$ : concentration of the i-th ion in mg $l^{-1}$

$z_i$ : charge of the i-th ion

$w_i$ : molar weight of the i-th ion in g

At ionic strengths higher than 0.1 meq $l^{-1}$, concentrations have to be corrected for the activity of single ions in the solution (A.P.H.A., A.W.W.A and W.E.F 1995). At Ic values < 0.1 meq $l^{-1}$, the difference between measured and calculated conductivity should not exceed 2% (MILES and YOST 1982).

## 2.4.3 Alkalinity (ALK) and Acid Neutralisation Capacity (ANC)

Within this report "alkalinity" (ALK) is used for **measured** values. Various methods that may be applied are described in detail in the ICP Waters Programme Manual (1996). Intercalibration exercises (2.2) have shown poor results for ALK measurements, certainly because the methodologies used in the single countries and laboratories are different. Nevertheless, measurements made by a certain laboratory with the same method over many years seem to be quite reliable. If measured ALK concentrations were available, they were used for trend analyses.

Acid neutralisation capacity (ANC) is a **definition**, based on the principle of electroneutrality and expressed by the following equation ({ } = concentrations in μeq $l^{-1}$):

$$ANC = \{Ca^{2+}\} + \{Mg^{2+}\} + \{Na^+\} + \{K^+\} - \{SO_4^{2-}\} - \{Cl^-\} - \{NO_3^-\}$$

The term ANC includes all cations and anions whose concentrations are pH dependent. For rivers and lakes with high ammonium concentrations, $NH_4^+$ should be considered (only influenced by $H^+$ at high pH). If all parameters for calculating ANC were available (and no ALK values), ANC was used for trend analyses.

## 2.5 Criteria Set for New Data Base

Data are not excluded from the main data base at NIVA without approval by the originator. The objective is to create good quality datasets for efficient use. To meet this objective, the ICP Waters data base has been devided into two parts, including a complete set of all reported information and a "new" subset of selected and quality controlled datasets which can be used, e.g., for statistical analyses.

The following criteria for establishing the new data base were applied:

-       Information on sites where data reporting ended in 1993 have not been read into the new data base.

-       For streams and lakes with more than one sampling site, only one was chosen. The new site description data (see 2.2) and national reports including detailed information on the monitoring sites have been used to make this choice.

-       For lakes where data of several depths have been reported, only is was chosen (0.5 to 1.0 m below surface).

-       Parameters important for statistical analyses were checked "manually" site by site to find outliers and to check the continuity of time series. Corrected data were read into the new data base.

-       For many sites ion balances cannot be calculated because one or more parameters are missing. Since the methods for analysing, e.g., sulphate and nitrate concentrations in water are quite reliable, datasets including at least $SO_4$, $NO_3$, Cl, pH, Ca and Mg were read into the new data base.

# 3. Trends in Surface Water Chemistry

## 3.1 ICP Waters Sites Chosen for Trend Analyses

The sampling schedules for sites included in ICP Waters vary substantially, from weekly or monthly, to quarterly in many countries, to one or two samples per year at some sites in some countries. In order to standardize the data used in this report, we chose quarterly sampling as a reasonable goal (achievable for most sites) and subsampled the data from more frequently sampled sites to produce a quarterly dataset. Sites with fewer than quarterly samples were not included in this analysis.

It was also evident from the outset that most sites exhibit curvilinear, rather than monotonic, trends. Often the slope of curves changes (e.g. from positive to negative, or vice-versa) in ca. 1990. For this reason, and also to enable a comparison of trends between decades, we decided to perform the tests separately on data from the 1980s and 1990s.
We also chose to exclude sites that are known to exhibit climate-driven developments, e.g., Nevins Lake in the U.S. (WEBSTER et al. 1990) and some of the moorland pools in the Netherlands (VAN DAM and BUSKENS 1993).

For many sites report of data started in 1991/92 and not enough data for trend analyses were available. At some of those sites water chemistry is obviously influenced strongly by bedrock material (e.g., by pyrite, gypsum). In these cases we are not sure wether it is possible at all to relate changes in $SO_4$ concentrations to decreasing (or increasing) sulphate deposition.

Only sites with more than *10 observations* were considered in either decadal dataset. Trend tests on 58 sites with data from the 1980s, and 68 sites with data from the 1990s could be performed.

## 3.2 Additional ICP Monitoring Sites Included in Trend Analyses

In cooperation with the International Co-operative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP Monitoring) several ICP-IM sites were included into the trend analyses (e.g., KLEEMOLA and KLEEMOLA and FORSIUS 1996). The results are summarized in **Annex B**.

## 3.3 Statistical Methods Used for Trend Analyses

### 3.3.1 Nonparametric Seasonal Kendall Test (SKT)

To analyse trends in alkalinity (ALK), acid neutralization capacity (ANC), base cations (SBC = $\Sigma(Ca^{2+} + Mg^{2+} + Na^+ + K^+)$), sulphate ($SO_4^{2-}$), nitrate ($NO_3^-$) and hydrogen ion ($H^+$) the nonparametric Seasonal Kendall test (SKT) was used (HIRSCH et al. 1982). The SKT has become a standard for

detecting site-specific trends in water quality data, because it can accommodate the non-normality, missing and censored data, and seasonality that are common in data of this type. Nevertheless, it is in a statistical sense a powerful trend test (LOFTIS and TAYLOR 1989). One limitation of the SKT is that it detects only monotonic trends. They need not be linear, but they must proceed in only one direction (increasing or decreasing). The SKT analyses the data within seasonal blocks, and compares the rank value for a single seasonal observation to the rank values of subsequent data from that same season. A Z statistic is produced for each season, and if the trends are homogenous among them, the seasonal Z statistics can be combined to produce an overall trend result.

The SKT does not estimate the slopes of trends. Slopes were calculated according to the method of SEN (1968), using the median of all between-year differences in the variable of interest. As with Z statistics, the slope values can be produced separately for each season, or combined to create an overall trend magnitude.

## 3.3.2 Regional Trend Meta-Analyses

In order to infer regional trends from the separate trend tests performed by SKT, we employ a variation of meta-analysis (HEDGES and OLKIN 1985). The term meta-analysis describes a family of techniques for analysing the summary statistics of a series of individual tests or studies, in order to determine whether the combined results have more significance than the individual tests. The methodology for combining test results has its roots in the work of Fisher (FISHER 1932), who recognized that p values from independent tests could be combined and tested against a chi-square distribution.

The procedure for combining SKT results was first presented by VAN BELLE and HUGHES (1984). The general concept is to perform a test on the combined Z statistics from individual SKT tests, with the goal of determining whether the distribution of individual trend tests deviates from the distribution that would be expected from chance. In its simplest form, meta-analysis involves squaring the individual Z statistics, taking a sum, and testing the result against a chi-square distribution with degrees of freedom equal to the number of sites subjected to the trend test.

Just as homogeneity of trend results among seasons is a concern when combining seasonal Z statistics from a single site into an overall trend result, the homogeneity of trends among different sites and decades is a concern in meta-analysis. If sites do not exhibit the same or similar trends, then it would be misleading to combine them. We test for homogeneity by attributing the variance among the Z statistics (in this case, from 60-70 sites and 4 seasons, or a total of 240 to 280 Z values, depending on the decade) to each of the possible sources of interest (region, decade, site and season). In practice, this is accomplished by submitting the Z statistics to an analysis of variance, and testing the resulting

sums of squares against a chi-square distribution (MATTSON et al., in press, VAN BELLE and HUGHES 1984). If any of these variables is significant in the analysis of variance, it indicates that the trends are not homogenous with respect to that variable. For example, most of the tests reported below are first segregated by region and decade; if the site variable in such a test is significant, it indicates that attributing differences in the trends to different regions and different decades is not sufficient, because significant differences between sites still exist and valid regional trends cannot be determined.

## 3.4 Development in Surface Water Chemistry at the Single Sites

The trend results presented by country are primarily an illustration of the differences between the two decades, 1980s and 1990s, in each country and at each site. Regional developments are summarized in chapter 3.5. Where no statistical analyses could be performed, short information on developments in water chemistry is given. The new site descriptions are also included (rather detailed) to allow comparison of chemistry results and site specific surroundings at the different ICP Waters locations.

The "80s" include the years 1983/85 - 1989 and the "90s" the years 1990 - 1994/95. Where seasalt corrected concentrations were used a * is added to the chemical formula (e.g., $SO_4$*). Within the figures, ** and * indicate that the probability that the observed change is by chance is < 0.1% or < 0.5% (e.g., z = 3.53**). Results of *all* SKT analyses, including the p values, are listed in **Annex C** (see also **Annex B** for the ICP-IM sites).

## 3.4.1 Austria

Data of three Austrian sites were read into the new database (**Table 2**). For all other sites reporting stopped in 1991. One sample is taken every autumn, only data of the 50 or 100 cm depth are used in this report. For two sites, DOS-K and MUT-T, no values of 1991/92 - 1994 are available. All data series could not be used for trend analyses (not enough data for the 90s; see also PSENNER 1989).

**Table 2.** The Austrian sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|---------------|-------------------|--------------|---------------------|
| Kärnten | Dösener See *(DOS-K)* | L | 1989-95 | N |
| Tirol | Mutterberger See *(MUT-T)* | L | 1989-95 | N |
| | Schwarzsee ob Sölden *(SOS-T)* | L | 1989-95 | N |

*Kärnten*

The elevation of **Dösener See** is 2281 meter above sea level. The catchment is 3.0 $km^2$ wide and lake area is 0.1 $km^2$. Precipitation averages 1400 mm $yr^{-1}$ (no runoff). Granite and gneiss are the dominant bedrock types. Soils are very shallow, about 10 cm.

$SO_4$ concentration seem to decrease beween 1989 (81 $\mu eq$ $l^{-1}$) and 1995 (38 $\mu eq$ $l^{-1}$), whereas there is no change in $NO_3$ values (mean = 23 $\mu eq$ $l^{-1}$). Measured alkalinity is obviously increasing; it was 23 $\mu eq$ $l^{-1}$ in 1989 and 87 $\mu eq$ $l^{-1}$ in 1995.

*Tirol*

**Mutterberger See** is 0.04 $km^2$ wide, the surroundig catchment 0.2 $km^2$. Its altitude is 2483 m above sea level. Berock is crysterlline.

The four $SO_4$ measurements (1989, 91, 92 and 95) show almost the same results.The mean value is 24 $\mu eq$ $l^{-1}$. The highest ALK value (measured) can be found in 1989 (17 $\mu eq$ $l^{-1}$), the lowest in 1991 ($\mu eq$ $l^{-1}$). In 1995 ALK is 3 $\mu eq$ $l^{-1}$. $NO_3$ concentrations in autumn are almost the same over the years (mean = 12 $\mu eq$ $l^{-1}$).

**Schwarzsee ob Solden** is elevated at 2799 m. Yearly precipitation avarages 1500 mm. Lake area is 0.03 $km^2$, and its mean depth is 10 m. The whole catchment is 0.2 $km^2$ wide.

One water sample is taken per year (in autumn). Measured alkalinity had dropped below zero in 1992. ALK obviously recovered until 1995, although $SO_4$ concentrations seem to increase during this period (**Figure 3**). No change in $NO_3$ concentrations can be seen (**Figure 3**).

Schwarzsee ob Sölden (Austria)
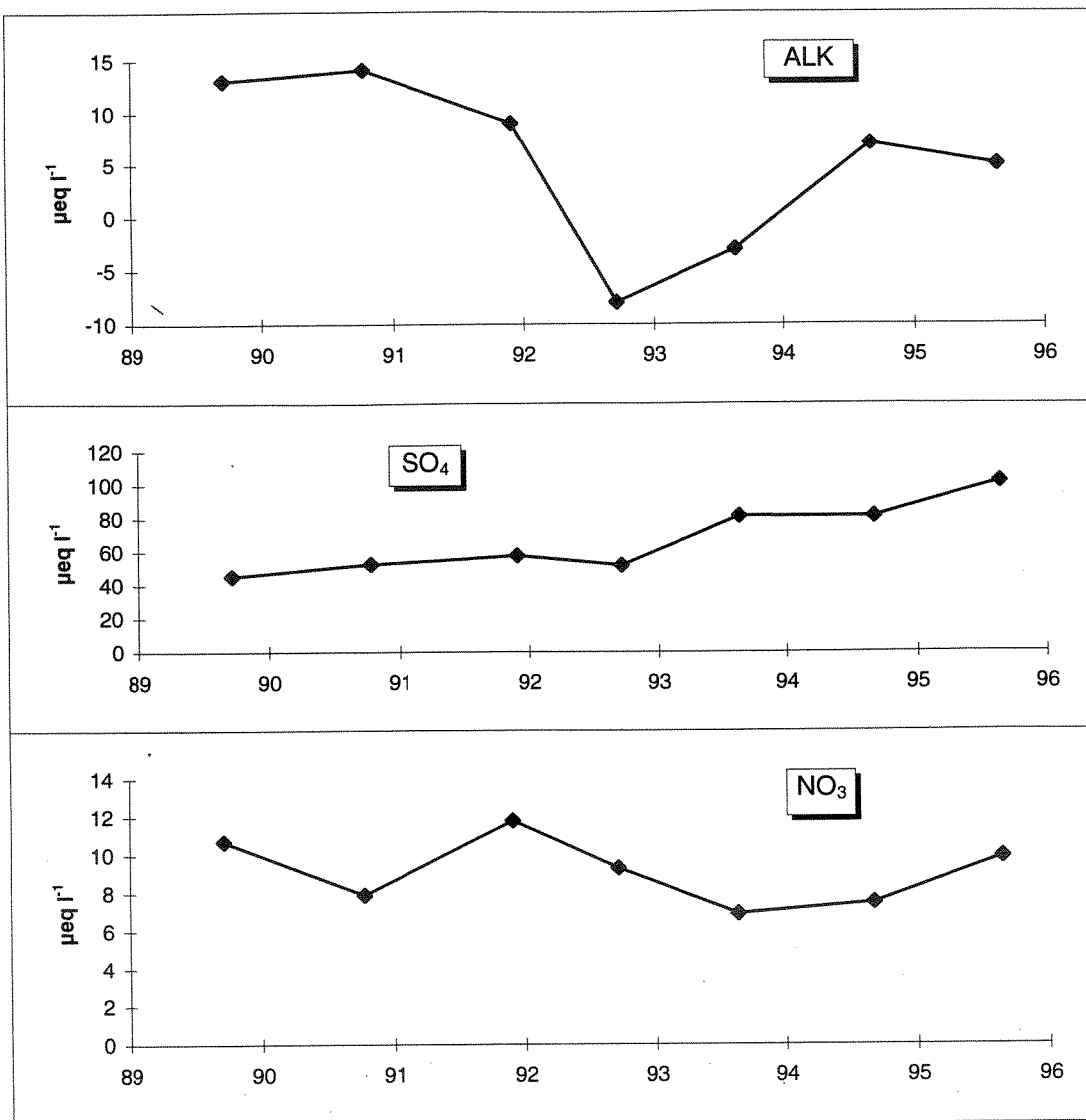


**Figure 3.** ALK, SO$_4$ and NO$_3$ at Schwarzsee ob Sölden, Austria.

### 3.4.2 Czech Republic and Slovak Republic

All data reported from Czech Republic and mentioned in this report are based on samples taken in the High Tatra Mountains in the Slovak Republic.

**Table 3.** The Slovakian sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|---------------|-------------------|--------------|---------------------|
| Tatra Mountains | Batizovske pleso *(BA-1)* | S | 1987-95 | N |
| | Starolesnianske pleso *(VS-15)* | S | 1987-95 | N |
| | Male Hincovo pleso *(ME-2)* | S | 1987-95 | N |
| | Slavkovske pleso *(SL-2)* | S | 1987-95 | N |
| | Jamske pleso *(ST-1)* | S | 1987-95 | N |
| | L'adove pleso *(VS-4)* | S | 1987-95 | N |

No results of 1991 and 1992 are available. Nevertheless, the data series are interesting because they start in 1987, and samples were taken regularily (in autumn) during all the other years until 1995.

*Tatra Mountains*

The dominant bedrock types at the High Tatra Mountains are granite and gneiss. Soils are shallow, 10 to maximal 50 cm deep. Up to 90% of the catchment areas are exposed to bedrock. The area is mostly covered with dwarf mountain-pine.

The development of $SO_4$ and ALK-G concentrations is very similiar at all sites. Although there are not enough data for trend analyses, $SO_4$ seems to decline between 1987 and 1995 (examples: **Figure 4** and **Figure 5**). Alkalinity shows the opposite tendency: values are increasing. ALK concentrations are just above or just below 0 µeq $l^{-1}$. In generell, $NO_3$ concentrations are low (up to 70 µeq $l^{-1}$).

Batizvske pleso (Slovak Republic)
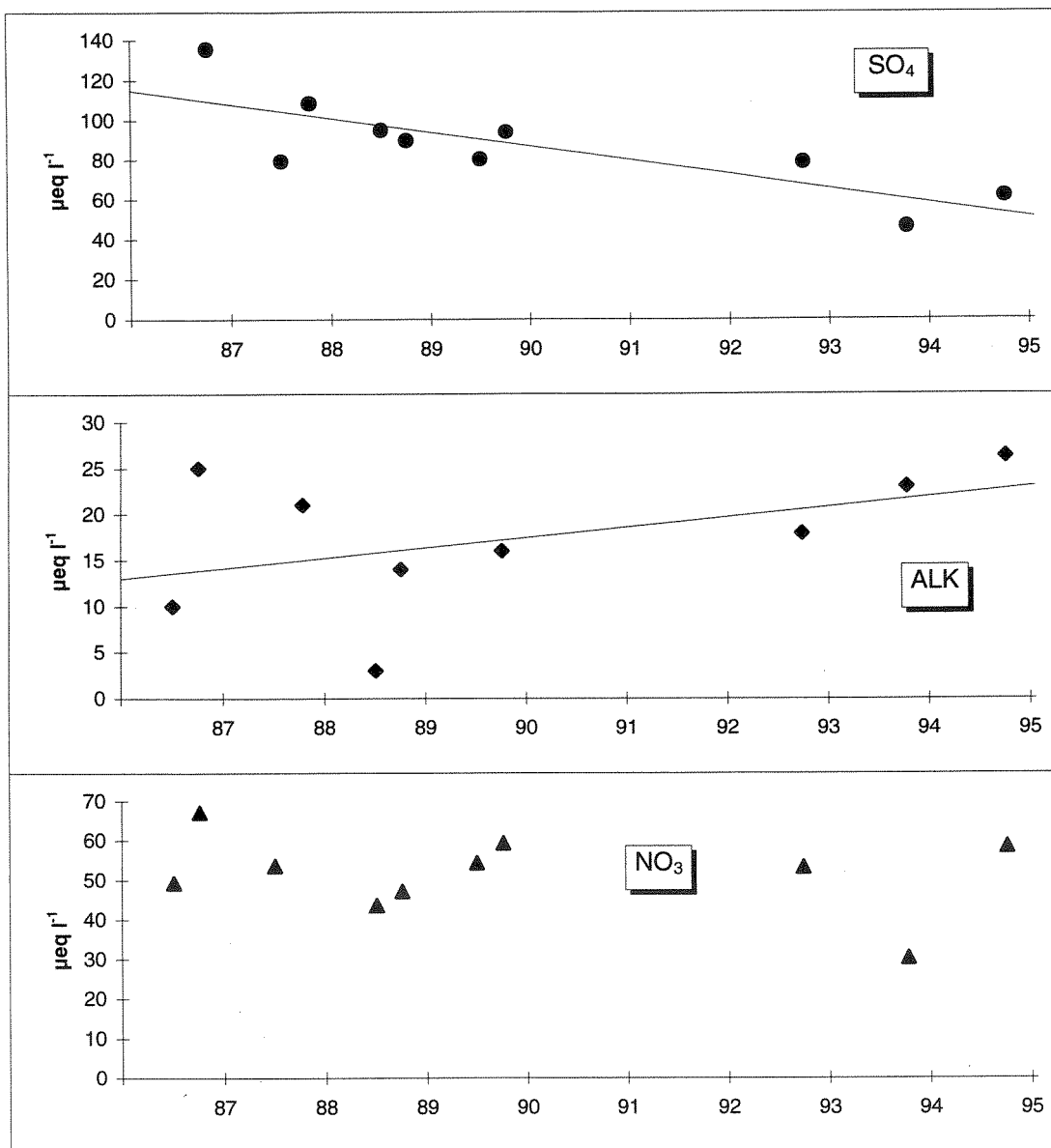


**Figure 4.** SO$_4$, ALK-G and NO$_3$ concentrations at Batizvske pleso, Slovak Repiublik.

Slavkovske pleso (Slovak Republic)
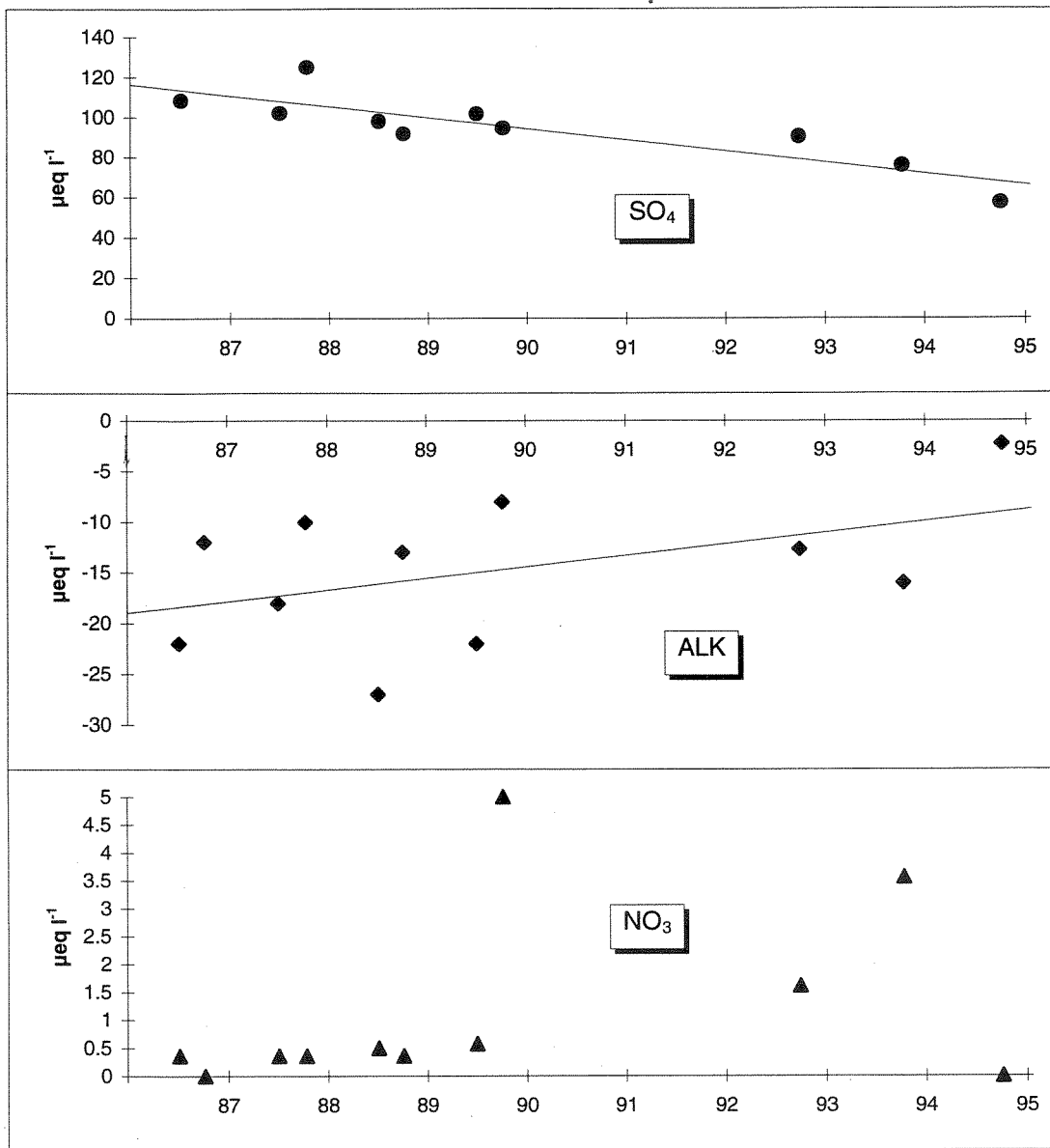


**Figure 5.** $SO_4$, ALK-G and $NO_3$ concentrations at Slavkovske pleso, Slovak Republic.

### 3.4.3 Denmark

Data of two Danish sites, located in the same region, have been reported to the Programme Center at NIVA. For both sites trend analyses were run for the 90ies only (not enough data during the 80ies).

**Table 4.** The Danish sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Sepstrup Sande | Skaerbaek, Station B (SK110B) | S | 1987-1995 | Y |
| Sepstrup Sande | Skaerbaek, Station F (SK038F) | S | 1984-1995 | Y |

*Sepstrup Sande*

Samples of two stations, B and F, have been taken at **Skaerbaek** river. **Station B** has an elevation of 75 m and **F** of 67 m. The catchment areas are 2.0 and 5.0 km$^2$, respectively. At both sites mean annual precipitation is 825 mm and runoff averages 365 mm yr$^{-1}$. The area is underlain by sand with podsolic soils (50 - 100 cm deep). The surrounding of station B is covered by 10% with coniferous (mainly spruce) forest, 80% are heather and grassland and 10% wetlands and bogs. At station F 30% of the vegetation are trees, 45% is heather and grassland and 5% are wetlands and bogs.

The SO$_4$ and SBC concentrations are declining significantly at both stations during the 1990s (**Figure 6** and **Figure 7**). There is a positive trend in ALK and pH in the 1980s. NO$_3$ concentrations are much higher at station F compared to station B (averages = 203 and 79 µeq l$^{-1}$). The NO$_3$ curves show a seasonal pattern but values during summer never drop below 40 (B) or 100 (F) µeq l$^{-1}$. At both stations there is a negative trend in H$^+$ concentration in the 1980s (see also REBSDORF et al. 1991).

Skaerbaek, Station B
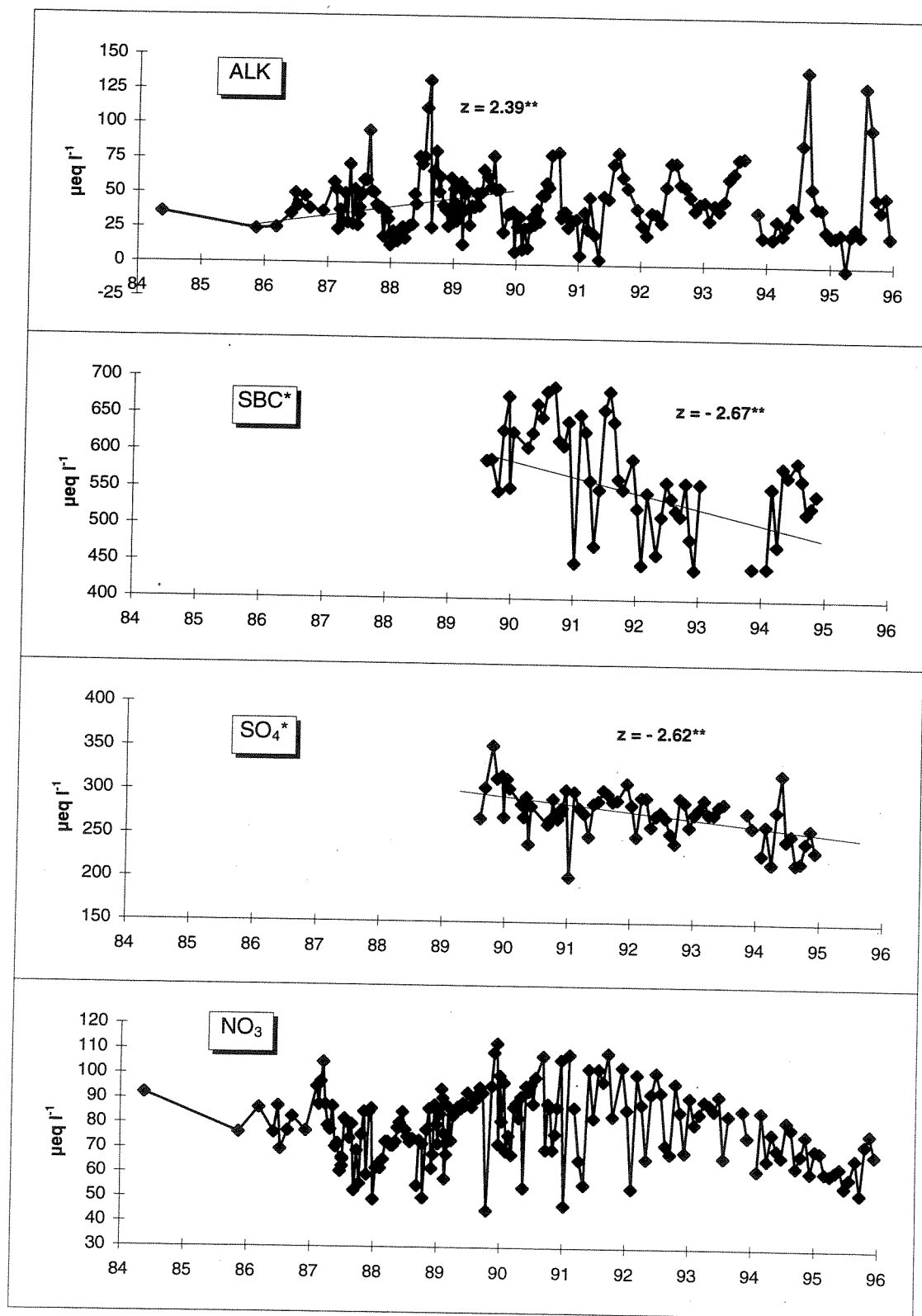


**Figure 6.** ALK, SBC*, SO₄*, NO₃ concentrations at Skaerbaek, Station B, Denmark.

Skaerbaek, Station F
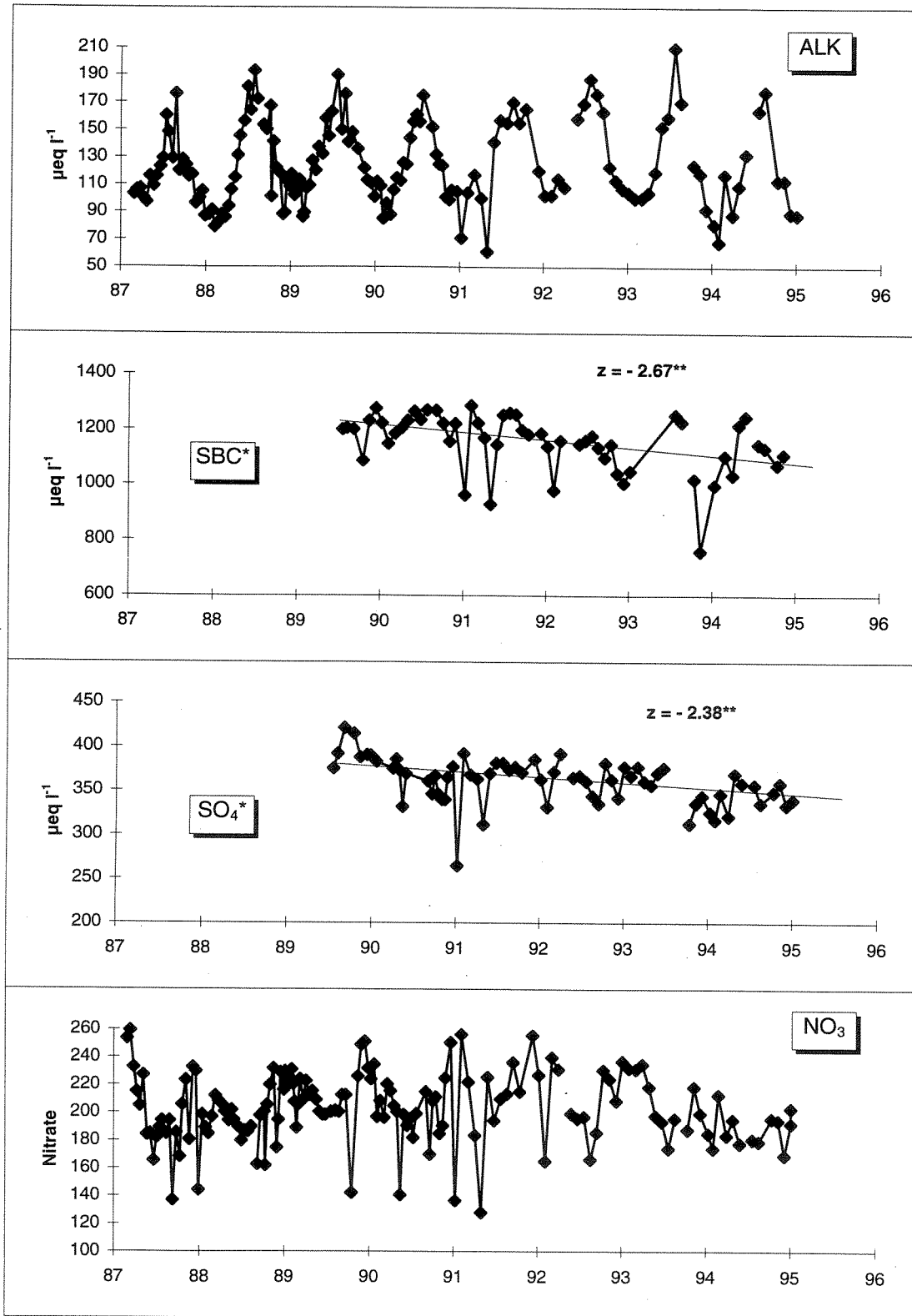


**Figure 7.** ALK, SBC*, SO$_4$*, NO$_3$ concentrations at Skaerbaek, Station F, Denmark.

## 3.4.4  Finland

Data of five ICP Waters sites in Finland have been reported to the Programme Center at NIVA. One criterium for choosing data for trend analyses was to use near surface samples if data of more than one depth are available. In case of the Finnish data, samples from 1 m depth were chosen. Dominant bedrock type at all Finnish sites is granite and gneiss.

**Table 5.** Finnish ICP Waters sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Southeastern Finland | Hirvilampi (SF01) | L | 1988-95 | Y (90s) |
| | Vuorilampi (SF02) | L | 1988-95 | Y (90s) |
| | Maekilampi (SF03) | L | 1990-95 | Y (90s) |
| Central Lapland | Suopalampi (SF05) | L | 1988-95 | N |
| | Vasikkajaervi (SF06) | L | 1988-95 | N |

*Southeastern Finland*

The elevation of **Hirvilampi** is 34 m, and the catchment is 4.0 $km^2$ wide. Average lake depth is 3.3 m, lake area is 0.08 $km^2$. 50% of the catchment is covered with coniferous forest, 40% is exposed to bedrock and 13% are wetland and heather. Yearly precipitation averages 770 and runoff 330 mm.

The $SO_4$ concentrations at Hirvilampi are significantly decreasing in the 1990s (**Figure 8.**).

Precipitation, runoff and bedrock types at **Vuorilampi** are the same as at Hirvilampi. Most of the 0.6 $km^2$ catchment is covered with coniferous forest (60%). Catchment area in 0.37 $km^2$, lake area 0.03 $km^2$ (average depth = 0.7 m) and elevation 32 m above sea level.

There are no significant changes in $SO_4$ and ALK concentrations at Vuorilampi. $NO_3$ peaks can be found in 1985 and 1988 (about 550 µeq $l^{-1}$).

**Maerkilampi** is a 0.03 $km^2$ lake surrounded by coniferous forest (70% of the 0.62 $km^2$ catchment area). Its elevation is 52 m. Mean runoff is 330 mm and precipitation 770 mm $yr^{-1}$.

There is a negative trend in $SO_4$ concentrations during the 1990s. The lake was limed in 1990 by a local landowner, which explains the significant increase in ALK. $NO_3$ concentrations show a typical seasonality.

Acidic deposition in southeastern Finland is still relatively high, especially due to high $SO_2$ emissions in Russia and Estonia. MANNIO and VUORENMAA (1995) analyzed data of 171 monitoring sites throughout Finland (sampled every autumn since 1990 and in 1987). They found a general decline in sulphate concentrations, compensated by a significant rise in both alkalinity and pH.

*Central Lapland*

The lakes **Suopalampi** and **Vasikkajaervi** are located in Lapland, where acidic deposition is low (elevations: 225 and 242 m above sea level). Mean precipitation (560 mm) and runoff (330 mm) are the same at both sites. Lake area of Suopalampi is 0.39 km$^2$ (average depth: 1.5 m) and Vasikkajaervi is 1.58 km$^2$ wide (average depth: 5.0 m). Most of the catchment areas are covered with wetlands and bogs.

No significant changes in water chemistry can be observed (1988 - 1995).

From 1997 onwards two additional lakes will be included into the ICP Waters Programme: **Kakkisenlampi** (North Karelia) and **Vitsjön** (South Savo). Both are clearwater lakes, sensitive to acidification, and have been intensively monitored since the 1980s.

## 3.4.5 France

For the first time data from France have been reported to the Programme Center at NIVA.

**Table 6.** The French site included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| French Pyrénées | Lake Aube | L | 1991-94 | N |

**Lake Aubé** is located in southern France (Pyrénées mountains). It's elevation is 2091 m above sea level, and the main bedrock type is granite. Catchment area is 0.77 km$^2$, most of it is exposed to bedrock. The site is included into the European Community's AL:PE / MOLAR project (WATHNE and PATRICK 1994). The lake is 0.9 km$^2$ in area and (maximum depth = 45 m). Average yearly runoff is > 1000 mm.

The $SO_4$, ALK and $NO_3$ concentrations have not changed during the period 1991 - 1994. Mean $SO_4$ concentrations are 26 µeq l$^{-1}$, alkalinity and $NO_3$ values average 13 and 9 µeq l$^{-1}$.

Hirvilampi, Finland
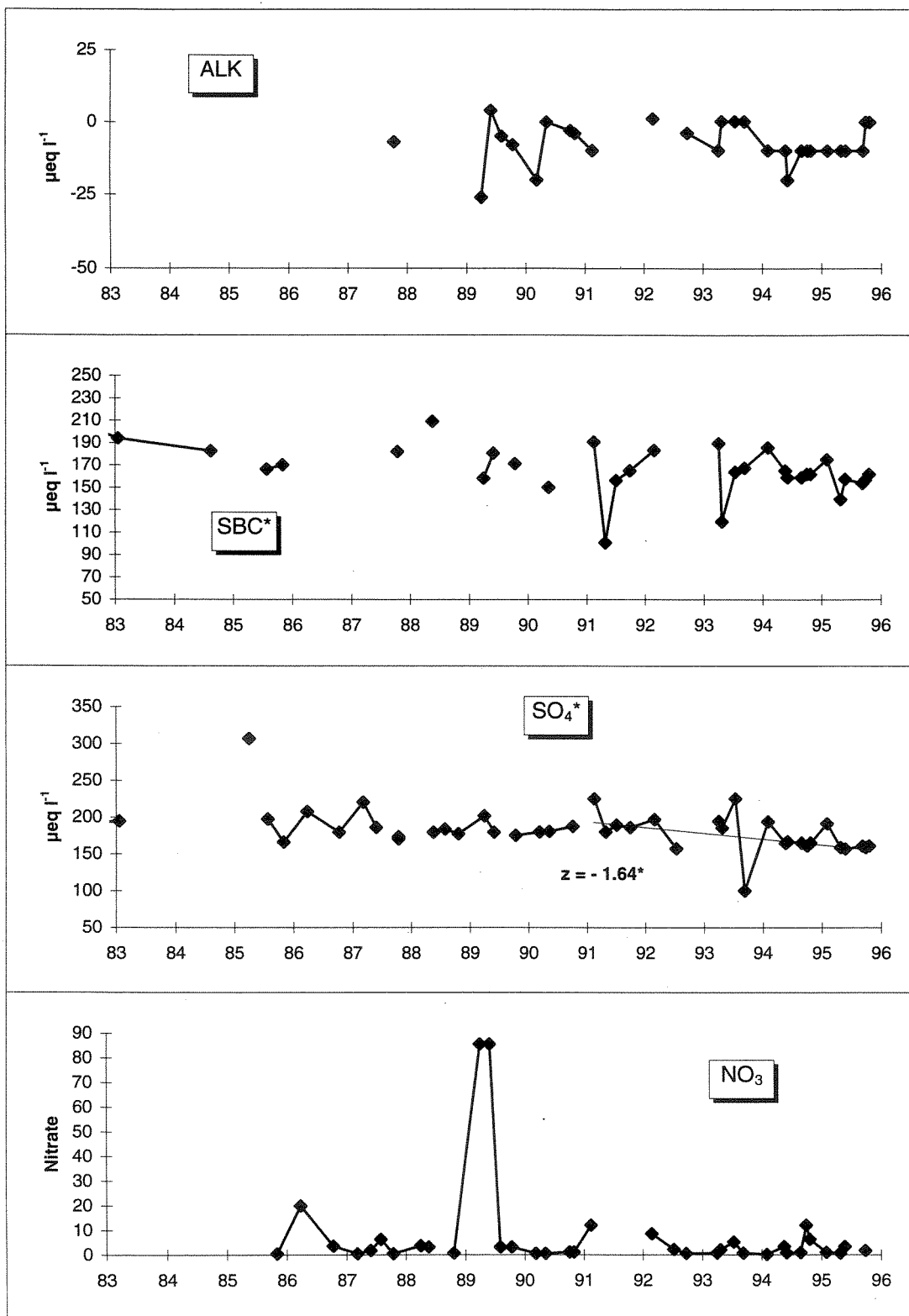


**Figure 8.** ALK, SBC*, SO$_4$*, NO$_3$ concentrations at Hirvilampi, Finland.

## 3.4.6 Germany

Data of 79 sites in Germany have been reported to the Programme Center at NIVA. Information on 27 of them was read into the new ICP Waters database. Water chemistry at the German monitoring sites has been analyzed by 14 different laboratories. In most cases not all parameters necessary to calculate an ion balances or to run all statistical analyses were available. Where ALK had not been measured, calculated ANC was taken (= $\Sigma$ SBC - $\Sigma$ SAA). If Na and K values were not available, Mg+Ca was plotted instead of SBC (no trend analyses).

**Table 7.** The German ICP Waters sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Bayerischer Wald | Vorderer Schachtenbach (Vor32) | S | 1984-94 | Y |
| | Hinterer Schachtenbach (Hin31) | S | 1984-94 | Y |
| | Seebach (See30) | S | 1984-94 | Y |
| | Große Ohe (Gro33) | S | 1984-94 | Y |
| Oberpfälzer Wald | Waldnaab (Wal2) | S | 1986-89; 1992-94 | N |
| Fichtelgebirge | Eger (Ege1) | S | 1984-94 | Y |
| | Röslau (Roe2) | S | 1984-94 | Y |
| Erzgebirge | Große Pyra (Gro2) | S | 1992-94 | N |
| | Rote Pockau (Rot3) | S | 1992-94 | N |
| | Wilde Weißeritz (Wi14) | S | 1993-94 | N |
| | Wolfsbach (Wol1) | S | 1992-94 | N |
| Elbsandsteingebirge | Taubenbach (Tau5) | S | 1993-94 | N |
| Schwarzwald | Dürreychbach (Due2) | S | 1987-94 | Y(90s) |
| Odenwald | Schmerbach (Sch3) | S | 1985-94 | N |
| Hunsrück | Gräfenbach (Gra5) | S | 1984-94 | Y(90s) |
| | Traunbach (Tra1) | S | 1984-94 | Y |
| Taunus | Rombach (Rom2) | S | 1987-94 | Y(90s) |
| Rothaargebirge | Elberndorfer Bach (Elb1) | S | 1986-89; 1992-94 | N |
| | Zinse (Zin2) | S | 1986-89; 1992-94 | N |
| Kaufunger Wald | Nieste (Nie5) | S | 1988-94 | Y(90s) |
| Harz | Dicke Bramke (Dic) | S | 1989-94 | Y(90s) |
| | Große Soese (GroS2) | S | 1990-94 | Y(90s) |
| | Große Bode (Gro14) | S | 1986-94 | Y |
| | Große Schacht (Sch9) | S | 1986-89; 1992-94 | N |
| Sächsische Tieflandbuch | Heidelbach (Hei7) | S | 1992-94 | N |
| | Ettelsbach (Ett6) | S | 1992-94 | N |
| Lauenburgische Seenplatte | Pinnsee (Pin3) | L | 1986-89; 1992-94 | N |

The brief site descriptions are based on information reported recently to the Programme Center at NIVA and on SCHNEBÖGL et al. (1996). At almost all German sites soil depth is about 100 cm. Thus, soils are relatively deep, especially compared to those in Scandinavia and Canada.

*Bayerischer Wald (Bayern)*

The main bedrock type at Bayerischer Wald is gneiss and granite. The elevation of the ICP Waters sites **Vorderer Schachtenbach, Hinterer Schachtenbach, Seebach** and **Große Ohe** is about 770 m above sea level. The area is covered with 70 % coniferous, 28 % deciduous forest and 2 % grassland and moors. Average precipitation is 1430 mm $yr^{-1}$. The mean soil type is dystric cambisol ($\approx$ 100 cm deep).

Although slopes of most $SO_4$ curves are negative over the whole observation period (1983/84-94), decrease in sulphate concentrations is not statistically significant (**Figure 9**). No change in pH can be observed. At Seebach $NO_3$ concentrations show a negative trend during the 90s. Nitrate is significantly increasing at Hinterer Schachtenbach from 1983 to 1994. In generell, $NO_3$ concentrations show a strong seasonality at all three sites. Minium values during the vegetation period exceed a concentration of about 30 µeq $l^{-1}$. Ca+Mg values do not seem to change.

*Oberpfälzer Wald (Bayern)*

Bedrock, soil type and depth at Oberpfälzer Wald are the same as at Bayerischer Wald. The **Waldnaab** catchment close to the Czech border is mainly covered by spruce (98%). The elevation is 727 m and the mean precipitation 1066 mm $yr^{-1}$. Since measurements of 1990 and 1991 are missing, data of Waldnaab could not be used for trend analyses.

*Fichtelgebirge (Bayern)*

Two ICP Waters sites are located at Fichtelgebirge: **Eger** and **Röslau** (660 m above sea level). The predominant bedrock types are phyllite and quartzite. Soils are about 100 cm deep, and 95% of the catchments is covered with coniferous forest (spruce, pine, larch). Precipitation is 850 - 1000 mm $yr^{-1}$. Only during the 80ies $SO_4$ concentrations at Röselau are significantly decreasing (slope negative at Eger). $NO_3$ values show a positive trend at Eger during the 80ies. The seasonal pattern is pronounced at Röselau while it is "damped" at Eger, with several peak values up to 140 µeq $l^{-1}$. Mg+Ca concentrations seem to stay nearly constant at both sites.

Vorderer Schachtenbach, Germany
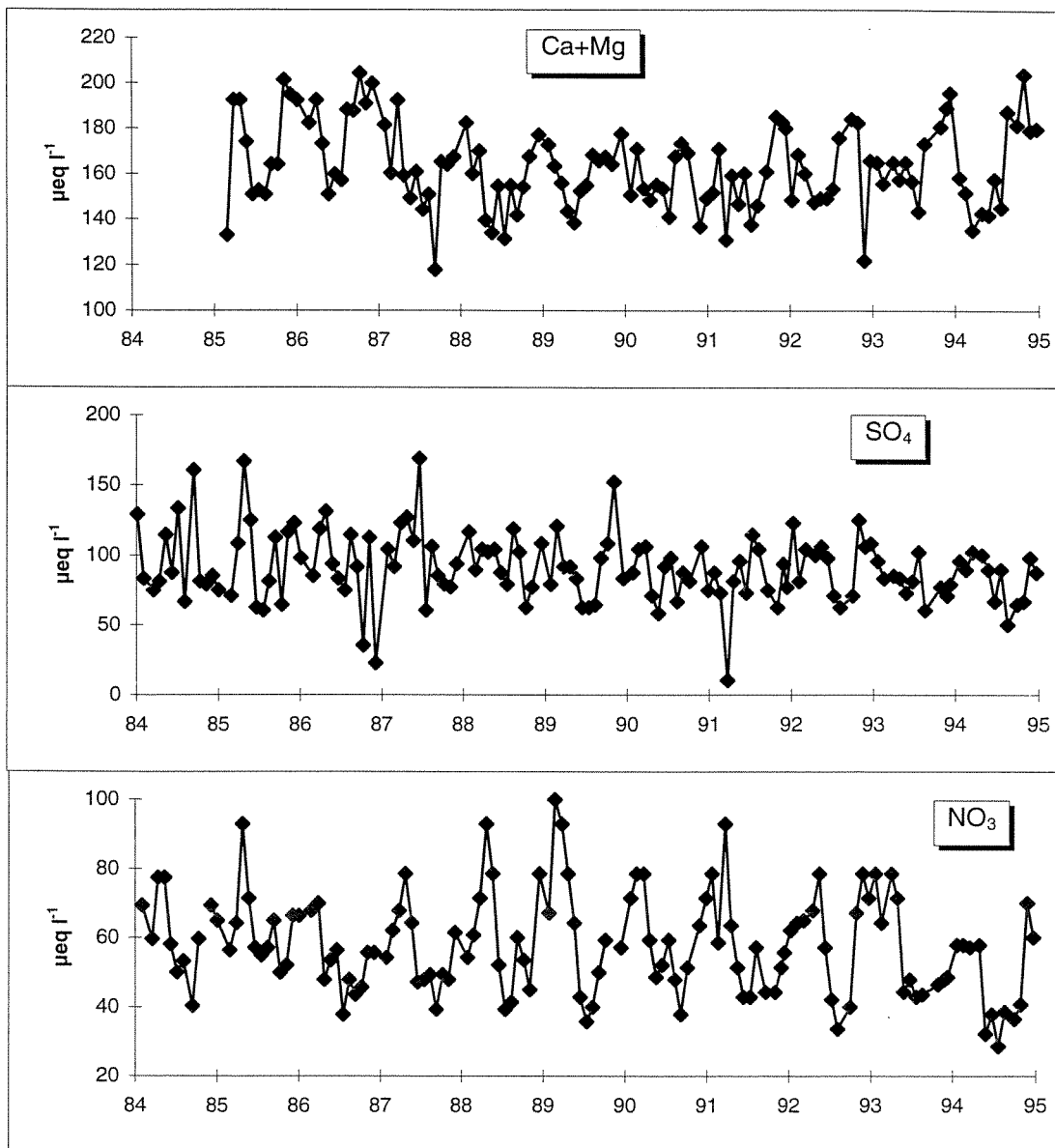


**Figure 9.** Ca+Mg, SO₄, and NO₃ concentrations at Vorderer Schachtenbach, Germany (Bayern).

*Erzgebirge (Sachsen-Anhalt)*

At Erzgebirge close to the Czech border four streams are monitored: **Große Pyra** (745 m; granite, gneiss), **Rote Pockau** (635 m; gneiss), **Wilde Weißeritz** (700 m; phyllite, granite, gneiss) and **Wolfsbach** (565 m; phyllite). The area is covered by 75 - 95% of coniferous forest, the rest is grass-, farmland, rocks or pasture. Water chemistry data have been reported regularly from 1992/93 onwards so that they could not be used for statistical analyses. Only very few samples had been analyzed in the 70ies and 80ies.

$SO_4$ concentrations at the Erzgebirge sites are relatively high (average in 1995 e.g.: 413, 881, 673 and 1015 $\mu$eq $l^{-1}$). A very high mean $NO_3$ values can be observed at Wolfsbach (1215 $\mu$eq $l^{-1}$ in 1995).

*Elbsandsteingebirge (Sachsen-Anhalt)*

As indicated by its name the main bedrock type at Elbsandsteingebirge is sandstone. **Taubenbach** is the only sampling site, established in 1993. Thus, statistical analyses could not be run. Most of the catchment, covered with coniferous forest, is located in the Czech Republic.

The average $SO_4$ concentration at Taubenbach in 1995 is 1042 $\mu$eq $l^{-1}$ ($NO_3$: 171 $\mu$eq $l^{-1}$).

*Schwarzwald (Baden Württemberg)*

The monitoring site **Dürreychbach** in located in the northern part of Schwarzwald at an elevation of 730 m. The main bedrock type is sandstone and average soil depth 100 cm. Mean yearly precipitation is 1740 mm $yr^{-1}$. The catchment is covered with spruce.

Since no data were reported before 1987 statistical analysis was restricted to the 90s (1990-94). $NO_3$ values were not used because the plotted curves show only 3 or 4 different concentration "levels". Although the slope of the $SO_4$ curve is negative, there is no significant decrease in $SO_4$ concentrations. While SBC values are significantly increasing, no change in pH and ANC can be seen.

*Hunsrück (Rheinland-Pfalz)*

**Gräfenbach** and **Traunbach** are the two streams being sampled at Hunsrück. The elevations are 515 and 620 m and the main bedrock type is quartzite. Averaging 1273 mm $yr^{-1}$, precipitation at Traunbach is much higher than at Gräfenbach (735 mm). Forest cover at both sites is 90%, mainly coniferous at Traunbach and 50% deciduous at Gräfenbach. 10% of the catchments is covered by heather, grassland or wetlands. For Gräfenbach there were not enough data available to run trend analyses for the 80ies.

There is no significant change in $SO_4$ concentrations at Gräfenbach during the 90s (negative slope). A strong downwards trend in $SO_4$ values can be observed at Traunbach especially during the 80ies (**Figure 10**). ANC and pH show an upwards trend during the 80ies, but both decrease again

(significantly) in the 90s. From 1990-94 $NO_3$ values decline significantly at Gräfenbach and Traunbach.

*Taunus (Hessen)*

The predominant bedrock at **Rombach** is quartzite (mean soil depth 100 cm). Average precipitation rate is 832 mm $yr^{-1}$ at an elevation of 570 m. The area is covered by 50% coniferous and 50% deciduous forest. Only data from 1990 - 1994 could be analyzed statistically. Significant changes found are decreasing $SO_4$ values and increasing ANC and SBC concentrations.

*Kaufunger Wald (Hessen)*

The elevation of the ICP Waters site **Nieste** is 320 m and mean precipitation is 875 mm. Sandstone is the predominant bedrock type (mean soil depth 100 cm). The ratio deciduous/coniferous forest is 50:50. Only data for the 90s were available for statistical analyses. There is no significant change in $SO_4$, $NO_3$, SBC, and $H^+$ concentrations. ANC shows a positive trend.

*Rothaargebirge (Nordrhein-Westfalen)*

Two streams at Rothaargebirge are included into the monitoring program: **Elberndorfer Bach** and **Zinse**. The elevation is 540 m above sea level and the main bedrock type is quartzite. Forest cover at both catchments is 90%, of which about 50% is coniferous and 40% deciduous. Precipitation averages 1450 mm $yr^{-1}$. Since data of 1990 and 1991 are missing at both sites, time series could not be used for trend analyses.

*Harz (Niedersachsen, Sachsen)*

The main bedrock type at **Dicke Bramke**, quartzite at **Große Soese**, granite and gneiss at **Große Bode** and limestone and granite at **Große Schacht**. At all sites the soils are about 100 cm deep. Precipitation ranges from 1150 to 1300 mm $yr^{-1}$, elevation from 370 - 765 m. The catchments are mainly covered with spruce. At Dicke Bramke ca. 10% are grassland and heather.

At Dicke Bramke and at Große Soese no significant changes in $SO_4$ concentrations can be observed. While there is a positive trend in $NO_3$ values at Dicke Bramke, they are significantly decreasing at Große Soese.

*Sächsische Tieflandbucht (Sachsen-Anhalt)*

Sächsische Tieflandbucht is influenced by the ice age. At **Ettelsbach** and **Heidelbach** the main bedrock type is sand and gravel. The Ettelsbach catchment is covered with coniferous (45%, mainly pine) and deciduous forest (55%, mainly oak). Its elevation is 185 m above sea level (132 m at

Heidelbach). At Heidelbach 85% of the vegetation is pine forest (15% deciduous). Averaging 600 mm yr$^{-1}$ (Ettelsbach) and 550 mm yr$^{-1}$ (Heidelbach) precipitation is not very high. Data of both streams have been reported to the Programme Center at NIVA since 1992. $SO_4$ concentrations are extremely high (up to 4032 µeq l$^{-1}$) and pH values may drop below 4.0 at both sides. $NO_3$ concentrations show a typical seasonality, and they are quite low (maxiumum about 19 µeq l$^{-1}$).

*Lauenburgische Seenplatte (Schleswig-Holstein)*

Sand is the main bedrock type in this part of Northern Germany. Pinnsee is a drainage lake with an average depth of 4.8 m and a lake area of 0.08 km$^2$. The catchment is covered with pine, spruce (80%) and beech, oak (20%).

Results of samples taken in 1 m depth have been read into the new ICP Waters data base. Unfortunately, no data of 1990 and 1991 have been reported. Thus, the results could not be used for statistical analyses. There does not seem to be a "visible" change in $SO_4$ concentrations from 1989 to 1994. The values are ranging from 113 to 161 µeq l$^{-1}$.

Traunbach, Germany
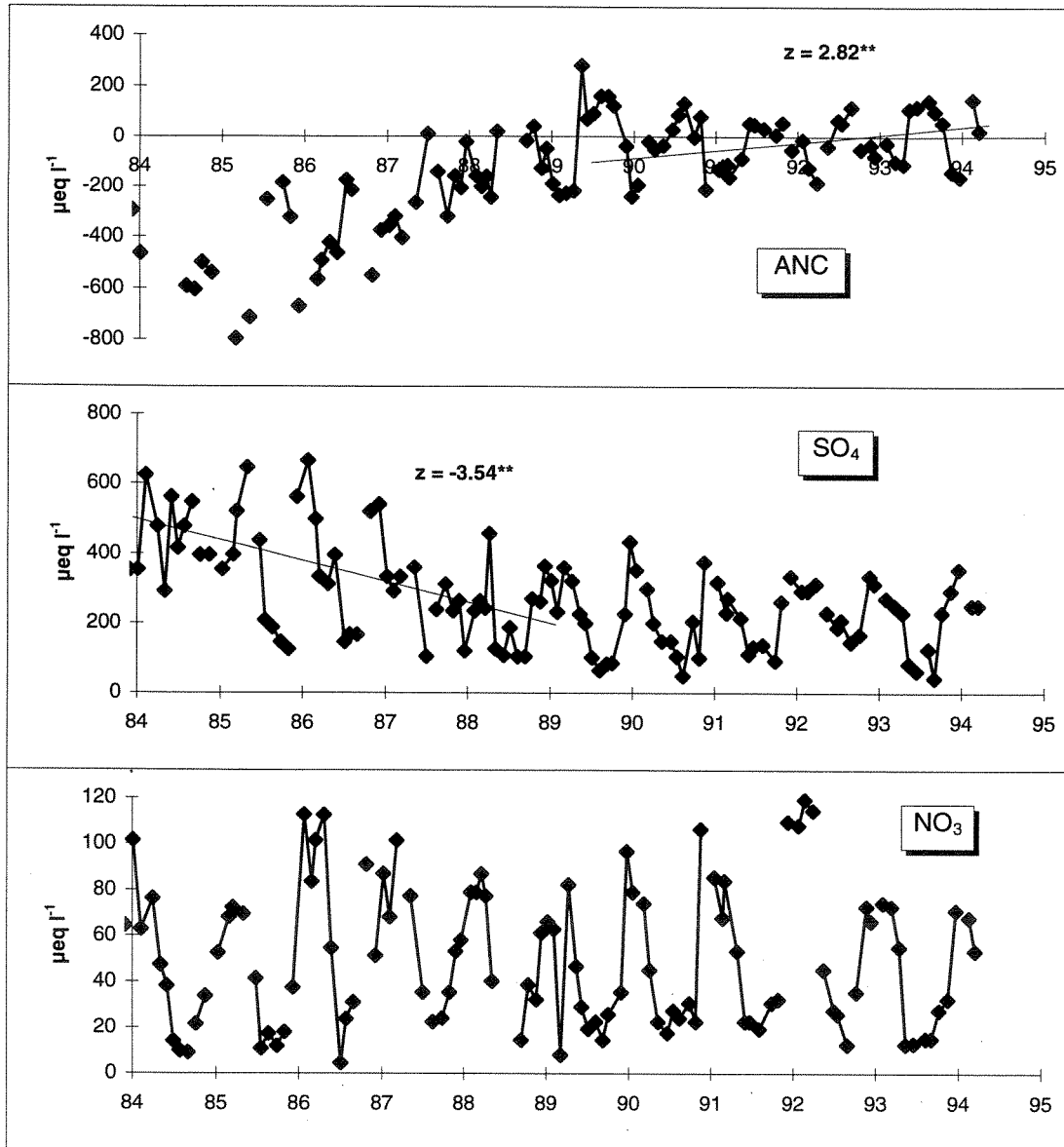


**Figure 10.** ANC, SO$_4$ and NO$_3$ concentrations at Traunbach, Germany (Hunsrück).

### 3.4.7 Hungary

Data of one site in Hungary (Matra Mountains) have been reported.

**Table 8.** The Hungrian site included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|---------------|-------------------|--------------|---------------------|
| Heves  | Csorret *(01)* | L | 1986-94 | N |

The elevation of **Csorret** is 534 m above sea level. The catchment is 8.4 km$^2$ wide. Average depth of the reservoir is 9.0 m (lake area = 0.1 km$^2$). Mean amount of rainfall is 650 mm yr$^{-1}$ and runoff averages 205 mm yr$^{-1}$. Andesite is the predominant bedrock material, and soils are shallow ($\approx$ 20 cm). 74% of the catchment is covered with forest, of which more than 80% is deciduous. 14% of the area are exposed to bedrock, the rest consists of grassland, pasture, wetland and bogs.

Chemistry data from 1986 onwards are available, but only very few parameters have been analyzed. Measured alkalinity is high in 1986, up to about 300 µeq l$^{-1}$ (**Figure 11**). In 1987 ALK declines by ca. 250 µeq l$^{-1}$ and ranges from 50 to 100 µeq l$^{-1}$ until 1994. SO$_4$ has only been measured since 1994. Mean concentration is 43 µeq l$^{-1}$. NO$_3$ values are high in 1986/87 (average = 346 µeq l$^{-1}$). No samples were analyzed between 1988 and 1990. NO$_3$ concentrations are still high in 1991, and they decrease in the 90s (average = 72 µeq l$^{-1}$ in 1994).



**Figure 11.** ALK concentrations at Csorret, Hungary.

## 3.4.8 Ireland

Data of three sites were read into the new database at NIVA. Only mid lake samples were considered. Sampling frequency was quite high from 1984 to 1989. No values at all exist for 1990 and 1991. Since 1992 samples have been taken very irregularily, i.e., 1-2 per year but not in the same month. Thus, trend analyses could not be performed.

**Table 9.** The Irish sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|------|-----|------|-----|
| Wicklow | Glendalough *(GLE10)* | L | 1984-95 | N |
| Donegal | Lough Veagh *(VEA10)* | L | 1988-95 | N |
| Galway | Lough Maumwee *(MAU10)* | L | 1984-95 | N |

*Wicklow*

The catchment of **Glendalough** Lake is 29% afforested (mostly coniferous), 39% heather and grassland and 27% exposed to bedrock. The catchment is 18.0 $km^2$ in area (lake area = 0.38 $km^2$; mean depth = 16.8 m). Its altitude is 133 m above sea level. The main bedrock type is schist, and soils are on average 30 cm deep. Mean precipitation is 2078 mm $yr^{-1}$ and mean runoff 1655 mm $yr^{-1}$.

There does not seem to be a change in $SO_4$, ALK and SBC concentrations during the 80ies. ALK is relatively high (measured by end point titration; see HOVIND 1996). $NO_3$ values decrease during the period of examination (**Figure 12**). Using the Wilcoxon Signed Rank test to compare the median values for the period 1984-85 and 1988-89 indicates a statistically significant decline (BOWMAN 1997, *pers. com.*; BOWMAN et al. 1996).

*Donegal*

**Lough Veagh** is elevated at 43 m above sea level. Main bedrock type is granite and average soil depth 30 cm. 90% of the vegetation covering the 35 $km^2$ catchment is heather and grassland. Only 4.5% are deciduous trees (lake area = 2.3 $km^2$). Precipitation averages 1400 mm and runoff 950 mm per year. Mean $SO_4$ concentration (1988-95) is 33 $\mu$eq $l^{-1}$. Average $NO_3$ values are also very low: 3 $\mu$eq $l^{-1}$. ALK is about 120 $\mu$eq $l^{-1}$, corresponding to a high pH range of 5.3 to 6.5.

*Galway*

**Lough Maumwee** is a lake of 0.3 $km^2$. The catchment is 4.3 $km^2$ in area, and average runoff is 1700 mm $yr^{-1}$ (mean depth = 2.3 m). Yearly precipitation averages 2150 mm. Catchment area is 4.3 $km^2$. Quartzite and granite are the main bedrock types, and mean soil depth is 50 cm. 80% of the Lough

Maumwee catchment is covered with heather and grassland. 11% are exposed bedrock with wetlands and bogs representing only 4% of the catchment area

$SO_4$ concentrations drop from peak values of 150 µeq $l^{-1}$ in 1984 to minimum values of almost zero in 1987 to 1990. In 1995, the concentrations have rised to 70 µeq $l^{-1}$ again. It seems as if regional factors (in climate?) influence sulphate dynamics more than input by atmospheric deposition. ALK is high (mean = 116 µeq $l^{-1}$), and $NO_3$ concentrations are close to zero during the vegetation period.

## Glendalough, Ireland
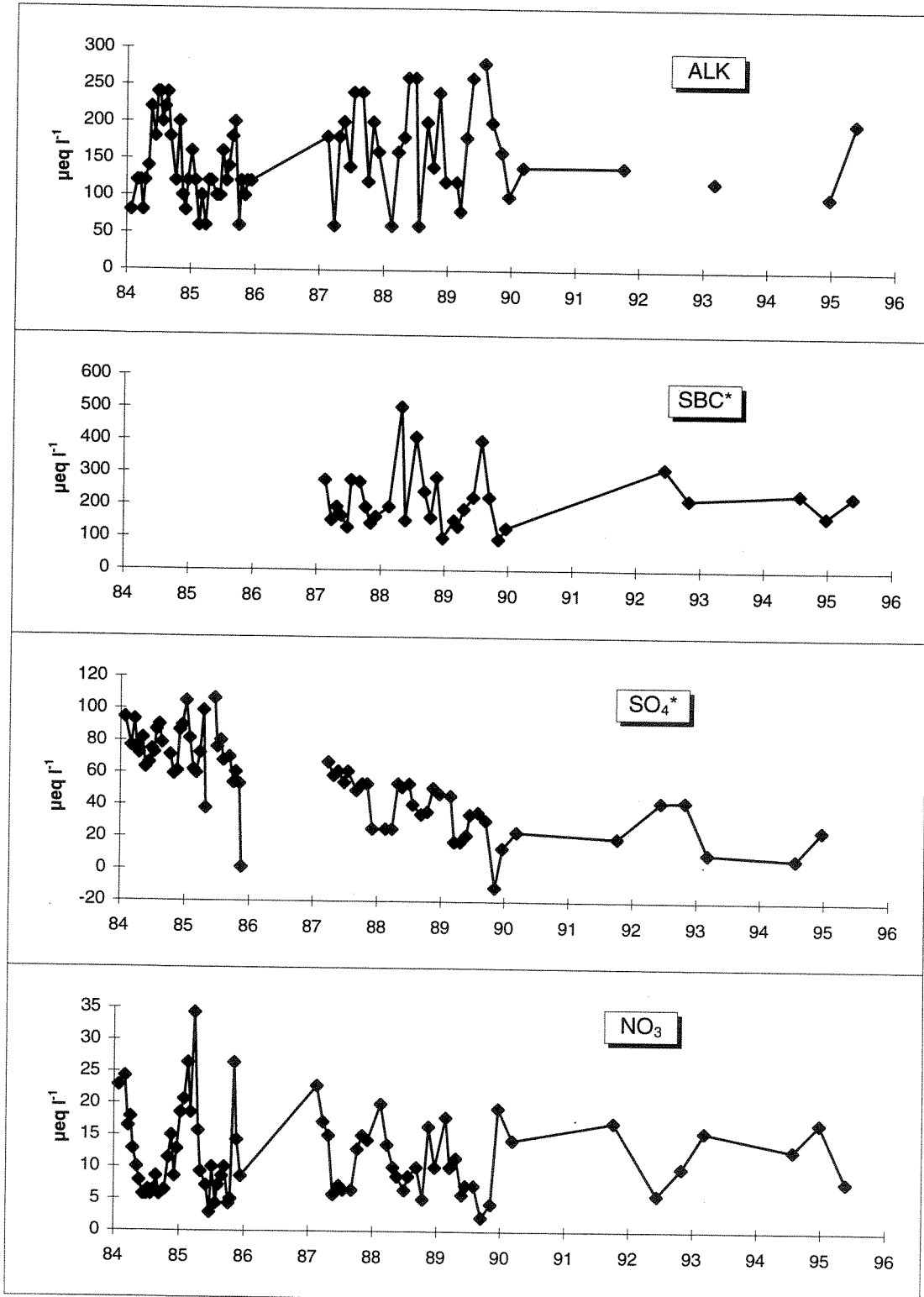


**Figure 12.** ALK, SBC*, SO₄* and NO₃ concentrations at Glendalough (midlake), Ireland.

### 3.4.9 Italy

Italy joined officially the UN ECE ICP on Waters Programme only in 1995. Nevertheless, long-term data series derived from research activities of the Instituto Italiano Di Idrobiologia (National Research Council) could be used for the Nine Year Report.
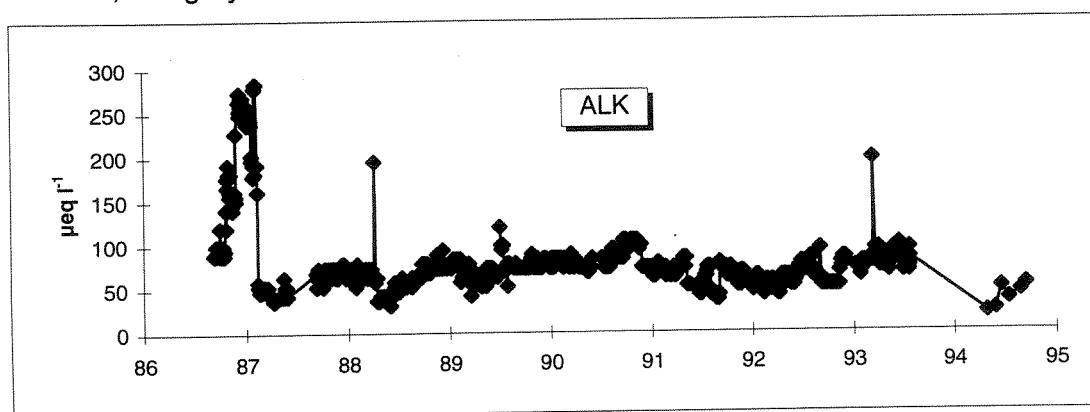
**Table 10.** The Italian sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|-------------|-------------------|--------------|---------------------|
| Piemonte | Pellino (ITA1) | S | 1984-95 | Y |
| Piemonte | Pellesino (ITA2) | S | 1986-95 | Y |
| Piemonte | Cannobino (ITA3) | S | 1979-95 | Y |
| Piemonte | Lago Paione Inferiore (ITA4) | L | 1978-95 | N |
| Piemonte | Lago Paione Superiore (ITA5) | L | 1978-95 | N |
| Piemonte | Lago Mergozzo (ITA6) | L | 1970-95 | N |

The sites are located in North-Western Italy, close to the Swiss border. Atmospheric deposition of pollutants is high in this area (MORSELLO and MARCHETTO 1996).

Two high altitude sites, Lago Paione Inferiore and Lago Paione Superiore, have been intensively investigated within the AL:PE/MOLAR project (EU's Environment and Climate Programme; e.g., WATHNE and PATRICK 1994). River Cannobino has been monitored within the framework of a study on eutrophication of Lake Maggiore. Rivers Pellinio and Pellesino have been studied as part of a project on the recovery of lake Orta.

For all sites measured alkalinity values are available and ion-balances can be used as quality control. Most of the Italian surface waters are not endangered by acid deposition. Carbonate is buffering, and ALK and pH values are rather high. Only a number of high altitude lakes in the Alps and Appenines are sensitive to acidification and some of them are acidified.

*Piemonte*

The catchment of river **Pellino** (17.5 $km^2$) is covered with deciduous forest (83%) and unfertilized pasture (14%). Its elevation is 260 m above sea level. Precipitation is 2000-2100 mm per year (runoff = 1300-1400 mm $yr^{-1}$). Typical bedrock is granite, paragneiss and micaschist.

Pellino (Italy)
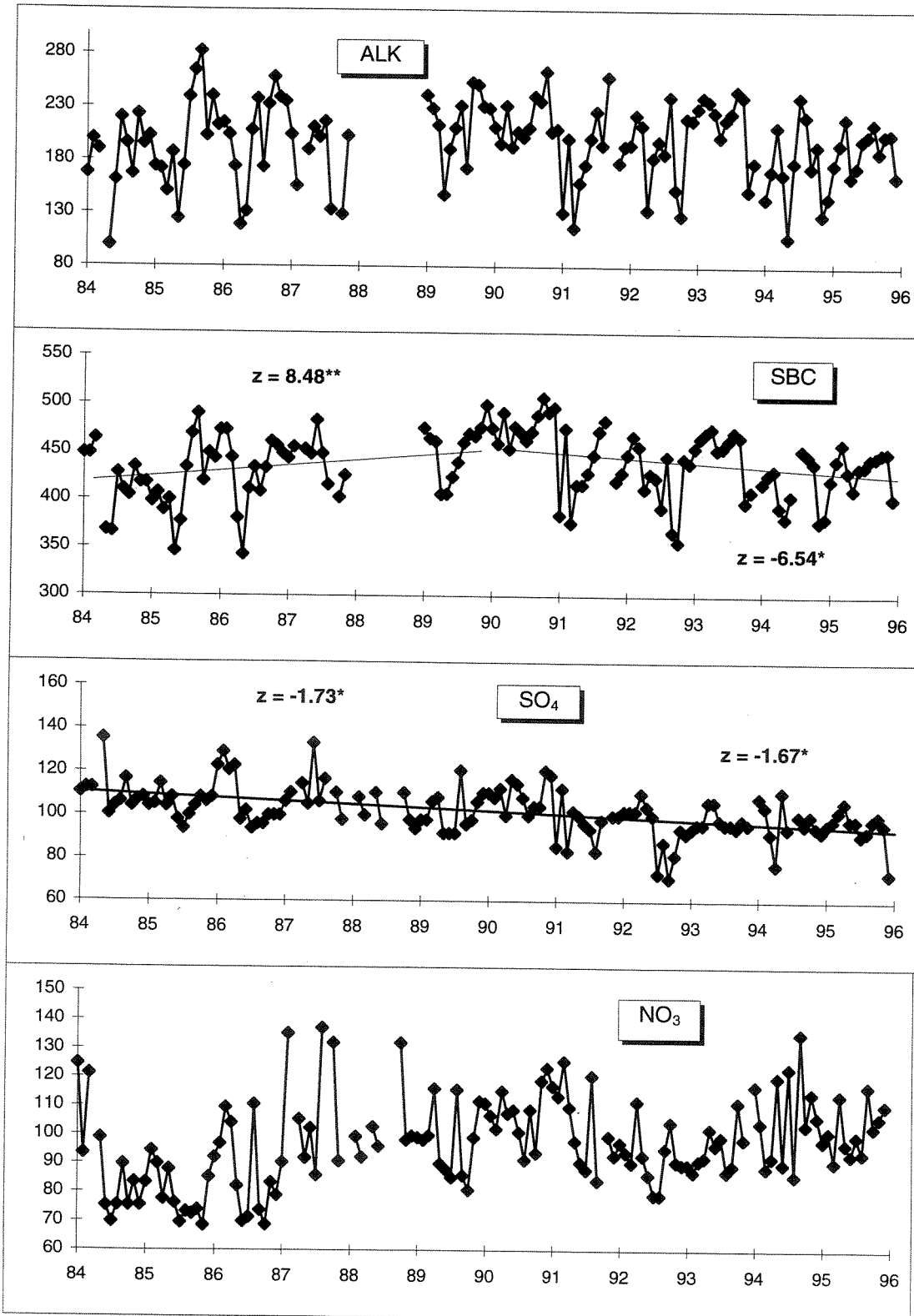


**Figure 13.** ALK, SBC, $SO_4$ and $NO_3$ concentrations at River Pellino, Italy.

$SO_4$ concentrations are significantly decreasing during the 80s and 90s (**Figure 13**). During the 80s SBC trends are positive while they become negative during the 90s. The pH values at Pellino (1984-95) range from 6.6 to 7.6.

River **Pellesino** is located at an elevation of 290 m. Precipitation ranges from 2000 to 2200 mm (average runoff = 1300 - 1400 mm). Most of the area is forested (78%; deciduous), 19% are unfertilized pasture (catchment area = 3.4 $km^2$).

Data of Pellesino were not used for trend analyses, although enough data are available both for the 80s and 90s. Average $SO_4$ concentration in the 80s is 83 $\mu eq\ l^{-1}$ whereas it is 79 $\mu eq\ l^{-1}$ during the 90s. A visual inspection of the data suggests a negative trend in $SO_4$ in the 90s. $NO_3$ concentrations seem not to change during the period of examination (average = 99 $\mu eq\ l^{-1}$). pH values are high, they range from pH 6.6 - 7.6.

Forest cover at river **Cannobino** is 45% (43% coniferous). Main bedrock type is acidic metamorphic and basic rock. Alpine meadows, bare rocks and pature cover the rest of the catchment (elevation = 193 m). Mean precipitation is 2400 mm $yr^{-1}$ and mean runoff 1450 mm.

The only significant changes at Cannobino are increasing $SO_4$ and $NO_3$ concentrations during the 80s (**Figure 14**). The $SO_4$ slope becomes negative during the 90s. ALK as well as pH values are high (mean ALK = 214 $\mu eq\ l^{-1}$, pH range = 6.7 - 8.9).

Both **Lago Paione Inferiore** and **Lago Paione Superiore** are high elevation lakes (2002 or 2269 m above sea level). The catchment areas are 1.14 or 0.55 $km^{-2}$ and most of the catchment is exposed to bedrock (90-93%). Average precipitation is 1450 and 1400 mm. Both lakes have no runoff and are 7.4 and 5.1 m deep.

The lake water concentrations at Lago Paione Inferiore and Lago Paione Superiore could not be used for trend analyses because not enough data for the 90s were available (only two samples in 1995: July and October). Both sites are characterized by low mean ALK values: 0.03 and 0.003 $\mu eq\ l^{-1}$, respectively (WATHNE and PATRICK 1994). Thus, both sites are sensitive to acidification.

The elevation of **Lake Mergozzo** is 194 m. Precipitation averages 2100 mm and runoff 1300 mm. Mean lake depth is 45.4 m, and it is 1.8 $km^{-2}$ wide. 69% of the catchment (10.4 $km^{-2}$) are covered with deciduous forest. At Lago Mergozzo only one sample per year was taken during 1980 to 1995 (mostly in January). Thus, trend analyses was not carried out.

Cannobino (Italy)
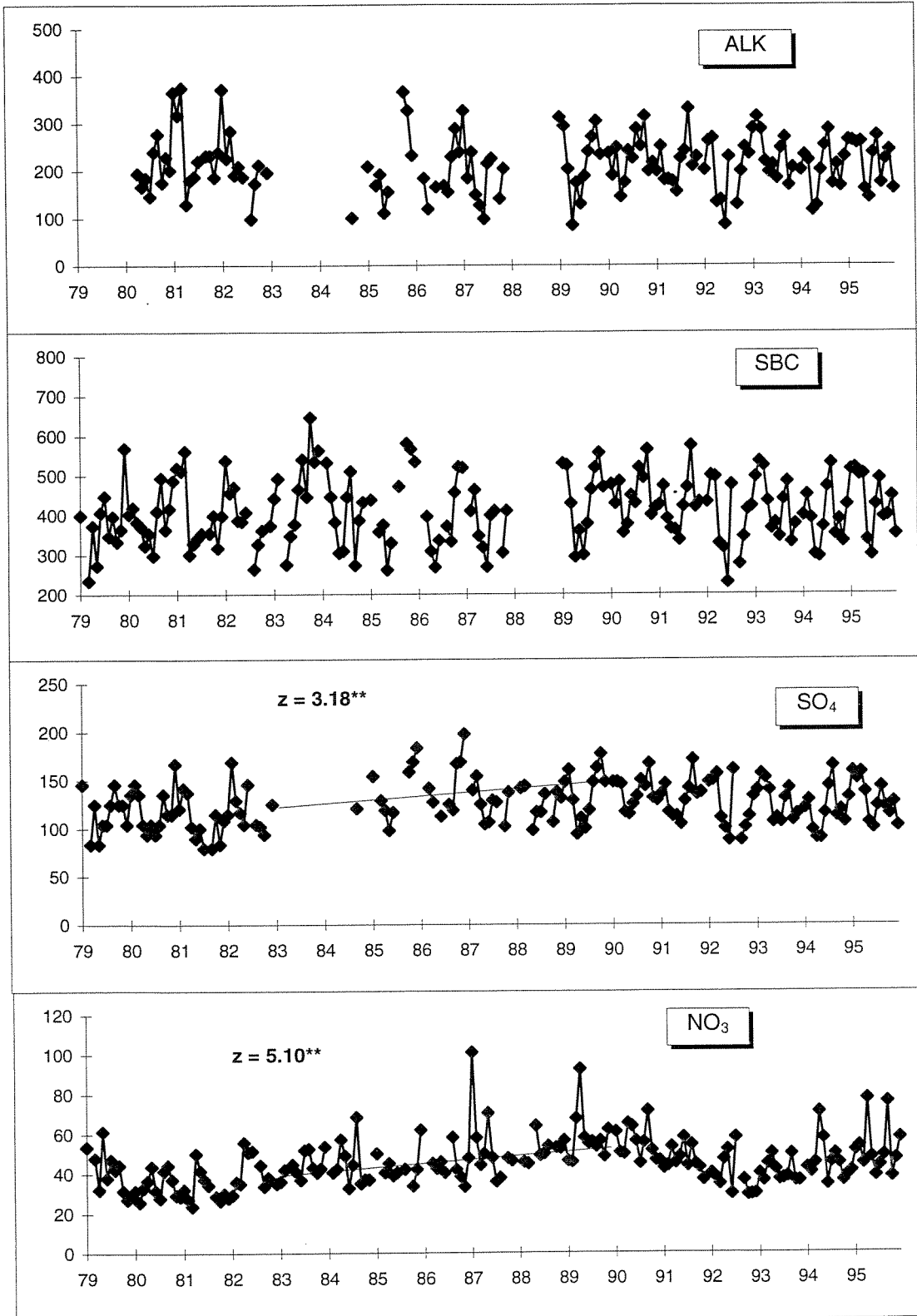


**Figure 14.** ALK, SBC, $SO_4$ and $NO_3$ concentrations at River Cannobino, Italy.

## 3.4.10 Lativa

Data of 12 Latvian sites have been reported to the Programme Center at NIVA. At most places sampling started in 1992 so that results of four years are available today. However, most of the Latvian rivers and lakes are not endangered by acid atmospheric deposition. Alkalinity (or ANC) is high, and pH values exceed 7.0 or even 8.0. Actually, many catchments are influenced by agricultural activities: 15 to 51% of the area may be covered with fertilized farmland. This, of course, has huge effects on, e.g., nitrogen and $SO_4$ concentrations in water. Thus, it was decided to include only three sites into this report. Not enough data were available to run statistical analyses.

**Table 11.** The Latvian sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Latgale | Daugavpils (DAUG-1) | S | 1992-95 | N |
| Vidzeme | Zoseni (ZOS-T) | S | 1992-95 | N |
| Kemeri | Zvirbuli (ZVIR) | S | 1992-95 | N |

*Latgale*

Average precipitation at **Daugavpils** is 633 mm and mean runoff 223 mm yr$^{-1}$. The site's altitude is 86 m above sea level. The catchment is 64500 km$^2$ wide. Main bedrock types are dolomite, clay and gypsum. 28 % of the catchment is covered with forest, 6 % with bogs.

Calculated ANC is extremely high at this site (maximum = 3373 µeq l$^{-1}$). This might be due to the buffering effect of dolomitic (and gypsum) bedrock material. $SO_4$ concentrations seem to be "normal", and $NO_3$ values are relatively high. Since no information on landuse in the surrounding area is available, it is not clear if water chemistry is mainly influenced by acid atmospheric deposition.

*Vidzeme*

**Zoseni** is another typical Latvian stream. Here, 51% of the 33 km$^2$ catchment are fertilized farmland. The rest of the area is mainly covered with forest. Zoseni's altitude is 173 m above sea level. Precipitation averages 727 and runoff 321 mm yr$^{-1}$. Catchment area is 33.5 km$^2$. Bedrock consists mainly of clay, dolomite and gypsum. $SO_4$ concentrations are comparable to those at Daugavpils (**Figure 15**) and $NO_3$ values seem to decrease during the period 1992-94. ANC and pH are quite high (pH ≈ 7.8 - 8.2).

*Kemeri*

The main "bedrock" type at **Zvirbuli** is peat. The catchment's elevation is 8 m, and catchment area 1.6 km$^2$. Average precipitation is 681 mm and runoff 5.4 mm yr$^{-1}$. The former wetland is now covered with pine, heather and rosemary.

Data of 1992 to 1995 are available (3 samples per year). SO$_4$ concentrations average 147 µeq l$^{-1}$. NH$_4$ is the predominant inorganic N compound (mean = 27 µeq l$^{-1}$). The pH ranges from 3.5 - 4.3 and average ANC is low (-123 µeq l$^{-1}$).

Zoseni (Latvia)
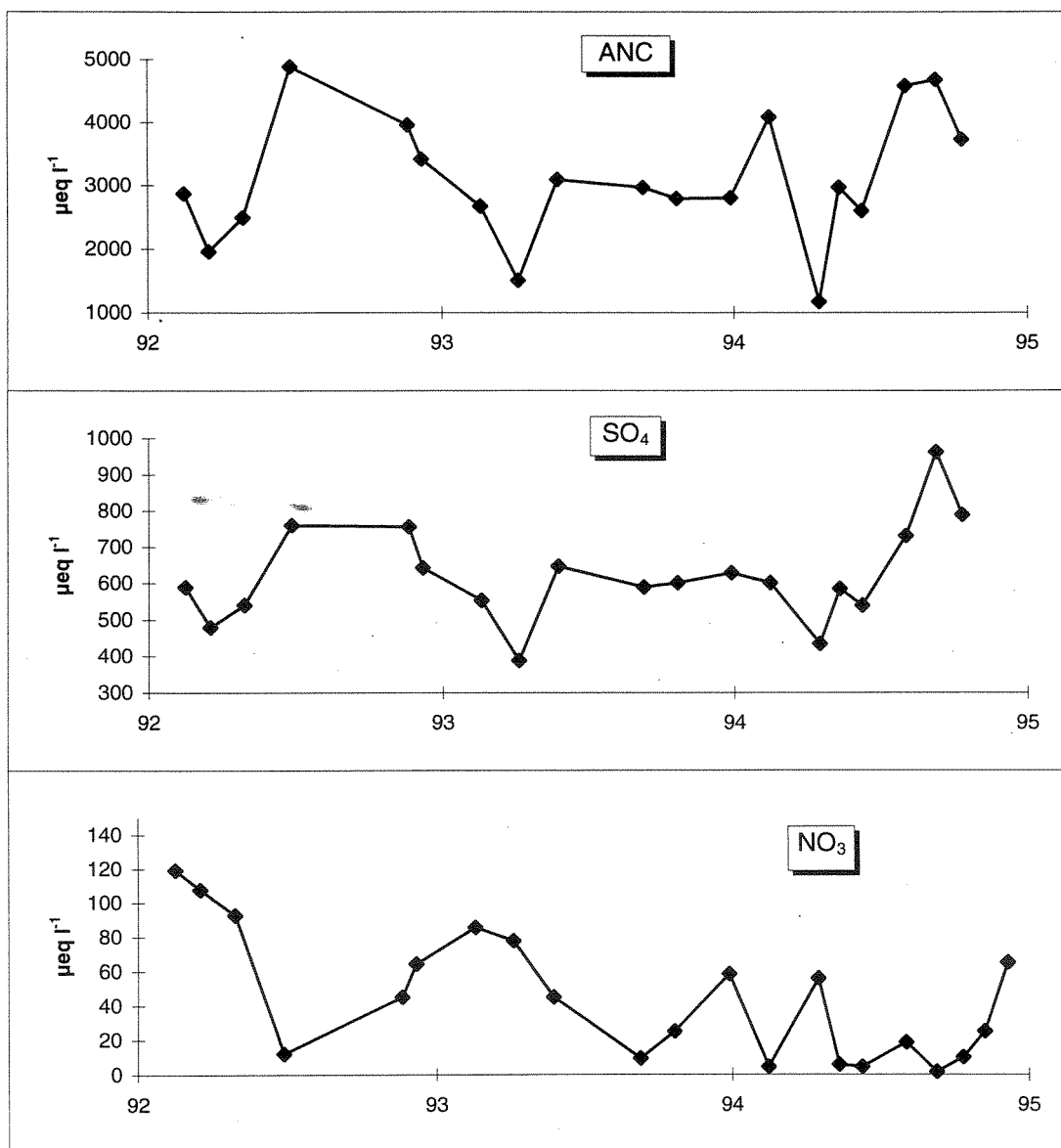


**Figure 15.** ALK, SO$_4$ and NO$_3$ concentrations at Zoseni, Latvia.

### 3.4.11 Norway

Data of seven sites are included into the new database at the Programme Centre, all of them could be used for trend analyses. Typical for many of the Norwegian sites are curvilinear trends especially in alkalinity (ALK), i.e., declining concentrations from 1983 to 1990 and increasing values from 1991 to 1995. The ICP Waters sites summarized in **Table 12** are part of a national monitoring program (SKJELKVÅLE 1996).

**Table 12.** The Norwegian sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|------|-------------------|--------------|---------------------|
| Aust-Agder | Birkenes (*BIE01*) | S | 1972-1995 | Y |
| | Tovdalselva (*OVELV 7 1*) | S | 1980-1995 | Y |
| Buskerud | Langtjern (*LAE01*) | L | 1972-1995 | Y |
| Finnmark | Dalelva (*DALELV 1*) | L | 1988-1995 | Y |
| Oppland | Aurdøla (*OVELV 90 1*) | S | 1986-1995 | Y |
| Rogaland | Vikedalselva (*OVELV 32 9*) | S | 1982-1995 | Y |
| Sogn og Fjordane | Gaular (*OVELV 57 3*) | S | 1980-1995 | Y |
| | Nausta (*OVELV 34 1*) | S | 1980-1995 | Y |
| | Trodøla (*OVELV 34 5*) | S | 1984-1995 | Y |
| Telemark | Storgama (*STE01*) | L | 1974-1995 | Y |

$SO_4$ deposition caused by long-range transport is highest in southern Norway. Thus, the $SO_4$ concentrations are also high in this region. The mean values at Birkenes are 106 µeq $l^{-1}$ for the 80s (1983-90) and 88 µeq $l^{-1}$ for the 90s (1991-95). At Nausta in western Norway average $SO_4$ concentrations are much lower: 21 µeq $l^{-1}$ in the 80s and 16 µeq $l^{-1}$ in the 90s. While alkalinity is negative at Birkenes (80s = -42 and 90s = -36 µeq $l^{-1}$), it is positive at Nausta (80s = 12 and 90s = 15 µeq $l^{-1}$). $NO_3$ concentrations are low at most ICP Waters sites. The curves show a typical seasonality, mostly with values near zero during the vegetation period.

*Aust-Agder (southern Norway)*

The **Birkenes** and **Tovdalselva** catchments are located in southern Norway (elevation: 250 and 500 m above sea level). The catchments are 0.41 and 1888 $km^2$ in area. Main bedrock type is granite and gneiss. At Birkenes most of the catchment is covered with forest (80 % coniferous and 10 % deciduous). Soils are on average 50 cm deep. 49 % of the area at Tovdalselva is exposed to bedrock, 45% is afforested (mostly coniferous). At both sites precipitation averages 1400 and runoff 1100 mm per year.

$SO_4$ concentrations at Tovdalselva decrease significantly both during the 80s and the 90s (**Figure 16**). At Birkenes a downwards trend can only be observed in the 80s. The decline in $SO_4$ is accompanied by significant increases in ALK at both catchments during the 80s as well as during the 90s. From 1983 to 1989 there is a negative trend in SBC values, while the slopes are positive in the 90s (significant increase at Tovdalselva). Trends in $H^+$ concentrations are negative at Tovdalselva (80s and 90s). At both sites $NO_3$ concentrations are increasing sgnificantly in the 80s, and at Birkenes a negative trend during the 90s can be observed. While $NO_3$ values at Birkenes are near zero in summer, the usually do not drop below the 5 µeq $l^{-1}$ line at Tovdalselva (**Figure 16**).

*Buskerud (south-eastern Norway)*

The catchment area of **Langtjern** is 4.8 $km^2$, most of it is covered with heather and grassland (69 %) and wetlands (16 %). The lake's elevation is ca. 630 m above sea level. It is 0.22 $km^2$ in area and on average 2.0 m deep. Main bedrock type is gneiss and soils are very shallow (ca. 50 cm). Precipitation averages 685 and runoff 550 mm $yr^{-1}$.

During the whole period of examination $SO_4$ concentrations are declining significantly (mean value in the 80s = 61 µeq $l^{-1}$, in the 90s = 54 µeq $l^{-1}$). ALK shows a negative trend in the 80s and a positive in the 90s (average 80s = 15, 90s = 32 µeq $l^{-1}$). SBC are only significantly decreasing during the 80s. The same is true for $H^+$ in the 90s.

*Finnmark (northern Norway)*

The elevation of the **Dalelva** catchment ranges from 10 - 241 m above sea level. Gneiss is the predominant bedrock type, and mean soil depth is 50 cm. Catchment area is 3.2 $km^2$. Precipitation averages 540 mm, and runoff 360 mm per year. Most of the catchment is covered with heather and grassland (ca. 60%).

$SO_4$ values are significantly decreasing both in the 80s and 90s (mean 90s = 97 µeq $l^{-1}$). The ALK, SBC and $NO_3$ trends are negative during the 80s and positive during the 90s. ALK is relatively high, 49 µeq $l^{-1}$ on average in the 90s. $H^+$ shows the opposite development: a significant increase in the 80s and a decline in the 90s.

Tovdalselva (Norway)
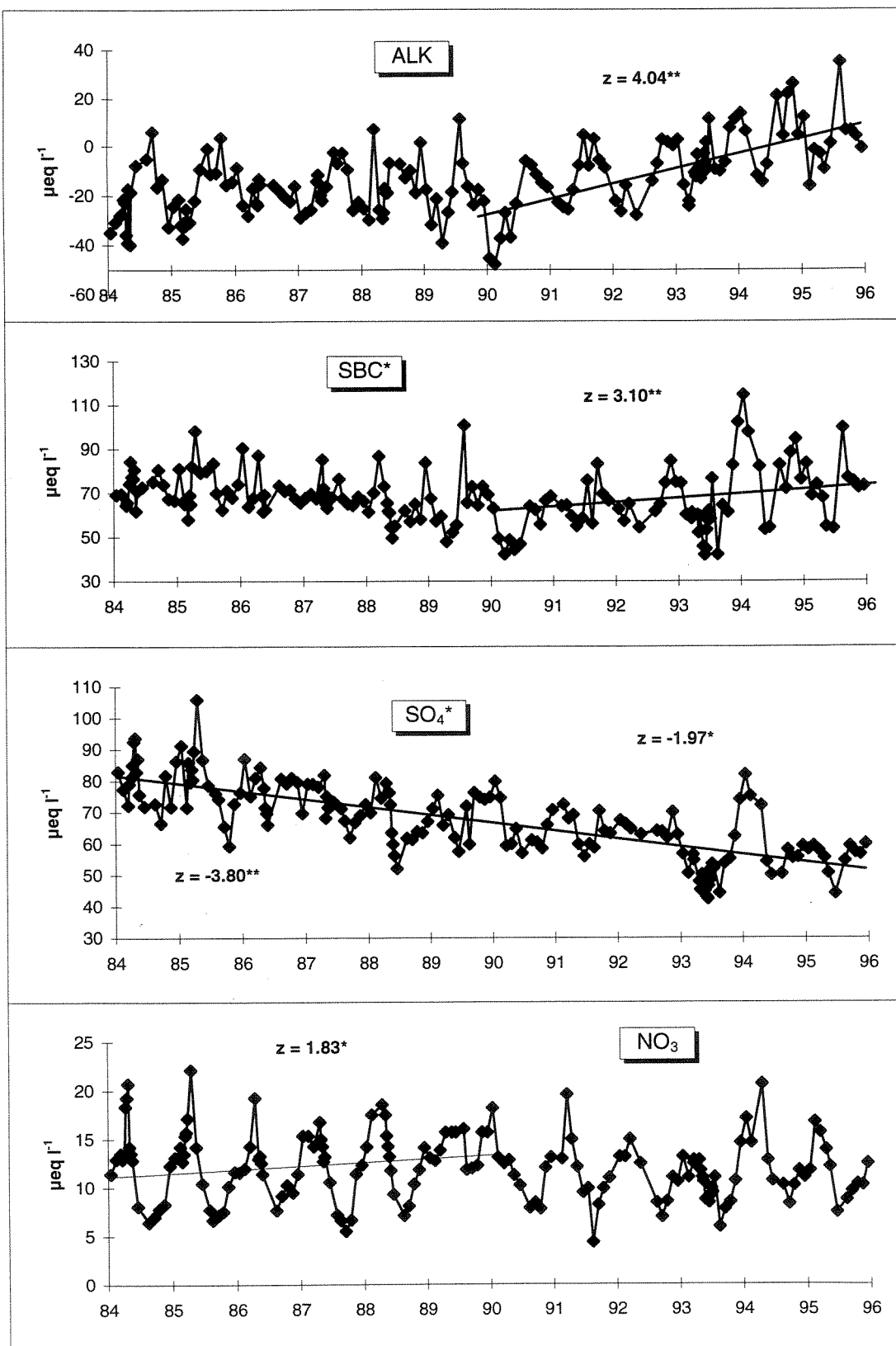


**Figure 16.** ALK, SBC, $SO_4$ and $NO_3$ concentrations at Tovdalselva, southern Norway.

*Oppland (south-eastern Norway)*

**Aurdøla** is a stream draining a 278 km$^2$ catchment (elevation 320 - 1285 m above sea level). Average precipitation is 685 mm yr$^{-1}$ (runoff = 550 mm yr$^{-1}$). Main bedrock type is granite. About half of the catchment area is afforested (46 % exposed to bedrock), two thirds are coniferous forest, one third deciduous.

A negative trend in SO$_4$ concentrations can only be observed during the 90s, accompanied by significantly increasing ALK and SBC values.


*Rogaland (south-western Norway)*

The catchment of **Vikedalselva** is 119 km$^2$ in area. The stream's elevation is 60 - 1118 m above sea level. Granite/gneiss is the predominante bedrock type. Precipitation averages 1800 mm per year. Most of the catchment is exposed to bedrock (about 65 %), 24% is covered with forest (mostly coniferous). Small areas with wetlands and bogs can be found.

At Vikeldalselva decreases in SO$_4$ are significant in the 80s, but not in the 90s. ALK and SBC show positive trends in the 90s and H$^+$ a negative one.


*Sogn and Fjordane (western Norway)*

Three streams are located in the Sogn and Fjodane region: **Gaular**, **Nausta** and **Trodøla**. The elevation of these streams ranges from 0 to 1290 m above sea level. Forest cover at Trodøla and Nausta is 50% (ratio deciduous to coniferous = 1:1), the rest of the area is exposed to bedrock (mainly gneiss). At Nausta 74 % of the catchment are not covered with soils (main bedrock type = gneiss). 19 % of the area are afforested and about half of the trees are deciduous, the other half coniferous. Precipitation is quite high at all three sites (yearly average = 3600 mm) and runoff averages 2400 mm. The catchment area of Trodøla and Gaular is 10 km$^2$, and it is 273 km$^2$ at Nausta.

No significant changes in SO$_4$ concentrations (1983-1995) can be observed at Gaular and Nausta, while there is a negative trend at Trodøla during the 80s. Average SO$_4$ value at Nausta, e.g., is 19 µeq l$^{-1}$. During the 90s ALK is significantly increasing at Nausta, Trodøla and Gaular (at Gaular also in the 80s). At Nausta mean ALK is 12 µeq l$^{-1}$ during the 80s and 15 µeq l$^{-1}$ in the 90s. There is a negative SBC trend at Gaular in the 80s. At Gaular and Nausta H$^+$ increases in the 90s (at Nausta also in the 80s). Only at Gaular NO$_3$ values are significantly increasing in the 90s. After 1989 the NO$_3$ concentration drops seldom below 3 µeq l$^{-1}$ during the vegetation period.

*Telemark (south-eastern Norway)*

The catchment area of **Storgama** is 0.6 km$^2$ (elevation: 580 - 690 m above sea level). Precipitation averages 1050 mm yr$^{-1}$ and runoff 950 mm yr$^{-1}$. Only 11 % of the catchment area is afforested (deciduous : coniferous = 1:1), 22 % are covered with heather and grassland and 18 % are exposed to bedrock (granite, gneiss).

The SO$_4$ concentrations at Storgama are decreasing significantly both during the 80s and 90s (**Figure 17**). This decline is accompanied by a significant rise in ALK in the 90s, while SBC values decrease during this period. The H$^+$ trend is negative during the 80s and positive in the 90s.

Storgama (Norway)
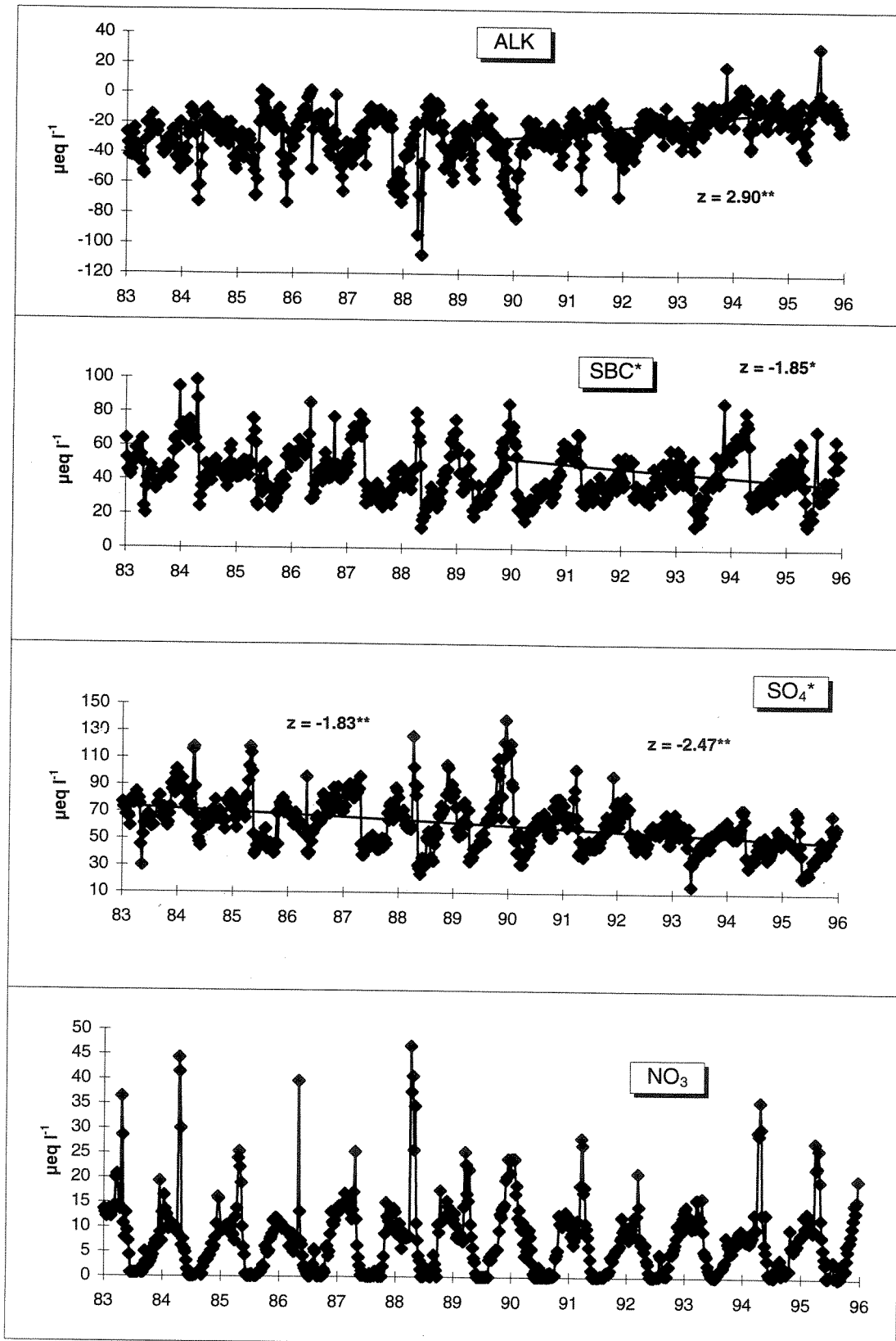


**Figure 17.** ALK, SBC, $SO_4$ and $NO_3$ concentrations at Storgama, south-eastern Norway.

## 3.4.12 Poland

Poland has reported data of two ICP Waters sites, both of them in the Tatra Mountains. Trend analyses was not possible because sampling did not begin before 1992. Ion balances can be calculated and used as quality control.

**Table 13.** The Polish sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Tatra Mountains | Dlugi Staw Gasienicowy (DLUGI) | L | 1992-95 | N |
| | Zielony Staw Gasienicowy (ZIELON) | L | 1992-95 | N |

*Tatra Mountains*

The dominant bedrock types at both Polish sites are granite and gneiss. Soils are shallow, 10 to maximal 50 cm deep. 72 - 92% of the catchment areas are exposed to bedrock. The catchment is covered sparcely with dwarf mountain-pine.

**Dlugi Staw Gasienicowy** is a lake, 0.02 $km^2$ in area, draining a catchment of 0.7 $km^2$. Its average depth is 5.1 m. Mean yearly precipitation is 1837 mm, and runoff is high: 1654 mm $yr^{-1}$.

During the last three years there was obviously no change in $SO_4$ concentrations (**Figure 18**). The same is true for $NO_3$ . Nitrate values show a seasonality but even in summer they do not drop below 30 $\mu eq$ $l^{-1}$, whereas alkalinity may almost be zero. Average pH is 5.9 and may drop to 5.1 during snowmelt periods. ALK seems to become relatively high when $SO_4$ and $NO_3$ concentrations are also increasing. The $NO_3$ dynamics at both Polish catchments is described in detail by RZYCHON and WORSZTYNOWICZ (1995).

Precipitation at **Zielony Staw Gasienicowy** averages 1795 and runoff 1544 mm $yr^{-1}$. The lake's mean depth is 7.6 m and its area 0.04 $km^2$. The whole catchment is 0.5 $km^2$ wide.

$SO_4$, $NO_3$ and ALK concentrations are almost a steady line (1992-95; **Figure 19**). $NO_3$ values show a seasonality, but are never lower than 10 $\mu eq$ $l^{-1}$.
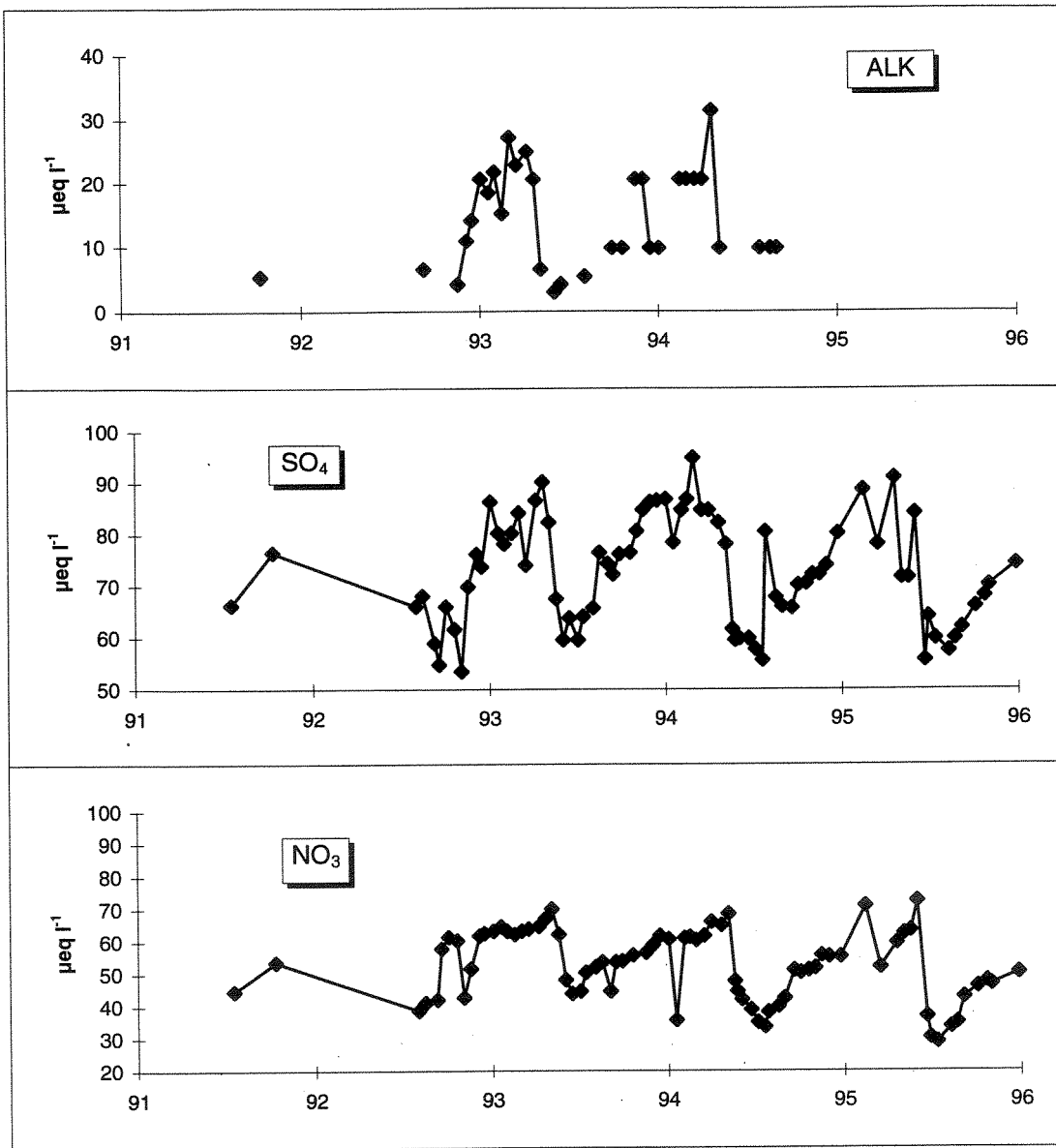
Dlugi (Poland)



**Figure 18.** ALK, $SO_4$ and $NO_3$ concentrations at Dlugi Staw, Poland.

Zielony (Poland)
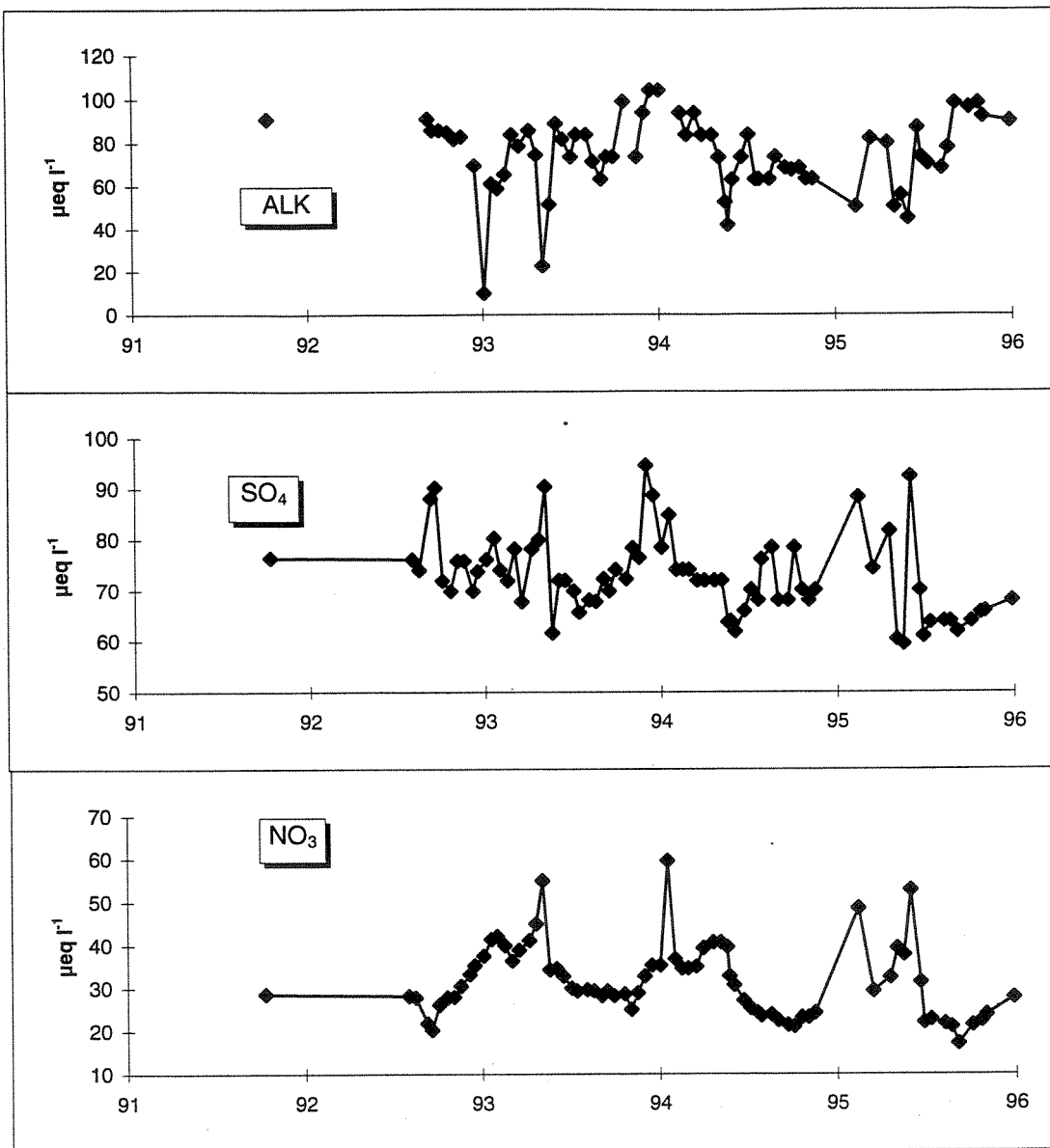


**Figure 19.** ALK, SO₄ and NO₃ concentrations at Zielony Staw, Poland

## 3.4.13 Russia

Water chemistry data of two sites are available at NIVA, but only those of Baikal were read into the new database. The $SO_4$ concentrations at Kola (northern Russia, close to the Finnish border) are extremely high. They range from 1190 to 2660 µeq $l^{-1}$, and alkalinity is also very high (about 160 µeq $l^{-1}$). For Baikal there were not enough data available to run trend analyses.

**Table 14.** The Russian site included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|---------------|-------------------|--------------|---------------------|
| Baikal | Baikal (*BAL2*) | L | 1991-94 | N |

### Baikal

$SO_4$ concentrations at Baikal did not change during the period 1991 to 1994 (**Figure 20**). The same is true for measured alkalinity (ALK) showing relatively high mean values of 69 µeq $l^{-1}$. $NO_3$ concentrations are low, showing a typical seasonality with concentrations close to zero during the vegetation period (average = 2.2 µeq $l^{-1}$).

## 3.4.14 Sweden

Data of 9 Swedish sites were read into the new database. For S01 to S04 data from 1988 to 1990 are missing. Thus, not for all sites separate trend analyses for the 80s and 90s could be run. Seasalt corrected values and measured ALK were used (see also PERSSON 1996).

**Table 15.** The Swedish sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|--------|---------------|-------------------|--------------|---------------------|
| Eastern Sweden | Delaangeraan, Iggesund (*S01*) | R | 1984-95 | Y |
| | Tvaeringen (*S05*) | L | 1984-95 | Y |
| | Stensjoen (*S06*) | L | 1985-95 | Y |
| Southeastern Sweden | Stroemsborg (*S03*) | R | 1985-95 | N |
| | Brunnsjoen (*S08*) | L | 1984-95 | Y |
| | Fiolen (*S09*) | L | 1984-95 | Y |
| | Storasjoen (*S10*) | L | 1984-95 | Y |
| Southwestern Sweden | Haersvatn (*S04*) | L | 1984-95 | Y |
| | Fraecksjoen (*S11*) | L | 1984-95 | Y |

## Baikal (Russia)
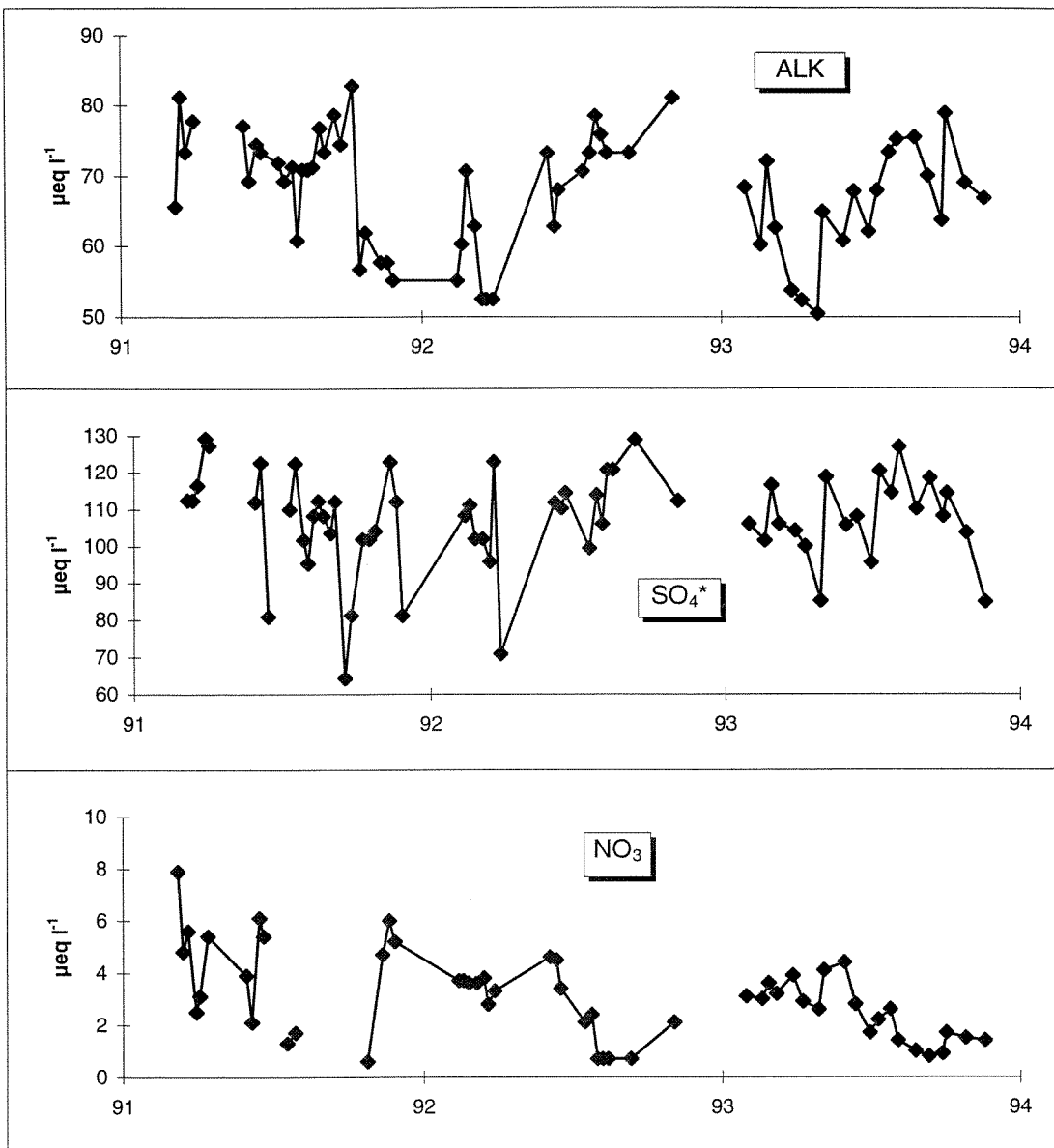


**Figure 20.** ALK, $SO_4$ and $NO_3$ concentrations at Baikal, Russia.

*Eastern Sweden*

**Delaangeraan** is a river draining a catchment of 1992 km$^2$.

$SO_4$* concentrations are significantly decreasing during the 90s (**Figure 21**). While ALK and SBC* show a negative trend during the 80s, ALK is increasing between 1991 and 1995. The opposite is true for $H^+$ values. A significant increase in $NO_3$ in the 80s and negative trend in the 90s can be observed. $NO_3$ values are close to zero during the vegetation period, except in 1986 and 1987 where they do not drop below 5 $\mu$eq l$^{-1}$.

Granite and gneiss are the main bedrock type at **Tvaeringen**. The catchment area is 32.6 km$^2$ and the lake is 0.38 km$^2$ in area. Elevation of Tvaeringen is 308 m above sea level. 83% of the area is covered with forest, most of it conferous (6% wetlands and bogs).

$SO_4$* as well as ALK concentrations are decreasing significantly during the 90s. $H^+$ values show a positive trend during the 80s. SBC* concentrations decline significantly from 1991-1995. The $NO_3$ curve shows a typical seasonality with higher peak values during the 80s compared to the 90s.

**Stensjoen** is a 3.7 km$^2$ catchment at an elevation of 268 m. The lake is on average 4.2 m deep and 0.6 km$^2$ in area, the whole catchment area is 3.7 km$^2$. 68% of the catchment are forests, 19% wetlands and bogs.

$SO_4$* and SBC* concentrations at Stensjoen show a negative trend during the 90s.

*Southeastern Sweden*

**Stroemsborg** river has a mean yearly runoff of 260 mm. The elevation of the 247 km$^2$ catchment is 168 m. Total forest cover is 87%.

There were not enough data available to run trend analyses. The $NO_3$ curve shows a typical seasonality with peak values of ca. 30 $\mu$eq l$^{-1}$.

**Haersvatn** is a 0.19 km$^2$ lake at an elevation of 129 m (mean depth = 5 m). Mean yearly runoff is 570 mm and average depth 5.7 m. The catchment is 2.2 km$^2$ in area and by 68% covered with forest. 13% are wetlands and bogs.

$SO_4$* and $NO_3$ concentrations are significantly decreasing during the 90s (**Figure 22**). Minium $NO_3$ values during summer are about 5 $\mu$eq l$^{-1}$ (seasonal pattern).

85% of the 2.7 km$^2$ catchment area at **Brunnsjoen** are covered with forest. The elevation of the lake (0.1 km$^2$) is 98 m. Runoff averages 220 mm yr$^{-1}$.

$SO_4$* concentrations decrease significantly during the 90s. $NO_3$ values, significantly decreasing between 1984 and 1990, are low during the vegetation period ($\approx$ 3 $\mu$eq l$^{-1}$).

Elevation of **Fiolen** is 226 m above sea level. Catchment area is 5.5 km$^2$ and lake area 1.6 km$^2$. Mean lake depth is 3.8 m and yearly runoff 315 mm. Only 40% of the catchment is covered with forest, 19% is farmland (rest = heather and bogs).

While there is a positive trend in $SO_4$* concentrations in the 80s, the $SO_4$* curve during the 90s is almost a straight line at a relatively high level ($\approx$ 200 $\mu$eq $l^{-1}$). ALK and SBC* values are significantly declining during the 80s. $NO_3$ concentrations show a positive trend during this period. $H^+$ values are significantly decreasing in the 90s.

**Storasjoen** is a lake at an elevation of 252 m above sea level. The catchment is 3.0 $km^2$ in area. It is covered by 83% with forest, 90% is coniferous. Lake area is 1.8 $km^2$, and its mean depth is 1.8 m. Runoff averages 330 mm $yr^{-1}$.

$NO_3$ values show a positive trend during the 90s, although concentrations stay close to zero during the vegetation periods.


*Southwestern Sweden*

Catchment area of **Fraecksjoen** is 4.3 $km^2$. Average runoff of the 0.3 $km^2$ lake is 500 mm $yr^{-1}$. (mean depth = 6.0 m). Forest covers 79% of the catchment, 8% of the area are wetlands and bogs. The altitude of Fraecksjoen is 58 m.

As at many other Swedish ICP Waters sites, $SO_4$* concentrations are significantly decreasing during the 90s. At Fraecksjoen this development is accompanied by a positive trend in ALK values. SBC* show a negative, $NO_3$ a positive trend during the 80s.

Delaangersaan (Sweden)
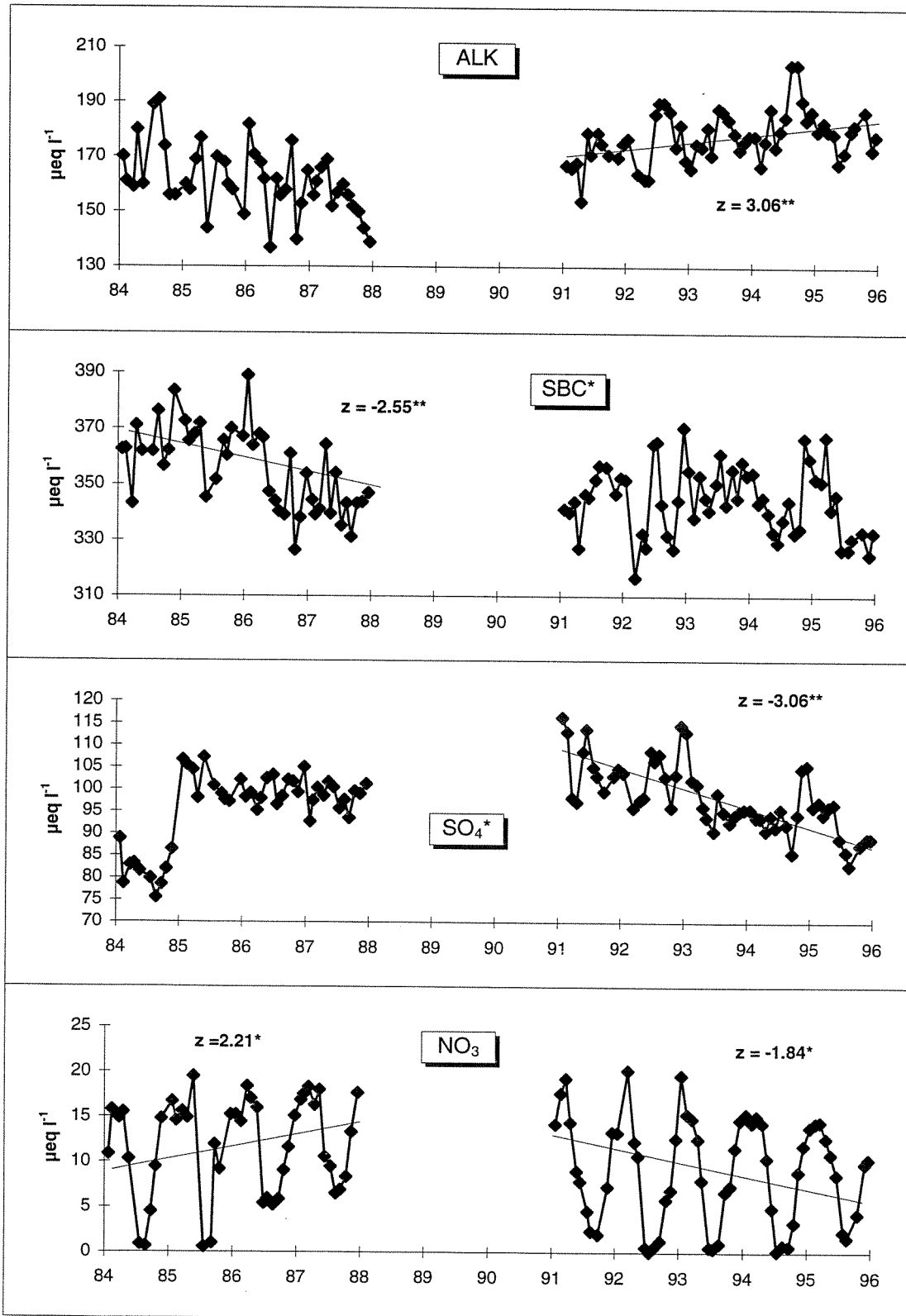


**Figure 21.** ALK, SBC*, SO$_4$* and NO$_3$ concentrations at Delaangeraan.

Haersvatn (Sweden)
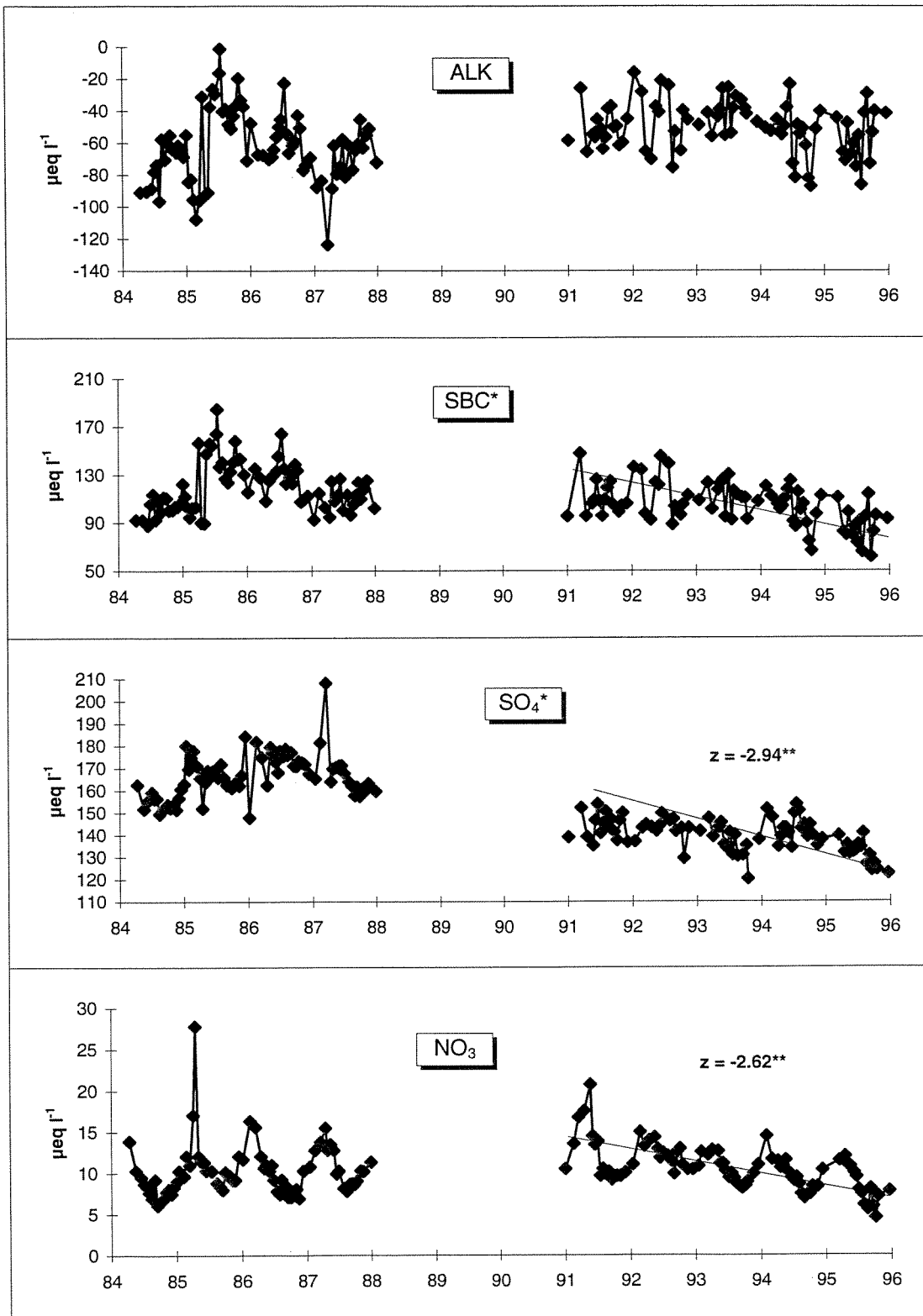


**Figure 22.** ALK, SBC*, SO$_4$* and NO$_3$ concentrations at Haersvatn.

### 3.4.15 The Netherlands

The national monitoring program in The Netherlands was stopped in 1994. Nevertheless, data of 1995 were reported to the Programme Centre, based on a scientific cooperation between AquaSense TEC Wageningen and NIVA. Results of five ICP Waters sites have been reported. Since water chemistry is very similar at all three sampling sites at Achterste Goorven, only station E with the longest data series (1984-95) was chosen to be included into the new database.

Additional to high acidic atmospheric deposition, $SO_4$ and nitrogen concentrations are also influenced by catchment-internal reduction/oxidation processes, induced by short-term changes in climate.

**Table 16.** The Dutch ICP Waters sites included into this report.

| Region | Site (Code) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Noord-Brabant | Achterste Goorven (AGE) | L | 1984-95 | N |
| Gelderland | Gerritsfles (GER) | L | 1984-95 | Y [*] |
| Drenthe | Kliplo (KLI) | L | 1984-95 | N |

[*] When trend analyses were run only data until 1994 were available.

For all sites ion-balanced were calculated and used as quality control, e.g., **Figure 23**.
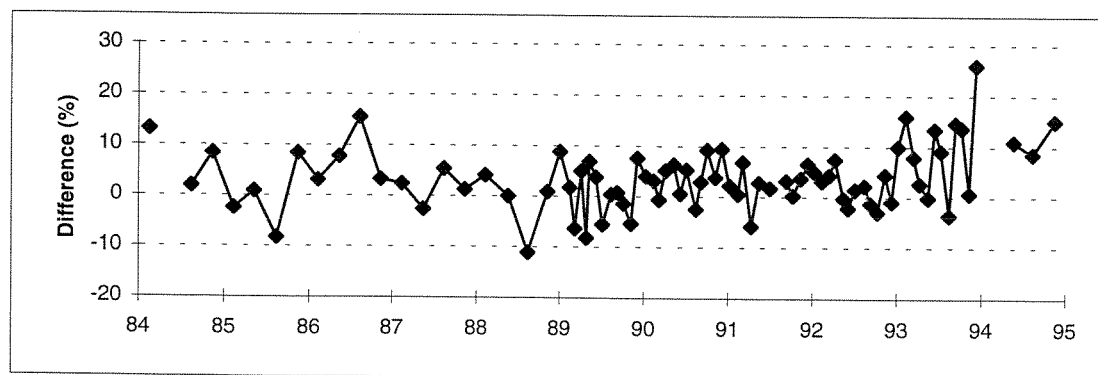


**Figure 23.** Ion-balance Gerritsfles (Netherland).

*Noord-Brabant*

**Achterste Goorven** is a small lake of 0.02 $m^{-2}$ at an elevation of only 8 m. The mean yearly runoff is 265 mm (average precipitation = 794 mm). The podsolic soils, underlain by sand, are about 100 cm deep. The pond is surrounded by a sparce pine/oak forest, interrupted by areas of heather and grassland.

The bottom of Achterste Goorven may dry up during warm summers (VAN DAM et al. 1996). Reduced sulphur and nitrogen compounds are being oxidized during these periods. At rewetting this may overrule the effects of acid deposition on sulphate and nitrate concentrations in lake water and influence as well other parameters (**Figure 24**). $SO_4$* and SBC* concentrations increase dramatically after 1990, accompanied by a decrease in ALK. $NH_4$ is the predominent inorganic nitrogen species.

*Gelderland*

The catchment of lake **Gerritsfles** (0.06 km$^2$) is mainly covered with heather, grassland and bogs with some pine and oak trees. Small patches of drift sand are covering about 10% of the area. The sandy soils are podsols ($\approx$ 100 cm deep). Precipitation averages 870 mm yr$^{-1}$, runoff 188 mm. The elevation of Gerritsfles is 40 m. Although the water level was low during the warm summers in 1986, 89 and 90, the lake did not dry up completely.

Data of Gerritsfles were used for trend analyses. For the 90s a downwards trend in $SO_4$* concentrations can be observed (**Figure 25**). ALK shows a significant increase during the 90s. H$^+$ concentrations are increasing in the 80s and declining in the 90s. $NO_3$ shows a negative trend in the 80s.

*Drenthe*

**Kliplo** is a very small lake (0.007 km$^2$) surrounded by birch trees. Its elevation is 12 m above sea level. 40% of the catchment area is covered with heather, 30 % is drift sand and 5% bog. Mean precipitation is 892 mm yr$^{-1}$ and runoff 248 mm yr$^{-1}$. During dry years up to 20% of the pool bottom may be exposed to the atmosphere, which seems to have a big influence on the shape of $SO_4$* and nitrogen curves.
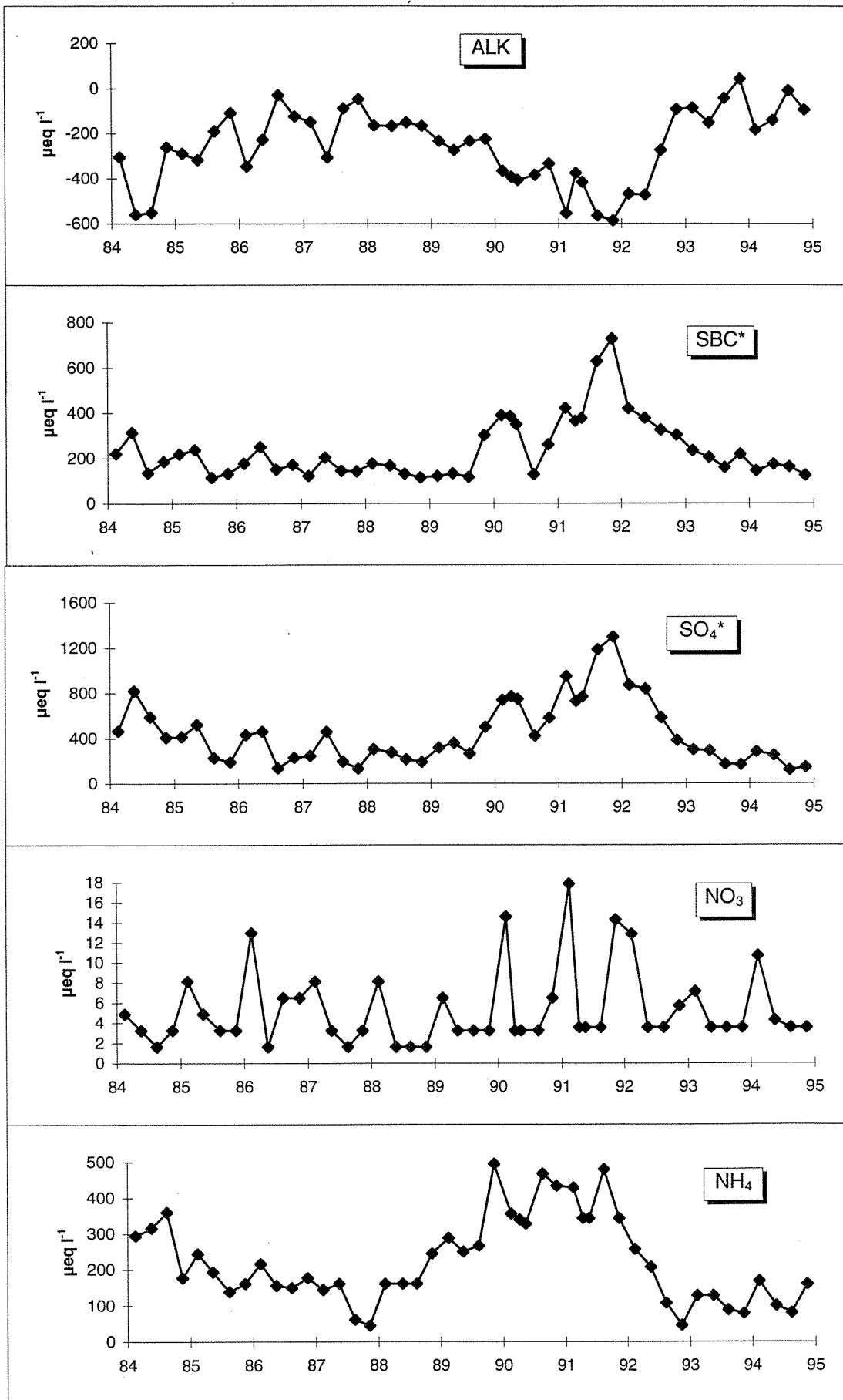
**Figure 24.** ALK, SBC\*, SO$_4$\*, NO$_3$ and NH$_4$ concentrations at Achterste Goorven E., The Netherlands.
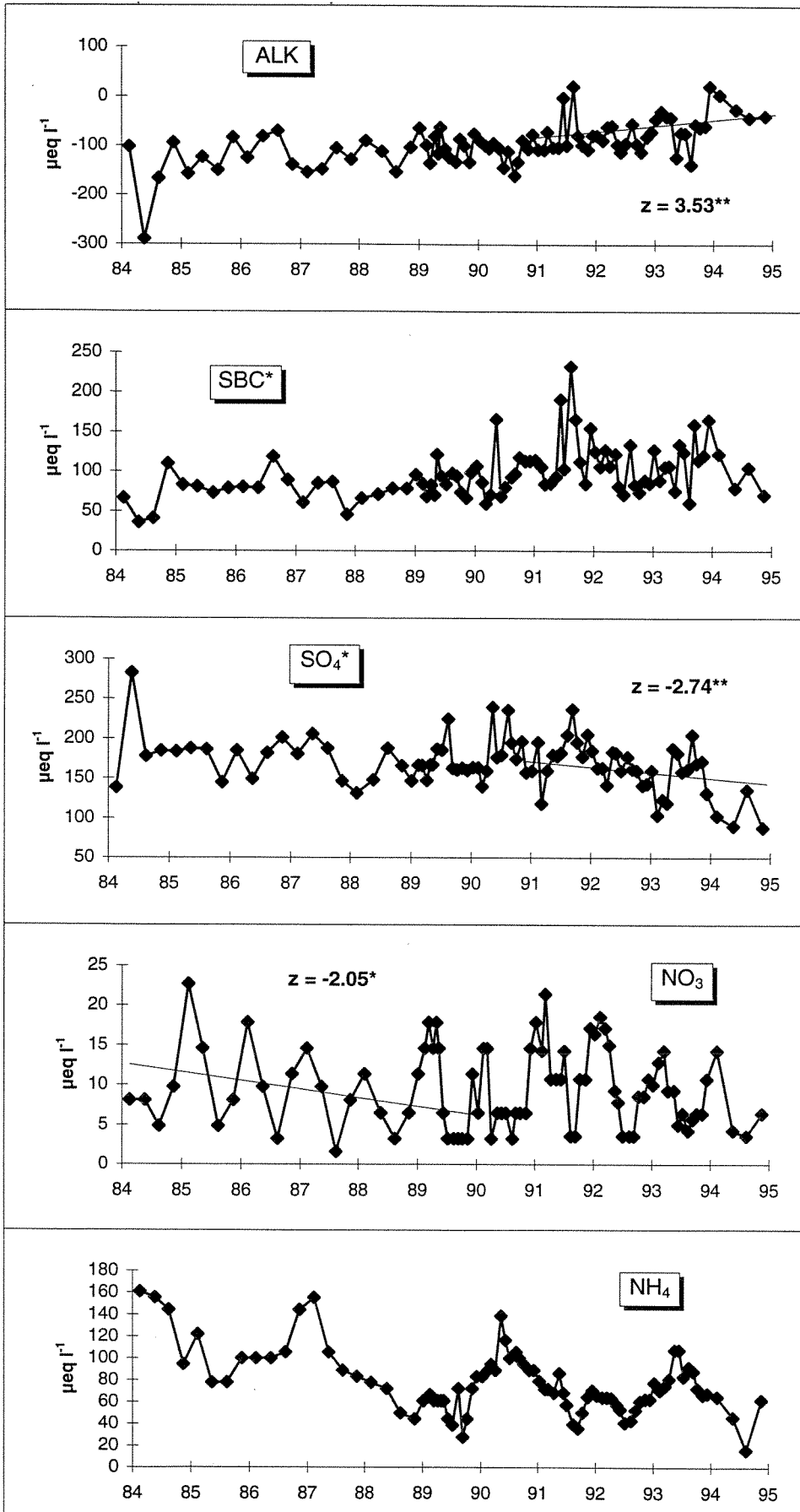
**Figure 25.** ALK, SBC*, SO$_4$*, NO$_3$ and NH$_4$ concentrations at Gerritsfles, The Netherlands.

### 3.4.16 United Kingdom

Data of all 6 ICP Waters sites in the U.K. have been read into the new database. Four samples per year are analyzed (spring, summer, autumn, winter). Trend analyses for the 90s could be run. The British ICP sites are part of a national monitoring program and described in detail by RENSHAW (1995).

**Table 16.** British ICP Waters sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Wales | Llyn Llagi (*UK15*) | L | 1988-95 | Y (90s) |
| England | Scoat Tarn (*UK10*) | L | 1988-95 | Y (90s) |
| Northern Ireland | Blue Lough (*UK21*) | L | 1990-95 | Y (90s) |
| Scotland | Round Loch of Glenhead (*UK07*) | L | 1988-95 | Y (90s) |
| | Lochnagar (*UK04*) | L | 1988-95 | Y (90s) |
| | Loch Coire nan Arr (*UK01*) | L | 1988-95 | Y (90s) |

Ion-balances of all sites could be calculated. If the difference was larger 20%, the values were not used for statistical analyses. Because monitoring started in 1988/89 trend analyses of $SO_4*$, $NO_3$, SBC*, ALK and $H^+$ concentrations were only run for the 90s (seasalt corrected values).

*Wales*

**Llyn Llagi** is the Welsh ICP Waters site. Average soil depth is 88 cm (podsols, gleys), and mean lake depth 5.8 m. The lake is 0.06 $km^2$ in area. Main bedrock types are slates and shales. Precipitation averages 2436 mm $yr^{-1}$ and runoff is high: 2192 mm $yr^{-1}$. The elevation is 380 m above sea level.

Trend analyses do not show any change in $SO_4*$ concentrations (mean = 40 µeq $l^{-1}$). ALK values decrease significantly (average: 49 µeq $l^{-1}$ in 1990 and 8 µeq $l^{-1}$ in 1994/95).

*England*

**Scoat Tarn** is located in the Lake District at an elevation of 602 m (mean precipitation 1616 mm $yr^{-1}$). The lake is 0.05 $km^2$ in area, and its average runoff is 1454 mm (depth: ≈ 10 m). Peat rankers are typical of the area. They are only 33 cm deep (volcanic bedrock).

Scoat Tarn is the only European ICP Waters catchment considered for trend analyses where the $SO_4*$ concentrations are significantly increasing during the 90s (**Figure 26**). SBC* and ANC are not changing. There is a negative trend in $H^+$ concentrations.

## Scoat Tarn (United Kingdom)
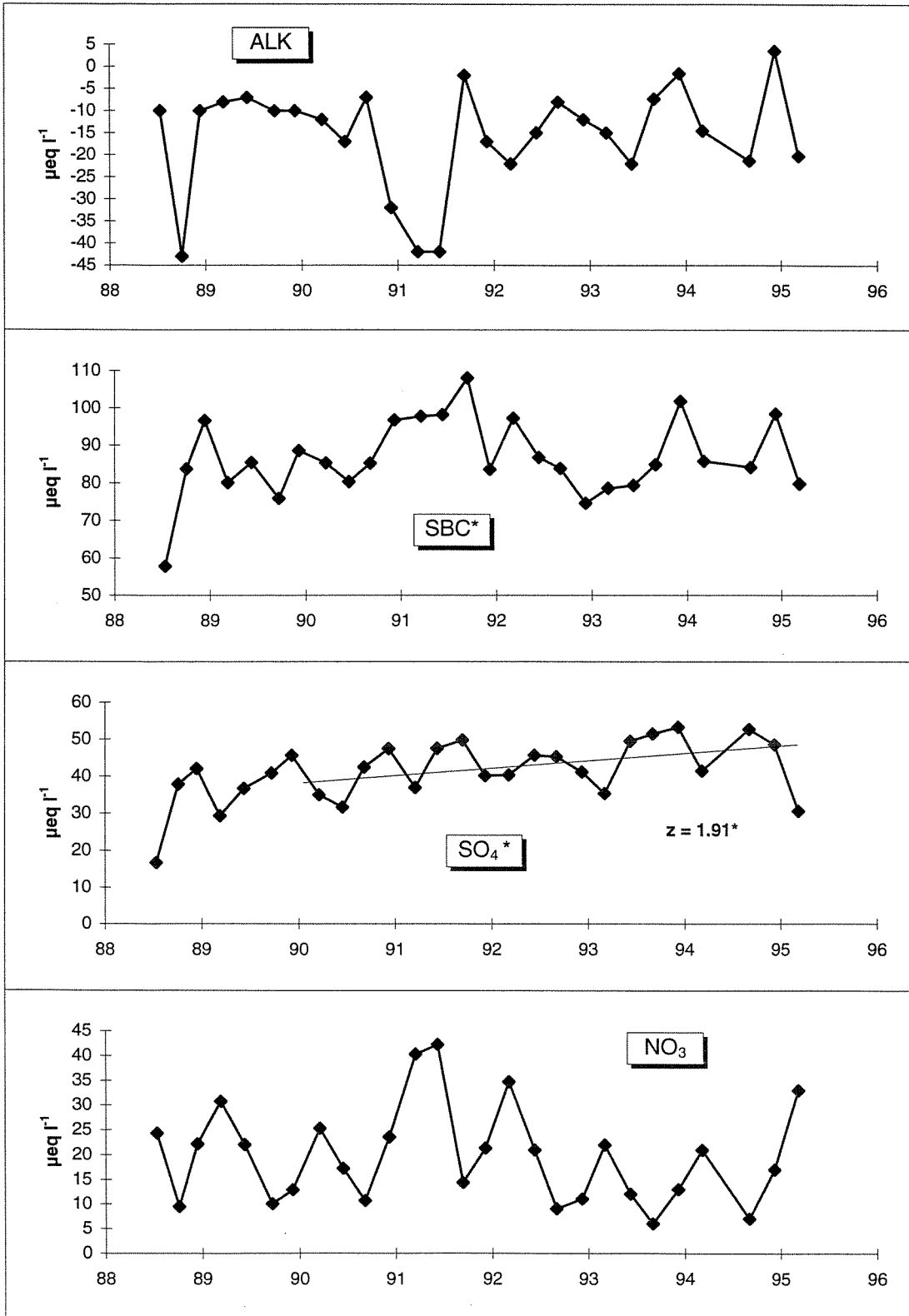


**Figure 26.** ALK, SBC\*, SO$_4$\* and NO$_3$ concentrations at Scoat Tarn, United Kingdom.

*Northern Ireland*

**Blue Lough**'s altitude is 340 m above sea level. Main bedrock type is granite covered with 40 cm deep blanket peat. The lake's mean depth is 1.7 m, lake area is 0.12 km$^2$ and runoff averages 934 mm yr$^{-1}$ (precipitation = 1038 mm yr$^{-1}$).

The only trend detected is an increase in H$^+$ concentrations. Average values for SO$_4$*, ALK, SBC* and NO$_3$ are 70, -38, 128 and 24 µeq l$^{-1}$.

*Scotland*

Three ICP sampling sites (lakes) are in Scotland. **Round Loch of Glenhead** in the Galloway region is located in a moorland area underlain by tonalite and tonalite granite (altitude = 295 m). The lake is 0.12 km$^2$ in area. The peaty podsols are about 88 cm deep. Precipitation averages 2223 mm and runoff 2001 mm yr$^{-1}$. **Lochnagar** is also surrounded by moorland (soils = peat, about 110 cm deep; elevation = 785 m). Lochagar is 0.10 km$^2$ in area. Main bedrock type is granite, average precipitation 928 mm and runoff 835 mm yr$^{-1}$. 1% of the catchment area of **Loch Coire nan Arr** is covered with coniferous forest, the rest is moorland (elevation = 125 m; size = 0.12 km$^2$). The peaty soils are underlain by sandstone. Mean precipitation is high: 2928 mm yr$^{-1}$. Runoff averages 2632 mm yr$^{-1}$.

At Lochnagar SO$_4$* and SBC* concentrations are significantly decreasing while the NO$_3$ values are significantly increasing (**Figure 27**). At the two other sites there is no trend in SO$_4$* concentrations, the curves have a positive slope. Average SO$_4$* values at Round Loch of Glenhead are 48 µeq l$^{-1}$ (ALK = -17 µeq l$^{-1}$) and at Loch Coire nan Arr 14 µeq l$^{-1}$. (ALK = 56 µeq l$^{-1}$).

## Lochnager (United Kingdom)
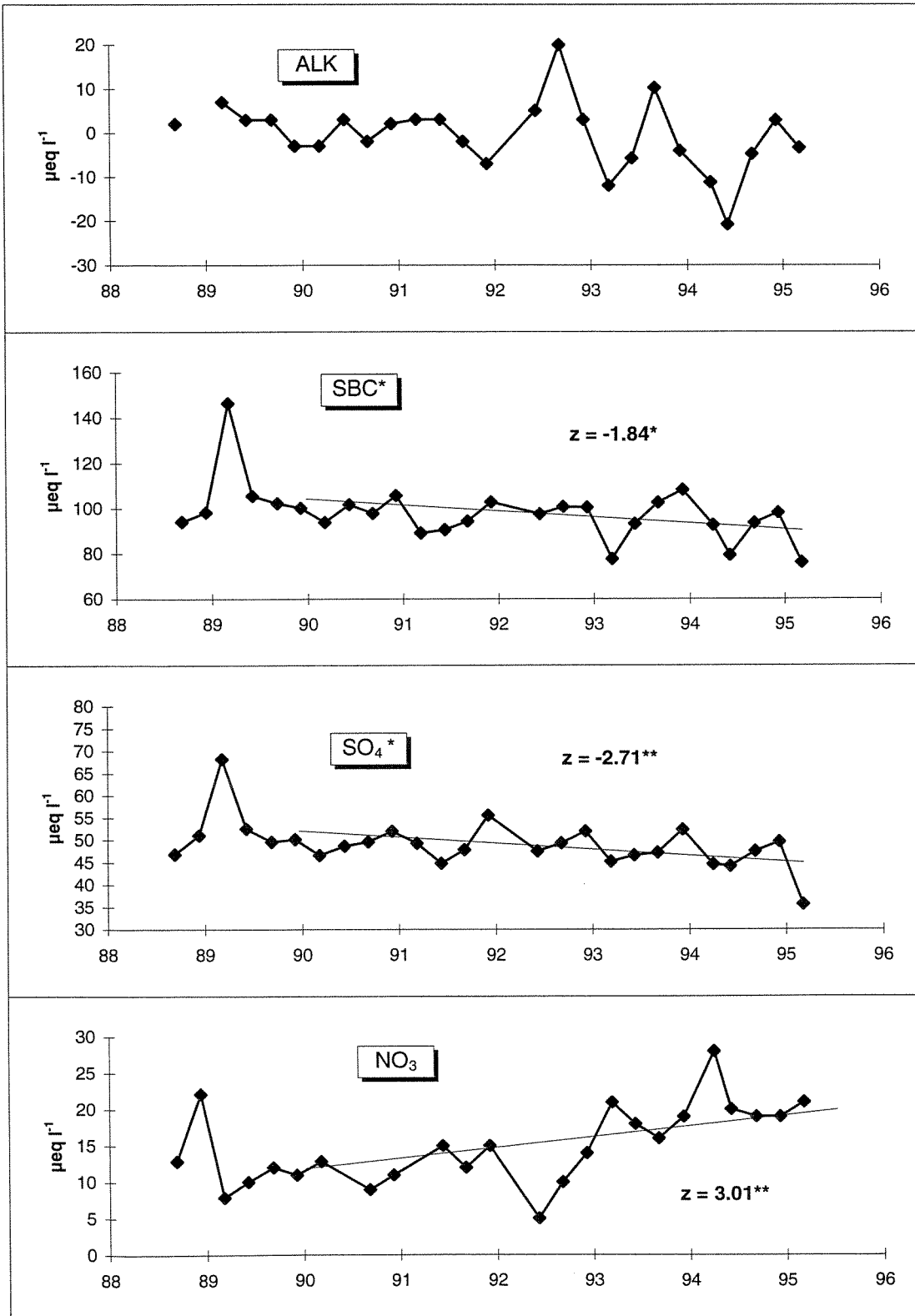


**Figure 27.** ALK, SBC*, SO₄* and NO₃ concentrations at Lochagar, United Kingdom.

## 3.4.17 Canada

Data for thirteen Canadian sites were included in this report. Nine of them were used for statistical analyses. Lakes in the the Kenora and Dryden districts of northwestern Ontario (CA111 - CA373) were not considered because no data for 1993-95 were available.

**Table 17.** The Canadian sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Nova Scotia | Beaverskin Lake *(C14)* | L | 1970-94 | N |
| | Kejimkujik Lake *(C13)* | L | 1970-94 | Y(80s) |
| | Mount Tom Lake *(C10)* | L | 1980-94 | N |
| | Mountain Lake *(C11)* | L | 1970-94 | N |
| Ontario, Algoma | Batchwana Lake *(C01)* | L | 1981-94 | Y |
| | Wishart Lake *(C02)* | L | 1980-94 | Y |
| | Little Turkey Lake *(C03)* | L | 1980-94 | Y |
| | Turkey Lake *(C04)* | L | 1980-94 | Y |
| Quebec | Lac Veilleux *(C05)* | L | 1982-94 | Y (80s) |
| | Lac Josselin *(C06)* | L | 1982-94 | Y (80s) |
| | Lac Bonneville *(C07)* | L | 1982-94 | Y |
| | Lac Laflamme *(C08)* | L | 1982-94 | Y |
| | Lac Macleod *(C09)* | L | 1982-94 | N |

Ion-balances could be calculated for the Ontario and Quebec sites and were used as quality control. Seasalt corrected concentrations were taken for trend analyses.

*Nova Scotia*

Average soil depth at all four ICP Waters sites in Nova Scotia is about 100 cm.

**Beaverskin Lake** is 0.061 km$^2$ in area (mean depth = 2.2 m), and catchment area is 1.00 km$^2$. Precipitation averages 1350 mm yr$^{-1}$ and runoff 850 mm yr$^{-1}$. The lake's elevation is 120 m (main bedrock type = greywacke and quartzite). Forest cover is 100% (20% deciduous and 80% coniferous). The last harvest was approximately 100 years ago.

There were not enough data available to run trend analyses. The average SO$_4$* concentrations in the 80s and 90s are almost the same (34 and 36 µeq l$^{-1}$). ALK values range from -4.8 to 30.8 µeq l$^{-1}$.

The elevation of **Kejimkujik Lake** is 90 m above sea level, and the main bedrock type is granite and slate. Yearly pecipitation averages 1350 mm (runoff = 800 mm). Kejimkujik Lake is 24.4 km$^2$ in area

and on average 4.4 m deep (catchment area = 723 km$^2$). 80% of the catchment is afforested (60% conferous, 40% deciduous). The forest is of mixed age (intermittent harvesting in small parcels). The rest of the area is mainly covered with wetlands and bogs.

SO$_4$* concentrations show a positive trend during the 80s (not enough data for trend analyses in the 90s). Average SO$_4$* values in the 80s = 34 and in the 90s = 35 µeq l$^{-1}$. No statistics for ALK could be run, mean ANC values in the 80s and 90s are 26 and 17 µeq l$^{-1}$. SBC and H$^+$ concentrations are decreasing significantly during the 80s. H$^+$ shows a positive trend in the 80s.

Mount **Tom Lake** is on average 2.00 m deep and 0.14 km$^2$ in area (elevation = 120 m). Average Precipitation is 1350 and runoff 850 mm per year. Half of the catchment (= 0.60 km$^2$) is covered with forest (ratio coniferous: deciduous = 1:4). The forest was harvested about 100 years ago. Some wetlands and bogs can be found.

Average SO$_4$* concentration in the 80s is 27 µeq l$^{-1}$ (ALK = 15) and in the 90s 21 µeq l$^{-1}$ (ALK = 12). NO$_3$ values range from 0.7 to 3.6 µeq l$^{-1}$.

Catchment area at **Mountain Lake** is 8.00 km$^2$ (lake area = 1.37 km$^2$). Average precipitation and runoff are 1350 and 850 mm. Mountain Lake's elevation is 120 m above sea level (average depth = 4.3 m). Total forest cover is 50% (25% deciduous and 75% coniferous), and the last harvest was approximately 100 years ago. 15% of the catchment area are wetlands and bogs.

SO$_4$* values are very similar to those at the other Nova Scotian sites (80s: 36 µeq l$^{-1}$; 90s: 38 µeq l$^{-1}$). ANC concentrations are around 3 µeq l$^{-1}$, and NO$_3$ ranges from 0.7 to 2.1 µeq l$^{-1}$.


*Ontario (Algoma)*

At all ICP Waters sites in this region, greenstone and diorite are the dominant bedrock types, and soil depth averages 100 cm. The last forest harvest was 80-100 years ago.

**Batchawana Lake** drains a catchment area of 0.86 km$^2$ (elevation = 497 m). Weekly samples are taken at the outlet of the lake (0.06 km$^2$). Average lake depth is 3.3 m and average runoff 720 mm yr$^{-1}$. Mean precipitation is 1200 mm yr$^{-1}$. 79% of the catchment is covered with forest, mostly deciduous.

SO$_4$* concentrations at Batchawana Lake are significantly decreasing during the 80s and 90s. Negative trends can be observed for the SBC values during and 90s. NO$_3$ values are relatively low (average = 18 µeq l$^{-1}$) and show a typical seasonal pattern with peaks in spring snow melt and concentrations near zero during the vegetations period.

Average yearly runoff at **Wishart Lake** is 770 mm (mean depth = 2.2 m). It is located at an elevation of 388 m with a mean precipitation rate of 1200 mm yr$^{-1}$. The 3.44 km$^2$ catchment is covered with forest (84%), 95% of it deciduous.

Wishart Lake (Ontario, Canada)
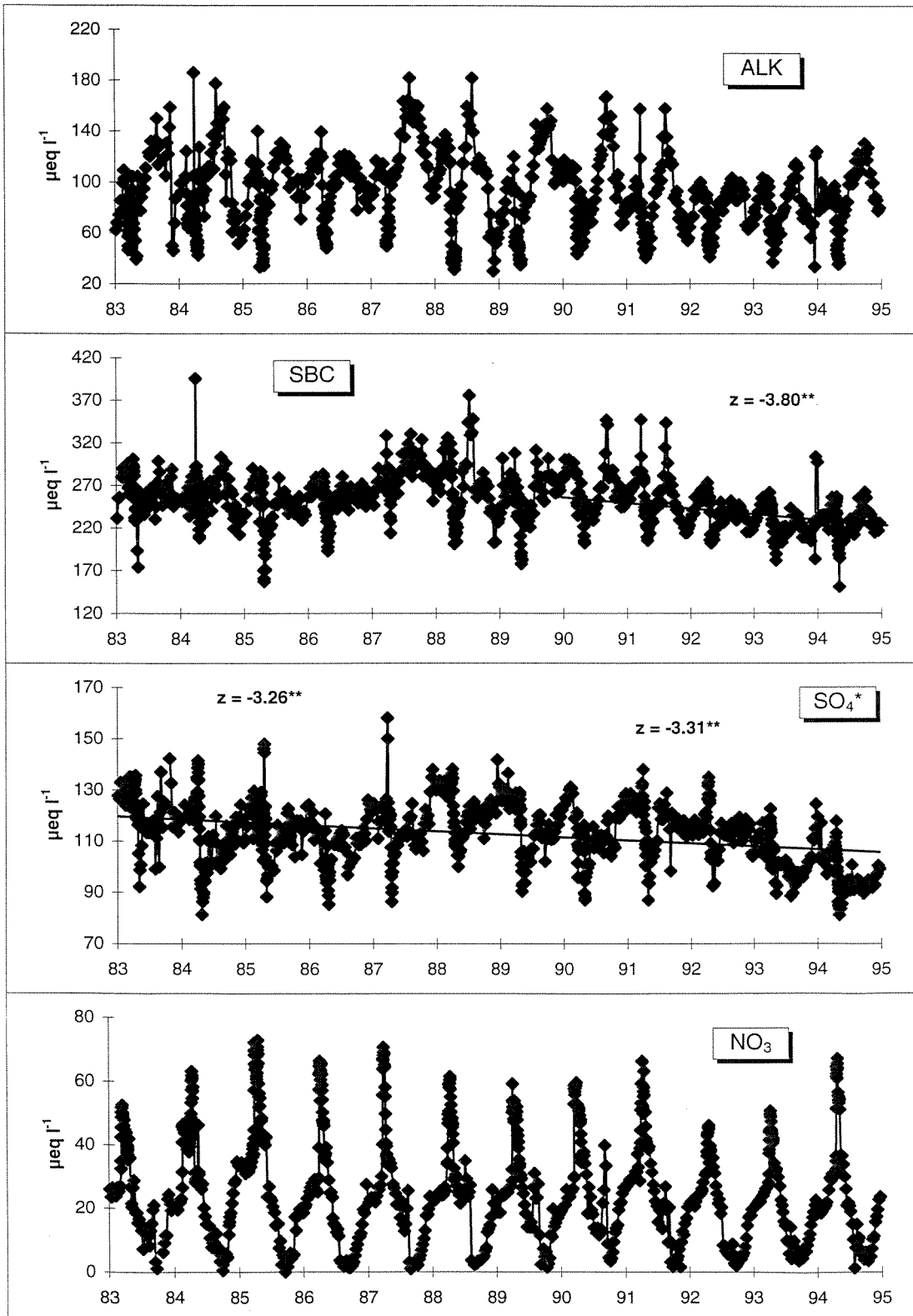


**Figure 28.** ALK, $SO_4$*, SBC and $NO_3$ concentrations at Wishard Lake, Ontario, Canada.

Sulphate concentrations show a negative trend both during the 80s and 90s (**Figure 28**). No changes in ALK can be observed, although SBC concentrations are significantly decreasing during the 90s. $NO_3$ concentrations reach the "zero line" in summer, although yearly means are higher than at Batchawana Lake.

**Turkey Lake** is elevated at 372 m. Receiving the same amount of precipitation as Little Turkey Lake (1200 mm $yr^{-1}$), runoff is only 870 mm $yr^{-1}$. Deciduous forest is the predominant vegetation type. Catchment and lake areas are 8.03 or 0.52 $km^2$. Mean lake depth is 12.2 m.

$SO_4$* concentrations decrease significantly both in the 80s and 90s (**Figure 29**). The upward step in $SO_4$* has been attributed to the influence of drought (JEFFRIES et al. 1995) and was a widespread observation throughout central Ontario. ALK and SBC trends are negative during the 90s.

**Little Turkey Lake** is about 6 m deep and surrounded by deciduous forest, covering almost 80% of the catchment area of 4.91 $km^2$. Runoff averages 920 and precipitation 1200 mm $yr^{-1}$. Lake area 0.19 $km^2$ and levation 375 m above sea level.

$SO_4$* concentrations are significantly decreasing during the 90s. ALK shows a positive trend in the 80s and SBC concentrations a negative one during the 90s. The $NO_3$ curve shows the same pattern as at Turkey Lake.


*Quebec*

Main bedrock type at **Lac Veilleux** is granite and gneiss. Soils are greater than 100 cm deep. 97% of the catchment is covered with forest, two thirds are coniferous. 25% of the trees are mature, 20% young and 55% were cut > 25 years ago. The catchment is 0.45 $km^2$ in area, lake area is 0.08 $km^2$. Precipitation averages 1650 mm $yr^{-1}$. The lake's elevation is 716 m.

No significant change in $SO_4$* (or any other) concentrations can be observed. For the 90s not enough data were available to run trend analyses. Nevertheless, $SO_4$* concentrations seem to decrease during this period (from about 65 to 55 µeq $l^{-1}$). ALK drops only once below zero, in 1990 and values range from -4 to 39 µeq $l^{-1}$. $NO_3$ concentrations do not exceed 10 µeq $l^{-1}$.

The catchment of **Lac Josselin** is underlain by granite and gneiss (soil depth > 60 cm). Mean precipitation is 1650 mm $yr^{-1}$. The lake is elevated at 671 m above sea level. 57% of the 1 $km^2$ catchment is covered with heather and grassland, the rest with deciduos and coniferous trees. 10% of the forest are mature, 20% young and 10% were cut > 25 years ago. Lake area is 0.25 $km^2$.

Only water chemistry data of the 80s could be used for trend analyses and no significant changes can be found during this period. $SO_4$*, ALK and $NO_3$ concentrations and their changes over time are very similar to those at Lac Veilleux.

Turkey Lake (Ontario, Canada)
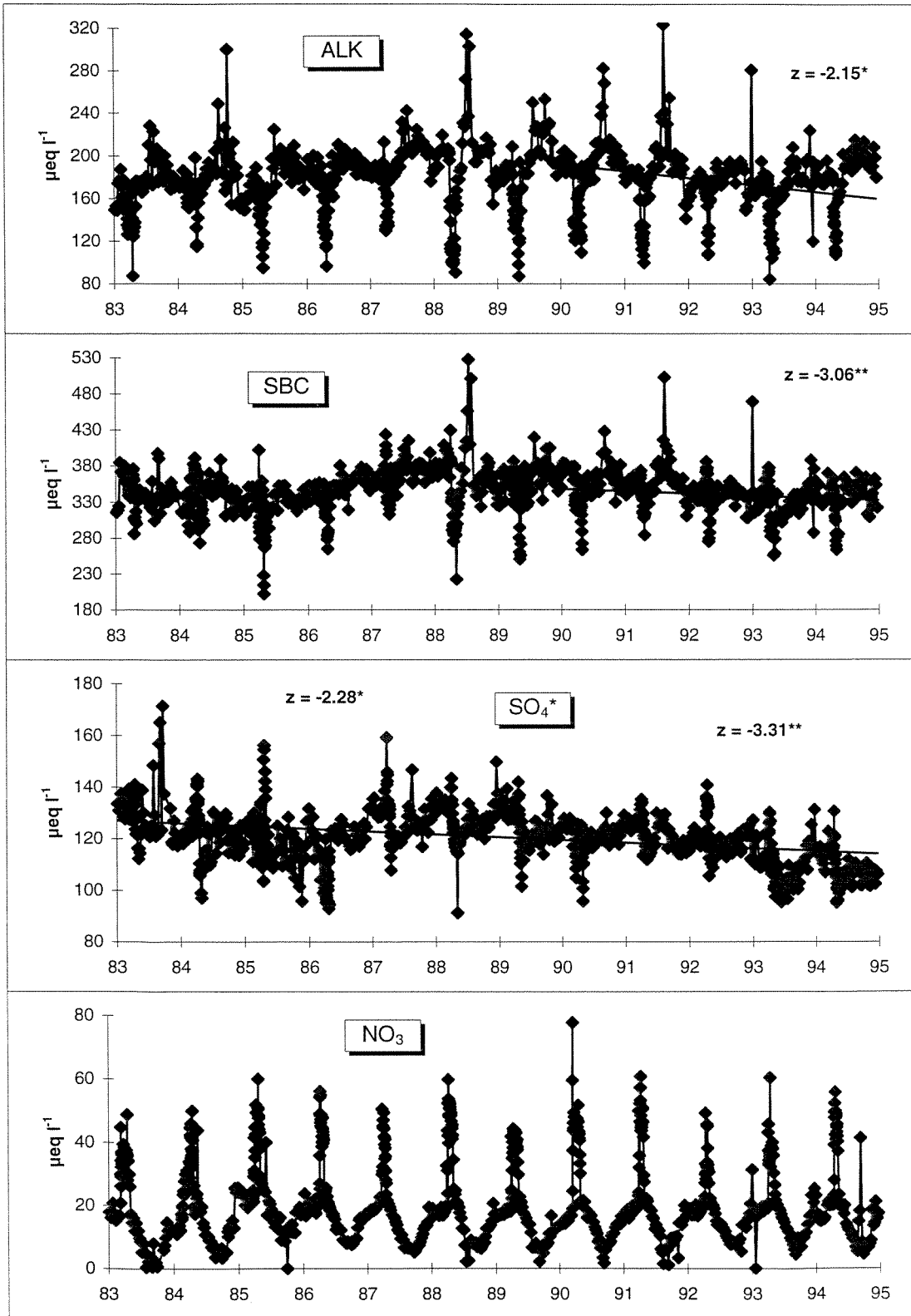


**Figure 29.** ALK, $SO_4$*, SBC and $NO_3$ concentrations at Turkey Lake, Ontario, Canada.

89% of the vegetation at **Lac Bonneville** catchment is coniferous forest (4% heather/grassland, 5% wetlands, 2% rocks). 40% of the forest is mature, the rest was cut more than 25 years ago. Again, main bedrock type is granite and gneiss. The soils are on average 60 cm deep. The catchment is 0.86 $km^2$ in area (lake area = 0.10 $km^2$). The lake's altitude is 808 m. Mean precipitation is 1650 mm $yr^{-1}$.

$SO_4$* concentrations are significantly decreasing during the 90s. While the trend in SBC is negative during the 80s, there is no significant change during the 90s. $NO_3$ concentrations are very low (average 80s = 4.2 µeq $l^{-1}$ and 90s = 3.2 µeq $l^{-1}$). The curve shows a typical seasonality. Peak values in spring seem to disappear from 1991 onwards.

**Lac Laflamme** is 0.06 $km^2$ in area and on average 2.1 m deep (elevation = 777 m). Main vegetation type covering the 0.68 $km^2$ catchment is coniferous forest, only a few deciduous trees can be found (3%). Last harvesting was in 1947 (average stem age = 50 years). 9% of the area are covered with wetlands and bogs. Precambrian gneiss is the main bedrock type, and the soils are about 110 cm deep. There is a significant decrease in $SO_4$* concentrations during 90s (**Figure 30**). No change in ALK (and SBC) values can be found. $NO_3$ concentrations are significantly decreasing during the 80s and 90s. The $NO_3$ curve shows a very strong seasonality with concentrations near zero during the vegetation period.

Elevation at **Lac Macleod** is 975 m above sea level. Main bedrock type is granite and gneiss, and soil depth is greater than 100 cm. Catchment area is 0.88 $km^2$ (lake = 0.10 $km^2$ in area). Precipitation averages 1650 mm $yr^{-1}$. 85% of the area is covered with coniferous forest (15% heather and grassland).

Not enough data were available to run trend analyses. The slope of the $SO_4$ curve is negative during the 90s (average concentration = 65 µeq $l^{-1}$; 80s: 80 µeq $l^{-1}$). There does not seem to be a change in ALK, the values are on average 9 (80s) and 7 (90s) µeq $l^{-1}$. $NO_3$ concentrations are close to zero. The curve does not show any seasonality (one peak of 10 µeq $l^{-1}$ in 1990).

Lac Laflamme (Quebec, Canada)
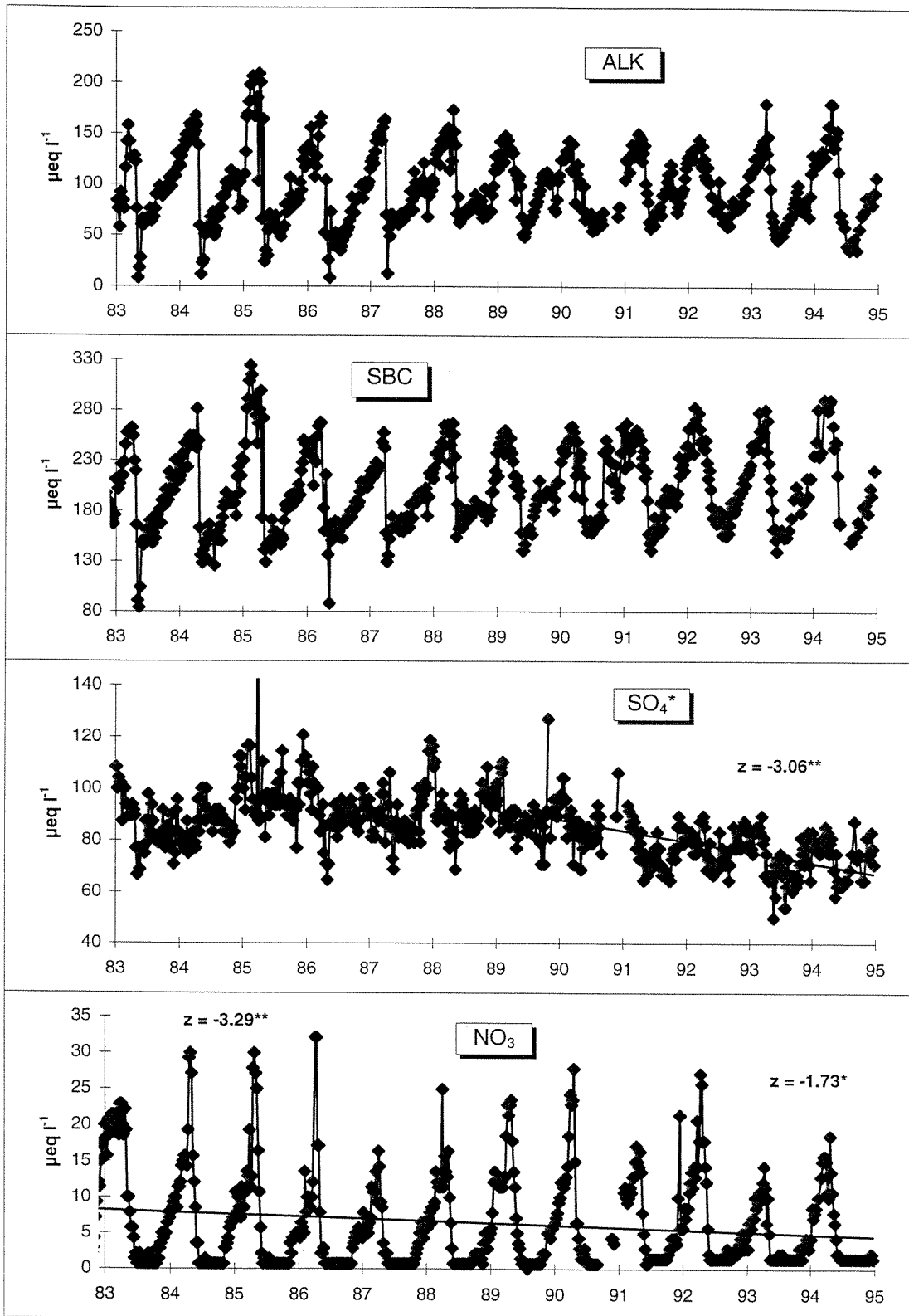


**Figure 30.** ALK, SO$_4$*, SBC and NO$_3$ concentrations at Lac Laflamme, Quebec, Canada.

### 3.4.18 United States of America

Data of all sites listed in **Table 18** were read into the new database (21 sites). 16 of them could be used for trend analyses.

**Table.18.** The U.S.A. sites included into this report.

| Region | Site (*Code*) | Lake/Stream L / S | Data Read In | Statistics YES / NO |
|---|---|---|---|---|
| Colorado | Seven Lakes (*4E2009*) | L | 1985-94 | N |
| | Summit Lake (*4E2060*) | L | 1985-94 | N |
| | Sunlight Lake (*4E2070*) | L | 1985-94 | N |
| | White Dome Lake (*4E2071*) | L | 1985-94 | N |
| Maine | Little Long Pond (*1E1132*) | L | 1982-94 | Y |
| | Tilden Pond (*1E1133*) | L | 1982-94 | Y |
| Michigan | Andrus (*2B3082*) | L | 1983-94 | Y |
| | Buckeye (*2B2102*) | L | 1983-94 | Y |
| | Johnson (*2B1047*) | L | 1983-94 | Y |
| Minnesota | Cruiser (*2A2063*) | L | 1983-94 | Y |
| New York | Arbutus (*1A1052*) | L | 1983-94 | Y |
| (Adirondacks) | Constable (*1A1017*) | L | 1982-94 | Y |
| | Dart Lake (*1A1106*) | L | 1982-94 | Y |
| | Heart Lake (*1A1102*) | L | 1982-94 | Y |
| | Lake Rondaxe (*1A1110*) | L | 1982-94 | Y |
| | Moss Lake (*1A1109*) | L | 1982-94 | Y |
| | Otter Lake (*1A2078*) | L | 1982-94 | Y |
| New York (Catskills) | East Branch Never Sink (*143010*) | S | 1983-94 | N |
| Wisconsin | Luna (*2C2062*) | L | 1983-94 | Y |
| | Nichols (*2C1069*) | L | 1983-94 | Y |
| | Sand (*2C1068*) | L | 1983-94 | Y |

*Colerado*

For the ICP Waters sites in Colerado there were not enough data available to run trend analyses. About 50% of the catchment areas are exposed to bedrock. 20-45% are covered with heather and grassland (rest = lakes). Elevation ranges from 3145 m above sea level (Summit Lake) to 3826 m (White Dome Lake). Average yearly precipitation is 813 mm (Seven Lakes and Summit Lake).

Catchment area of **Seven Lakes** is 0.67 km$^2$. Main bedrock type is Quartz Marzonite. The lake is ca. 0.18 km$^2$ in area. There is no difference in mean $SO_4$* concentrations in the 80s and 90s ($\approx$ 11 µeq l$^{-1}$;

ALK $\approx$ 44 $\mu$eq l$^{-1}$). The pH values are relatively high (pH 6.0 - 7.1) and NO$_3$ concentrations low (mostly < 0.1 $\mu$eq l$^{-1}$).

**Summit Lake** is on average 1.0 m deep and 0.02 km$^2$ in area (catchment area = 0.11 km$^2$). Felsic gneiss is the predominant bedrock type. Mean SO$_4$* values in the 80s are 20 and in the 90s 19 $\mu$eq l$^{-1}$. Alkalinity ranges from 40 - 91 $\mu$eq l$^{-1}$ (pH around 7.0). NO$_3$ concentrations are very low, the highest value measured since 1985 is 14 $\mu$eq l$^{-1}$.

Catchment and lake areas of **Sunlight Lake** are 0.75 and 0.05 km$^2$ (main bedrock type = granite). Average SO$_4$* concentrations in the 80s are much lower than those in the 90s: 39 and 58 $\mu$eq l$^{-1}$, respectively. Both ALK and pH are high (ALK = 35 $\mu$eq l$^{-1}$ in the 80s and 46 in the 90s; pH $\approx$ 7.0). Maximum in NO$_3$ concentration is 11 $\mu$eq l$^{-1}$ (minimum = 0.7 $\mu$eq l$^{-1}$).

Lake and catchment areas at **White Dome Lake** are 0.02 and 0.4 km$^2$. Average SO$_4$* concentrations are almost the same in the 80s and 90s (36 and 37 $\mu$eq l$^{-1}$). Only for September 1988 a negative value for ALK can be observed. Otherwise concentrations range from 17.0 - 0.0 $\mu$eq l$^{-1}$. During the vegetation period NO$_3$ concentrations stay on a level of about 6 $\mu$eq l$^{-1}$.


*Maine*

The main bedrock type at both sites in Maine is granite. Most of the area is afforested: 95% at Little Long Pond and 100% at Tilden Pond (deciduous). Precipitation is about 1300 mm and average runoff 635 mm at both sites. The elevation is $\approx$ 70 m above sea level. **Little Long Pond** is on average 11.5 m deep. Catchment and lake areas are 2.38 and 0.25 km$^2$. **Tilden Pond** is 0.15 km$^2$ in area (catchment area = 0.49 km$^2$). Mean depth of the lake is 4.4 m.

While there is no significant change in SO$_4$* concentrations during the 80s, a negative trend in the 90s can be observed at both sites (**Figure 31**). In the 80s SBC values are significantly increasing at Tilden Pond (Fig. 3.29). At Little Long Pond and Tilden Pond there is a positive trend in ALK values in the 80s. At both sites NO$_3$ concentrations are very low (a few peaks up to 2 $\mu$eq l$^{-1}$).


*Michigan*

**Andrus** is a seepage lake located at an elevation of 190 m (catchment area = 1.86 km$^2$). The lake is on average 4.3 m deep (lake area = 0.13 km$^2$). Mean precipitation is 1247 mm yr$^{-1}$. 93 % of the catchment is covered with deciduous forest.

Tilden Pond (Maine, USA)
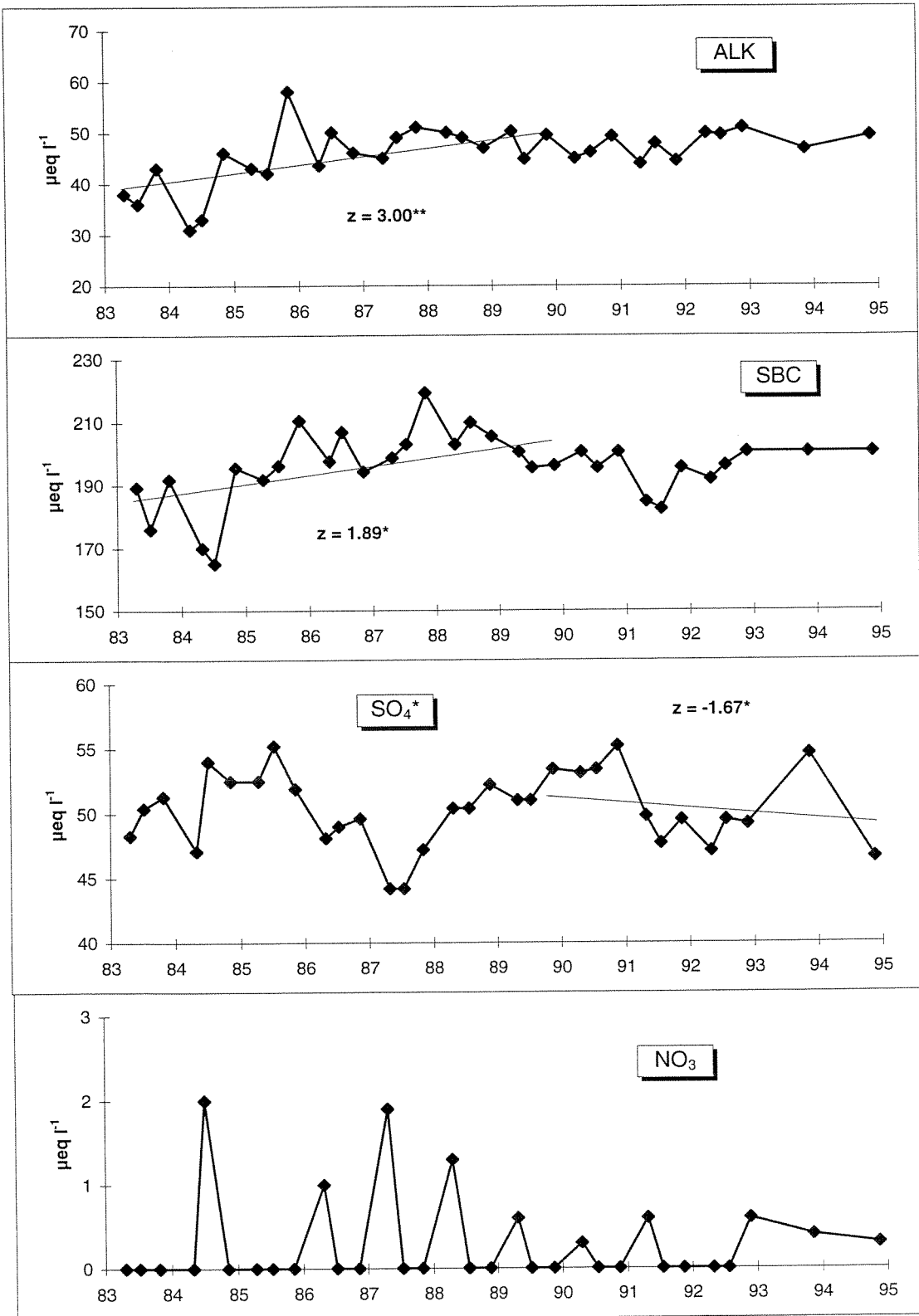


**Figure 31.** ALK, $SO_4^*$, SBC and $NO_3$ concentrations at Tilden Pond, Maine, U.S.A.

There is a negative trend in $SO_4$*, ALK and SBC concentrations during the 90s (mean values: 88, 19 and 138 $\mu$eq $l^{-1}$). $NO_3$ values are near zero during the vegetation periods, and pH ranges from 5.5 to 6.5.

**Buckeye** has a lake area of 0.48 $km^2$ (mean depth = 4.2 m). The catchment is 3.28 $km^2$ in area, precipitation averages 498 mm and runoff 381 mm per year. Most of the catchment is afforested (deciduous trees). The lake's elevation is 273 m above sea level.

In the 80s $SO_4$* and $H^+$ concentrations are decreasing significantly. Average $SO_4$* concentration in the 80s is 75 $\mu$eq $l^{-1}$ and in the 90s $\mu$eq $l^{-1}$. ALK shows a positive trend during this period while it is decreasing again in the 90s (average 1994 = 157 $\mu$eq $l^{-1}$). SBC is also decreasing in the 90s. $NO_3$ are low and show a seasonality (mean 80s = 2.4 and 90s = 0.9 $\mu$eq $l^{-1}$).

Average precipitation at **Johnson** is 1211 mm (seepage lake). The lake is on average 2.9 m deep and 0.17 $km^2$ in area (elevation = 252 m above sea level). 78 % of the catchment are afforested (deciduous forest). The rest of the area are lakes.

$SO_4$* concentrations are significantly decreasing during the 90s (average = 108 $\mu$eq $l^{-1}$). $NO_3$ values are low and show typical seasonal variations (mean 1983-94 = 1.6 $\mu$eq $l^{-1}$).


*Minnesota*

**Cruiser** is a 1.19 $km^2$ catchment located at an elevation of 379 m. The lake is 0.48 $km^2$ in area and precipitation averages 735 mm $yr^{-1}$ (runoff = 127 mm). 40% of the catchment area are lakes, 60 % deciduous forest. Mean lake depth is 14.1 m.

Sulphate values show a negative trend in the 80s (average during this period = 56 $\mu$eq $l^{-1}$). No change in ALK can be observed (mean 1983-94 = 123 $\mu$eq $l^{-1}$), while SBC concentration increase significantly in the 80s. Average pH values are close to pH 7. $NO_3$ concentrations are very low (typical seasonality).


*New York (Adirondack Mountains)*

Seven ICP Waters sites are located in the Adirondack Mountains. Main bedrock type in this region is gneiss and granitic gneiss. Deciduous forest is the predominat vegetation type (81 to 95% of total forest cover).

The **Arbutus** catchment is 3.42 $km^2$ in area (lake area = 0.49 $km^2$). Precipitation averages 916 mm and runoff 635 mm per year. The lake's depth is 2.1 m on average, and its elevation 513 m above sea level.

A negative trend in $SO_4$* concentrations can be observed for the 90s (average = 130 $\mu$eq $l^{-1}$). SBC as well as $NO_3$ show significant increases in 80s and a decrease in the 90s. The $H^+$ trend is negative in the 80s, but positive in the 90s. $NO_3$ concentrations are low.

The elevation of lake **Constable** is 582 m above sea level. The catchment is 13.8 $km^2$ in area (lake area = 0.21 $km^2$). Precipitation averages 906 mm per year and runoff 762 mm.

In the 80s $SO_4$* and ALK concentrations at Constable are significantly decreasing (**Figure 32**). SBC and $NO_3$ trends are negative in the 90s. $NO_3$ values are significantly increasing in the 80s (regarding changes in seasonality see **Figure 32**).

The altitude of **Dart Lake** is 535 m. Catchment area is 106 $km^2$, lake area 0.58 $km^2$. Yearly precipitation averages 904 mm and runoff 762 mm. The mean depth of the lake is 4.2 m.

A significant decrease in $SO_4$* concentrations can be observed in the 80s (average 80s = 129 µeq $l^{-1}$ and 90s =114 µeq $l^{-1}$). ALK concentrations are also decreasing in the 80s (increasing the in 90s), while there is a positive trend in SBC in 80s and negative in 90s. $H^+$ is significantly decreasing in the 80s as well as in the 90s. $NO_3$ values show a negative trend in the 90s. The pattern of the nitrate curve is very similar to the one observed for Constable (Fig. 3.30). The level of peak values as well as the "bottom line" during the vegetation period increase from 1983 to 1990 and then decrease again. The same pattern can be observed at all sites located in the Adirondack Mountains. At Dart Lake the $NO_3$ values in spring/summer do not drop below 10 µeq $l^{-1}$.

**Heart Lake** is draining a 0.60 $km^2$ catchment, elevated at 659 m above sea level. The lake is 0.11 $km^2$ in area and on average 5.4 m deep.

$SO_4$* shows a negative trend during the 90s only (average concentration = 89 µeq $l^{-1}$). ALK values are significantly decreasing in the 80s (mean = 35 µeq $l^{-1}$) and SBC concentrations in the 90s. $H^+$ increases significantly in the 80s and decreases in 90s.

The elevation of **Lake Rondaxe** is is 524 m above sea level (catchment area = 139.4 $km^2$). It is on average 3.4 m deep and 0.68 $km^2$ in area. Mean yearly precipitation is 901 mm and runoff 762 mm.

During the 80s a negative trend in $SO_4$* concentrations can be observed (average 80s = 125 and 90s = 113 µeq $l^{-1}$). $NO_3$ concentrations are significantly decreasing in the 90s.

Precipitation at **Moss Lake** averages 904 mm $yr^{-1}$ (runoff = 762 mm). The lake is draining a 12.48 $km^2$ catchment (elevation = 535 m). Mean depth of Moss Lake is 5.8 m and lake area is 0.45 $km^2$.

During the 80s a downwards trend in $SO_4$* can be found, accompanied by significantly increasing SBC concentrations (average $SO_4$ in 80s =131 µeq $l^{-1}$). $NO_3$ is significantly decreasing during the 90s.

**Otter Lake** is the seventh lake located in the Adirondack Mountains (elevation = 502 m above sea level). It is 0.16 $km^2$ in area and on average 1.3 m deep. Precipitation is about 863 mm $yr^{-1}$ (runoff = 508 mm $yr^{-1}$).

## Constable Pond (Adirondacks, U.S.A.)
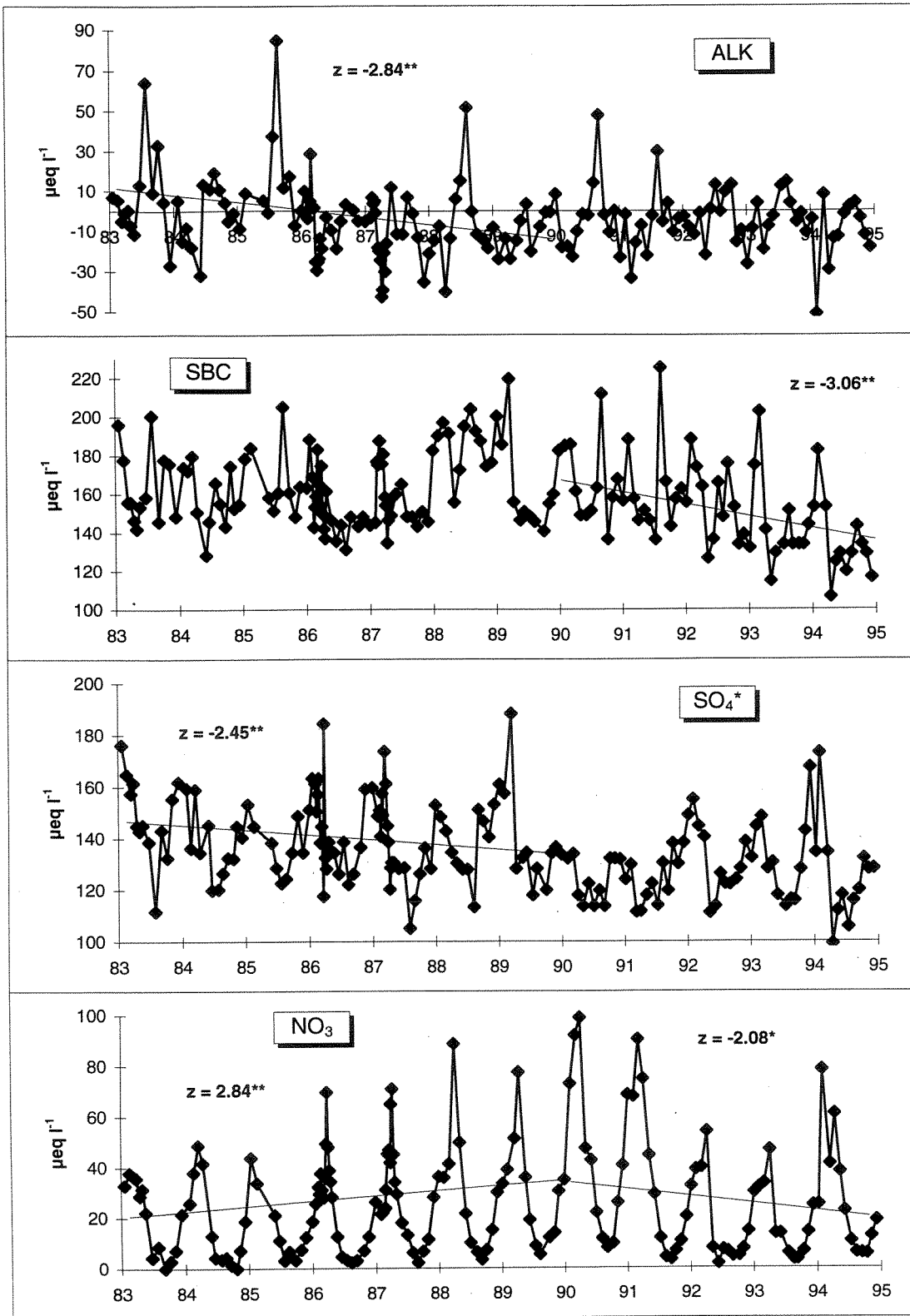


**Figure 32.** ALK, $SO_4^*$, SBC and $NO_3$ concentrations at Constable Pond, Adirondacks, U.S.A.

Sulphate concentrations at Otter Lake show a negative trend in the 90s. ALK values decline significantly in the 80s (mean ALK in 80s = 3.2 $\mu$eq $l^{-1}$). SBC decreases in the 90s. $NO_3$ curves show a positive trend in the 80s, and it is negative in the 90s (compare Fig 3.30).

*New York (Catskills)*

**East Branch Never Sink** is a river in the Catskill Mountains, draining a 34.57 $km^2$ catchment. Forest cover is 100% (95% deciduous and 5% coniferous). Main bedrock type is sandstone and shale, and the streams elevation is 613 m above sea level.

There were not enough data available to run trend analyses. Average $SO_4$* concentrations in the 80s are 122 $\mu$eq $l^{-1}$ and in the 90s 118 $\mu$eq $l^{-1}$. Alkalinity is negative: -7.6 (80s) and -2.9 $\mu$eq $l^{-1}$ (90s). $NO_3$ values are relatively high (average 1983-93 = 24 $\mu$eq $l^{-1}$). The development in $NO_3$ concentrations shows the same pattern that can be observed at the sites in the Adirondack Mountains (increases in the 80s and decreases in the 90s). The pH values range from 4.7 - 5.6.

*Wisconsin*

Average depth of the seepage lake **Luna** is 4.0 m and mean precipitation at the 0.8 $km^2$ catchment is 884 mm per year (elevation = 517 m). The lake is 0.26 $km^2$ in area. 67% of the its surroundings are covered with deciduous forest (rest = lakes).

$SO_4$* concentrations do not significantly change at Luna, and the mean values in the 80s and 90s are 88 and 84 $\mu$eq $l^{-1}$. A negative trend in ALK and SBC can be observed in the 90s (average during this period: 17 $\mu$eq $l^{-1}$). SBC are increasing in the 80s. $NO_3$ concentrations are very low (mean 1983-94 = 0.9 $\mu$eq $l^{-1}$), and the curve shows a typical seasonality (pH = 5.8 - 6.5).

**Nichols** is also a seepage lake (area = 0.15 $km^2$; average depth = 2.0 m). The catchment is 0.91 $km^2$ in area. Precipitation averages 885 mm $yr^{-1}$ and runoff 254 mm $yr^{-1}$. Nichols' elevation is 495 m above sea level. 84% of the catchment is covered with deciduous forest, 38% are lakes.

During the 90s $SO_4$* concentrations are significantly decreasing at Nichols (**Figure 33**). There is no trend in alkalinity (average 80s = 25 and 90s = 36 $\mu$eq $l^{-1}$). $H^+$ values are significantly decreasing in the 80s (pH around 6.0). For the 90s a positive trend in $NO_3$ in the 90s cam be observed.

Average precipitation and runoff at **Sand** are the same as at Nichols. Forest cover (deciduous) at this site is only 60% (38% lakes). The seepage lake is 0.15 $km^2$ in area (catchment area = 0.39 $km^2$).

$SO_4$* shows a negative trend in the 90s, but ALK and SBC are also decreasing.

Nichols Lake (Wisconsin, U.S.A.)
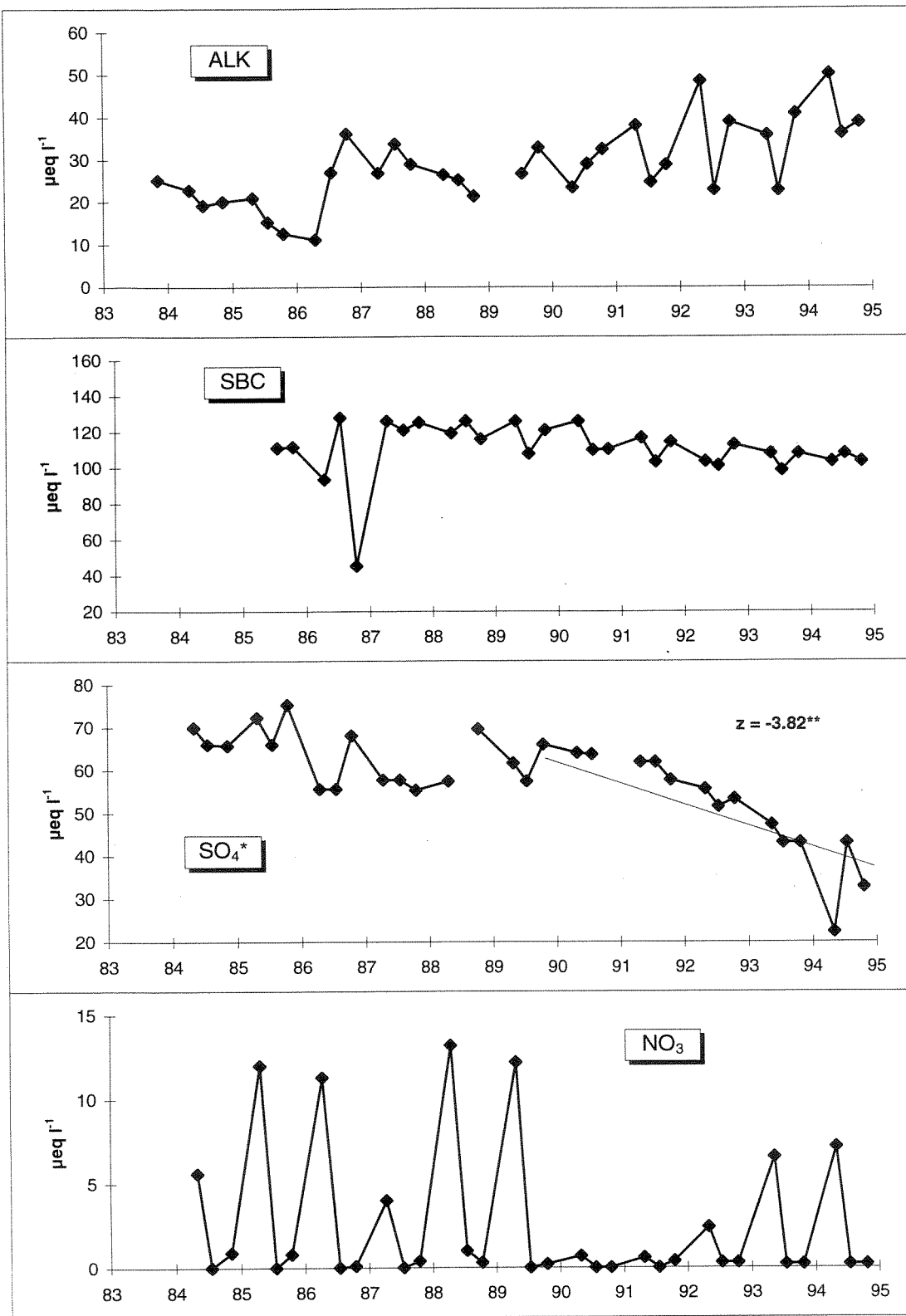


**Figure 33.** ALK, $SO_4$*, SBC and $NO_3$ concentrations at Nichols Lake, Wisconsin, U.S.A.

## 3.5 Regional Trends in Surface Water Chemistry

One of the most striking results of the regional trend meta-analysis is the large differences between countries and between decades (**Table 19**). In almost all cases, the amount of variation between sites within a country was sufficiently large that a country-by-country analysis is not a good presentation of the results (i.e., the differences between sites within a country were as large or larger than the differences between countries when all sites were combined). We present the trend results by country here primarily as an illustration of the differences between the two decades in each country, rather than as a valid summary of each country's results. Many of the results (particularly for $SO_4^{2-}$, ALK and SBC) reinforce our original impression that curvilinear trends are common, and that 1990 seems to be a common inflection point. These results are also supported by the more valid regional results we present below.

One aim of the regional meta-analysis is to group together those sites whose trends are similar. We could perform this grouping via a cluster analysis or other multivariate technique, but we chose instead to cluster the sites geographically. This grouping has the advantage of allowing us to test a number of important regional hypotheses, for example: (1) are the regional decreases in deposition concentrations of $SO_4^{2-}$, observable in many parts of Europe and North America, reflected in surface water $SO_4^{2-}$ trends; (2) do regional decreases in $SO_4^{2-}$ lead to recovering values of surface water ANC; (3) are $NO_3^-$ concentrations, known to be increasing at many intensive research sites in Europe and North America, increasing on a regional level; and (4) are base cation concentrations changing in response to lower $SO_4^{2-}$ concentrations, and are the changes consistent with potential recovery from acidification.

We chose the following regions to conduct this regional analysis: (1) the Nordic Countries (Finland, Sweden and Norway); (2) Europe (Italy, Germany, Netherlands and Denmark); (3) the U.K.; (4) Midwestern North America (Wisconsin, Minnesota, Michigan and Ontario); (5) the Adirondack Mountains (New York) and Quebec; and (6) the north Atlantic coast of North America (Vermont, Maine and Nova Scotia). With a few exceptions, the assignments of sites to one of these regions was straight-forward; the Danish sites were grouped with Europe rather than the Nordic Countries largely on the basis of the chemistry of the two Danish sites (more typical of Europe than of Scandinavia).

The results of the regional analysis are shown in **Tables 20** through **Table 24**. As mentioned above, these are the results of performing an analysis of variance on Z statistics from the seasonal Kendall

**Table 19.** Results of regional trend analyses by *country and decade*. In almost all cases, the amount of between-site variation is too high and/or the sample size too small for these to be valid estimates of regional changes. They are presented solely for the purpose of illustrating between-country and between-decade differences in trends.

| Country | Decade | Number of Sites* | $\Delta$ ALK $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ SO$_4^{2-}$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ NO$_3^-$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ SBC $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ H$^+$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ |
|---|---|---|---|---|---|---|---|
| Canada | 1980s | 12 | +0.8 (n.s.) | -0.4 (p < 0.10) | +0.0 (n.s.) | -0.9 (p < 0.10) | -0.01 (p < 0.001) |
| | 1990s | 6 | +0.8 (n.s.) | -4.8 (p < 0.001) | -0.1 (n.s.) | -2.5 (p < 0.001) | -0.00 (n.s.) |
| Denmark | 1980s | 2 | +2.5 (p < 0.05) | +0.0 (n.s.) | -1.0 (n.s.) | +0.0 (n.s.) | -0.16 (p < 0.05) |
| | 1990s | 2 | -0.6 (n.s.) | -6.5 (p < 0.05) | -3.6 (p < 0.01) | -47.2 (p < 0.01) | -0.01 (n.s.) |
| Finland | 1980s | 1 | | -1.1 (n.s.) | +0.0 (n.s.) | -1.5 (n.s.) | -0.13 (n.s.) |
| | 1990s | 4 | +5.1 (n.s.) | -3.4 (p < 0.05) | +0.0 (n.s.) | -0.4 (n.s.) | -0.17 (n.s.) |
| Germany | 1980s | 7 | | -8.3 (p < 0.001) | +1.4 (p < 0.001) | -4.8 (n.s.) | -0.09 (n.s.) |
| | 1990s | 13 | +12.7 (p < 0.001) | -5.1 (p < 0.001) | -2.4 (p < 0.001) | +1.9 (n.s.) | -0.00 (n.s.) |
| Italy | 1980s | 2 | +1.2 (n.s.) | -0.1 (n.s.) | +2.9 (p < 0.001) | +3.9 (n.s.) | -0.00 (n.s.) |
| | 1990s | 2 | -4.5 (n.s.) | -2.1 (p < 0.10) | -0.3 (n.s.) | -6.4 (p < 0.10) | -0.00 (n.s.) |
| Netherlands | 1980s | 1 | +3.3 (n.s.) | +0.8 (n.s.) | -0.5 (p < 0.10) | +0.9 (n.s.) | +6.29 (p < 0.05) |
| | 1990s | 1 | +16.9 (p < 0.01) | -16.0 (p < 0.05) | -0.8 (n.s.) | +1.9 (n.s.) | -2.27 (n.s.) |
| Norway | 1980s | 7 | -1.2 (p < 0.001) | -0.8 (p < 0.001) | +0.1 (p < 0.05) | -1.6 (p < 0.001) | +0.10 (p < 0.01) |
| | 1990s | 7 | +4.1 (p < 0.001) | -1.6 (p < 0.001) | +0.1 (n.s.) | +2.1 (p < 0.001) | -0.44 (p < 0.001) |
| Sweden | 1980s | 9 | -1.8 (p < 0.05) | +0.5 (n.s.) | +0.1 (n.s.) | -7.7 (p < 0.001) | +0.01 (n.s.) |
| | 1990s | 9 | +1.4 (p < 0.10) | -3.8 (p < 0.001) | -0.2 (p < 0.05) | -1.9 (p < 0.01) | -0.01 (p < 0.01) |
| U.K. | 1990s | 6 | +0.2 (n.s.) | -0.0 (n.s.) | +0.2 (p < 0.001) | -1.5 (n.s.) | -0.14 (p < 0.10) |
| U.S. | 1980s | 17 | +0.2 (n.s.) | -1.4 (p < 0.001) | +0.2 (p < 0.001) | +1.2 (p < 0.001) | -0.02 (p < 0.001) |
| | 1990s | 17 | +0.1 (n.s.) | -1.3 (p < 0.001) | -0.1 (p < 0.001) | -3.3 (p < 0.001) | -0.00 (p < 0.01) |

\* The number of sites for each decade varies for each variable tested. Listed are the number of sites that had sufficient data for SO$_4^{2-}$ tests; this variable generally had the most complete data, and therefore the largest sample sizes.

tests. The outcome of this analysis is a partitioning of the variance in these Z statistics among all of the possible factors that might contribute to it.

In the analyses presented here, the primary sources of variation among the Z statistics are regions, decades, sites and seasons. Our goal in performing this analysis of variance is to account for the majority of variability in Z scores by subdividing the sites appropriately into decades (because trends are clearly different between decades) and regions; this has the effect of removing "noise" from the regional trend signal by grouping together only those sites that have similar trends, and increases the likelihood that we will be able to detect significant regional trends. Results for each variable are discussed separately below.

It is important to recognize that this analysis determines only whether sites in each region are behaving similarly with respect to acidification. It indicates the central tendency of trends in each region, but it does not test whether the sites chosen are truly representative of the regions in which they reside. If the sites in ICP Waters are regionally-representative, then the regional trends presented below should represent valid regional phenomena. But there is no way, using only the trend information analysed here, to determine whether the sites included in ICP-Waters are representative of the population of sites in their regions. Homogeneity among the trends at individual sites in the ICP regions (as is generally observed in all of the results discussed below) gives us some confidence that the sites illustrate trends that are generalizable to the region level, but we do not explicitly test this assumption in the results presented in this report.

### 3.5.1 Sulphate

Decreases in $SO_4^{2-}$ concentrations are nearly universal, and have been throughout the period of record (**Table 20**). The U.K. (in the 1990s) and Atlantic North America (in the 1980s) are the only regions not to show significant decreasing regional trends, although even in these two non-significant cases, the slopes of the changes in $SO_4^{2-}$ are negative. Every region for which data for both decades is available exhibit larger decreases in the 1990s than in the 1980s, suggesting that emissions controls in both North America and Europe have accelerated the rates of decline of sulphur deposition. On the basis only of $SO_4^{2-}$ trends, we would expect to see recovery in ALK values in the 1990s in all regions except the U.K.

**Table 20.** Results of regional trend meta-analysis for $SO_4^{2-}$. Slope values are $\mu eq\, l^{-1}\, yr^{-1}$. Regional trends that are significant at $p<0.05$ are shown in bold.

| Region | Decade | Number of Sites | Trend Sum of Squares | p | Slope |
|---|---|---|---|---|---|
| **Adirondacks and Quebec** | **1980s** | **12** | **12.74** | **0.000** | **-0.63** |
| | **1990s** | **9** | **13.56** | **0.000** | **-1.62** |
| **Europe** | **1980s** | **12** | **6.48** | **0.011** | **-1.26** |
| | **1990s** | **18** | **22.39** | **0.000** | **-4.50** |
| **Midwestern North America** | **1980s** | **12** | **29.03** | **0.000** | **-1.46** |
| | **1990s** | **12** | **64.28** | **0.000** | **-4.25** |
| **Nordic Countries** | **1980s** | **17** | **16.98** | **0.000** | **-0.83** |
| | **1990s** | **20** | **67.24** | **0.000** | **-2.95** |
| Atlantic North America | 1980s | 6 | 0.04 | 0.849 | -0.00 |
| | **1990s** | **7** | **6.36** | **0.012** | **-2.42** |
| U.K. | 1990s | 6 | 0.08 | 0.772 | -0.01 |

**Table 21.** Results of regional trend meta-analysis for $NO_3^-$. Slope values are $\mu eq\, l^{-1}\, yr^{-1}$. Regional trends that are significant at $p<0.05$ are shown in bold.

| Region | Decade | Number of Sites | Trend Sum of Squares | p | Slope |
|---|---|---|---|---|---|
| **Adirondacks and Quebec** | **1980s** | **12** | **25.72** | **0.000** | **+0.46** |
| | **1990s** | **12** | **28.67** | **0.000** | **-1.57** |
| **Europe** | **1980s** | **12** | **15.39** | **0.000** | **+1.37** |
| | **1990s** | **17** | **32.81** | **0.000** | **-2.38** |
| Midwestern North America | 1980s | 12 | 2.98 | 0.084 | +0.04 |
| | 1990s | 12 | 0.45 | 0.501 | -0.00 |
| **Nordic Countries** | **1980s** | **17** | **6.06** | **0.014** | **+0.13** |
| | 1990s | 20 | 0.30 | 0.586 | +0.01 |
| Atlantic North America | 1980s | 6 | 0.04 | 0.832 | +0.00 |
| | 1990s | 7 | 0.30 | 0.586 | -0.00 |
| **U.K.** | **1990s** | **6** | **18.38** | **0.000** | **+0.17** |

## 3.5.2 Nitrate

The 1980s were characterized by regional increases in $NO_3^-$ concentrations in almost all regions (**Table 21**); with the exception of the U.K. (where $NO_3^-$ is increasing in the 1990s), these trends have disappeared in the 1990s. In some cases there is evidence of a regional $NO_3^-$ decline in the 1990s (e.g., Adirondacks/Quebec and Europe). In most regions there have not been decreases in nitrogen deposition that would account for widespread declines in surface water $NO_3^-$. It seems likely that some other regional-scale phenomenon is responsible for the trends in the 1990s. One possibility is subtle changes in climate, or in climatic extremes, that can have large effects on watershed nitrogen cycling (e.g., LÜKEWILLE and WRIGHT 1997; MITCHELL et al. 1996) have produced a temporary hiatus in the long-term increase in $NO_3^-$ concentrations that most nitrogen researchers expect to occur in the future. If this is the case, then the trend results we observe for the 1990s (1990-1994) may not be representative of long-term patterns.

## 3.5.3 Alkalinity (ALK)

Regional trend results for ALK suggest recovery from acidification in some regions, and lack of recovery in others (**Table 22**). In the European and Nordic sites ALK is increasing regionally. The European sites also exhibited increases in ALK in the 1980s, but the rate of increase is accelerated in the 1990s. A very similar pattern is suggested for Atlantic North America (although the regional 1990s trend is not significant). By using data from a larger number of sites in the Northeastern U.S., STODDARD et al. (in press) were able to show that recovery in ALK is now widespread in this region of North America. In the Nordic Countries, there was evidence of regional acidification in the 1980s, but regional recovery (increasing ALK) in the 1990s. The remaining regions (Adirondacks and Quebec, Midwestern North America and the U.K.) all show either no recovery or further acidification.

## 3.5.4 Strong Base Cations (SBC)

Much of the explanation for the lack of recovery in ALK (despite widespread regional declines in $SO_4^{2-}$ and $NO_3^-$) seems to lie in the base cation trends (**Table 23**). While strong base cations (SBC) declines are expected to result from declines in acid anion concentrations (i.e., some of the cation loss from watersheds is driven by the mobile anions $SO_4^{2-}$ and $NO_3^-$; as concentrations of these ions decline, the amounts of base cations mobilized from watershed soils will also decline), cation decreases that are of the same magnitude as anion decreases will lead to little or no recovery in ALK. Both regions with declining $SO_4^{2-}$ that fail to show recovery in the 1990s (Adirondacks/Quebec and Midwestern North America) are characterized by strongly declining base cation concentrations (-4.1

**Table 22.** Results of regional trend meta-analysis for *ALK*. Slope values are $\mu eq\,l^{-1}\,yr^{-1}$. Regional trends that are significant at $p<0.05$ are shown in bold.

| Region | Decade | Number of Sites | Trend Sum of Squares | p | Slope |
|--------|--------|-----------------|----------------------|------|-------|
| **Adirondacks and Quebec** | **1980s** | **12** | **19.24** | **0.000** | **-1.39** |
|  | 1990s | 9 | 3.38 | 0.066 | +0.53 |
| **Europe** | **1980s** | **5** | **3.93** | **0.047** | **+2.50** |
|  | **1990s** | **13** | **16.72** | **0.000** | **+7.94** |
| **Midwestern North America** | **1980s** | **11** | **6.26** | **0.012** | **+0.71** |
|  | **1990s** | **7** | **10.65** | **0.001** | **-1.60** |
| **Nordic Countries** | **1980s** | **14** | **26.92** | **0.000** | **-1.41** |
|  | **1990s** | **19** | **35.28** | **0.000** | **+2.50** |
| **Atlantic North America** | **1980s** | **5** | **13.29** | **0.000** | **+1.37** |
|  | 1990s | 2 | 0.39 | 0.531 | +1.14 |
| U.K. | 1990s | 6 | 0.20 | 0.653 | +0.15 |

**Table 23.** Results of regional trend meta-analysis for *SBC*. Slope values are $\mu eq\,l^{-1}\,yr^{-1}$. Regional trends that are significant at $p<0.05$ are shown in bold.

| Region | Decade | Number of Sites | Trend Sum of Squares | p | Slope |
|--------|--------|-----------------|----------------------|------|-------|
| **Adirondacks and Quebec** | **1980s** | **12** | **4.00** | **0.046** | **+0.39** |
|  | **1990s** | **12** | **18.73** | **0.000** | **-3.41** |
| Europe | 1980s | 6 | 0.43 | 0.513 | +0.00 |
|  | 1990s | 11 | 1.34 | 0.247 | -6.25 |
| Midwestern North America | 1980s | 12 | 1.08 | 0.298 | +0.94 |
|  | **1990s** | **12** | **53.33** | **0.000** | **-4.79** |
| **Nordic Countries** | **1980s** | **20** | **58.29** | **0.000** | **-2.42** |
|  | 1990s | 20 | 0.01 | 0.913 | -0.05 |
| Atlantic North America | 1980s | 6 | 0.03 | 0.873 | -0.13 |
|  | 1990s | 7 | 0.56 | 0.454 | -0.89 |
| U.K. | 1990s | 6 | 2.35 | 0.125 | -1.46 |

and -3.1 $\mu$eq $l^{-1}$ $yr^{-1}$, respectively). In the Nordic Countries, declining base cations have previously been suggested as the reason that ALK recovery was not occurring in the 1980s (KIRCHNER and LYDERSEN 1995). Our results suggest that base cations are no longer declining in the Nordic countries, and regional recovery in ALK is occurring (**Table 22**).

### 3.5.5 Hydrogen Ion ($H^+$)

All of the hydrogen ion trends that are detectable at a regional level are consistent with the ALK trends observed within each region (**Table 24**). In regions where ALK is recovering, hydrogen ion concentrations are either decreasing or unchanged. Because large increases in hydrogen ion concentrations would only be expected in recovering sites with ALK values below zero, not all regions with ALK recovery exhibit decreases in hydrogen ion concentration (e.g., especially where sites represent a mixture of acidic and non-acidic surface waters).

**Table 24.** Results of regional trend meta-analysis for $H^+$. Slope values are $\mu$eq $l^{-1}$ $yr^{-1}$. Regional trends that are significant at p<0.05 are shown in bold.

| Region | Decade | Number of Sites | Trend Sum of Squares | p | Slope |
|---|---|---|---|---|---|
| Adirondacks and Quebec | 1980s | 12 | 1.70 | 0.193 | +0.00 |
| | **1990s** | **12** | **7.09** | **0.008** | **-0.01** |
| Europe | 1980s | 12 | 2.35 | 0.126 | -0.05 |
| | 1990s | 18 | 0.00 | 0.947 | -0.00 |
| **Midwestern North America** | **1980s** | **4** | **38.14** | **0.000** | **-0.02** |
| | 1990s | 4 | 3.03 | 0.082 | -0.00 |
| **Nordic Countries** | **1980s** | **20** | **6.08** | **0.014** | **+0.01** |
| | **1990s** | **22** | **25.88** | **0.000** | **-0.15** |
| **Atlantic North America** | **1980s** | **6** | **14.84** | **0.000** | **-0.02** |
| | 1990s | 7 | 3.00 | 0.083 | +0.01 |
| U.K. | 1990s | 6 | 2.63 | 0.105 | -0.14 |

# 4. Assessment of Nitrogen Leaching

The significant decrease in sulphate concentrations at European and North American ICP Waters sites can be explained by a decline in sulphur deposition due to successful emission reduction measures (chapter 3). At many catchments increasing nitrate concentrations during the 1980s and decreasing values during the 1990s can be observed, although there is no significant upwards or downwards trend in nitrogen (N) deposition. This pattern cannot be explained by other changes in the catchments, such as point sources, agriculture, clearcutting and forest fire. Hence, dose/response relationships are much more complicated for nitrogen than for sulphur. Changes in N deposition may not always be directly correlated with changes in $NO_3$ leaching. The "nitrogen (N) status" of an ecosystem, or changes in N status, seem to be an important indicator of N saturation.

This chapter is a summary and update of results (and discussions) presented by TRAAEN and STODDARD (1995) and STODDARD and TRAAEN (1995).

## 4.1 Nitrate in Runoff

Reports of increased nitrate concentrations in runoff 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 et al. 1988). Increasing trends in $NO_3$ concentrations were also revealed in the US EPA's Long-term Monitoring Project. Nine of 15 drainage lakes monitored in the Adirondack area showed increases in nitrate concentrations, ranging from 0.5 to 2.0 $\mu eq\ l^{-1}\ yr^{-1}$ in the period 1982 -1991 (DRISCOLL & VAN DREASON 1993). Further, 5 of 8 streams in the Catskill area exhibited significant upward trends in nitrate, ranging from 1.3 to almost 3 $\mu eq\ l^{-1}\ yr^{-1}$ in the period 1983 -1989 (MURDOCH & STODDARD 1992). These trends have partly disappeared in the 1990s (STODDARD et al., in press), a phenomenon that can also be observed at many ICP Waters sites (chapter 3).

Nevertheless, any excess nitrate leaving a catchment will contribute to acidification of the watershed by removing base cations from soil and by mobilising aluminium and $H^+$ ions. $NO_3$, when accompanied by $H^+$ instead of base cations, will reduce the acid neutralising capacity (ANC; see chapter 2.4.3).

The frequency distribution for 5 intervals of yearly average $NO_3$ concentrations for ICP Waters sites from each of the participating countries were calculated (**Table 25**). Most of the data are from

TRAAEN and STODDARD (1995). In addition sites from Hungary, Latvia, Italy and Poland were considered. Data from all 169 sites are combined in **Figure 34**. Unweighted mean values, as used in this report, underestimate actual yearly 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

Thirty percent of the ICP sites have yearly average concentrations of nitrate below 5 µeq $l^{-1}$. Most of these sites are situated in the Nordic countries (except Denmark), Ireland, Canada, U.S. and Russia. More than half of the sites have nitrate concentrations above 10 µeq $l^{-1}$. Due to the episodic pattern of nitrate at many sites, the significance of nitrate in acidification is often greater than shown by average figures (see also 4.4). Nitrate concentrations greater than 50 µeq $l^{-1}$ are found at 27% of the sites, mainly in Germany (27 out of 30 German sites), but also in Latvia (8 sites), Austria (3 sites), Belgium (2 sites) and Denmark (2 sites), Italy (2 sites) and Hungary (1 site). Sites in The Netherlands differ from most other sites in having ammonium as the main form of inorganic N. All Dutch sites have $NH_4$ concentrations greater than 100 µeq $l^{-1}$. If conditions for nitrification should improve, the $NH_4$ content of these sites has a great acidification potential.

In total, the results suggest that more than 50% of the ICP Waters sites have $NO_3$ concentrations above those expected from watersheds with no impact from N deposition (STODDARD 1994).
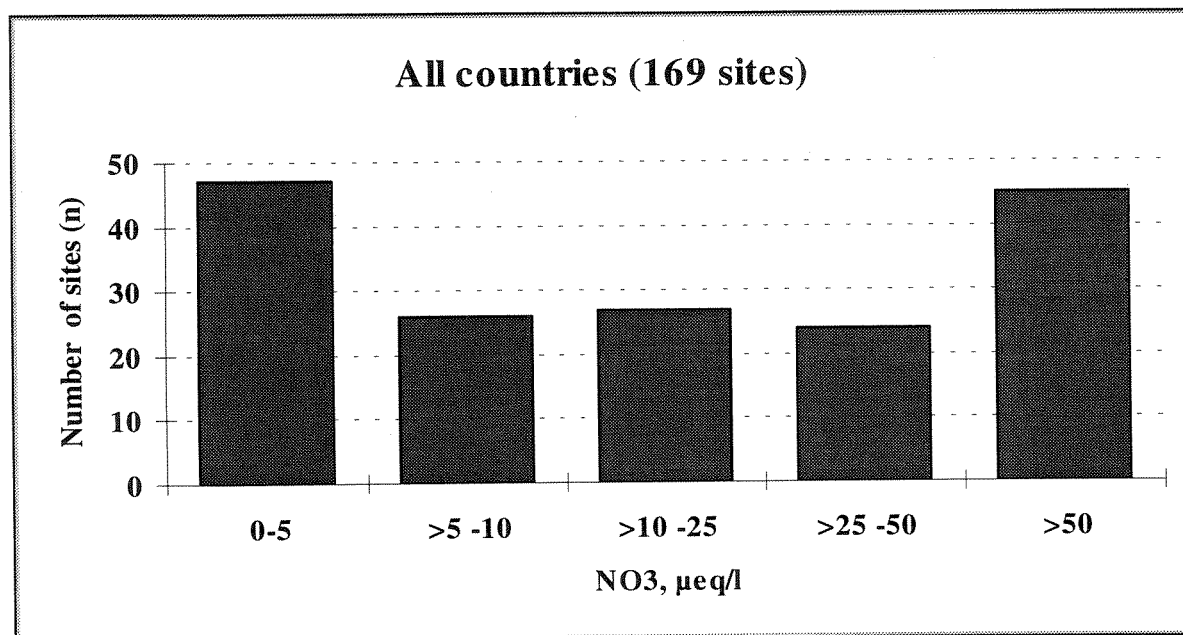


**Figure 34.** Frequency distribution of yearly average nitrate concentrations at 169 ICP Waters sites.

**Table 25.** Frequency distribution of yearly average $NO_3$ concentrations ($\mu$eq $l^{-1}$) at ICP Waters sites.

| Interval | 0-5 | >5 -10 | >10 -25 | >25 -50 | >50 | sum |
|---|---|---|---|---|---|---|
| Norway | 4 | 5 | 1 | 0 | 0 | 10 |
| Sweden | 3 | 5 | 3 | 0 | 0 | 11 |
| Finland | 6 | 0 | 0 | 0 | 0 | 6 |
| Denmark | 0 | 0 | 0 | 0 | 2 | 6 |
| Germany | 1 | 0 | 0 | 2 | 27 | 30 |
| The Netherlands | 0 | 4 | 1 | 0 | 0 | 5 |
| Belgium | 0 | 0 | 1 | 5 | 2 | 8 |
| Austria | 0 | 0 | 8 | 0 | 3 | 11 |
| UK | 1 | 1 | 4 | 0 | 0 | 6 |
| Ireland | 7 | 6 | 4 | 1 | 0 | 18 |
| Canada | 13 | 1 | 2 | 2 | 0 | 18 |
| USA | 9 | 4 | 2 | 8 | 0 | 23 |
| Russia | 2 | 0 | 0 | 0 | 0 | 2 |
| Latvia | 1 | 0 | 1 | 2 | 8 | 12 |
| Italy | 0 | 0 | 0 | 3 | 2 | 5 |
| Hungary | 0 | 0 | 0 | 0 | 1 | 1 |
| Poland | 0 | 0 | 0 | 2 | 0 | 2 |
| Sum | 48 | 26 | 27 | 24 | 45 | 169 |
| % | 28 | 15 | 16 | 14 | 27 | 100 |

## 4.2 Relative Importance of Nitrate in Acidification

The importance of nitrate in acidification relative to sulphate can be illustrated by the concentration of $NO_3$ divided by the sum of non-marine sulphate and $NO_3$ ($\mu$eq $l^{-1}$): $KNS = NO_3^- / (SO_4^{2-}* + NO_3^-)$, $mmol_c$ $mmol_c^{-1}$. The frequency distribution of KNS values for sites in different ICP Waters countries was calculated (**Table 26; Figure 35**).

Nitrate constitutes more than 10% of the non-marine acid anions in 63 % of the ICP-sites. 5 percent have KNS values > 0.5, i.e., at these sites nitrate makes a greater contribution to acidification than sulphate. These sites are situated in Germany, Austria, Ireland and Italy. The results show that sulphate is generally the most important acid anion in acidification, but nitrate does contribute significantly at a majority of the ICP sites.

The importance of nitrate as acidifying anion may be fairly constant or vary with seasons, depending upon the degree of N saturation in the catchment and the sulphate concentration. At Birkenes, southern Norway, $NO_3$ amounts to 18% of the acid non-marine anions in spring, but has no importance in the summer season. In Constable Lake, USA, the importance of nitrate varies from 38% in spring to 4% in summer. Vorderer Schachtenbach, Germany, shows only minor seasonal changes. The KNS-values lies between 0.35 and 0.45 all year, suggesting a late stage of nitrogen saturation (see chapter 4.4).

Seasonal changes in the relative importance of $NO_3$ in acidification are also shown in an assessment of Canadian lakes (JEFFRIES et al. 1995): mean KNS values for lakes having high concentration of nitrate (>15 $\mu$eq $l^{-1}$) were usually in the range 0.12 to 0.19 in the period January to May, while the values in the period June to October were usually well below 0.1.

**Table 26.** Relative importance of $NO_3$ in acidification at ICP Waters sites; $KNS = NO_3^- / (SO_4^{2-*} + NO_3^-)$, $\mu$eq l $\mu$eq $l^{-1}$.

| Interval | 0 - 0.05 | >0.05 - 0.1 | >0.1 - 0.25 | >0.25 - 0.5 | >0.5 | sum |
|---|---|---|---|---|---|---|
| Norway | 3 | 1 | 3 | 3 | | 10 |
| Sweden | 5 | 6 | | | | 11 |
| Finland | 4 | 1 | | | | 5 |
| Denmark | | | | 2 | | 2 |
| Germany | 1 | 6 | 36 | 19 | 3 | 65 |
| The Netherlands | 3 | 2 | | | | 5 |
| Belgium | | 1 | 7 | | | 8 |
| Austria | 1 | | 4 | 5 | 3 | 13 |
| UK | | 1 | 4 | 1 | | 6 |
| Ireland | | | 2 | 1 | 1 | 4 |
| Canada | 6 | 4 | | | | 10 |
| USA | 10 | 3 | 9 | | | 22 |
| Russia | 2 | | | | | 2 |
| Latvia | 2 | 7 | 2 | 1 | | 12 |
| Hungary | | 1 | | | | 1 |
| Poland | | | | 2 | | 2 |
| Italy | | | | 3 | 2 | 5 |
| Sum | 37 | 33 | 67 | 37 | 9 | 183 |
| % | 20 | 18 | 37 | 20 | 5 | 100 |
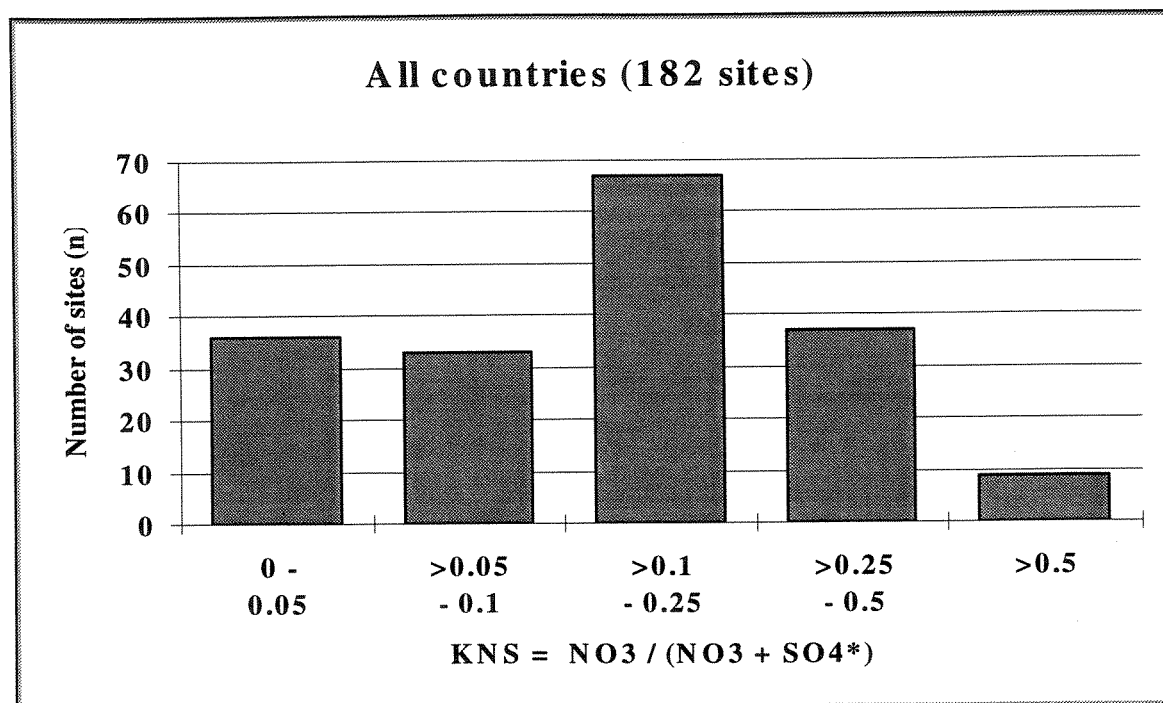
**Figure 35.** Relative importance of nitrate in acidification at 182 ICP sites.

## 4.3 Nitrogen Deposition and Inorganic Nitrogen in Runoff

Empirical data show that there is a relationship between nitrogen deposition and nitrogen loss from forested ecosystems in Europe. According to DISE and WRIGHT (1995) N retention capacity within many forests is reached at an atmospheric N input of 9-10 kg N ha$^{-1}$ yr$^{-1}$. Intermediate N deposition between 9 and 25 kg N ha$^{-1}$ yr$^{-1}$ may lead to significant amounts of $NO_3$ leached to seepage or surface water. These thresholds are based on data from monitoring plots across a gradient of current N deposition in Europe and from several experimental sites (ENSF catchments (Evaluation of Nitrogen and Sulphur Fluxes); NITREX project, e.g., TIETEMA & BEIER 1995).

Input/output data for nitrogen are not available for most of the ICP Waters sites. However, one may get a general view of N leakage by comparing the concentrations in runoff ($NO_3$ + $NH_4$) and total (wet + dry) N deposition fluxes (**Figure 36**). Most runoff data used in this report are from 1990 (TRAAEN and STODDARD 1995). For sites included in the ICP Waters data base after 1994 (Latvia, Italy, Poland and Hungary), yearly average concentrations for 1990, 1991 or 1992 were used; choosing the year with the most complete data set. Deposition fluxes are values for the EMEP grid where the respective ICP Water sites are situated (data from Meteorological Synthesising Centre-West of EMEP, Norwegian Meteorological Institute).

The ICP Waters data (**Figure 36**) show a pattern confirming the general concept that watersheds have threshold values for N deposition above which increased N leakage occurs. Some catchments have a pronounced nitrogen leakage at a total N deposition as low as 7 - 11 kg N ha$^{-1}$ yr$^{-1}$. In the case of the Latvian sites an influence of agriculture (e.g., pasure land) on water quality cannot be excluded. Some other catchments have very high annual precipitation (1200 -2500 mm yr$^{-1}$). These sites may have a high output *flux* of N in runoff even if the concentrations are moderate. They are mainly situated in Norway, Ireland, Italy and the UK. TRAAEN and STODDARD (1995) calculated output fluxes for ICP Waters sites by subtracting an estimated evapotranspiration of 300 mm yr$^{-1}$ from EMEP yearly average precipitation values. The results stressed that there might be N leakage even below total deposition values of 10 kg N ha$^{-1}$ yr$^{-1}$ at these sites. This might be due to short contact time between water and the soil's root zone during heavy rainfall. STODDARD (1994) observed a threshold for N-leakage at a wet deposition as low as 2.5 kg N ha$^{-1}$ yr$^{-1}$, corresponding to a wet + dry deposition of approximately 5 kg N ha$^{-1}$ yr$^{-1}$.
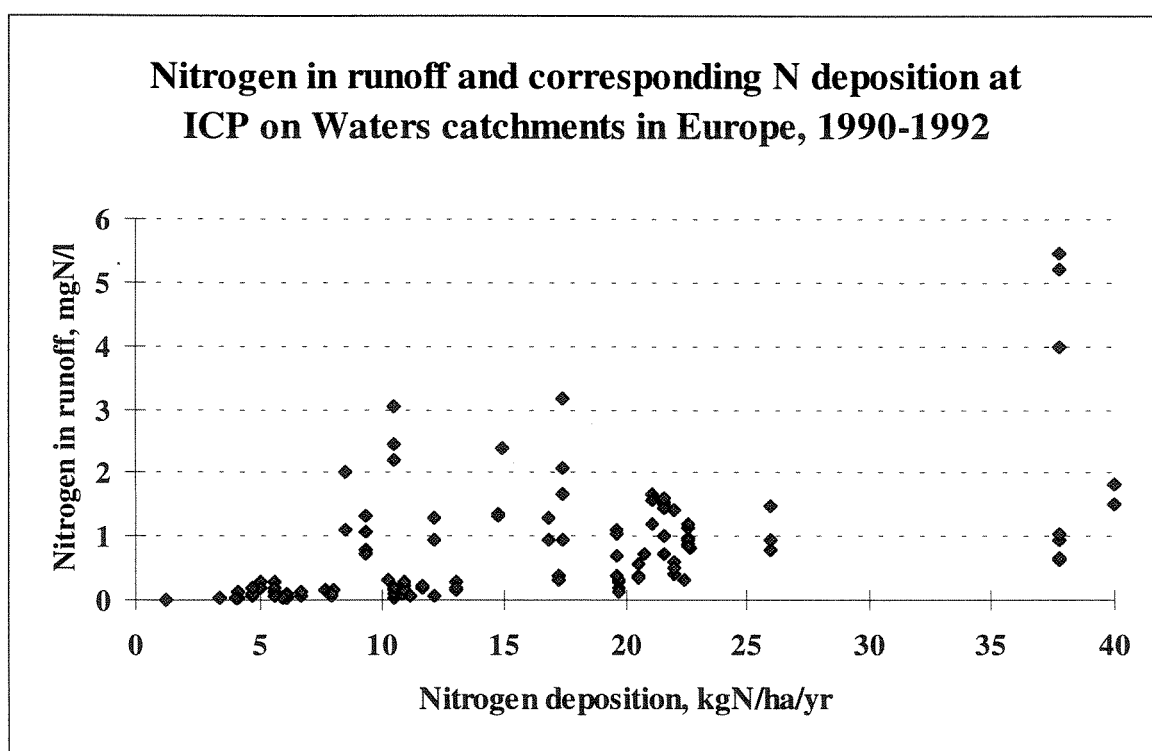


**Figure 36.** Nitrogen (NO$_3^-$ + NH$_4^+$) concentration in runoff and total nitrogen deposition fluxes (wet + dry) for European ICP on Waters sites in 1990-1992. The deposition data are 1990 values for the EMEP grids in which the respective sites are situated (data from Norwegian Meteorological Institute). Most runoff data are yearly averages for 1990.

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 N deposition fluxes, and finally, the site selection does not necessarily include the most sensitive sites with respect to N leakage.

Despite these uncertainties, the data in **Figure 37** are in reasonably good agreement with data from DISE and WRIGHT (1995) mentioned above.

The same is true for recent results of the ICP Integrated Monitoring Programme. Catchment and plot-scale input/output budgets for N were calculated for several sites (FORSIUS et al. 1996). Site specific runoff or soil water output fluxes (kg N $ha^{-1}$ $yr^{-1}$) were compared with site specific weighted throughfall and bulk deposition (kg N $ha^{-1}$ $yr^{-1}$). A clear relationship between $NH_4$ + $NO_3$ deposition and leaching of inorganic N (mostly $NO_3$) can be observed. $NO_3$ leaching occurs rarely at deposition levels < 8-10 kg N $ha^{-1}$ $yr^{-1}$.

Even if there is a strong correlation between the current N deposition and N runoff, there are several other factors influencing N leakage. Empirical data show that N uptake depends strongly on forest age and soil N status. The yearly N demand of an aggrading forest is relatively high and decreases as the trees reach maturity (EMMETT et al. 1993). In soils with large N pools, receiving additional N input through atmospheric deposition, microbial N demands may easily be fulfilled and other factors such as labile organic carbon may limit soil microbial processes (JOHNSON 1992, JOSLIN et al. 1992). The likelihood that the watershed will leach $NO_3$ is higher than at sites with a low soil N pool.

## 4.4 Classification of Nitrogen Saturation

Nitrogen is the growth-limiting nutrient in many natural 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 plants and micro-organism growth. N saturation is defined by persistent losses of nitrate ($NO_3$) and/or ammonium ($NH_4$) in streamflow or groundwater discharge which may be accompanied by increases in N mineralization and nitrification in soil (ABER et al. 1989).

STODDARD (1994) has elaborated the term *persistent* by addressing monthly variations in $NO_3$ leaching from catchments and thus by considering the seasonality of biological N turnover processes.

He names different stages in a hypothetical time course which ecosystems can pass through if atmospheric N deposition remains high or increases.

The N cycle at **Stage 0** is dominated by forest and microbial uptake governing the seasonal $NO_3$ pattern of runoff water. $NO_3$ concentrations are very low during most of the year, and measurable concentrations can only be found during snowmelt. At **Stage 1**, this pattern is amplified: the switch from physical to nutrient limitation in spring is postponed. Substantial $NO_3$ may leave the watershed during extreme hydrological events. In **Stage 2**, the seasonal onset of N limitation is even further delayed so that biological demand does not longer control $NO_3$ 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 $NH_4$ in soils, results in an increase in $NO_3$ baseflow concentrations to levels as high as those found in deposition. **Stage 3** is characterised by a lack of any coherent seasonal pattern in $NO_3$ output. Mineralization of stored N can add substantially to $NO_3$ output in surface waters which may, together with gaseous emissions ($N_2O$), 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: TRAAN and STODDARD 1995, STODDARD and TRAAN 1995). Clear criteria for sites with frequent samples (**Table 27**) and infrequent samples were set (**Table 28**).

**Table 27.** N saturation stage criteria for sites with **frequent** samples. The criteria are based on monthly average $NO_3^-$ concentrations.

| Stage | Criteria |
|-------|----------|
| **Stage 0** | $\geq 3$ months in the growing season with $NO_3 \leq 3$ $mmol_c$ $m^{-3}$ and peak value $< 20$ $mmol_c$ $m^{-3}$. |
| **Stage 1** | 1-2 months in the growing season with $NO_3 \leq 3$ $mmol_c$ $m^{-3}$, or $\geq 3$ months in the growing season with $NO_3 \leq 3$ $mmol_c$ $m^{-3}$ and peak value $\geq 20$ $mmol_c$ $m^{-3}$. |
| **Stage 2** | No month with $NO_3 \leq 3$ $mmol_c$ $m^{-3}$ and $\geq 3$ months in the growing season with $NO_3 \leq 50$ $mmol_c$ $m^{-3}$. |
| **Stage 3** | $< 3$ months with $NO_3 < 50$ $mmol_c$ $m^{-3}$. |
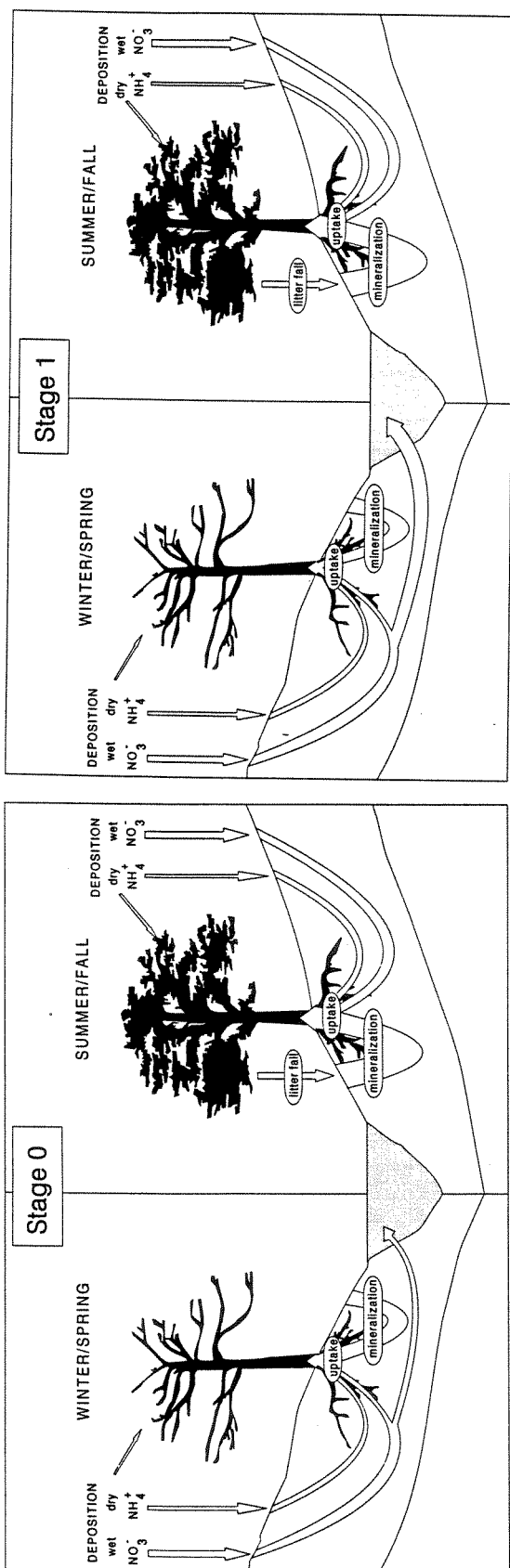
**Figure 37a.** Schematic representation of the watershed N cycle at **Stage 0** (a) and **Stage 1** (b) of N saturation; examples at ICP Waters sites. The size of arrows is roughly equivalent to the sizes of fluxes for each process. Cycles are devided into winter/spring (dormant) and summer/fall (growing) season.
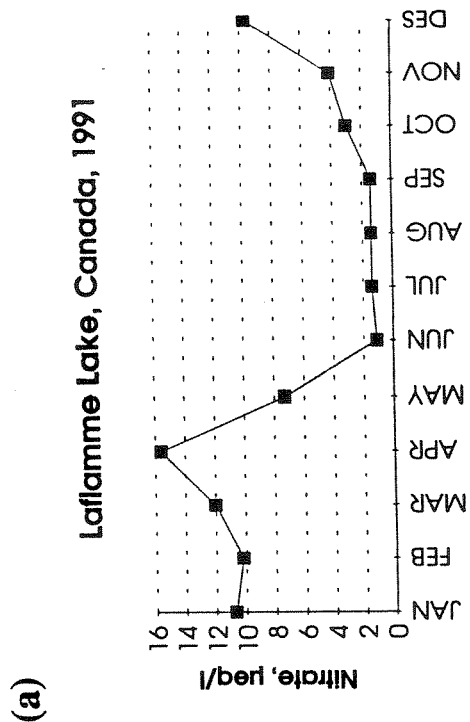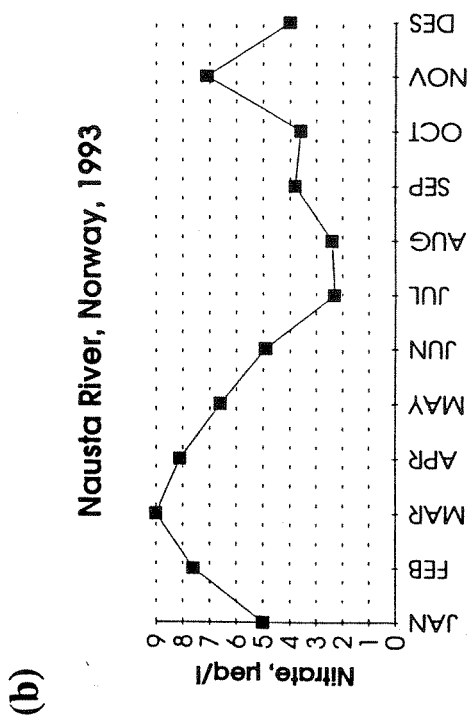
**Table 28.** N saturation stage criteria for sites with **infrequent** samples (equally spaced in time, at least 2 samples in the growing season).

| Stage | Criteria |
|---|---|
| **Stage 0** | At least $\geq 2$ values in the growing season with $NO_3 \leq 3$ mmol$_c$ m$^{-3}$ and peak value $\leq 10$ mmol$_c$ m$^{-3}$. |
| **Satge 0/1** | 1 value in the growing season $\leq 3$ mmol$_c$ m$^{-3}$ and peak value $\leq 10$ mmol$_c$ m$^{-3}$, or $\geq$ values in the growing season $\leq 3$ mmol$_c$ m$^{-3}$ and paek value $> 10$ mmol$_c$ m$^{-3}$, but $< 20$ mmol$_c$ m$^{-3}$. |
| **Stage 1** | $\leq 1$ value in the growing season $\leq 3$ mmol$_c$ m$^{-3}$ and peak value $> 20$ mmol$_c$ m$^{-3}$. |
| **Stage 1/2** | $\geq 1$ value in the growing season $\leq 5$ mmol$_c$ m$^{-3}$, but $> 3$ mmol$_c$ m$^{-3}$. |
| **Stage 2** | $\geq 2$ values in the growing season $> 5$ mmol$_c$ m$^{-3}$, but $\leq 50$ mmol$_c$ m$^{-3}$. |
| **Tage 2/3** | At least 1 value in the growing season $> 50$ mmol$_c$ m$^{-3}$, and at least 1 value $\leq 50$ mmol$_c$ m$^{-3}$. |
| **Stage 3** | All values in the growing season $> 50$ mmol$_c$ m$^{-3}$. |

Typical examples for all 3 nitrogen saturation stages at different ICP Waters sites are summarized in **Figure 37a** and **Figure 37b** (from STODDARD and TRAAN 1995).

Further. examples of seasonal variation of $NO_3$ concentration from each country and suggested classification of N saturation were given by TRAAEN and STODDARD (1995).

Additional countries, Latvia, Italy, Hungary and Poland have recently (1995/96) started reporting data to the ICP Waters Programme Centre. The criteria for N saturation stages (**Table 27 and 28**) were also applied to these new datasets (**Figure 38 to Figure 41**).

A summary of the N saturation status of ICP on Waters catchments is shown in **Table 29** Included are 144 sites analysed by TRAAEN and STODDARD (1995) and all 19 Latvian sites, Italy, Poland and Hungary.
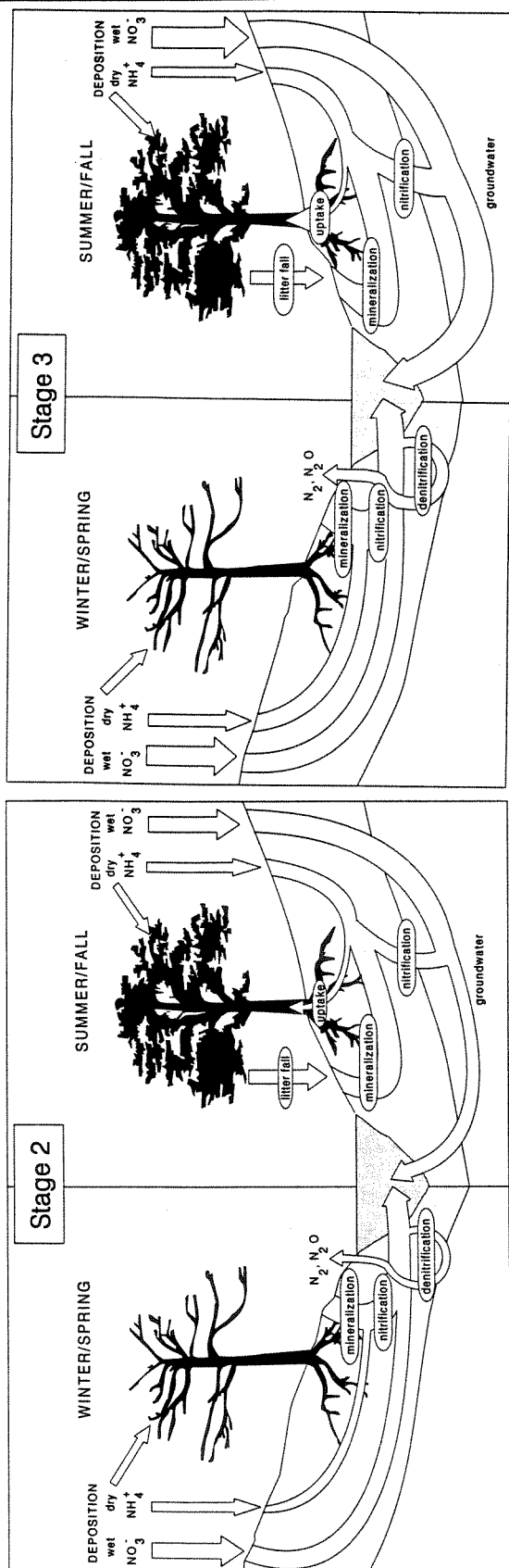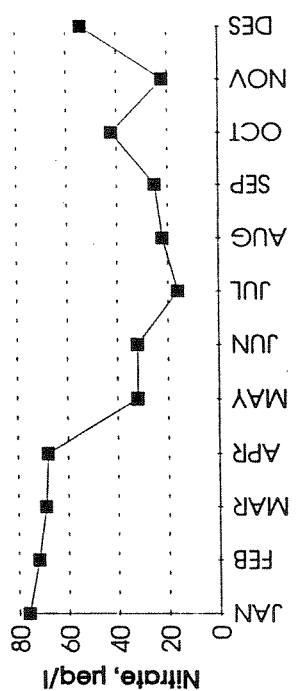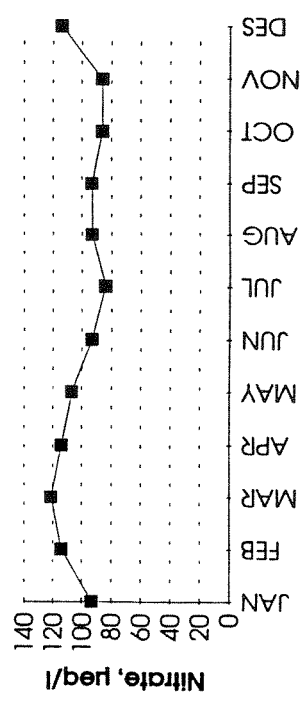
**Figure 37b**. Schematic representation of the watershed N cycle at **Stage 2** (c) and **Stage 3** (d) of N saturation; examples at ICP Waters sites. The size of arrows is roughly equivalent to the sizes of fluxes for each process. Cycles are devided into winter/spring (dormant) and summer/fall (growing) season.

**Figure 38.** Stages of nitrogen saturation based on the annual pattern of nitrate concentrations (monthly averages). JELG-2: Lielupe, below Jelgava. SOZ-T: Tulija, Zoseni. DAUG-1: Daugava, above Daugavpilis.
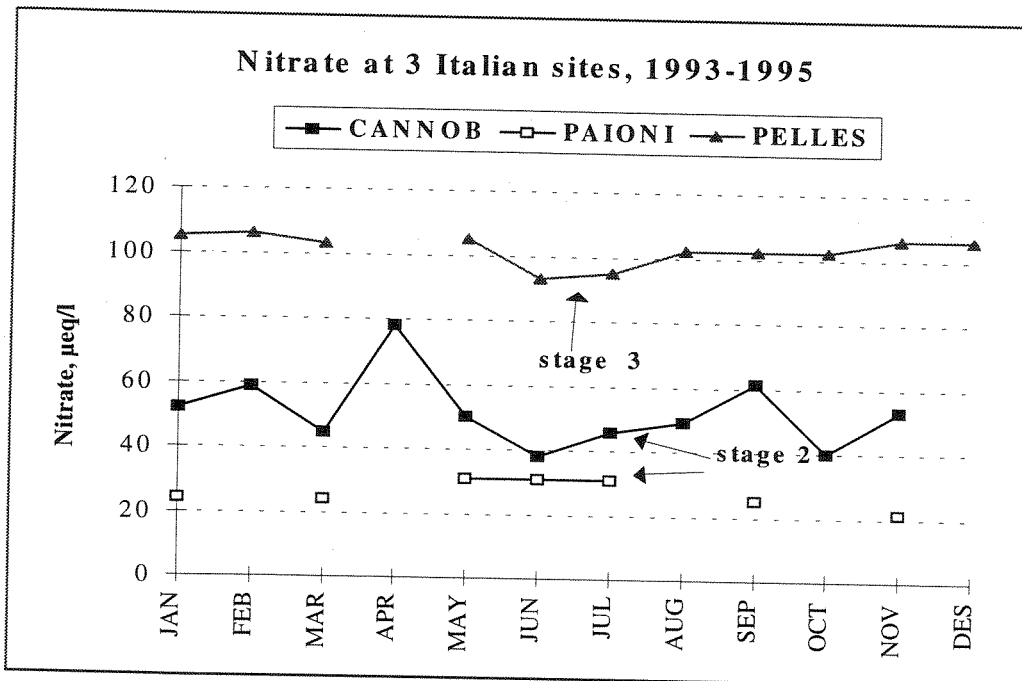


**Figure 39.** Stages of nitrogen saturation based on the annual pattern of nitrate concentrations (monthly averages). CANNOB: River Cannobino. PAIONE: Lake Paione Inferiore. PELLES: River Pellesino.
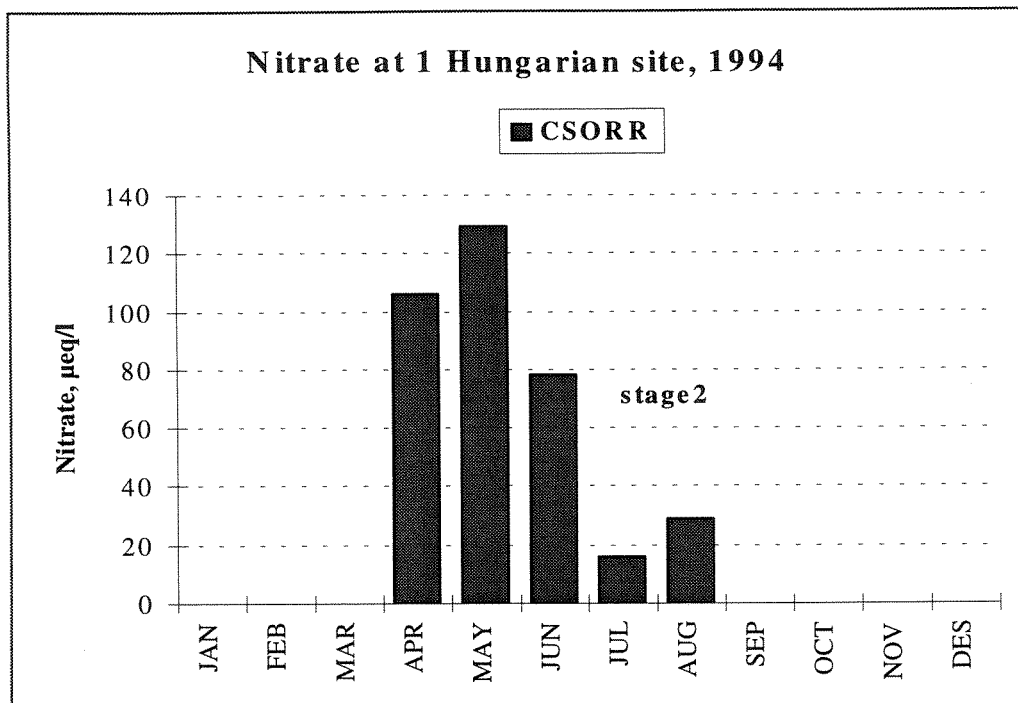
**Figure 40.** Stages of nitrogen saturation based on the annual pattern of nitrate concentrations (monthly averages). CSORR: Csorret Lake, Matra mountains.
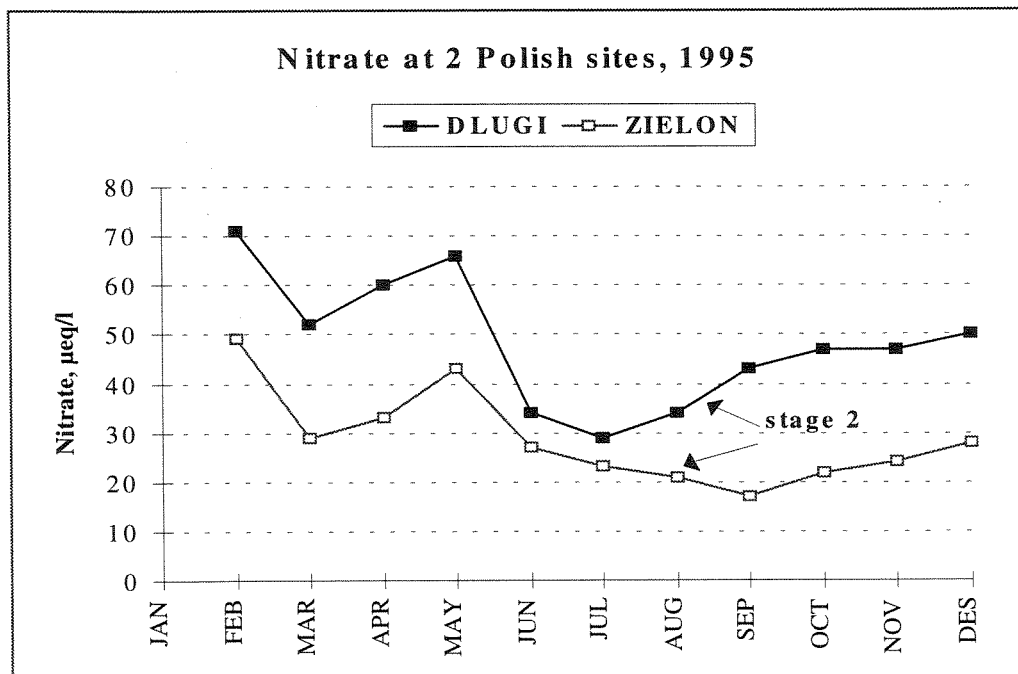


**Figure 41.** Stages of nitrogen saturation based on the annual pattern of nitrate concentrations (monthly averages). DLUGI: Dlugi lake, Tatra mountains. ZIELON: Zielony lake, Tatra Mountains.

**Table 29.** Number of ICP catchments within each stage of nitrogen saturation. For explanation see text.

| Country | Stage 0 | Stage 0/1 | Stage 1 | Stage 2 | Stage 2/3 | Stage 3 | Total |
|---|---|---|---|---|---|---|---|
| Norway | 4 | | 4 | 2 | | | 10 |
| Sweden | 6 | | 3 | 2 | | | 11 |
| Finland | 4 | 2 | | | | | 6 |
| Denmark | | | | | | 2 | 2 |
| Germany | | 1 | | 8 | 1 | 20 | 30 |
| Netherlands | | | | | | 5 | 5 |
| Belgium | | | | 7 | | 1 | 8 |
| Austria | | | | 3 | | 3 | 6 |
| UK | 1 | 2 | | 3 | | | 6 |
| Ireland | 10 | | 5 | 3 | | | 18 |
| Canada | 9 | 6 | | 3 | | | 18 |
| USA | 8 | 4 | 3 | 7 | | | 22 |
| Russia | 2 | | | | | | 2 |
| Italy | | | | 3 | | 2 | 5 |
| Hungary | | | | 1 | | | 1 |
| Latvia | 1 | | 1 | 6 | 2 | 2 | 12 |
| Poland | | | | 2 | | | 2 |
| **Total** | **45** | **15** | **16** | **50** | **3** | **35** | **164** |

More than half (88 out of 163) of the ICP catchments exhibit a high degree of nitrogen saturation (Stage 2 or 3). Such sites are found in all participating countries except Finland and Russia. All ICP on Waters sites in Denmark, Belgium, The Netherlands, Austria, Italy, Poland and Hungary, and 29 out of 30 German sites and 10 out of 12 Latvian sites show Stage 2 or Stage 3 patterns.

## 4.5 Relationship between N Deposition and N Saturation Stages

There is a clear connection between N deposition values and stage classification at European ICP Waters sites (**Figure 42**). At Stage 0 and 1 catchments only 8 out of 36 have N deposition values above 10 kg $ha^{-1}$ $yr^{-1}$, while at Stage 2 only 6 out of 36 have deposition values below 10 kg $ha^{-1}$ $yr^{-1}$. 22 out of 32 Stage 3 sites have deposition values above 20 kg $ha^{-1}$ $yr^{-1}$. These results suggest that a threshold value of N deposition, above which significant N saturation can exist, occurs at 7 kg $ha^{-1}$ $yr^{-1}$.
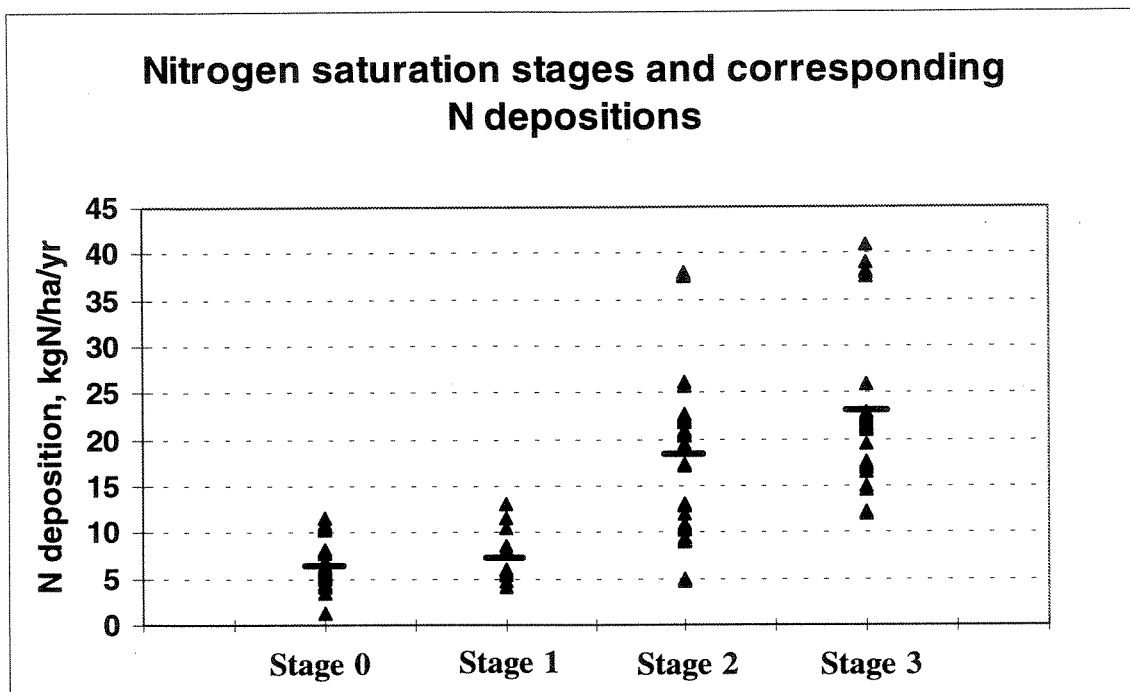
**Figure 42.** Relationship between nitrogen deposition (wet + dry, estimated from EMEP grid data) and stages of nitrogen saturation for 102 European ICP on Waters sites. Average values are indicated by horizontal bars (e.g. Stages 0/1 and 2/3 are not included).

However, it should be reiterated that deposition is only one factor influencing $NO_3$ leaching from catchments. Nitrogen demands on the part of vegetation (controlled largely by forest age and health) and on the part of soils (controlled largely by pool sizes for nitrogen and carbon) must be satisfied before N saturation can occur. Thus, the site's **"nitrogen status"** has a significant influence on the timing and extent of response to (changes in) N deposition.

Because information on forest status and soil chemistry is unavailable for most ICP on Waters catchments, the data in **Figure 42** indicate only a strong influence of rates of N deposition on the stage of N saturation, but cannot precisely identify what an appropriate threshold value of N deposition is.

## 4.6 Indicators of Nitrogen Status

Results of monitoring programs and manipulation experiments (e.g., NITREX) in Europe and the USA suggest that $NO_3$ leaching as well as N in throughfall are significantly correlated with N concentrations in current needles, needle litterfall and forest floor (MCNULTY et al. 1991, TIETEMA and BEIER 1995, GUNDERSEN et al. 1996). Even at high N deposition, changes in N

status of forests may only need to occur in these compartments, while changes in mineral soil, e.g., are marginal. Adding or removing (roof) N to different NITREX manipulation sites, GUNDERSEN et al. (1996) found that changes in $NO_3$ leaching were small at low N status sites, but substantial at high N status sites. C/N ratios in compartments such as forest floor may be reliable indicators of N status that are easy to measure.

FORSIUS et al. (1996) combined statistics on ICP Integrated Monitoring data with results of the EXMAN and NITREX manipulation projects (TIETEMA and BEIER 1995). They found that inorganic N output fluxes from catchments and plots are highly correlated with N deposition, N concentration in current year needles and organic matter and N flux in litterfall. FORSIUS et al. (1996) point out that such statistical relationships, elaborated at intensively studied sites, in combination with regional monitoring data (e.g., from ICP Waters and ICP Forests) are useful tools to link process level data with regional-scale questions.

## 4.7 Climate and Hydrology

Seasonal changes in runoff may not only be driven by biological activity (and deposition), but also by short-term changes in climate. At Birkenes, an ICP Waters monitoring site in southern Norway, the mean winter temperature (1 January to 31 March) can be above or below 0°C. This has a large influence on hydrological flow patterns during winter and spring months and thereby also on the concentrations and fluxes of chemical components including $NO_3$. The difference in runoff patterns of $NO_3$ between "cold" and "warm" years is discussed in detail by LYDERSEN (1995). When winter air temperature is higher than 0°C most of the precipitation is rain, passing the catchment slowly by subsurface flow. High $NO_3$ streamwater concentrations are observed during spring snow-melt following cold winters. During relatively warm years the highest N output fluxes occur in late autumn and during the first three months of the year (limited biological activity), and the yearly fluxes are generally higher than in years with cold winters. The maximum fluxes in years with snow occur during melt in spring.

Changes in soil temperature themselves can have large effects on N cycling in forested ecosystems (e.g., MITCHELL et al. 1996, WRIGHT and LÜKEWILLE 1996, LÜKEWILLE and WRIGHT 1997). Thus, the N saturation status of a catchment may be enhanced directly, e.g., by increases in temperature and N release, as well as more indirectly by changes in seasonal runoff patterns. This is of additional significance with regard to global change scenarios predicting an increase in ambient air

temperature in this area by 8 °C in winter and 3 °C in summer within the next 50 years (global circulation models; HOUGHTON 1994, MITCHELL et al. 1995).

# 5. Effects of Acidification on Aquatic Fauna: Geographical Extent, Dose/Response, and Long-term Trends

## 5.1 Background

The biological monitoring of the ICP Waters Programme consist of national activities of the participating countries. Monitoring practice varies from country to country with regard to time-series and type of studies. In this report we concentrate on *invertebrate studies*. The longest biological monitoring series are from Norway, starting in 1981 and 1982. Other countries with several years of monitoring are Germany, Sweden, the United Kingdom and Ireland, while newcomers such as Latvia and Poland have just started to report biological data to the Programme Centre. So far 397 data sets, consisting of invertebrates and water chemistry have been evaluated for the geographical extent and dose / response relationships. Trends can only be assessed for sites with the long-term data series.

Invertebrates respond to different compounds in the water. One main task of the ICP Waters Programme is to compare results from different regions in Europe and North America. It was therefore important to harmonize sampling and data handling when the program started in 1985. Since the geographical distribution of many species is restricted, universal indicator species or communities are usually not available. The strategy for monitoring biological effects was therefore to develope methods based on the influence of acidified water on the various species/communities. Many invertebrates are sensitive to acid water and become extinct at different levels of acidity. Presence or absence of such species indicates both current and recent past acidity (age/lifetime of certain organisms/species) . This knowledge has been utilized in a simple model, calculating an acidification index (ICP Waters Programme Mannual 1988 and 1996), which gives a number between 0 and 1, indicating strong to low/no acidification. The index is a universal number, comparable between regions and independent of the geographical distribution of invertebrates (see chapter 5.2).

Another way of assessing acidity based on biological material is the use of numerical relationships between species communities and pH or other characteristics of acidified waters. A relatively long tradition on analyzing diatom communities in relation to water quality exists (e.g., BATTARBEE et al. 1988, BIRKS et al. 1990). Similar methods have also been developed for invertebrates (LARSEN et al. 1996). When calibrated to local conditions, different invertebrate assemblages can be related to pH with a resolution of about 0.3 pH units.

Both methods mentioned above have advantages and disadvantages. For the presence/absence method the most important task is a representative sampling of animals at the site either by use of qualitative or quantitative methods. Taxonomists need to have good knowledge at least of the sensitive fauna. The method is most useful for sites with pronounced (seasonal) changes in water acidity leading to shifts between tolerance limits of important key organisms. Other parameters than identifying sensitive species have low or no influence on the evaluation, as long as they do not modify the tolerance limit. Ca and TOC can change the tolerance level (HÄMÄLÄINEN and HUTTUNEN 1990, LIEN et al. 1996), and should be taken into account when setting the limits for a specific locality.

For multi-variate analyses quantified samples of the total invertebrate fauna and thorough taxonomic work are necessary. These analyses are very important because they take into account all available environmental parameters and give information on the importance of certain parameters within a data set. They are, however, not only sensitive to the quality of the taxonomic work, but also to the quality of the sampling method used, season of sampling as well as the habitat at the site. It is time consuming to prepare the data sets for running multi-variate analyses which makes them more costly than evaluations using the acidification index.

Species can be absent for other reasons than acidified water. Especially during the earliest stages of acidification (sublethal water quality for sensitive species) the presence/absence approach is of low or none value. At these stages, multi-variate numerical techniques are the only useful method. In combination with training sets for a certain region these techniques can also be used for trend analyses.

This report includes an extensive summary of biological results obtained so far in the ICP Waters Programme. The presentation will mostly focus on findings published in international journals by LIEN et al. (1996), RADDUM and SKJELKVÅLE (1995), RADDUM and FJELLHEIM (1996), LARSEN et al. (1996), with addition of new analyses.

Results from studies on the presence/absence (acidification index) of sensitive species are the basis for evaluating the **geographical extent** of acidification. The presence/absence studies in combination with water chemistry data from the ICP Waters Programme are also the basis for assessing **dose/response** relationships. In this evaluation the multivariate analyses are of high importance for verifying the validity of indicator species as well as for detecting new indicators. The **trend analyses** are based both on presence/absence and on multi-variate techniques, where they can be applied.

## 5.2 Presence/Absense Studies and Acidification Index

The relationship between surface water chemistry and freshwater fauna has been evaluated in several ways. One approach is to describe empirical relationships between water chemistry parameters and the presence-absence of organisms (RADDUM et al. 1988, FJELLHEIM and RADDUM 1990). We used this method on data from a broad survey of lakes and rivers in southern Norway, for which water chemistry and information on invertebrate populations are available (LIEN et al 1996).

The absence of some species of invertebrates from lakes with low pH and high Al does not necessarily demonstrate that the populations had been lost over time or that a change to conditions of low pH and high Al caused their elimination. However, several lines of evidence taken together might indicate a causal relationship between changing water chemistry (towards lower pH and higher Al) and the loss of biota. From different countries studies on empirical relationships between fauna and water chemistry as well as laboratory bioassays indicate response to lowered pH and other chemical characteristics associated with acidification (RADDUM 1979, MATTHIAS 1983, ENGBLOM and LINGDELL 1983, HÄMÄLÄINEN and HUTTUNEN 1990. LIEN et al. 1996). All these studies show that methods based on presence/absence, or tolerance limits, are useful for mapping regional differences in acidity and are also suitable for monitoring purposes.

Based on presence/absence studies an acidification index has been developed to assess damage to invertebrates in freshwater. This index is based on knowledge of critical limits for different species (RADDUM and FJELLHEIM 1984, RADDUM et al. 1988, FJELLHEIM and RADDUM 1990). A score (index) is given for a certain invertebrate assemblage by using a range from 0 to 1. Index 1 means that one or more species with a low tolerance for acidified water can be found (tolerating a pH down to 5.5). These species are indicators of little potential damage to invertebrate populations. An index of 0.5 indicates that none of the most sensitive species is present, but the locality shows species that are moderately sensitive to acidification (tolerating a pH down to 5.0). Such communities should be considered as moderately damaged by acidification. An acidification index of 0.25 indicates that the site lacks all the sensitive species mentioned above, but contains species with a tolerance towards pH down to 4.7. For localities showing only organisms with high tolerance for acidic water (pH < 4.7), the acidification index is 0.

In several studies the index has been used as a tool for assessing the degree of acidification of surface waters. BOWMAN (1991) showed that the acidification status indicated by the macro-invertebrate communities in **Ireland** was in good agreement with that suggested by chemical water analyses.

In **Sweden** statistical analyses were performed to classify lakes by means of the acidification index and the TWINSPAN method (JOHNSON et al. 1993). Generally, the two methods were quite comparable, and the results were best when comparing samples taken on stony shores.

In **Finland** HÄMÄLÄINEN and HUTTUNEN (1990) evaluated the tolerance limit (TL), or presence/absence approach, by using the weighted averaging (WA) method in order to assess acidity in streams. They found that predictions based on both TL and WA were highly significant correlated to streamwater pH in autumn.

**Norwegian** and **German** material was used for multi-variate numerical analysis to quantify the relationship between invertebrate species and water chemistry (LARSEN et al. 1996). Detrendened canonical correspondence analysis (DCCA) showed that pH and aluminium were the most important factors to explain changes in fauna (see also chapter 5.6). The possibility to predict pH by looking at invertebrate assemblages was examined by means of weighted averaging (WA) regression and calibration, and weighted averaging partial-least-squares regression (WA-PLS). pH could be predicted with an accuracy of 0.3 pH units. The study also showed that the abundance of many species increases with pH, and that there is a very good agreement with the respective acidification indexes. The analysis came also up with new species which are probably sensitive to acidification, and which can be included into the list of sensitive species used in the presence/absence approach.

The conclusion from these international studies is that the acidification index is a suitable tool for both mapping acidification damages and monitoring purposes. The method is most appropriate for surface waters where acidification is evident. Very early stages of acidification, when the most sensitive species are still present, are difficult to detect by this method. Furthermore, the index is not a useful tool at strongly acidified localities, where changes may occur among the most tolerant species. In both cases multi-variate techniques are better approaches. The advantage of the presence/absence method is its simplicity allowing countries with differing monitoring practice to participate. However, all mentioned methods are available and in use within the ICP Waters program.

## 5.3 Geographical Extent of Acidification

In most cases the bedrock at the ICP Waters sites is highly to moderately sensitive to acidification (**Figure 43**). The buffer capacity is lowest in western Norway, where mean yearly calcium (Ca) concentrations vary from 7.5 to 25.0 $\mu$eq $l^{-1}$. In the United Kingdom and Ireland Ca concentrations from 12.5 to 130.0 $\mu$eq $l^{-1}$ can be observed, while they range from 10.0 to 430.0 $\mu$eq $l^{-1}$ in central Europe. The range of mean yearly pH at sites in Norway, United Kingdom and Ireland is 4.6 -7.2, in

Sweden 4.4 - 6.9 and in central Europe 4.0 - 7.4. The sensibility to acidification at the ICP Waters sites covers a wide range (from very low to very high).
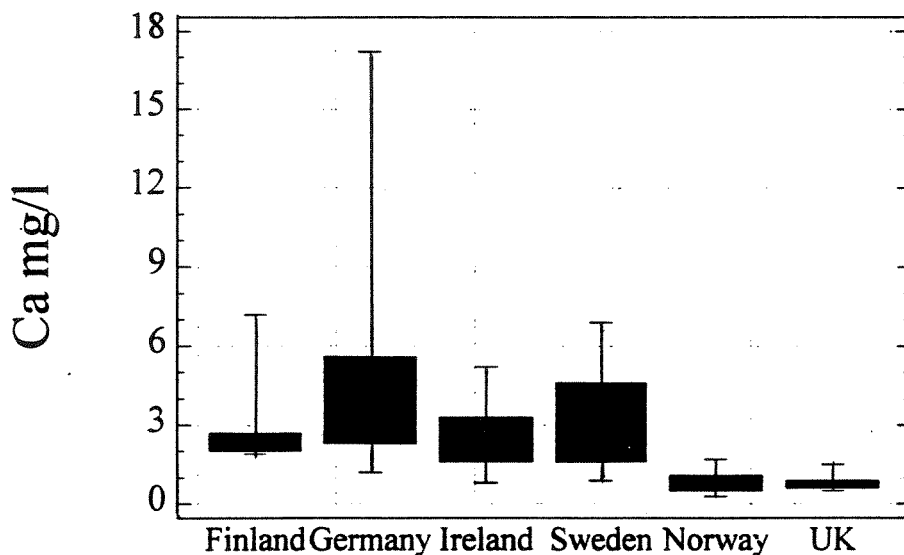


**Figure 43.** Mean annual values for Ca for the period 1988 - 1992 for participating countries in ICP Waters .

The acidification indexes for different UN ECE countries is shown in Figures 2 - 4. We have divided the biological material into two parts, i. e., samples collected before and after 1990. The aim is to investigate the extent of acidification and possible regional changes for the two time periods on a broad scale. Choosing 1990 allows to divide the material from most of the countries in fairly even parts.

The **Canadian** data sets, sampled in 1987 and 1990/91, are from the experimental lake area (ELA). All sensitive invertebrate species are present, indicating low acidification in that region (**Figure 44**). In 1987 some of the most sensitive species were missing, while the 1990/91 samples did not show any signs of acidification.

Both before and after 1990 acidification was also low at the monitored **Irish** watersheds. A few sites were clearly acidified, but the mean values indicate low acidity. The catchments of the acidified sites have been extensively reforested.
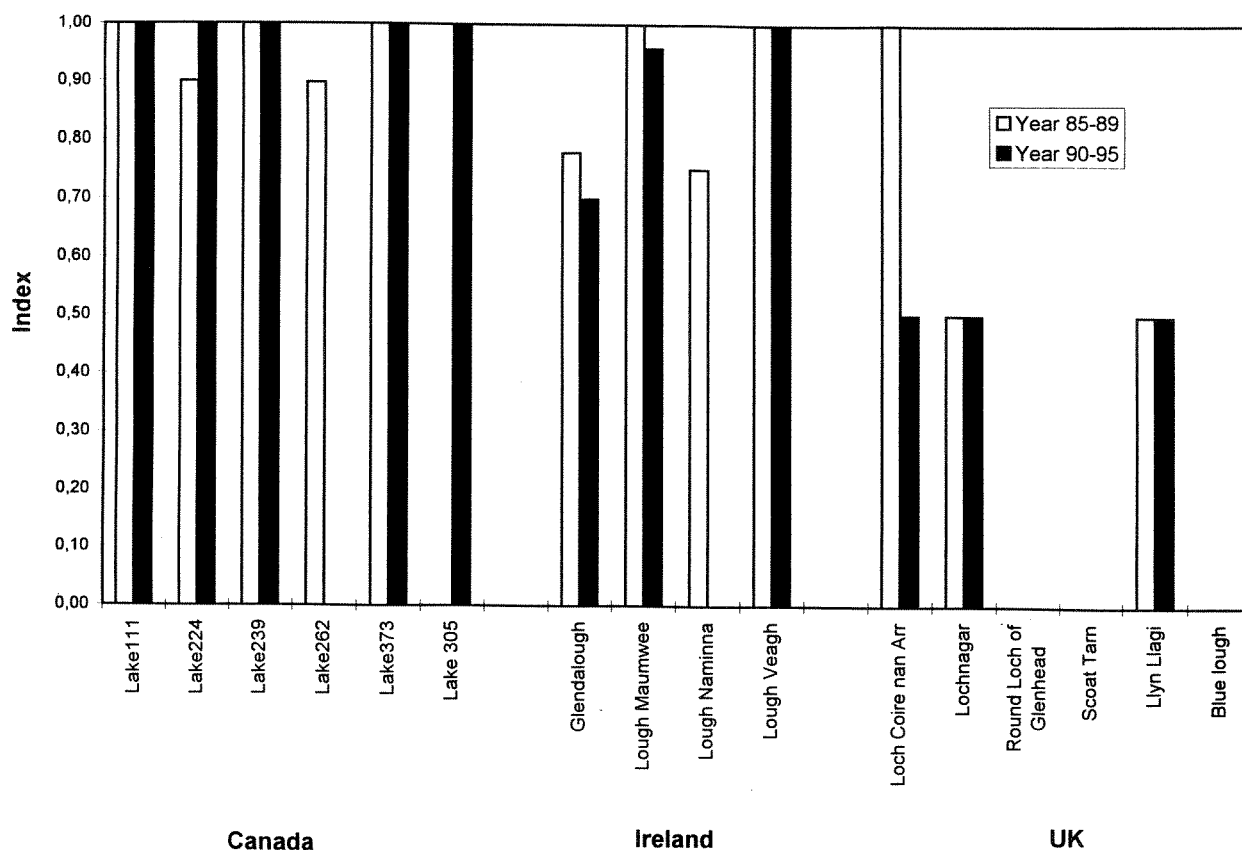
**Figure 44.** Mean acidification index for sites in Canada, Ireland and UK befor and after 1990.

For the **United Kingdom** material of 1988 - 1995 is available, and no changes in acidification index can be found, except for Loch Coire nan Arr. In this lake several of the most sensitive species were abundant in 1989, while none of them were found in 1994, and the index dropped from 1 to 0.5. The other lakes had an index of either 0.5 or 0 in both years, and are thus not damaged by acidification. The lakes in the UK represent strongly to slightly acidic sites.

In **Austria**, Stingelbach is the only watershed where sensitive species had probably some minor problems in 1989, while no such difficulties seemed to occur in 1990 (**Figure 45**).

For **Belgium** biological data of 1988 and 1990 are available, showing a mean acidification index of 0.42 and 0.40, respectively. The investigated watersheds were rather acidic both in 1981 and 1990.
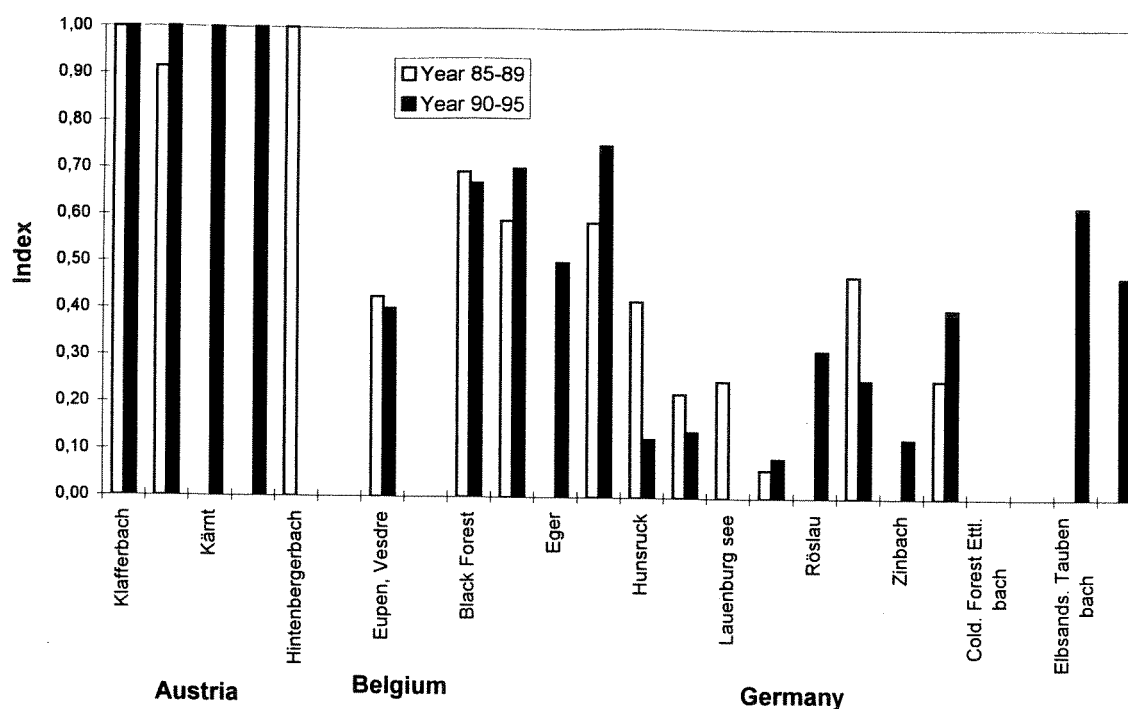
**Figure 45.** Mean acidification index for sites in Austria, Belgium and Gemany before and after 1990.

The monitoring in **Germany** covers a high number of watersheds, and data series are of different lengths. The longest ones (from 1984 onwards) are from Black Forest, East Bavaria and Hunsrück. At Harz monitoring started in 1986 and in Kaufunger Wald and Odenwald in 1988. The latest data included are from 1994. The number of sites in each watershed vary. In general, the different regions in Germany are moderately to highly acidified (see some examples in **Figure 45**). The situation before and after 1990 has changed for some watersheds. A higher mean index after 1990 was estimated for East Bavaria, Harz and Taunus. In Odenwald the acidification index is close to zero, strongly acidified, and the small improvement (1990-94) is due to only one sample and is thus not very convincing. In the Black Forest the situation has not changed. Decreasing acidification indexes after 1990 can be observed for Hunsrück, Kaufunger Wald and Rothaargebirge. High to moderate acidification is also typical for sites included into the ICP Waters Programme after 1990. For watersheds with samples taken before and after 1990 the mean index was 0.41 and 0.39, respectively, indicating no change for the whole area of Germany.

In **Norway** the biological monitoring shows highly to slightly acidified watersheds (**Figure 46**). The division of indexes into before and after 1990 indicates improvements in the Farsund area. A small increase in mean acidification index can also be observed for Nausta. The other monitored sites show similar results for the periods before and after 1990. A more detailed analysis of trends in some of these watersheds will be discussed in chapter 5.4.
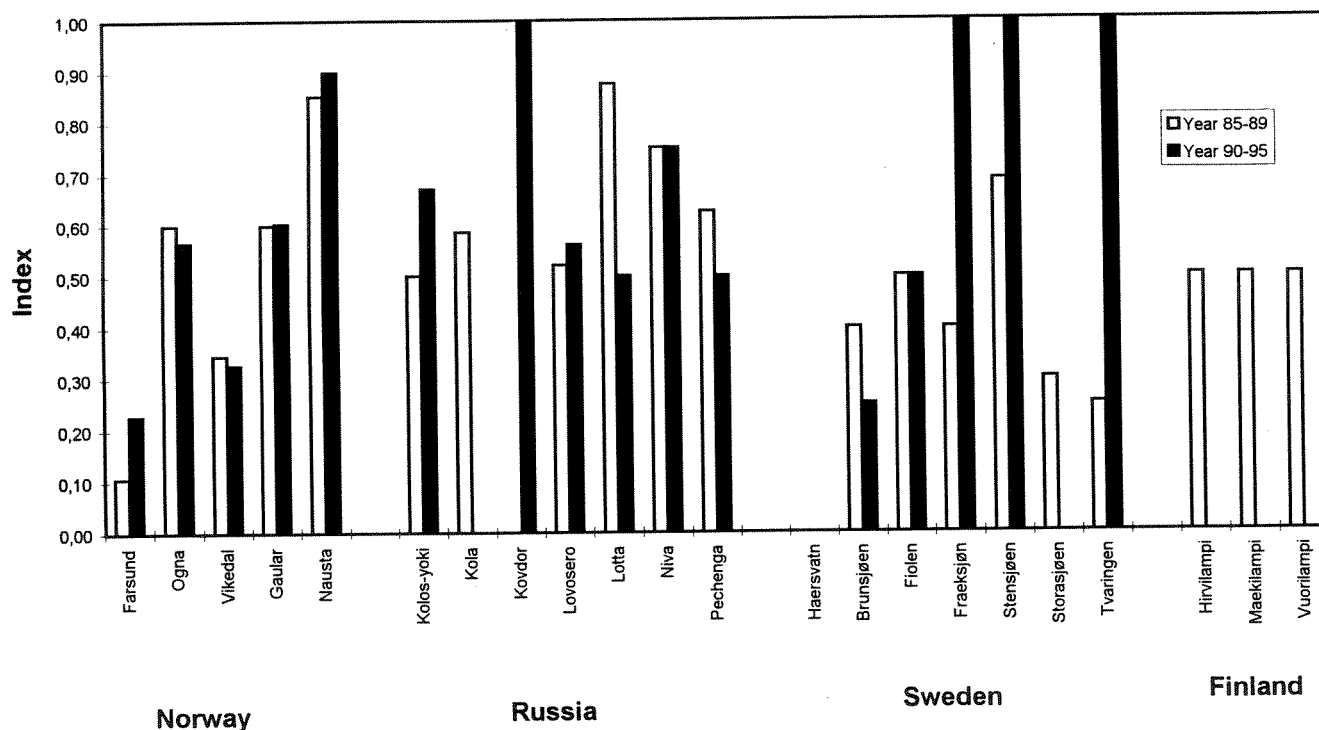


**Figure 46.**    Mean acidification index for sites in Norway, Russia, Sweden and Finland before and after 1990.

For western **Russia** acidification indexes of 238 different sites have been reported. However, the raw material from the single locations is not available. The indexes indicate low acidification in most regions of Russia. An index of 1 was estimated for 87% of the sites, while indexes 0.0, 0.25 and 0.5 constituted 3%, 3% and 7%, respectively. In subregions Kola and Karelia 6% or 2% of the sites got index 0.0. However, material from the most north west of Russia was delivered recently (for 1988, 1989, 1990) and thus could also be analyzed (**Figure 46**). In general, these watersheds are little to moderately acidified. In the river systems Lotta and Pechenga the data indicate increasing acidity

from 1989 to 1990. Since this is based on two consecutive years, yearly variations should be taken into account.

From **Sweden** biological material from seven lakes is available, ranging from highly to little acidification. In **Figure 46** the mean acidification index is shown for observations before and after 1990. The improvement indicated for Fræksjøn, Stensjøn and Tvaeringen during the last period is of low significance, since data only from one time sampling is included in the Figure.

The **Finish** material is from lakes only and sampled before 1990. With an index of 0.5, all lakes were clearly acidic (**Figure 46**).

## 5.4 Dose/response Relationships

The mean values of chemical parameters are of low importance for evaluating the effects of acidified water on invertebrates. Invertebrates are usually affected during episodes with high concentrations of toxic agents. Therefore, water chemistry data from episodes were chosen for those sites, where variation in water chemistry was considerable (most extreme data were excluded). For investigating the dose/response relationships we used these peak concentrations and related them to the acidification index.

The data were harmonized according to methods described in the ICP Waters Programme Manual (1987 and 1996). For further harmonization and data quality control, intercalibrations were performed both for water chemistry (chapter 2.2) and biology.

The highest and lowest number of species/taxa were recorded in Germany and Ireland with 223 and 104 taxa, respectively. For Norway and Sweden 161 and 219 taxa were noted. Sites with an index value of 1 were most frequent in Ireland and least frequent in Sweden and Germany (**Figure 47**). In Norway, the proportion of sites with an index of 1 and 0 were about equal, while sites with an index of 0.25 and 0.5 were relatively few.

The mean pH and ANC at the different acidification indexes in Norway and Germany are shown in **Figure 48** and **Figure 49** (from LIEN et al. 1996 and RADDUM and SKJELKVÅLE 1995). In Norway clear water sites with acidification indexes of 1.0 and 0.0 had a mean pH of 5.8 and 4.9, while ANC was 21 and -21 µeq l$^{-1}$, respectively. In Germany sites with an index of 1 had a mean pH of 6.8 and an ANC of 150 µeq l$^{-1}$, while the corresponding values at index 0 were 4.65 (pH) and -32

µeq l[-1] (ANC). This shows that the ranges of both pH and ANC are much higher in Germany than in Norway.
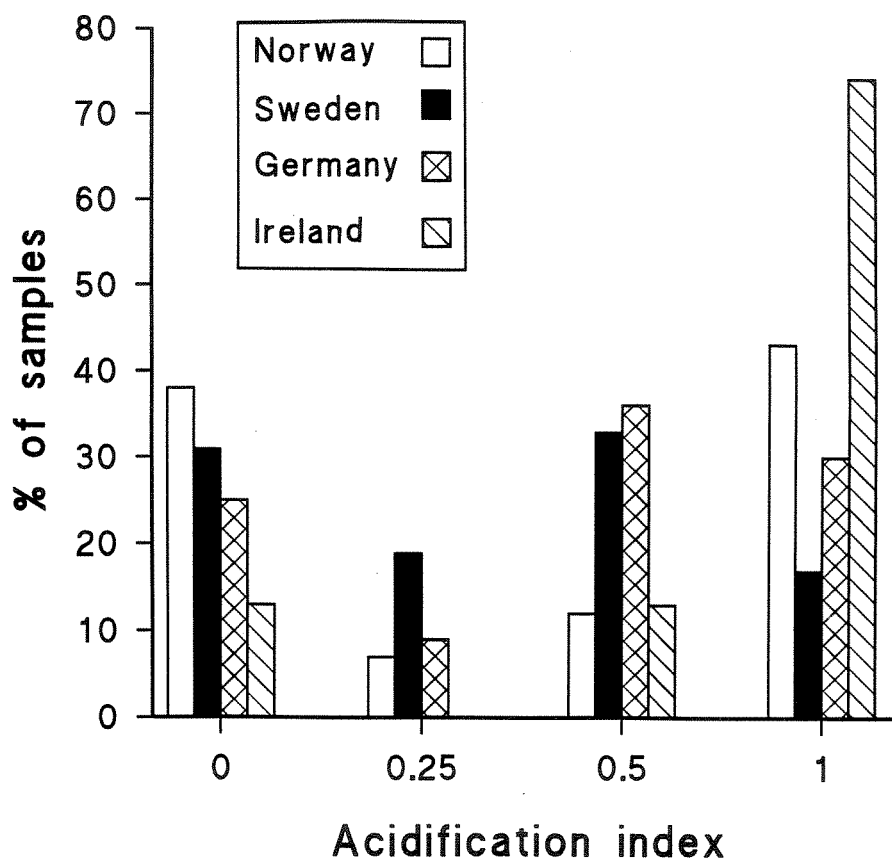


**Figure 47.** Distribution of samples at different acidification indexes.

**Table 30.** Mean number of species recorded at different acidification indexes.

| Country/index | 0 | 0.25 | 0.5 | 1.0 |
|---|---|---|---|---|
| Norway | 6.43 +/- 2.85 | 9.0 +/- 2.06 | 8.31 +/- 3.05 | 9.33 +/-2,77 |
| | n = 53 | n = 9 | n = 16 | n = 60 |
| Sweden | 10.73+/-6.34 | 14.86+/-5.15 | 19.67+/-6.43 | 26.83+/-9.56 |
| | n = 11 | n = 7 | n = 12 | n = 6 |
| Irland | 12.4+/-5.4 | | 16.5+/-4.7 | 17.3+/-6.1 |
| | n = 6 | | n = 6 | n = 32 |
| Germany | 4.83+/-2,57 | 8.16+/-2.11 | 14.47+/-6.48 | 25.08+/-16.74 |
| | n = 53 | n = 18 | n = 74 | n = 63 |

The number of species at the different acidification levels were different in Norway, Sweden, Ireland and Germany (**Table 30**). At an index of 1 the number was lowest in Norway and highest in Germany and Sweden. At an index of 0 the sites in Germany had the lowest occurrence of species. The relative decrease of species from index 1 to 0 was 28, 31, 60 and 81% for Ireland, Norway Sweden and Germany, respectively (RADDUM and SKJELKVÅLE 1995).
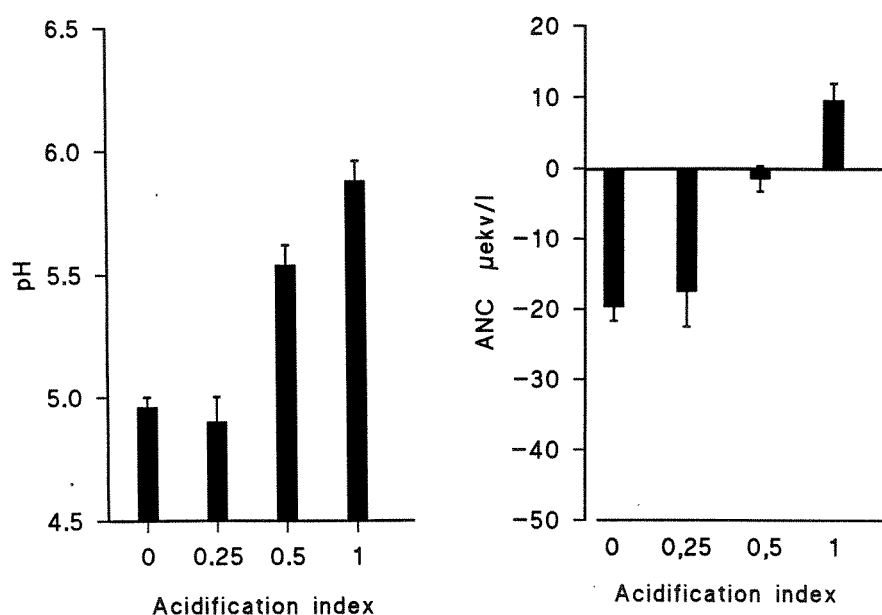


**Figure 48.** Acidification index against mean pH (left) and ANC (right) in Norway. Standard deviation is indicated.

The results of the ICP Waters monitoring underline that the critical limits of chemical compounds set for invertebrates vary in accordance with the original water chemistry and the fauna's adaptation to their surroundings. In Norway and Ireland pH drops very seldom below 4.5 (acidification index 0) while in central Europe pH values of less than 4 may occur (minimum values as low as pH 3.2; SKJELKVÅLE et al. 1994). The critical pH for the acidification index of 0 is 4.7. The exceedance of this limit is therefore much higher at the most acidic sites in Germany compared to those in Norway. This might be one important factor explaining the strong decrease in species number in the most acidified regions in Germany. Many German surface waters not affected by acidic atmospheric deposition have a high pH and high calcium levels. Species such as snails and mussels, having critical limits with respect to pH and Ca, are living at those sites (ØKLAND 1986). Consequently, effects of acidification will be relatively higher in these areas than in clearwaters found, e.g., in Norway. Species living at clearwater sites are under immense selection pressure for tolerance of oligotrophic

water. Euryecious species are favoured, and the difference in diversity between the acidification levels is reduced (see data of Ireland and Norway). This kind of selection pressure is typical for surface waters with low pH and conductivity is lower in central Europe. This may be one additional moment for explaining the strong reduction in number of index 0 species at German sites, compared with the situation in Norway.
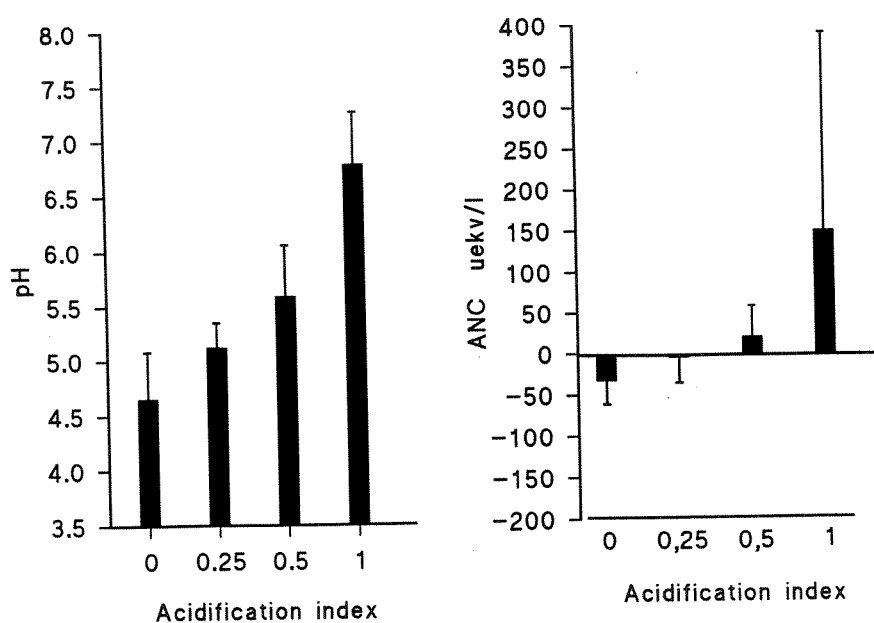
**Figure 49.** Acidification index against mean pH (left) and ANC (right) in Germany. Standard deviation is indicated.

Obviously, adaptations to different water qualities influence the acid tolerance of species in different regions. This observation is supported by the fact that clearwater sites with an index of 1 in Norway had a mean pH of 5.8 (ANC = 10 µeq $l^{-1}$) while the mean pH for sites in Germany with an index of 1 was 6.8 (ANC = 150 µeq $l^{-1}$; Lien et al. 1996). To protect fish and invertebrates in acidified surface waters with low Ca concentrations of 7.5 - 50 µeq $l^{-1}$, LIEN et al. (1996) proposed an ANC $\leq$ 20 µeq $l^{-1}$. This limit is, however, much lower than the mean ANC for sites with an index of 1 in Germany. Due to the high variation in water chemistry in different regions of Germany it is difficult to assess an ANC limit for central Europe. Nevertheless, the most sensitive fauna was very seldom found in waters with pH $\leq$ 6 (RADDUM and SKJELKVÅLE 1995). We therefore propose pH 6 as a critical limit for protection of the most sensitive fauna. Based on this pH level an ANC limit was determined by using a correlation analysis including the German pH and ANC data (SKJELKVÅLE et al. 1994). The analysis showed that pH 6 corresponds with an ANC of $\approx$ 50 µeq $l^{-1}$ (**Figure 50**). In acidified regions

of central Europe a critical ANC limit of $\leq 50$ µeq $l^{-1}$ is therefore proposed to protect the most sensitive organisms. We also feel that the ANC limit in other areas with originally high pH and high calcium concentrations in surface waters should be 50 µeq $l^{-1}$ rather than 20 µeq $l^{-1}$. One example is sites in southern Sweden.



**ANC vs. pH on German Stations**

$y = 62.5x - 325.1$

$r^2 = 0.37$
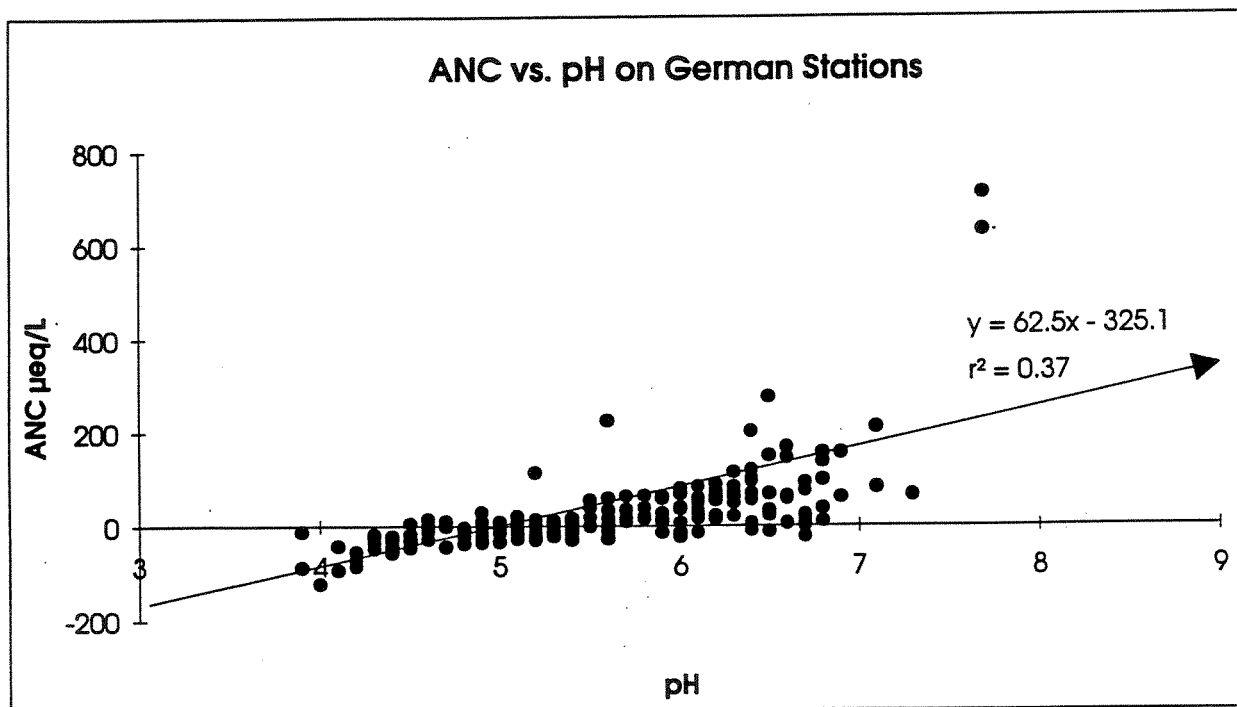
**Figure 50.** Correlation between pH and ANC in the German data set of sites with biological data.

## 5.5 Long-term Trends in Acidification Index

To detect long-term trends within the invertebrate fauna composition the acidification index was calculated for ICP Waters sites with long and regular monitoring series. For some of the river systems in Norway monitoring dates back to 1981. The data show that the spring samples generally indicate the highest acidification (low index) due to release of acids during snow smelt. During fall the acidity is normally lower, indicated by a higher index. Due to the difference between the seasons the trend analysis is performed separately on spring and fall data.

Generally, low index values were observed at the start of monitoring. The indexes increased until 1988, afterwards they decreased until 1990. Since then the idexes have risen again. Therefore, RADDUM and FJELLHEIM (1995) chose two periods for correlation analyses between time and

index: 1981 - 1988 and 1989 - 1994. Data from Norwegian watersheds were taken (**Figure 51, Figure 52** and **Table 31**). At Farsund, the spring values show a slightly negative development (r = -0.58) in acidification index during 1981 - 1988, while no change was found for the second period. At River Vikedal the change in acidification index was positive (r = 0.57) during spring in the first period, while it was similar to that at Farsund from 1989 - 1994. At Nausta a significantly positive trend was found for the first period during spring (r = 0.84, p = 0.03). For the second period acidification indexes increased slightly.

**Table 31.** Correlation analysis of acidification index and time for spring and fall during two periods. For trend directions, see Figure 5.9 and Figure 5.10.

| Watershed | Period 1981 - 1988 | | | | Period 1989 - 1994 | | | |
| | Spring | | Fall | | Spring | | Fall | |
| | r | p | r | p | r | p | r | p |
|---|---|---|---|---|---|---|---|---|
| Farsund | 0.58 | 0.16 | 0.17 | 0.71 | 0.06 | 0.89 | 0.76 | 0.04 |
| Vikedal | 0.57 | 0.18 | 0.82 | 0.02 | 0.28 | 0.58 | 0.58 | 0.23 |
| Nausta | 0.84 | 0.03 | 0.15 | 0.77 | 0.65 | 0.16 | 0.84 | 0.03 |

During fall there was no trend at Farsund in the first period (until 1990), but for 1989 - 1994 a significantly positive trend was observed (r = 0.76, p = 0.04). By including the 1995 data this positive trend became stronger (r = 0.907, p = 0.004). At River Vikedal a significant trend existed for the first period (r = 0.82, p = 0.02), while there was only a slight increase during the second period. At River Nausta there was no trend during fall from 1981 - 1988, while a significant positive trend was found in the second period (r = 0.84, p = 0.03). Addition of the 1995 data did not change the correlation since the acidification index was 1 both in 1994 and 1995 and cannot increase further.

Correlation analyses for the whole period (1981 - 1994) show a positive, but not significant, change of the acidification indexes for Farsund, Vikedal and Nausta either for spring and or fall (correlation factors in the range of 0.5 to 0.74).
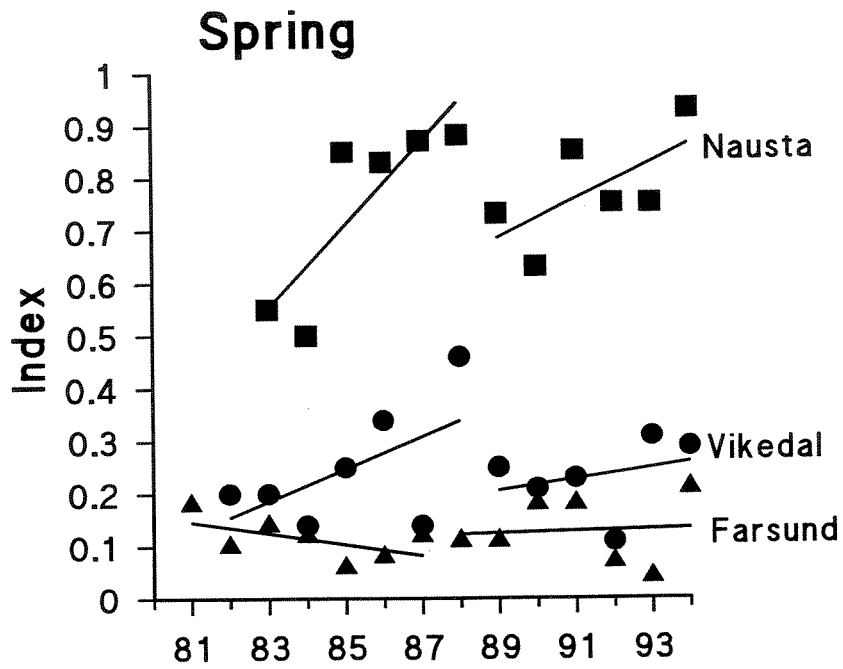
**Figure 51.** The acidification index for Nausta, Vikedal and Farsund during spring.
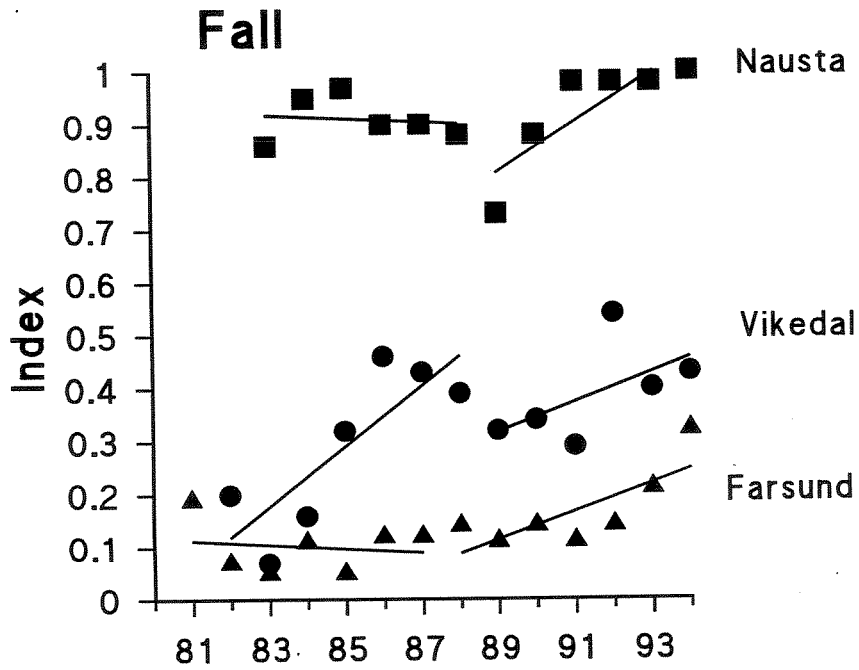


**Figure 52.** The acidification index for Nausta, Vikedal and Farsund during fall.

## 5.6 Trend Analysis by Use of Multi-variate Techniques

Because $SO_2$ (and heavy metal) emissions in Europe and North America have significantly decreased since the 1980s, attention now focuses on the recovery of flora and fauna in acidified lakes and rivers. Diatoms are good ecological indicators of lake water pH. For two Scottish lochs diatom studies give strong evidence that acidification is reversible (BATTARBEE et al.. 1988). Studies by LARSEN et al. (1996) show that invertebrates are as good predictors of modern pH as diatoms. We therefore wish to look for possible recovery trends in invertebrate data sets collected within the ICP Waters Programme. Since the acidification index shows improvements for Farsund and Nausta (Norway; chapter 5.5) we chose those sites for multi-variate statistical analysis.

The data we used consist of two sets:

1.  Invertebrates sampled from the Farsund river system in south-western Norway in the framework of the ICP Waters monitoring program. The material was sampled at seven stations in spring and in autumn, from 1981 to 1995 (exceptions: spring and autumn 1982; 1983).

2.  A data set consisting of 180 samples of invertebrates and water chemistry data of 21 unlimed lake-river systems in western Norway. The animals were sampled from 1988 to 1990 simultaneously with the water samples that were analyzed for pH, calcium, alkalinity (ALK), total aluminium and conductivity. The training set was established by LARSEN et al. (1996) to quantify the relationship between invertebrates and water chemistry by using canonical correspondence analysis (CCA) and detrended canonical correspondence analysis (DCCA). For information about this technique see TER BRAAK and VERDONSCHOT (1995).

To look for possible recovery trends in the Farsund invertebrate data set the same DCCA analysis was run as for the training set (LARSEN et al. 1996), but with the Farsund invertebrate abundance data included as passive samples in the analysis. This means that the Farsund invertebrate samples are positioned on the basis of their fauna composition and similarities with the training set biology into the DCCA space of the training set (see BIRKS et al. 1990). This gives the posibility to explore whether any trend in the invertebrate assemblage, through fourteen years of monitoring, reflect any changes of pH in the water.
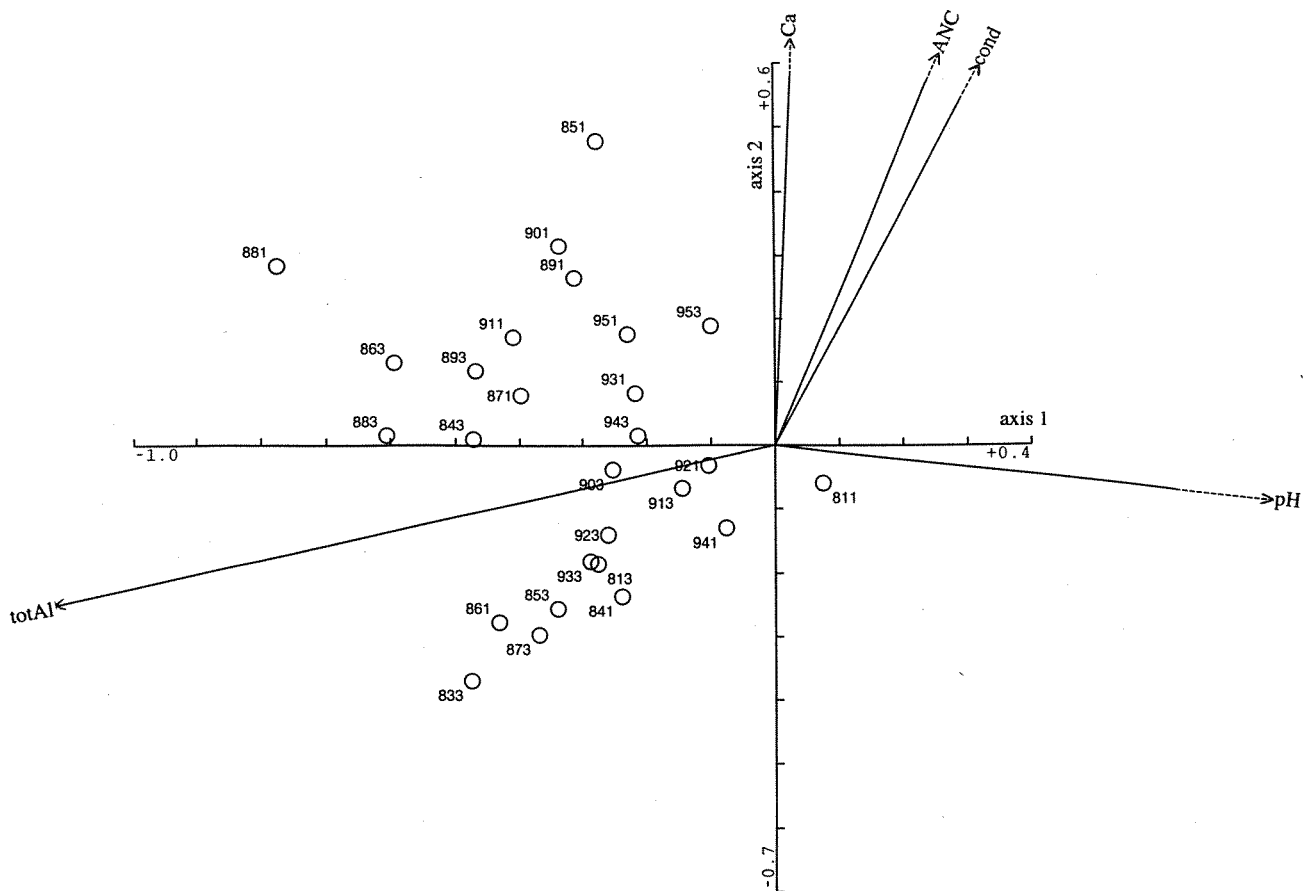
**Figure 53.**   Biplot with the Farsund samples included as passive samples in the DCCA analysis of the training set. Only the Farsund localities shown. See text for more information.

**Figure 53** shows the DCCA analysis results for the fauna of the Farsund river system placed in a system of coordinates in relation to the chemistry data in the training set. The arrows indicate in what direction the environmental variables increase. pH rises to the right along axis 1, and decreases to the left (the arrow can be elongated to the left along axis 1). The origin represents the average pH. The invertebrate samples in **Figure 53** are described by three numbers. The first two indicate the year the invertebrate assemblage was sampled, and the third number distinguishes between spring (= 1) or fall (= 3). The biological samples can be projected in 90° angles to the water chemistry arrows. Samples projected more on the right side of the pH arrow represent a higher pH than samples located more on

the left side. The figure makes it possible to see how the invertebrate fauna reflects different pH values and aluminium concentrations during the 14 years of sampling.

DCCA axis 2 (y-axis) shows a statistically significant correlation with calcium. Seven taxa are characterizing the calcium rich part of the gradient, but their occurence is low in the training set. The samples from Farsund (passive samples) are therefore not related to axis 2 (calcium gradient).

Based on **Figure 53**, **Figure 54** and **Figure 55** show the "projected" pH for spring and fall of the different years. It seems as if the sample of spring 1981 is related to a higher pH value than the rest of the Farsund data. The following years the invertebrate samples are related to more acid conditions, accompanied by higher aluminium concentrations. The highest $H^+$ concentrations is indicated for spring 1988. From 1989 onwards pH values have increased and aluminum concentrations decreased. The conditions seem to have improved considerably from 1991 to 1995, showing the highest pH values of the whole sampling period (exception: first sample in 1981).
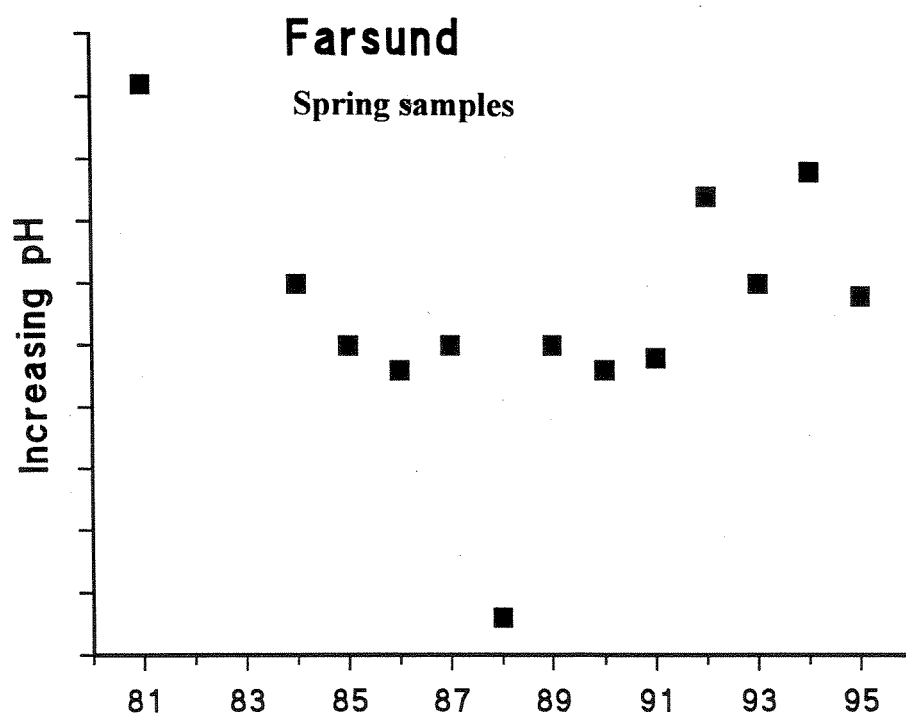


**Figure 54.** Estimated relative pH values, based on the invertebrate community for the different years during spring. Highest and lowest pH cover a pH range of 0.4 units.
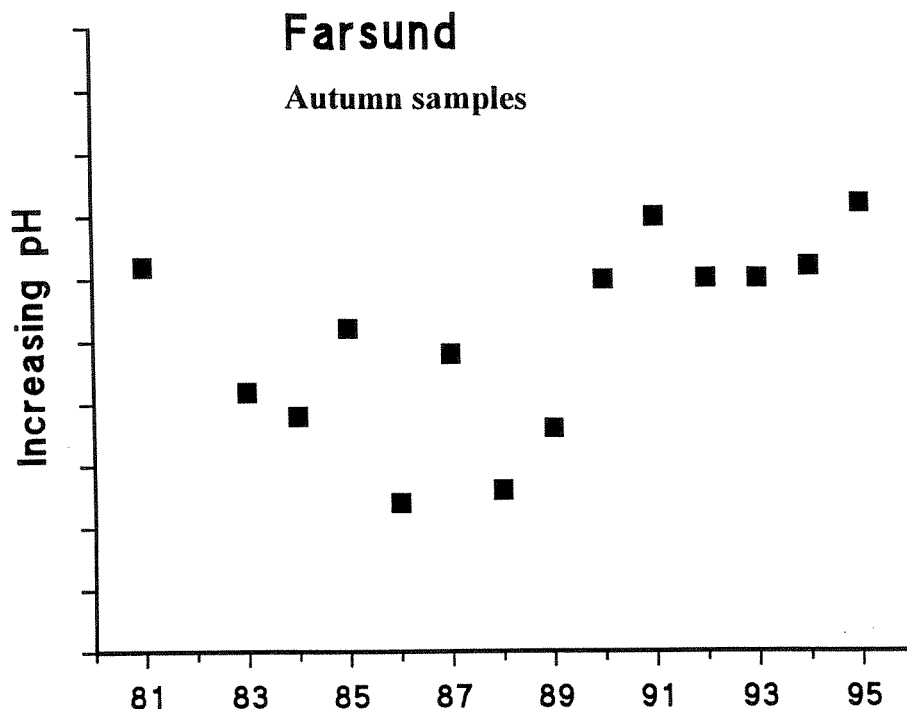
**Figure 55.** Estimated relative pH values, based on the invertebrate community for the different years during autumn. Highest and lowest pH cover a pH range of 0.25 units.

The same type of analysis described above was applied to the Nausta data. In comparison to Farsund, water acidification at Nausta is low. First, a simple correspondence analysis (CA) was performed using the whole data set with no chemical parameters included (**Figure 56**). This analysis separated clearly the fall and spring samples, underlining the importance of sampling dates. Annual seasonality has to be considered when running trend analyses.

Compared with the training set the data from the station in Nausta with the best water quality are scattered on the right side of the y-axis, indicating high pH values (**Figure 57**). The data at the more acidic site, station Trodøla, are located inbetween those of the best station and the training set data. at this stage it is not clear if this technique shows any significant change in acidification of the watershed.

In the future, the DCCA technique will be applied to other ICP Waters data sets. This is possible as soon as training sets for the different regions are available. In this connection we performed multi-variate analysis using the invertebrate material and water chemistry data from Germany as an attempt to establish a training set for central Europe. In the German data set pH shows a high correlation ($r = 0.787$) with DCCA axis 1, followed by conductivity ($r = 0.709$), calcium ($r = 0.703$) and total

aluminium (r = -0.564). Compared to the Norwegian data set the German one shows a large range in conductivity values and calcium concentrations, which has an influence on the invertebrate communities. This phenomenon leads to a second gradient that is much more important in the German set than in the Norwegian one. It becomes more complicated to assess dose/responds relationships and may be one reason for the lower importance of aluminium in the German data compared to the Norwegian ones.
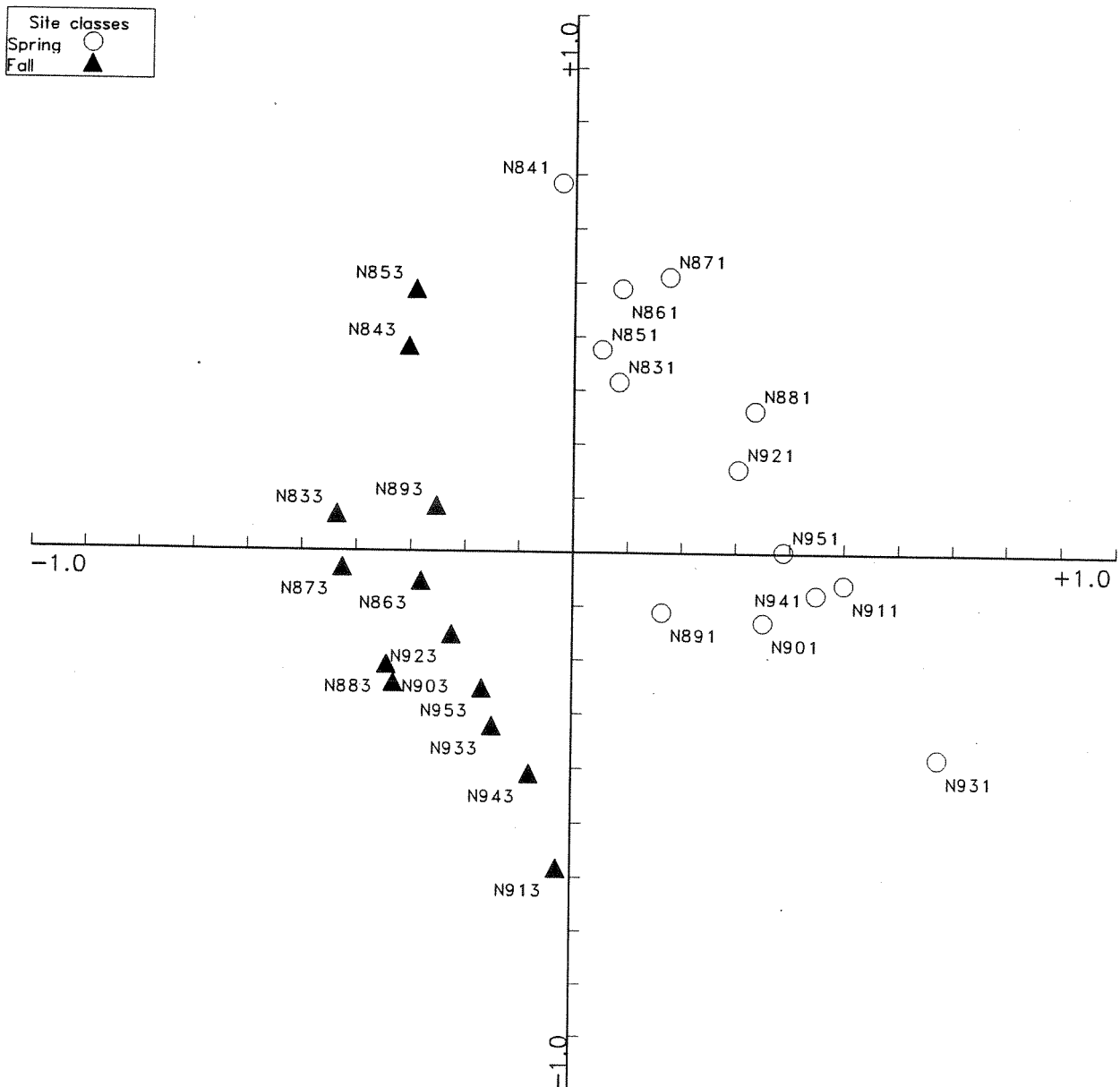


**Figure 56.**    CA, correspondence analysis applied to the total data set from the Nausta watershed without any water chemistry variabels included.
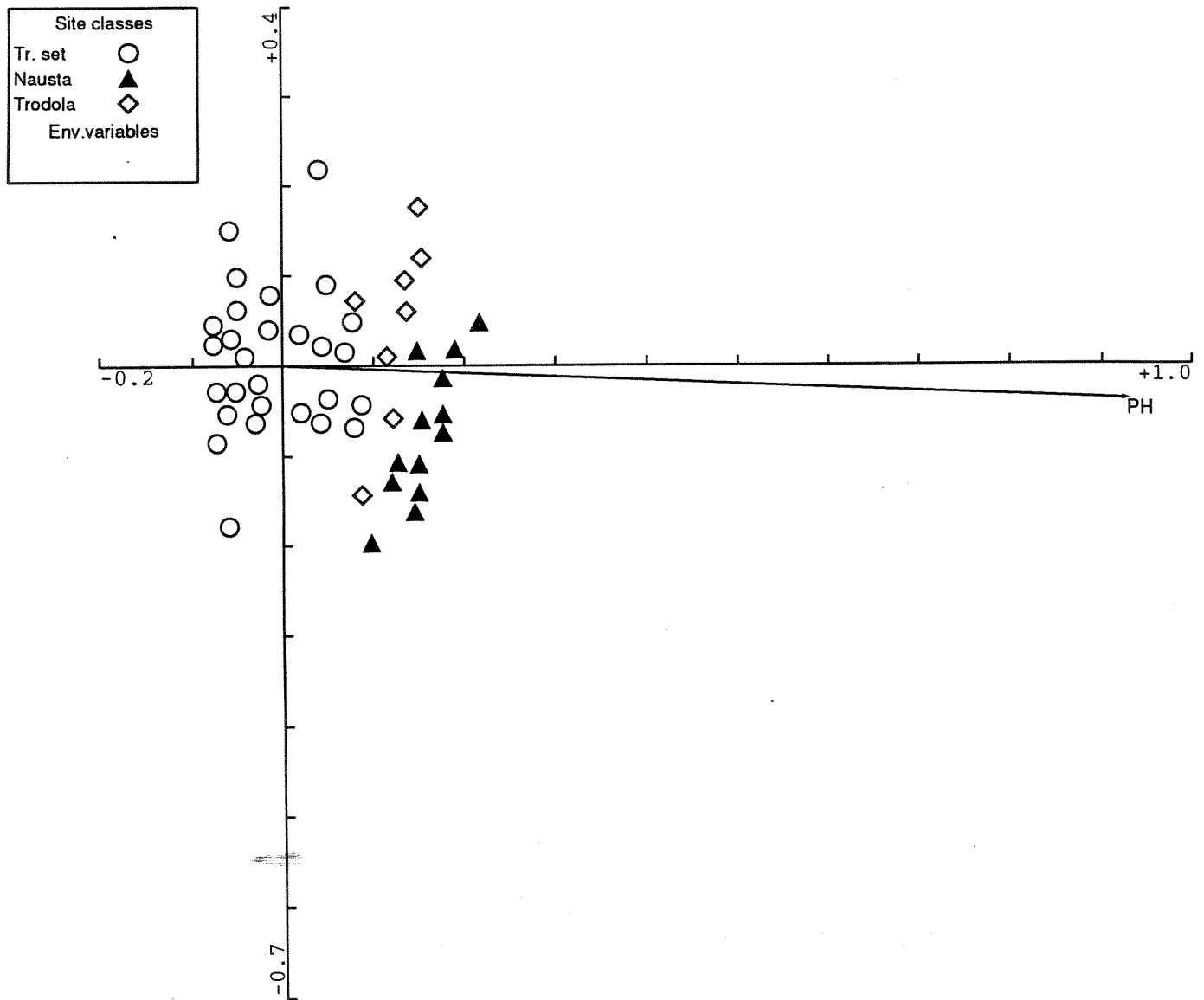
**Figure 57.** Biplot with the Nausta and Trodøla localities as pssive samples in the DCCA analysis of the training set. Of the environmental variables only pH is shown.

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# 8. Further Work

At its 12th meeting, held from 23 to 25 October 1996 in Silkeborg, Denmark, the ICP Waters Programme Task Force agreed on the following work-plan for the 1996 - 1997 period (from the minutes):

o    Intercalibration 1997;

o    Participitation in preparing the WGE Handbook;

o    Continuing the preparation of a workshop on biological assessment and monitoring (including its financing) in cooperation with ICP Integrated Monitoring (main topic of the workshop should be the assesment of various techniques for evaluating aquatic biota);

o    Preparation of the 1997 technical report on regional distribution, representiveness of sites in the network in relation to mapping of critical loads;

o    Further consideration of problems related to regional lake and river data (e.g. by an ad hoc group of experts, to be set up at the 13th meeting of the Task Force in 1997, and in cooperation with the National Focal Points);

o    Cooperation with the European Commission (EC) research programmes; consideration of possible application for a grant from the fifth framework programme;

o    Participitation in the preparation of the 1997 joint report on the effect-oriented activities;

o    Preparation for the presentation of ICP Waters on Internet;

o    Input to the workshop on heavy metals and persistent organic pollutants (Germany, November 1997).

# Annex A.

## Chemical and Biological Site Data for ICP Waters

**Table 32.** Sites in **bold** have been included in the new data base.

| Station name | File | Chemistry code | Year | Site data | Biology code | Biology year |
|---|---|---|---|---|---|---|
| **Austria, Kärnten, Dösener See** | **IA-AU** | **DOS-K** | **1989-1995** | x | | |
| Austria, Kärnten, Gradenbach | IA-AU | GRA-K | 1989-1991 | x | 2-1 | 1990-1991 |
| Austria, Käernten, Melniksee | IA-AU | MEL-K | 1988-1995 | x | | |
| Austria, Käernten, Wangenitzbach | IA-AU | WAN-K | 1990-1991 | x | 2-14 | 1990-1991 |
| Austria, Tirol, Gossenkölle See | IA-AU | GKS-T | 1988-1992 | x | | |
| Austria, Tirol, Hairlacher See | IA-AU | HAI-T | 1988-1992 | x | | |
| **Austria, Tirol, Mutterberger See** | **IA-AU** | **MUT-T** | **1988-1995** | x | | |
| **Austria, Tirol, Schwarzsee ob Sölden** | **IA-AU** | **SOS-T** | **1989-1995** | x | | |
| Austria, Upper Austria, Klafferbach, Rehberg | IA-AU | KLA1-0 | 1989-1991 | x | 15 1-5 | 1989-1991 |
| Austria, Upper Austria, Stinglbach vor Klafferbach | IA-AU | KLA2-0 | 1989-1991 | x | 16 1-3 | 1989-1991 |
| Austria, Upper Austria, Klafferbach, Brucke, Freundorf | IA-AU | KLA3-0 | 1989-1991 | x | 14 1-3 | 1989-1991 |
| Austria, Upper Austria, Ramenbach, Brucke Hinterberg | IA-AU | KLA4-0 | 1989-1991 | x | | 1990-1991 |
| | | | | | | |
| Belgium, Eupen, Getz river, Small dam | IA-BE | B03 | 1987-1991 | | 1-2 | 1988-1990 |
| Belgium, Eupen, Vesdre river, Bellesfort | IA-BE | B02 | 1987-1991 | | 1-1 | 1988-1990 |
| Belgium, Eupen, Vesdre, Dam | IA-BE | B01 | 1987-1991 | | | |
| Belgium, Jalhay, Gileppe river, Chemin des charbonniers | IA-BE | B06 | 1987-1991 | | 1-4 | 1988-1990 |
| Belgium, Jalhay, Helle river, Schornstein | IA-BE | B04 | 1987-1991 | | 1-3 | 1988-1990 |
| Belgium, Jalhay, Lake Gileppe | IA-BE | B05 | 1987-1991 | | 1-4 | 1990 |
| Belgium, Jalhay, Louba river, Les Hes | IA-BE | B07 | 1987-1991 | | 1-5 | 1988-1990 |
| Belgium, Jalhay, Soor river, Small dam | IA-BE | B08 | 1987-1991 | | 1-6 | 1988-1990 |
| | | | | | | |
| **Canada, Nova Scotia, Beaverskin Lake** | **IA-CA** | **C14** | **1970-1994** | x | | |
| **Canada, Nova Scotia, Kejimkujik Lake** | **IA-CA** | **C13** | **1970-1994** | x | | |
| Canada, Nova Scotia, Little Red Lake | IA-CA | C12 | 1979-1992 | x | | |
| **Canada, Nova Scotia, Mount Tom Lake** | **IA-CA** | **C10** | **1980-1994** | x | | |
| **Canada, Nova Scotia, Mountain Lake** | **IA-CA** | **C11** | **1970-1994** | x | | |
| **Canada, Ontario, Algoma Region. Batchawana Lake** | **IA-CA** | **C01** | **1980-1994** | x | | |
| **Canada, Ontario, Algoma Region, Little Turkey Lake** | **IA-CA** | **C03** | **1980-1994** | x | | |
| **Canada, Ontario, Algoma Region, Turkey Lake** | **IA-CA** | **C04** | **1980-1994** | x | | |
| **Canada, Ontario, Algoma Region, Wishart Lake** | **IA-CA** | **C02** | **1980-1994** | x | | |
| Canada, Ontario, Lake 111 | IA-CA | CA111 | 1987-1990 | x | 1 1-5 | 1988-1990 |
| Canada, Ontario, Lake 224 | IA-CA | CA224 | 1987-1990 | x | 2 1-5 | 1987-1991 |
| Canada, Ontario, Lake 239 | IA-CA | CA239 | 1987-1990 | x | 3 1-5 | 1987-1991 |
| Canada, Ontario, Lake 262 | IA-CA | CA262 | 1987-1990 | x | 4 1-5 | 1990 |
| Canada, Ontario, Lake 305 | IA-CA | CA305 | 1987-1990 | x | 6 1-5 | 1990-1991 |
| Canada, Ontario, Lake 373 | IA-CA | CA373 | 1987-1990 | x | 5 1-5 | 1990-1991 |
| **Canada, Quebec, Lac Laflamme** | **IA-CA** | **C08** | **1981-1994** | x | | |
| **Canada, Quebec, Parc de la Jacques-Cart., Lac Bonnevlle** | **IA-CA** | **C07** | **1981-1994** | x | | |
| **Canada, Quebec, Reserve faunique des Laur., Lac Macleod** | **IA-CA** | **C09** | **1982-1994** | x | | |
| **Canada, Quebec, Reserve faunique des Laur., Lac Josselin** | **IA-CA** | **C06** | **1982-1994** | x | | |
| **Canada, Quebec, Reserve faunique des Laur., Lac Veilleux** | **IA-CA** | **C05** | **1982-1994** | x | | |
| | | | | | | |
| **Czech Republic, Slovak Republic, Batizovske pleso** | **IA-CS** | **BA-1** | **1987-1995** | x | 21-8 | 1994 |
| **Czech Republic, Slovak Republic, Starolesnianske pleso** | **IA-CS** | **VS-15** | **1987-1995** | x | 21-3 | 1994 |
| **Czech Republic, Slovak Republic, Male Hincovo pleso** | **IA-CS** | **ME-2** | **1987-1995** | x | 21-11 | 1994 |
| **Czech Republic, Slovak Republic, Slavkovske pleso** | **IA-CS** | **SL-2** | **1987-1995** | x | 21-5 | 1994 |
| **Czech Republic, Slovak Republic, Jamske pleso** | **IA-CS** | **ST-1** | **1987-1995** | x | 21-1 | 1994 |
| **Czech Republic, Slovak Republic, L'adove pleso** | **IA-CS** | **VS-4** | **1987-1995** | x | 21-9 | 1994 |
| | | | | | | |
| **Denmark, Sepstrup Sande, Skaerbaek, Station B** | **IA-DA** | **SK110B** | **1976-1995** | x | | |
| **Denmark, Sepstrup Sande, Skaerbaek, Station F** | **IA-DA** | **SK038F** | **1987-1995** | x | | |
| | | | | | | |
| **Finland, Hirvilampi** | **IA-SF** | **SF01** | **1978-1995** | x | 2 3 | 1988 |
| Finland, Kivijärvi | IA-SF | SF04 | 1962-1995 | x | | |
| **Finland, Lapland, Suopalampi** | **IA-SF** | **SF05** | **1982-1995** | x | | |
| **Finland, Lapland, Vasikkajärvi** | **IA-SF** | **SF06** | **1992-1995** | x | | |
| **Finland, Mäkilampi** | **IA-SF** | **SF03** | **1978-1995** | x | 2 1 | 1988 |
| **Finland, Vuorilampi** | **IA-SF** | **SF02** | **1978-1995** | x | 2 2 | 1988 |

(continued)

**Table 32** (continued). Sites in **bold** have been included in the new data base.

| Station name | File | Chemistry code | Year | Site data | Biology code | Biology year |
|---|---|---|---|---|---|---|
| **France, Pyrenees, Lake Aubé** | **IA-FR** | **FA01** | **1991-1995** | x | | |
| Germany, Black Forest, Hüttenbächle | IA-GE | Hue2 | 1986-1991 | x | 7 2 | 1985-1990 |
| Germany, Black Forest, Kaltenbach | IA-GE | Kal4 | 1986-1991 | x | | 1986-1990 |
| Germany, Balck Forest, Kaltenbach | IA-GE | Kal5 | 1986-1989 | x | 7 5 | 1986 |
| Germany, Black Forest, Kleine Kinzig | IA-GE | Kle1 | 1986-1994 | x | 7 1 | 1987-1993 |
| **Germany, Colditzer Forst, Ettelsbach** | **IA-GE** | **Ett6** | **1992-1995** | x | **15-1** | **1992-1993** |
| **Germany, Dahlener Heide, Heidelbach** | **IA-GE** | **Hei7** | **1992-1995** | x | **16-1** | **1992-1993** |
| **Germany, East Bavaria, Grosse Ohe** | **IA-GE** | **Gro33** | **1979-1995** | x | **8 33** | **1983-1993** |
| **Germany, East Bavaria, Hinterer Schachtenbach** | **IA-GE** | **Hin31** | **1983-1995** | x | **8 31** | **1983-1993** |
| Germany, East Bavaria, Rachelsee | IA-GE | Rac40 | 1984-1993 | x | | 1983-1993 |
| **Germany, East Bavaria, Seebach** | **IA-GE** | **See30** | **1983-1995** | x | **8 30** | **1983-1993** |
| **Germany, East Bavaria, Vorderer Schachtenbach** | **IA-GE** | **Vor32** | **1983-1995** | x | **8 32** | **1983-1993** |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal1 | 1986-1990 | x | 8 1 | 1986 |
| **Germany, East Bavaria, Waldnaab** | **IA-GE** | **Wal2** | **1986-1995** | x | **8 2** | **1986-1993** |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal3 | 1986-1994 | x | 8 3 | 1986-1993 |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal5 | 1986-1987 | x | | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal8 | 1986-1994 | x | 8 8 | 1986-1993 |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal9 | 1986-1994 | x | 8 9 | 1986-1993 |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal11 | 1986-1990 | x | 8 1 | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal12 | 1986-1990 | x | 8 11 | |
| Germany, East Bavaria, Wlaldnaab | IA-GE | Wal14 | 1986-1994 | x | 8 14 | 1986-1993 |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal16 | 1986-1990 | x | 8 16 | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal17 | 1986-1987 | x | 8 17 | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal18 | 1986-1990 | x | 8 18 | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal15a | 1987-1987 | x | | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal17a | 1987-1990 | x | | |
| Germany, East Bavaria, Waldnaab | IA-GE | Wal17b | 1988-1990 | x | | |
| **Germany, Elbsandsteingebirge, Taubenbach** | **IA-GE** | **Tau5** | **1993-1995** | x | **13-1** | **1992-1993** |
| **Germany, Erzgebirge, Grosse Pyra** | **IA-GE** | **Gro2** | **1981-1995** | x | **14-1** | **1993** |
| Germany, Erzgebirge, Neunzehnhain | IA-GE | Neu8 | 1966-1993 | x | | 1993 |
| **Germany, Erzgebirge, Rote Pockan** | **IA-GE** | **Rot3** | **1979-1995** | x | **14-2** | **1993** |
| Germany, Erzgebirge, Sosa | IA-GE | Sos9 | 1953-1993 | x | 14-3 | 1993 |
| **Germany, Erzgebirge, Wilde Weisseritz** | **IA-GE** | **Wil4** | **1966-1995** | x | **14-4** | **1993** |
| **Germany, Erzgebirge, Wolfsbach** | **IA-GE** | **Wol1** | **1992-1995** | x | **14-5** | **1992-1993** |
| **Germany, Fichtelgebirge, Eger** | **IA-GE** | **Ege1** | **1982-1995** | x | **10-1** | **1992-1993** |
| **Germany, Fichtelgebirge, Röslau** | **IA-GE** | **Roe2** | **1982-1995** | x | **11-1** | **1992-1993** |
| Germany, Fichtelgebirge, Zinnbach | IA-GE | Zin3 | 1983-1994 | x | 12-1 | 1992-1993 |
| Germany, Harz, Alte Riefensbeek | IA-GE | AltR1 | 1986-1992 | x | 2-3 | 1986-1993 |
| Germany, Harz, Alte Riefensbeek | IA-GE | AltR2 | 1986-1991 | x | 2-4 | 1986-1990 |
| Germany, Harz, Alte Riefensbeek | IA-GE | AltR3 | 1986-1991 | x | 2-5 | 1986-1990 |
| **Germany, Harz, Dicke Bramke** | **IA-GE** | **Dic** | **1988-1995** | x | | **1988-1993** |
| **Germany, Harz, Grosse Bode** | **IA-GE** | **Gro14** | **1986-1995** | x | **2-14** | **1986-1993** |
| Germany, Harz, Grosse Soese | IA-GE | GroS1 | 1986-1992 | x | 2-6 | 1986-1990 |
| **Germany, Harz, Grosse Soese** | **IA-GE** | **GroS2** | **1986-1995** | x | **2-7** | **1986-1993** |
| Germany, Harz, Grosse Soese | IA-GE | GroS3 | 1986-1991 | x | 2-8 | 1986-1990 |
| Germany, Harz, Lange Bramke | IA-GE | LanL1 | 1988-1991 | x | 2-11 | 1986-1989 |
| Germany, Harz, Lange Bramke | IA-GE | LanL2 | 1988-1991 | x | 2-12 | 1986-1990 |
| Germany, Harz, Lange Bramke | IA-GE | LanL3 | 1988-1991 | x | | 1986-1990 |
| Germany, Harz, Mollentalbach | IA-GE | Mol10 | 1986-1990 | x | 2-10 | 1986-1990 |
| **Germany, Harz, Grosse Schacht** | **IA-GE** | **Sch9** | **1986-1995** | x | **2-9** | **1986-1993** |
| Germany, Harz, Varleybach | IA-GE | Var1 | 1986-1990 | x | | 1986-1990 |
| Germany, Harz, Varleybach | IA-GE | Var2 | 1986-1990 | x | | 1986-1990 |
| Germany, Harz, Warme Bode | IA-GE | War15 | 1986-1991 | x | | 1986-1990 |
| **Germany, Hunsrueck, Grafenbach** | **IA-GE** | **Gra5** | **1982-1995** | x | **9-5** | **1982-1993** |
| Germany, Hunsrueck, Schwollbach | IA-GE | Sch10 | 1982-1990 | x | 9-10 | 1982-1990 |
| **Germany, Hunsrueck, Traunbach** | **IA-GE** | **Tra1** | **1982-1995** | x | **9-1** | **1983-1993** |
| Germany, Hunsrueck, Traunbach | IA-GE | Tra2 | 1984-1990 | x | 9-2 | 1985-1990 |
| Germany, Kaufunger Wald, Nieste | IA-GE | Nie1 | 1987-1994 | x | 3-1 | 1987-1993 |
| Germany, Kaufunger Wald, Nieste | IA-GE | Nie2 | 1987-1990 | x | 3-2 | 1987-1990 |
| Germany, Kaufunger Wald, Nieste | IA-GE | Nie3 | 1987-1994 | x | 3-3 | 1987-1993 |
| Germany, Kaufunger Wald, Nieste | IA-GE | Nie4 | 1987-1990 | x | 3-4 | 1987-1990 |
| **Germany, Kaufunger Wald, Nieste** | **IA-GE** | **Nie5** | **1987-1995** | x | **3-5** | **1987-1990** |
| Germany, Lauenburg, Garrensee | IA-GE | Gar1 | 1986-1989 | x | 1-1 | 1986-1989 |
| Germany, Lauenbrug, Pinnsee | IA-GE | Pin3 | 1986-1993 | x | 1-3 | 1986-1993 |
| Germany, Lauenburg, Plötschersee | IA-GE | Plo2 | 1986-1989 | x | 1-2 | 1986-1989 |

(continued)

**Table 32** (continued). Sites in **bold** have been included in the new data base.

| Station name | File | Chemistry code | Year | Site data | Biology code | Biology year |
|---|---|---|---|---|---|---|
| Germany, Odenwald, Schmerbach | IA-GE | Sch1 | 1984-1994 | x | 6-1 | 1987-1993 |
| **Germany, Odenwald, Schmerbach** | **IA-GE** | **Sch3** | **1985-1995** | **x** | **6-2** | **1987-1993** |
| Germany, Odenwald, Schmerbach | IA-GE | Sch4 | 1988-1990 | x | 6-3 | 1988-1990 |
| Germany, Odenwald, Schmerbach, Hiltersklinger Teich | IA-GE | Sch2 | 1984-1990 | x | 6-4 | 1987-1993 |
| **Germany, Rothaargebirge, Elberndorfer Bach** | **IA-GE** | **Elb1** | **1986-1995** | **x** | **4-1** | **1986-1992** |
| **Germany, Rothaargebirge, Zinse** | **IA-GE** | **Zin2** | **1986-1995** | **x** | **4-2** | **1986-1992** |
| Germany, Taunus, Rombach | IA-GE | Rom1 | 1988-1990 | x | 5-1 | 1987-1992 |
| **Germany, Taunus, Rombach** | **IA-GE** | **Rom2** | **1986-1995** | **x** | **5-2** | **1987-1992** |
| Germany, Taunus, Rombach | IA-GE | Rom3 | 1986-1994 | x | 5-3 | 1987-1993 |
| Germany, Taunus, Rombach | IA-GE | Rom4 | 1987-1990 | x | 5-4 | 1987-1992 |
| Germany, Taunus, Silberbach | IA-GE | Sil5 | 1986-1990 | x | 5-5 | 1987-1990 |
| | | | | | | |
| **Hungary, Matra Mountains, Csorret reservoir** | **IA-HU** | **CSORR** | **1986-1994** | **x** | | |
| | | | | | | |
| Ireland, Doo Lough, Inflow 1 | IA-IR | DOO11 | 1987-1989 | x | | |
| Ireland, Doo Lough, Inflow 2 | IA-IR | DOO12 | 1987-1989 | x | | |
| Ireland, Doo Lough, Inflow 3 | IA-IR | DOO13 | 1987-1989 | x | | |
| Ireland, Doo Lough, Mid Lake | IA-IR | DOO10 | 1984-1989 | x | | |
| Ireland, Glendalough, Lake Upper, Inflow 1 | IA-IR | GLE11 | 1984-1995 | x | 1-1 | 1987-1995 |
| Ireland, Glendalough, Lake Upper, Inflow 2 | IA-IR | GLE12 | 1984-1995 | x | 1-3 | 1987-1995 |
| Ireland, Glendalough, Lake Upper, Inflow 3 | IA-IR | GLE13 | 1987-1995 | x | | |
| **Ireland, Glendalough, Lake Upper, Mid Lake** | **IA-IR** | **GLE10** | **1984-1995** | **x** | | |
| Ireland, Lough Maumwee, Inflow 1 | IA-IR | MAU11 | 1984-1995 | x | 2-1 | 1988-1995 |
| Ireland, Lough Maumwee, Inflow 2 | IA-IR | MAU12 | 1984-1995 | x | 2-2 | 1988-1995 |
| **Ireland, Lough Maumwee, Mid Lake** | **IA-IR** | **MAU10** | **1984-1995** | **x** | **2-3** | **1988-1995** |
| Ireland, Lough Naminna, Inflow 1 | IA-IR | NAM11 | 1987-1993 | x | 4-1 | 1988-1995 |
| Ireland, Lough Naminna, Mid Lake | IA-IR | NAM10 | 1987-1993 | x | 4-4 | 1988-1995 |
| **Ireland, Lough Veagh, Mid Lake** | **IA-IR** | **VEA10** | **1988-1995** | **x** | **5-5** | **1988-1995** |
| Ireland, Lough Veagh, Inflow 1 | IA-IR | VEA11 | 1988-1995 | x | 5-1 | 1988-1995 |
| Ireland, Lough Veagh, Inflow 2 | IA-IR | VEA12 | 1988-1995 | x | 5-2 | 1988-1995 |
| Ireland, Lough Veagh, Inflow 3 | IA-IR | VEA13 | 1988-1995 | x | 5-3 | 1988-1995 |
| Ireland, Lough Veagh, Inflow 4 | IA-IR | VEA14 | 1988-1995 | x | 5-4 | 1988-1995 |
| | | | | | | |
| **Italy, Lake Paione Inferiore** | **IA-ITA** | **ITA3** | **1978-1995** | **x** | | |
| **Italy, Lake Mergozzo** | **IA-ITA** | **ITA5** | **1970-1995** | **x** | | |
| **Italy, Lake Paione Superiore** | **IA-ITA** | **ITA4** | **1992-1995** | **x** | | |
| **Ital, River Cannobino** | **IA-ITA** | **ITA2** | **1971-1995** | **x** | | |
| **Italy, River Pellino** | **IA-ITA** | **ITA1** | **1984-1995** | **x** | | |
| **Italy, River Pellesino** | **IA-ITA** | **ITA6** | **1986-1995** | **x** | | |
| Italy, River Pescone | IA-ITA | ITA7 | 1984-1995 | x | | |
| | | | | | | |
| Latvia, Barta, Dukupiji | IA-LA | DUK-B | 1992-1995 | x | 3-1 | 1994-1995 |
| Latvia, Burtnieku Lake, hydrosite | IA-LA | BURT-1 | 1992-1995 | x | | |
| Latvia, Burtnieku Lake, 1.0 km from bank | IA-LA | BURT-2 | 1992-1995 | x | | |
| **Latvia, Daugava, 3 km above Daugapils** | **IA-LA** | **DAUG-1** | **1992-1995** | **x** | | |
| Latvia, Daugava, 1.5 km below Daugapils | IA-LA | DAUG-2 | 1992-1995 | x | | |
| Latvia, Gauja, 2.5 km above Valmiera | IA-LA | VALM-1 | 1992-1995 | x | 1-1 | 1994-1995 |
| Latvia, Gauja, 3.0 km below Valmiera | IA-LA | VALM-2 | 1992-1995 | x | 1-2 | 1994-1995 |
| Latvia, Liela Jugla, Zaki | IA-LA | ZAK-LJ | 1992-1995 | x | | |
| Latvia, Lielupe, 0.5 km above Jelgave | IA-LA | JELG-1 | 1992-1995 | x | 2-1 | 1994-1995 |
| Latvia, Lielupe, 1.0 km below Jelgave | IA-LA | JELG-2 | 1992-1995 | x | 2-2 | 1994-1995 |
| **Latvia, Tulija, Zoseni** | **IA-LA** | **ZOS-T** | **1992-1995** | **x** | | |
| **Latvia, Zvirbuli stream, hydrosite** | **IA-LA** | **ZVIR** | **1992-1995** | **x** | | |
| | | | | | | |
| **Norway, Aust-Agder, Birkenes stream, Birkenes** | **BIE01** | | **1972-1995** | **x** | | |
| **Norway, Aust-Agder, Tovdalselva, Boen bruk** | **OVELV** | **7 1** | **1980-1995** | **x** | | |
| **Norway, Buskerud, Langtjern, outflow** | **LAE01** | | **1972-1995** | **x** | | |
| **Norway, Finnmark, Dalelva, Jarfjord** | **DALELV** | **1** | **1988-1995** | **x** | | |
| **Norway, Oppland, Aurdoela, Aurdalsfjorden** | **OVELV** | **90 1** | **1986-1995** | **. x** | | |
| Norway, Rogaland,Ogna | | | | | 3 1-10 | 1984-1995 |
| **Norway, Rogaland, Vikedalselva, Vindafjord** | **OVELV** | **32 9** | **1982-1995** | **x** | 6 1-18 | 1987-1995 |
| **Norway, Sogn og Fjordane, Gaular, Eldalen.** | **OVELV** | **57 3** | **1980-1995** | **x** | 17 1-17 | 1985-1995 |
| **Norway, Sogn og Fjordane, Nausta, Naustdal** | **OVELV** | **34 1** | **1980-1995** | **x** | 20 1-20 | 1985-1995 |
| **Norway, Sogn og Fjordane, Trodøla** | **OVELV** | **34 5** | **1984-1995** | **x** | 20 7 | 1986-1995 |
| **Norway, Telemark, Storgama, Outflow** | **STE01** | | **1974-1995** | **x** | | |

(continued)

**Table 32** (continued). Sites in **bold** have been included in the new data base.

| Station name | File | Chemistry code | Year | Site data | Biology code | Biology year |
|---|---|---|---|---|---|---|
| **Norway, Vest-Agder, Farsund** | | | | | | |
| Gjaervollstadvatn and Saudlandsvatn | | | | | 2 1-7 | 1981-1995 |
| | | | | | | |
| **Poland, Tatra Mountains, Dlugi, Staw Gasienicowy** | IIA-PO | DLUGI | 1992-1995 | x | 22-3 | 1994 |
| **Poland, Tatra Mountains, Zielony, Staw Gasienicowy** | IIA-PO | ZIELON | 1992-1995 | x | 22-4 | 1994 |
| | | | | | | |
| Russia, Kola | IA-RU | KOL | 1991-1993 | x | | |
| **Russia, Baikal** | IA-RU | BAL2 | 1991-1994 | | | |
| | | | | | | |
| Sweden, Alsteraan, Getebo | IA-SV | S02 | 1985-1995 | x | | |
| Sweden, Alsteraan, Stroemsborg | IA-SV | S03 | 1985-1995 | x | | |
| Sweden, Anraasen, Haersvatn | IA-SV | S04 | 1984-1995 | x | | |
| **Sweden, Brunnsjoen** | IA-SV | S08 | 1984-1995 | x | 3-1 | 1986-1992 |
| Sweden, Delaangersaan, Iggersund | IA-SV | S01 | 1984-1995 | x | | |
| **Sweden, Fiolen** | IA-SV | S09 | 1984-1995 | x | 1-1 | 1986-1992 |
| **Sweden, Fraecksjoen** | IA-SV | S11 | 1984-1995 | x | 5-1 | 1986-1992 |
| Sweden, Haersvatn | IA-SV | S12 | 1984-1995 | x | 4-1 | 1986-1992 |
| Sweden, Moecklasjoen | IA-SV | S07 | 1984-1987 | x | | |
| **Sweden, Stensjoen** | IA-SV | S06 | 1985-1995 | x | 6-1 | 1986-1992 |
| **Sweden, Storasjoen** | IA-SV | S10 | 1984-1995 | x | 2-1 | 1986-1992 |
| **Sweden, Tvaeringen** | IA-SV | S05 | 1984-1995 | x | 7-1 | 1986-1992 |
| | | | | | | |
| The Netherlands, Achterste Goorven, station A | IA-NE | AGA | 1958-1995 | x | | |
| The Netherlands, Achterste Goorven, station B | IA-NE | AGB | 1925-1995 | x | | |
| **The Netherlands, Achterste Goorven, station E** | IA-NE | AGE | 1975-1995 | x | | |
| **The Netherlands, Gerritsfles** | IA-NE | GER | 1991-1995 | x | | |
| **The Netherlands, Kliplo** | IA-NE | KLI | 1958-1995 | x | | |
| | | | | | | |
| **UK, N. Ireland, Blue Loch** | IA-UK | UK21 | 1990-1995 | x | 9-1 | 1995 |
| **UK, England, Scoat Tarn** | IA-UK | UK10 | 1988-1995 | x | 5-3 | 1988-1995 |
| **UK, Scotland, Loch Coire nan Arr** | IA-UK | UK01 | 1988-1995 | x | 5-1 | 1988-1995 |
| **UK, Scotland, Lochnager** | IA-UK | UK04 | 1988-1995 | x | 2-2 | 1995 |
| **UK, Scotland, Round of Glenshead** | IA-UK | UK07 | 1988-1995 | x | 6-1 | 1988-1995 |
| **UK, Wales, Llyn Llagi** | IA-UK | UK15 | 1988-1995 | x | 7-1 | 1988-1995 |
| | | | | | | |
| **USA, Colorado, Seven Lakes** | IA-US | 4E2009 | 1985-1994 | x | | |
| **USA, Colorado, Summit Lake** | IA-US | 4E2060 | 1985-1994 | x | | |
| **USA, Colorado, Sunlight Lake** | IA-US | 4E2070 | 1985-1994 | x | | |
| **USA, Colorado, White Dome Lake** | IA-US | 4E2071 | 1985-1994 | x | | |
| **USA, Maine, Little Long Pond** | IA-US | 1E1132 | 1982-1994 | x | | |
| **USA, Maine, Tilden Pond** | IA-US | 1E1133 | 1982-1994 | x | | |
| **USA, Michigan, Andrus** | IA-US | 2B3082 | 1983-1994 | x | | |
| **USA, Michigan, Buckeye** | IA-US | 2B2102 | 1983-1994 | x | | |
| **USA, Michigan, Johnson** | IA-US | 2B1047 | 1983-1994 | x | | |
| **USA, Minnesota, Cruiser** | IA-US | 2A2063 | 1983-1994 | x | | |
| **USA, New York, Adirondack Mountain, Arbutus** | IA-US | 1A1052 | 1983-1994 | x | | |
| **USA, New York, Adirondack Mountain, Constable** | IA-US | 1A1017 | 1982-1994 | x | | |
| **USA, New York, Adirondack Mountain, Dart Lake** | IA-US | 1A1106 | 1982-1994 | x | | |
| **USA, New York, Adirondack Mountain, Heart Lake** | IA-US | 1A1102 | 1982-1994 | x | | |
| **USA, New York, Adirondack Mountain, Lake Rondaxe** | IA-US | 1A1110 | 1982-1994 | x | | |
| **USA, New York, Adirondack Mountain, Moss Lake** | IA-US | 1A1109 | 1982-1994 | x | | |
| **USA, New York, Adirondack Mountain, Otter Lake** | IA-US | 1A2078 | 1982-1994 | x | | |
| **USA, New York, Catskill M., East Branch Neversink, Mid.** | IA-US | 143010 | 1983-1993 | x | | |
| USA, New York, Catskill m., High Falls Brook | IA-US | 143105 | 1983-1993 | x | | |
| **USA, Wisconsin, Luna** | IA-US | 2C2062 | 1983-1994 | x | | |
| **USA, Wisconsin, Nichols** | IA-US | 2C1069 | 1983-1994 | x | | |
| **USA, Wisconsin, Sand** | IA-US | 2C1068 | 1983-1994 | x | | |

# Annex B.

## Comparison of Trends at ICP Integrated Monitoring Sites
## with ICP Waters Regional Trends

As a first attempt at integrating the results of another ICP program with ICP Waters, we conducted trends tests on all available data from sites in ICP Integrated Monitoring, using methods identical to those used for ICP Waters (see chapter 3.3). In most cases, data were available only for the 1990s, and our decision to collapse the data into quarterly values (to maintain comparability among all of the ICP Waters and ICP IM sites) resulted in fewer than 24 observations per site for conducting the seasonal Kendall tests. This illustrates one of the reasons that we embarked on the regional trend analysis for ICP Waters in the first place. When there are few observations on which to base a trend analysis, the probability of detecting significant trends is very small (i.e., the trend must be very strong to be significant). When the trend results from many sites are combined, as is done for the regional meta-analysis, it has the effect of multiplying the sample size and, if all of the site exhibit similar (homogeneous) trends, increasing the likelihood that a significant trend will be detected. Most of the ICP IM trends are not significant at the $p<0.05$ level (**Table 33**), and this may be as much a result of small sample sizes as of a lack of strong trends.

We have compared the ICP IM results with the meta-analysis results for each region (and decade) for which ICP IM data exist (**Table 33**). Because so few of the ICP IM results are significant ($p<0.05$), the amount of interpretation that we can make is limited. In the case of $SO_4^{2-}$, all of the significant ICP IM results are consistent with the regional trend results (trends are downward for both the 1980s and 1990s in Midwestern North America and in the Nordic Countries). Since the regional results for $SO_4^{2-}$ were the most dramatic (see chapter 3.5), and because sulphur deposition is such a large-scale phenomenon, it is not at all surprising that there is consistency within the ICP Waters sites and between the ICP Waters and ICP IM sites. In the case of base cations, the pattern observed for the Nordic Countries in the regional analysis is corroborated by the ICP IM results: base cations were declining strongly throughout the Nordic Countries in the 1980s, but have now either flattened out or a beginning to increase. Too few data are available to say whether the ICP IM sites also illustrate how the reversal in base cation trends has led to a recovery in ANC, as the ICP Waters regional analysis did.

For all other variables, there are too few significant results to make any conclusions. In the future, it may be possible to merge the two ICP datasets, and re-analyze the regional trends using the combined data. This could produce more reliable regional estimates of change, particularly for Europe, where few ICP Waters sites had adequate date for the seasonal Kendall tests.

**Table 33.** Comparison of regional trends results from ICP Waters sites with individual trends results for ICP IM sites (only regions with both ICP Waters and ICP IM sites are shown).

| Region | ICP IM Site | Decade | $\Delta$ ANC $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ SO$_4^{2-}$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ NO$_3^-$ $ueq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ SBC $ueq \cdot l^{-1} \cdot yr^{-1}$ |
|---|---|---|---|---|---|---|
| **Europe** | | **1980s** | **+2.5** (p < 0.05) | **-1.3** (p<0.05) | **+1.4** (p<0.001) | **+0.0** (n.s.) |
| **Europe** | | **1990s** | **+7.9** (p<0.001) | **-4.5** (p < 0.001) | **-2.4** (p < 0.001) | **-6.3** (n.s.) |
| | CH_1 | | | -11.7 (n.s.) | -0.8 (p<0.10) | -34.1 (n.s.) |
| | CZ_01_74 | | | -22.9 (p<0.10) | +17.0 (n.s.) | -3.4 (n.s.) | +23.2 (n.s.) |
| | CZ_01_75 | | | -34.8 (n.s.) | +48.6 (p<0.10) | +22.2 (n.s.) | +83.0 (n.s.) |
| | DE_01 | | | -2.9 (n.s.) | +0.4 (n.s.) | -1.3 (n.s.) | +11.7 (p<0.10) |
| | RU15 | | | -131.5 (p<0.10) | -36.3 (n.s.) | +1.7 (n.s.) | -141.9 (p<0.05) |
| **Midwestern North America** | | **1980s** | **+0.7** (p<0.05) | **-1.5** (p<0.001) | **+0.0** (p<0.10) | **-0.9** (n.s.) |
| | CA_01* | | | +1.7 (p<0.01) | -1.2 (p<0.05) | -0.0 (n.s.) | +2.1 (p<0.10) |
| **Midwestern North America** | | **1990s** | **+-1.6** (p<0.01) | **-4.3** (p < 0.001) | **-0.0** (n.s.) | **-4.8** (p<0.001) |
| | CA_01* | | | -1.6 (n.s.) | -3.7 (p < 0.001) | +0.1 (n.s.) | -6.3 (p < 0.001) |
| **Nordic Countries** | | **1980s** | **-1.4** (p<0.001) | **-0.8** (p < 0.001) | **+0.1** (p < 0.05) | **-2.4** (p<0.001) |
| | SE01_1 | | | | -8.6 (p<0.10) | +0.0 (n.s.) | -9.2 (p < 0.001) |
| | SE01_2 | | | | -5.7 (n.s.) | -0.3 (n.s.) | -3.1 (n.s.) |
| | SE02_1 | | | | +3.7 (n.s.) | -0.5 (n.s.) | -0.6 (n.s.) |
| | SE02_2 | | | | +2.1 (n.s.) | -0.0 (n.s.) | +5.0 (n.s.) |
| | SE11 | | | | +0.0 (n.s.) | +0.1 (n.s.) | -13.5 (n.s.) |
| | SE12 | | | | -5.0 (p < 0.001) | +0.5 (p<0.01) | -39.5 (p < 0.001) |

**Table 33.** Comparison of regional trends results from ICP Waters sites with individual trends results
*(cont.)* for ICP IM sites (only regions with both ICP Waters and ICP IM sites are shown).

| Region | ICP IM Site | Decade | $\Delta$ ANC $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ $SO_4^{2-}$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ $NO_3^-$ $\mu eq \cdot l^{-1} \cdot yr^{-1}$ | $\Delta$ SBC $\mu eq \cdot l^{-1} \cdot yr^{-1}$ |
|---|---|---|---|---|---|---|
| **Nordic Countries** | | **1990s** | **+2.5** ($p < 0.001$) | **-3.0** ($p < 0.001$) | **+0.0** (n.s.) | **-0.0** (n.s.) |
| | FI01 | | | -4.0 (n.s.) | +0.3 ($p<0.05$) | +0.5 (n.s.) |
| | FI03_01 | | +0.3 (n.s.) | -3.1 ($p < 0.001$) | +0.0 (n.s.) | -1.6 (n.s.) |
| | FI03_07 | | -0.1 (n.s.) | -1.1 ($p<0.05$) | +0.0 (n.s.) | -5.1 (n.s.) |
| | FI05 | | +1.0 (n.s.) | -0.7 (n.s.) | -0.0 (n.s.) | -11.6 ($p<0.10$) |
| | NO02a | | | +0.5 ($p<0.10$) | +0.1 (n.s.) | +2.5 ($p<0.01$) |
| | SE01_1 | | | -2.4 (n.s.) | -0.1 (n.s.) | +1.5 ($p<0.10$) |
| | SE01_2 | | | -3.0 (n.s.) | -0.3 (n.s.) | +0.2 (n.s.) |
| | SE02_1 | | | +6.2 (n.s.) | +0.3 (n.s.) | -1.9 (n.s.) |
| | SE02_2 | | | -0.7 (n.s.) | +0.4 (n.s.) | -3.3 (n.s.) |
| | SE11 | | | -6.0 ($p<0.05$) | -0.3 (n.s.) | 2.2 (n.s.) |
| | SE12 | | | -0.4 (n.s.) | -0.6 (n.s.) | +21.3 ($p<0.05$) |
| | SE13 | | -3.2 (n.s.) | | | |
| **United Kingdom** | | **1990s** | **+0.2** (n.s.) | **-0.0** (n.s.) | **+0.2** ($p < 0.001$) | **-1.5** (n.s.) |
| | GB_02 | | -4.6 ($p<0.01$) | | | |

\* ICP IM site CA01 (Turkey Lake) is also part of ICP Waters. Its results are included here for completeness, but a valid comparison between this site and the regional ICP Waters results cannot be made, because the data from this site contributed to the regional estimate.

# Annex C

## SO$_4$: Results of the Nonparametric Seasonal Kendall Test (SKT) for the Single ICP Waters Sites

Table 34. SO$_4$ concentrations.

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---------|------|-----------|----------|--------|---|---|---|-------|
| CANADA | C01 | BATCHAWANA | SO$_4$ | 80 | 37 | -1.90 | 0.029 | -1.29 |
| | C02 | WISHART | SO$_4$ | 80 | 40 | -3.26 | 0.001 | -2.19 |
| | C03 | LITLE_TURKEY | SO$_4$ | 80 | 39 | -2.50 | 0.006 | -1.27 |
| | C04 | TURKEY | SO$_4$ | 80 | 40 | -2.28 | 0.011 | -1.20 |
| | C05 | LAC_VEILLE | SO$_4$ | 80 | 15 | -0.13 | 0.449 | 0.00 |
| | C06 | LAC_JOSSELI | SO$_4$ | 80 | 13 | -1.09 | 0.142 | -2.21 |
| | C07 | BONNEVILLE | SO$_4$ | 80 | 28 | 1.36 | 0.088 | 1.26 |
| | C08 | LAFLAMME | SO$_4$ | 80 | 32 | 1.55 | 0.061 | 1.17 |
| | C09 | LAC_MACLEO | SO$_4$ | 80 | 15 | 0.00 | 0.500 | 0.00 |
| | C10 | MOUNT_TOM_LAKE | SO$_4$ | 80 | 15 | 0.00 | 0.500 | 0.00 |
| | C12 | RED_LAKE | SO$_4$ | 80 | 15 | 0.11 | 0.447 | 0.00 |
| | C13 | KEJUMKUJIK_LAKE | SO$_4$ | 80 | 36 | 2.37 | 0.009 | 0.52 |
| | C01 | BATCHAWANA | SO$_4$ | 90 | 20 | -4.04 | 0.000 | -6.29 |
| | C02 | WISHART | SO$_4$ | 90 | 20 | -3.31 | 0.000 | -5.07 |
| | C03 | LITLE_TURKEY | SO$_4$ | 90 | 20 | -3.55 | 0.000 | -5.55 |
| | C04 | TURKEY | SO$_4$ | 90 | 20 | -3.31 | 0.000 | -3.71 |
| | C07 | BONNEVILLE | SO$_4$ | 90 | 20 | -3.21 | 0.000 | -5.72 |
| | C08 | LAFLAMME | SO$_4$ | 90 | 20 | -3.06 | 0.001 | -3.22 |
| DENMARK | SK110B | SKAERBAEK_B | SO$_4$ | 90 | 20 | -2.62 | 0.004 | -3.17 |
| | SK038F | SKAERBAEK_F | SO$_4$ | 90 | 20 | -2.38 | 0.008 | -9.80 |
| FINLAND | SF01 | HIRVILAMPI | SO$_4$ | 80 | 13 | 0.00 | 0.500 | -1.05 |
| | SF01 | HIRVILAMPI | SO$_4$ | 90 | 18 | -1.64 | 0.050 | -3.20 |
| | SF02 | VUORILAMPI | SO$_4$ | 90 | 17 | -0.84 | 0.203 | -3.50 |
| | SF03 | MAEKILAMPI | SO$_4$ | 90 | 19 | -2.85 | 0.002 | -5.59 |
| GERMANY | EGE1 | EGER | SO$_4$ | 80 | 24 | -1.60 | 0.055 | -8.33 |
| | HIN31 | HINTERER_SCHACTENBACH | SO$_4$ | 80 | 28 | -1.13 | 0.131 | -3.26 |
| | ROE2 | ROESLAU | SO$_4$ | 80 | 24 | -2.35 | 0.009 | -17.18 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| GERMANY | SEE30 | SEEBACH | SO$_4$ | 80 | 24 | -0.28 | 0.390 | -0.78 |
| | TRA1 | TRAUNBACH | SO$_4$ | 80 | 27 | -3.54 | 0.000 | -48.32 |
| | VOR32 | VORDERER_SCHACHTENBACH | SO$_4$ | 80 | 24 | 0.00 | 0.537 | -0.09 |
| | ZIN2 | ZINSE | SO$_4$ | 80 | 24 | -2.16 | 0.015 | -12.03 |
| | DIC | DICKE_BRAMKE | SO$_4$ | 90 | 17 | -0.46 | 0.325 | -3.90 |
| | DUE6 | DUERREYCHBACH | SO$_4$ | 90 | 20 | -1.10 | 0.137 | -1.39 |
| | EGE1 | EGER | SO$_4$ | 90 | 20 | 0.00 | 0.500 | 0.35 |
| | GRA5 | GRAFENBACH | SO$_4$ | 90 | 20 | -1.73 | 0.042 | -9.89 |
| | GROS2 | GROSSE_SOESE | SO$_4$ | 90 | 20 | -1.43 | 0.077 | -5.09 |
| | HIN31 | HINTERER_SCHACTENBACH | SO$_4$ | 90 | 20 | 0.37 | 0.358 | 0.41 |
| | NIE5 | NIESTE | SO$_4$ | 90 | 18 | -1.37 | 0.087 | -11.68 |
| | ROE2 | ROESLAU | SO$_4$ | 90 | 20 | -0.99 | 0.164 | -5.44 |
| | ROM2 | ROMBACH | SO$_4$ | 90 | 19 | -2.63 | 0.003 | -16.83 |
| | SEE30 | SEEBACH | SO$_4$ | 90 | 20 | -0.12 | 0.452 | -0.46 |
| | TRA1 | TRAUNBACH | SO$_4$ | 90 | 20 | -1.10 | 0.137 | -8.91 |
| | VOR32 | VORDERER_SCHACHTENBACH | SO$_4$ | 90 | 20 | -0.12 | 0.452 | -0.49 |
| | ZIN2 | ZINSE | SO$_4$ | 90 | 20 | -1.84 | 0.032 | -8.91 |
| ITALY | ITA1 | PELLINO | SO$_4$ | 80 | 23 | -1.98 | 0.023 | -1.73 |
| | ITA2 | CANNOBINO | SO$_4$ | 80 | 36 | 3.18 | 0.001 | 1.63 |
| | ITA1 | PELLINO | SO$_4$ | 90 | 24 | -1.78 | 0.037 | -1.67 |
| | ITA2 | CANNOBINO | SO$_4$ | 90 | 24 | -1.22 | 0.112 | -2.43 |
| NETHERLANDS | GER | GERRITSFLES | SO$_4$ | 80 | 23 | 0.50 | 0.311 | 0.75 |
| | GER | GERRITSFLES | SO$_4$ | 90 | 19 | -2.74 | 0.002 | -16.03 |
| NORWAY | BIE01 | BIRKENES | SO$_4$ | 80 | 60 | -3.26 | 0.001 | -1.59 |
| | DALELV | DALELVA | SO$_4$ | 80 | 61 | -2.43 | 0.010 | -0.83 |
| | LAE01 | LANGTJERN | SO$_4$ | 80 | 61 | -2.43 | 0.010 | -0.83 |
| | OVE329 | VIKEDALSELVA | SO$_4$ | 80 | 22 | -1.79 | 0.036 | -0.42 |
| | OVE573 | GAULAR | SO$_4$ | 80 | 24 | -0.66 | 0.257 | -0.31 |
| | OVE901 | AURDOELA | SO$_4$ | 80 | 16 | -0.51 | 0.308 | -0.76 |
| | OVEL71 | TOVDALSELVA | SO$_4$ | 80 | 40 | -3.80 | 0.000 | -1.51 |
| | BIE01 | BIRKENES | SO$_4$ | 90 | 25 | -0.97 | 0.168 | -1.61 |
| | DALELV | DALELVA | SO$_4$ | 90 | 24 | -2.91 | 0.001 | -2.70 |
| | LAE01 | LANGTJERN | SO$_4$ | 90 | 24 | -3.10 | 0.000 | -3.45 |
| | OVE329 | VIKEDALSELVA | SO$_4$ | 90 | 25 | -0.44 | 0.332 | -0.46 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---------|------|-----------|----------|--------|-----|-------|-------|--------|
| NORWAY | OVE573 | GAULAR | SO$_4$ | 90 | 24 | -1.41 | 0.080 | -0.55 |
| | OVE901 | AURDOELA | SO$_4$ | 90 | 25 | -2.90 | 0.001 | -1.58 |
| | OVEL71 | TOVDALSELVA | SO$_4$ | 90 | 24 | -1.97 | 0.024 | -1.58 |
| SWEDEN | S01 | DALAANGERSAAN | SO$_4$ | 80 | 12 | -1.83 | 0.032 | -1.33 |
| | S04 | ANRAASEN | SO$_4$ | 80 | 15 | 1.47 | 0.071 | 4.12 |
| | S05 | TVAERINGEN | SO$_4$ | 80 | 16 | 0.27 | 0.394 | 0.48 |
| | S06 | STENSJOEN | SO$_4$ | 80 | 12 | -0.87 | 0.195 | -1.11 |
| | S08 | BRUNNSJOEN | SO$_4$ | 80 | 20 | -1.35 | 0.090 | -14.66 |
| | S09 | FIOLEN | SO$_4$ | 80 | 17 | -2.29 | 0.010 | -5.58 |
| | S10 | STORASJOEN | SO$_4$ | 80 | 13 | 0.59 | 0.282 | 1.10 |
| | S11 | FRAECKSJOEN | SO$_4$ | 80 | 17 | 0.25 | 0.401 | 0.97 |
| | S12 | HAERSVATN | SO$_4$ | 80 | 15 | 0.68 | 0.253 | 1.37 |
| | S01 | DALAANGERSAAN | SO$_4$ | 90 | 20 | -3.06 | 0.001 | -4.15 |
| | S04 | ANRAASEN | SO$_4$ | 90 | 20 | -2.08 | 0.018 | -1.99 |
| | S05 | TVAERINGEN | SO$_4$ | 90 | 24 | -3.29 | 0.000 | -3.27 |
| | S06 | STENSJOEN | SO$_4$ | 90 | 23 | -3.97 | 0.000 | -2.10 |
| | S08 | BRUNNSJOEN | SO$_4$ | 90 | 23 | -2.98 | 0.001 | -25.44 |
| | S09 | FIOLEN | SO$_4$ | 90 | 23 | 0.00 | 0.500 | -0.30 |
| | S10 | STORASJOEN | SO$_4$ | 90 | 23 | -1.59 | 0.056 | -4.04 |
| | S11 | FRAECKSJOEN | SO$_4$ | 90 | 23 | -2.18 | 0.014 | -4.90 |
| | S12 | HAERSVATN | SO$_4$ | 90 | 21 | -2.94 | 0.001 | -3.80 |
| U.K. | UK01 | LOCH_COIRE | SO$_4$ | 90 | 21 | 0.45 | 0.328 | 0.18 |
| | UK04 | LOCHNAGER | SO$_4$ | 90 | 20 | -2.71 | 0.003 | -0.73 |
| | UK07 | ROUND | SO$_4$ | 90 | 21 | 1.13 | 0.131 | 1.53 |
| | UK10 | SCOAT_TARN | SO$_4$ | 90 | 20 | 1.91 | 0.027 | 1.40 |
| | UK15 | LLYN_LLAGI | SO$_4$ | 90 | 20 | -0.12 | 0.453 | -0.20 |
| | UK21 | BLUE_LOCH | SO$_4$ | 90 | 17 | -0.30 | 0.395 | -0.40 |
| U.S.A. | 1A1-017S | CONSTABLE_POND_OUTLET | SO$_4$ | 80 | 31 | -2.45 | 0.007 | -1.23 |
| | 1A1-052O | ARBUTUS_LAKE | SO$_4$ | 80 | 28 | 0.68 | 0.250 | 0.22 |
| | 1A1-102O | HEART_LAKE | SO$_4$ | 80 | 31 | 0.00 | 0.500 | -0.02 |
| | 1A1-106O | DART_LAKE | SO$_4$ | 80 | 31 | -3.61 | 0.000 | -2.31 |
| | 1A1-109O | MOSS_LAKE | SO$_4$ | 80 | 31 | -2.19 | 0.014 | -1.45 |
| | 1A1-110O | LAKE_RONDAXE | SO$_4$ | 80 | 31 | -2.71 | 0.003 | -1.42 |
| | 1A2-078S | OTTER_LAKE_OUTLET | SO$_4$ | 80 | 31 | -4.00 | 0.000 | -1.71 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| U.S.A. | 1C1-101E | BRSO4H | SO$_4$ | 80 | 29 | -2.21 | 0.014 | -2.60 |
| | 1E1-132E | LITTLE_LONG_POND | SO$_4$ | 80 | 14 | -1.60 | 0.055 | -0.71 |
| | 1E1-133E | TILDEN_POND | SO$_4$ | 80 | 14 | 1.25 | 0.108 | 0.42 |
| | 2A2-063E | CRUISER | SO$_4$ | 80 | 18 | -1.80 | 0.036 | -1.36 |
| | 2B1-047E | JOHNSON | SO$_4$ | 80 | 18 | 1.08 | 0.141 | 0.90 |
| | 2B2-102E | BUCKEYE | SO$_4$ | 80 | 18 | -3.70 | 0.000 | -3.99 |
| | 2B2-106E | NEVINS | SO$_4$ | 80 | 18 | -1.08 | 0.141 | -1.93 |
| | 2B3-082E | ANDRUS | SO$_4$ | 80 | 18 | -1.59 | 0.056 | -3.28 |
| | 2C1-068E | SAND | SO$_4$ | 80 | 18 | -0.43 | 0.337 | -1.16 |
| | 2C1-069E | NICHOLS | SO$_4$ | 80 | 18 | -0.87 | 0.195 | -1.78 |
| | 2C2-062E | LUNA | SO$_4$ | 80 | 18 | -1.59 | 0.056 | -1.55 |
| | 1A1-017S | CONSTABLE_POND_OUTLET | SO$_4$ | 90 | 20 | 0.00 | 0.548 | -0.18 |
| | 1A1-052O | ARBUTUS_LAKE | SO$_4$ | 90 | 20 | -2.33 | 0.009 | -1.75 |
| | 1A1-102O | HEART_LAKE | SO$_4$ | 90 | 20 | -1.84 | 0.032 | -1.12 |
| | 1A1-106O | DART_LAKE | SO$_4$ | 90 | 20 | 0.00 | 0.548 | -0.03 |
| | 1A1-109O | MOSS_LAKE | SO$_4$ | 90 | 20 | -1.35 | 0.090 | -1.50 |
| | 1A1-110O | LAKE_RONDAXE | SO$_4$ | 90 | 20 | -0.12 | 0.452 | -0.65 |
| | 1A2-078S | OTTER_LAKE_OUTLET | SO$_4$ | 90 | 20 | -0.12 | 0.452 | -0.42 |
| | 1C1-101E | BRSO4H | SO$_4$ | 90 | 15 | -1.57 | 0.058 | -2.42 |
| | 1E1-132E | LITTLE_LONG_POND | SO$_4$ | 90 | 11 | -2.25 | 0.010 | -1.17 |
| | 1E1-133E | TILDEN_POND | SO$_4$ | 90 | 11 | -1.67 | 0.048 | -1.88 |
| | 2A2-063E | CRUISER | SO$_4$ | 90 | 14 | 0.15 | 0.440 | 0.25 |
| | 2B1-047E | JOHNSON | SO$_4$ | 90 | 14 | -3.44 | 0.000 | -5.63 |
| | 2B2-102E | BUCKEYE | SO$_4$ | 90 | 14 | -0.46 | 0.325 | -0.63 |
| | 2B2-106E | NEVINS | SO$_4$ | 90 | 14 | -2.93 | 0.001 | -6.45 |
| | 2B3-082E | ANDRUS | SO$_4$ | 90 | 14 | -3.24 | 0.000 | -6.04 |
| | 2C1-068E | SAND | SO$_4$ | 90 | 15 | -3.71 | 0.000 | -5.62 |
| | 2C1-069E | NICHOLS | SO$_4$ | 90 | 15 | -3.82 | 0.000 | -8.02 |
| | 2C2-062E | LUNA | SO$_4$ | 90 | 15 | 1.56 | 0.060 | 1.78 |

## ALK: Results of the Nonparametric Seasonal Kendall Test (SKT) for the Single ICP Waters Sites

Table 36. ALK concentrations.

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| CANADA | C01 | BATCHAWANA | ALK | 80 | 37 | -1.50 | 0.067 | -1.28 |
| | C02 | WISHART | ALK | 80 | 39 | 1.11 | 0.133 | 1.21 |
| | C03 | LITLE_TURKEY | ALK | 80 | 39 | 2.22 | 0.013 | 1.44 |
| | C04 | TURKEY | ALK | 80 | 40 | 2.82 | 0.002 | 1.66 |
| | C05 | LAC_VEILLE | ALK | 80 | 15 | 0.00 | 0.500 | 0.00 |
| | C06 | LAC_JOSSELI | ALK | 80 | 13 | 0.30 | 0.386 | 0.83 |
| | C07 | BONNEVILLE | ALK | 80 | 21 | -1.14 | 0.128 | -0.80 |
| | C08 | LAFLAMME | ALK | 80 | 32 | -1.79 | 0.036 | -2.02 |
| | C09 | LAC_MACLEO | ALK | 80 | 15 | 0.25 | 0.041 | 0.22 |
| | C10 | MOUNT_TOM_LAKE | ALK | 80 | 15 | 1.08 | 0.140 | 2.00 |
| | C12 | RED_LAKE | ALK | 80 | 15 | 0.85 | 0.196 | 1.30 |
| | C13 | KEJUMKUJIK_LAKE | ALK | 80 | 36 | 2.25 | 0.012 | 0.80 |
| | C01 | BATCHAWANA | ALK | 90 | 20 | -0.857 | 0.200 | -1.48 |
| | C02 | WISHART | ALK | 90 | 20 | -0.367 | 0.360 | -0.88 |
| | C03 | LITLE_TURKEY | ALK | 90 | 20 | -0.987 | 0.16 | -2.33 |
| | C04 | TURKEY | ALK | 90 | 20 | -2.148 | 0.015 | -3.58 |
| | C07 | BONNEVILLE | ALK | 90 | 20 | 1.35 | 0.090 | 1.00 |
| | C08 | LAFLAMME | ALK | 90 | 20 | 0.86 | 0.198 | 0.53 |
| DENMARK | SK110B | SKAERBAEK_B | ALK | 90 | 21 | -0.68 | 0.251 | -1.89 |
| | SK038F | SKAERBAEK_F | ALK | 90 | 21 | 0.57 | 0.287 | 0.63 |
| FINLAND | SF01 | HIRVILAMPI | ALK | 80 | --- | --- | --- | --- |
| | SF01 | HIRVILAMPI | ALK | 90 | 12 | 0.78 | 0.223 | 5.05 |
| | SF02 | VUORILAMPI | ALK | 90 | 12 | 0.00 | 0.500 | 2.50 |
| | SF03 | MAEKILAMPI | ALK | 90 | 13 | 2.10 | 0.015 | 12.91 |
| GERMANY | EGE1 | EGER | ALK | 80 | --- | --- | --- | --- |
| | HIN31 | HINTERER_SCHACTENBACH | ALK | 80 | --- | --- | --- | --- |
| | ROE2 | ROESLAU | ALK | 80 | --- | --- | --- | --- |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| GERMANY | SEE30 | SEEBACH | ALK | 80 | -- | -- | -- | -- |
| | TRA1 | TRAUNBACH | ALK | 80 | -- | -- | -- | -- |
| | VOR32 | VORDERER_SCHACHTENBACH | ALK | 80 | -- | -- | -- | -- |
| | ZIN2 | ZINSE | ALK | 80 | -- | -- | -- | -- |
| | DIC | DICKE_BRAMKE | ALK | 90 | 17 | 0.15 | 0.440 | 1.23 |
| | DUE6 | DUERREYCHBACH | ALK | 90 | 18 | 1.09 | 0.139 | 4.98 |
| | EGE1 | EGER | ALK | 90 | 20 | 1.84 | 0.032 | 7.94 |
| | GRA5 | GRAFENBACH | ALK | 90 | 20 | 0.95 | 0.172 | 8.99 |
| | GROS2 | GROSSE_SOESE | ALK | 90 | -- | -- | -- | -- |
| | HIN31 | HINTERER_SCHACTENBACH | ALK | 90 | -- | -- | -- | -- |
| | NIE5 | NIESTE | ALK | 90 | 18 | 2.46 | 0.006 | 34.91 |
| | ROE2 | ROESLAU | ALK | 90 | -- | -- | -- | -- |
| | ROM2 | ROMBACH | ALK | 90 | 19 | 2.74 | 0.002 | 52.43 |
| | SEE30 | SEEBACH | ALK | 90 | -- | -- | -- | -- |
| | TRA1 | TRAUNBACH | ALK | 90 | 20 | 2.82 | 0.002 | 16.46 |
| | VOR32 | VORDERER_SCHACHTENBACH | ALK | 90 | -- | -- | -- | -- |
| | ZIN2 | ZINSE | ALK | 90 | 19 | 2.74 | 0.002 | 52.43 |
| ITALY | ITA1 | PELLINO | ALK | 80 | 20 | 1.35 | 0.090 | 6.72 |
| | ITA2 | CANNOBINO | ALK | 80 | 27 | -0.87 | 0.193 | -4.38 |
| | ITA1 | PELLINO | ALK | 90 | 24 | -1.03 | 0.152 | -5.64 |
| | ITA2 | CANNOBINO | ALK | 90 | 24 | -0.47 | 0.321 | -3.40 |
| NETHERLANDS | GER | GERRITSFLES | ALK | 80 | 23 | 0.60 | 0.278 | 3.33 |
| | GER | GERRITSFLES | ALK | 90 | 19 | 3.53 | 0.000 | 16.85 |
| NORWAY | BIE01 | BIRKENES | ALK | 80 | 59 | -2.37 | 0.009 | -1.21 |
| | DALELV | DALELVA | ALK | 80 | 61 | -5.32 | 0.000 | -1.64 |
| | LAE01 | LANGTJERN | ALK | 80 | 61 | -5.32 | 0.000 | -1.64 |
| | OVE329 | VIKEDALSELVA | ALK | 80 | 22 | 0.00 | 0.541 | -0.04 |
| | OVE573 | GAULAR | ALK | 80 | 24 | -2.35 | 0.009 | -1.25 |
| | OVE901 | AURDOELA | ALK | 80 | 16 | -0.85 | 0.200 | -0.88 |
| | OVEL71 | TOVDALSELVA | ALK | 80 | 40 | 2.19 | 0.014 | 0.87 |
| | BIE01 | BIRKENES | ALK | 90 | 25 | 2.55 | 0.005 | 4.65 |
| | DALELV | DALELVA | ALK | 90 | 24 | 2.72 | 0.003 | 5.74 |
| | LAE01 | LANGTJERN | ALK | 90 | 24 | 2.72 | 0.003 | 4.13 |
| | OVE329 | VIKEDALSELVA | ALK | 90 | 25 | 3.08 | 0.001 | 3.23 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| NORWAY | OVE573 | GAULAR | ALK | 90 | 24 | 1.78 | 0.037 | 1.33 |
| | OVE901 | AURDOELA | ALK | 90 | 25 | 3.61 | 0.000 | 3.46 |
| | OVEL71 | TOVDALSELVA | ALK | 90 | 24 | 4.04 | 0.000 | 6.06 |
| SWEDEN | S01 | DALAANGERSAAN | ALK | 80 | 16 | -2.89 | 0.001 | -3.78 |
| | S04 | ANRAASEN | ALK | 80 | 15 | 0.00 | 0.500 | -0.09 |
| | S05 | TVAERINGEN | ALK | 80 | 17 | -0.27 | 0.394 | -1.79 |
| | S06 | STENSJOEN | ALK | 80 | 12 | 0.00 | 0.500 | -0.50 |
| | S08 | BRUNNSJOEN | ALK | 80 | -- | --- | --- | --- |
| | S09 | FIOLEN | ALK | 80 | 17 | -1.67 | 0.047 | -1.58 |
| | S10 | STORASJOEN | ALK | 80 | -- | --- | --- | --- |
| | S11 | FRAECKSJOEN | ALK | 80 | 16 | -0.27 | 0.394 | -2.17 |
| | S12 | HAERSVATN | ALK | 80 | 15 | -1.35 | 0.089 | -6.30 |
| | S01 | DALAANGERSAAN | ALK | 90 | 20 | 3.06 | 0.001 | 2.11 |
| | S04 | ANRAASEN | ALK | 90 | 20 | -0.37 | 0.358 | -1.02 |
| | S05 | TVAERINGEN | ALK | 90 | 24 | -2.83 | 0.002 | -5.38 |
| | S06 | STENSJOEN | ALK | 90 | 23 | 1.19 | 0.118 | 1.38 |
| | S08 | BRUNNSJOEN | ALK | 90 | 20 | 0.24 | 0.407 | 0.20 |
| | S09 | FIOLEN | ALK | 90 | 23 | 0.50 | 0.311 | 0.80 |
| | S10 | STORASJOEN | ALK | 90 | 22 | 0.83 | 0.206 | 2.98 |
| | S11 | FRAECKSJOEN | ALK | 90 | 22 | 2.21 | 0.013 | 1.78 |
| | S12 | HAERSVATN | ALK | 90 | 21 | 0.90 | 0.185 | 2.00 |
| U.K. | UK01 | LOCH_COIRE | ALK | 90 | 21 | -0.68 | 0.251 | -3.55 |
| | UK04 | LOCHNAGER | ALK | 90 | 20 | -0.62 | 0.269 | -0.40 |
| | UK07 | ROUND | ALK | 90 | 21 | 1.60 | 0.055 | 4.00 |
| | UK10 | SCOAT_TARN | ALK | 90 | 20 | 0.48 | 0.319 | 0.70 |
| | UK15 | LLYN_LLAGI | ALK | 90 | 20 | -2.86 | 0.002 | -10.10 |
| | UK21 | BLUE_LOCH | ALK | 90 | 18 | 0.42 | 0.339 | 1.71 |
| U.S.A. | 1A1-017S | CONSTABLE_POND_OUTLET | ALK | 80 | 31 | -2.84 | 0.002 | -2.94 |
| | 1A1-052O | ARBUTUS_LAKE | ALK | 80 | 28 | 0.98 | 0.166 | 1.67 |
| | 1A1-102O | HEART_LAKE | ALK | 80 | 31 | -2.58 | 0.005 | -1.93 |
| | 1A1-106O | DART_LAKE | ALK | 80 | 31 | -3.23 | 0.001 | -1.76 |
| | 1A1-109O | MOSS_LAKE | ALK | 80 | 31 | -0.39 | 0.349 | -1.03 |
| | 1A1-110O | LAKE_RONDAXE | ALK | 80 | 31 | -1.55 | 0.061 | -2.04 |
| | 1A2-078S | OTTER_LAKE_OUTLET | ALK | 80 | 31 | -3.74 | 0.000 | -3.30 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| U.S.A. | 1C1-101E | BRSO4H | ALK | 80 | 14 | 2.01 | 0.021 | 1.37 |
| | 1E1-132E | LITTLE_LONG_POND | ALK | 80 | 14 | 3.00 | 0.001 | 2.10 |
| | 1E1-133E | TILDEN_POND | ALK | 80 | 16 | -1.53 | 0.064 | -1.70 |
| | 2A2-063E | CRUISER | ALK | 80 | 17 | 0.12 | 0.454 | 0.15 |
| | 2B1-047E | JOHNSON | ALK | 80 | 18 | 2.60 | 0.004 | 2.43 |
| | 2B2-102E | BUCKEYE | ALK | 80 | -- | -- | -- | -- |
| | 2B2-106E | NEVINS | ALK | 80 | -- | -- | -- | -- |
| | 2B3-082E | ANDRUS | ALK | 80 | 18 | 0.43 | 0.337 | 0.43 |
| | 2C1-068E | SAND | ALK | 80 | 18 | 0.00 | 0.500 | 0.35 |
| | 2C1-069E | NICHOLS | ALK | 80 | 18 | 1.16 | 0.124 | 1.50 |
| | 2C2-062E | LUNA | ALK | 80 | 18 | 0.74 | 0.232 | 1.61 |
| | 1A1-017S | CONSTABLE_POND_OUTLET | ALK | 90 | 20 | -0.37 | 0.358 | -0.14 |
| | 1A1-052O | ARBUTUS_LAKE | ALK | 90 | 20 | -0.12 | 0.452 | -0.23 |
| | 1A1-102O | HEART_LAKE | ALK | 90 | 20 | -1.35 | 0.090 | -0.52 |
| | 1A1-106O | DART_LAKE | ALK | 90 | 20 | 2.82 | 0.002 | 1.22 |
| | 1A1-109O | MOSS_LAKE | ALK | 90 | 20 | 0.61 | 0.272 | 0.73 |
| | 1A1-110O | LAKE_RONDAXE | ALK | 90 | 20 | 1.59 | 0.056 | 2.51 |
| | 1A2-078S | OTTER_LAKE_OUTLET | ALK | 90 | 20 | 0.61 | 0.272 | 0.36 |
| | 1C1-101E | BRSO4H | ALK | 90 | -- | -- | -- | -- |
| | 1E1-132E | LITTLE_LONG_POND | ALK | 90 | 11 | 0.20 | 0.422 | 0.88 |
| | 1E1-133E | TILDEN_POND | ALK | 90 | 11 | 1.02 | 0.158 | 1.41 |
| | 2A2-063E | CRUISER | ALK | 90 | 14 | -0.62 | 0.270 | -0.65 |
| | 2B1-047E | JOHNSON | ALK | 90 | 14 | 0.15 | 0.440 | 1.04 |
| | 2B2-102E | BUCKEYE | ALK | 90 | 14 | -2.62 | 0.003 | -7.68 |
| | 2B2-106E | NEVINS | ALK | 90 | -- | -- | -- | -- |
| | 2B3-082E | ANDRUS | ALK | 90 | 14 | -1.70 | 0.044 | -1.60 |
| | 2C1-068E | SAND | ALK | 90 | 15 | -2.69 | 0.003 | -1.82 |
| | 2C1-069E | NICHOLS | ALK | 90 | 15 | 0.99 | 0.164 | 2.33 |
| | 2C2-062E | LUNA | ALK | 90 | 15 | -3.82 | 0.000 | -7.16 |

## SBC: Results of the Nonparametric Seasonal Kendall Test (SKT) for the Single ICP Waters Sites

Table 37. SBC concentrations.

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---------|------|-----------|----------|--------|-----|-------|-------|--------|
| CANADA | C01 | BATCHAWANA | SBC | 80 | 37 | -1.60 | 0.055 | -1.85 |
| | C02 | WISHART | SBC | 80 | 40 | -0.13 | 0.447 | -0.27 |
| | C03 | LITLE_TURKEY | SBC | 80 | 39 | 0.93 | 0.177 | 0.81 |
| | C04 | TURKEY | SBC | 80 | 40 | 1.48 | 0.070 | 2.05 |
| | C05 | LAC_VEILLE | SBC | 80 | 15 | -0.62 | 0.267 | -0.98 |
| | C06 | LAC_JOSSELI | SBC | 80 | 13 | -0.44 | 0.333 | -0.30 |
| | C07 | BONNEVILLE | SBC | 80 | 28 | -2.78 | 0.003 | -2.67 |
| | C08 | LAFLAMME | SBC | 80 | 32 | 0.06 | 0.475 | 0.08 |
| | C09 | LAC_MACLEO | SBC | 80 | 15 | -1.37 | 0.086 | -0.96 |
| | C10 | MOUNT_TOM_LAKE | SBC | 80 | 15 | -0.64 | 0.261 | -0.77 |
| | C12 | RED_LAKE | SBC | 80 | 15 | -0.74 | 0.229 | -1.79 |
| | C13 | KEJUMKUJIK_LAKE | SBC | 80 | 36 | -2.61 | 0.004 | -2.32 |
| | C01 | BATCHAWANA | SBC | 90 | 20 | -3.80 | 0.000 | -9.00 |
| | C02 | WISHART | SBC | 90 | 20 | -3.80 | 0.000 | -8.70 |
| | C03 | LITLE_TURKEY | SBC | 90 | 20 | -4.29 | 0.000 | -8.87 |
| | C04 | TURKEY | SBC | 90 | 20 | -3.06 | 0.001 | -6.29 |
| | C07 | BONNEVILLE | SBC | 90 | 20 | -1.10 | 0.137 | -0.72 |
| | C08 | LAFLAMME | SBC | 90 | 20 | -0.86 | 0.198 | -4.22 |
| DENMARK | SK110B | SKAERBAEK_B | SBC | 90 | 18 | -2.67 | 0.003 | -52.10 |
| | SK038F | SKAERBAEK_F | SBC | 90 | 18 | -2.67 | 0.003 | -42.26 |
| FINLAND | SF01 | HIRVILAMPI | SBC | 80 | --- | --- | --- | --- |
| | SF01 | HIRVILAMPI | SBC | 90 | 16 | -0.77 | 0.224 | -2.09 |
| | SF02 | VUORILAMPI | SBC | 90 | 15 | 0.15 | 0.440 | 0.64 |
| | SF03 | MAEKILAMPI | SBC | 90 | 16 | -0.31 | 0.382 | -0.70 |
| GERMANY | EGE1 | EGER | SBC | 80 | --- | --- | --- | --- |
| | HIN31 | HINTERER_SCHACTENBACH | SBC | 80 | --- | --- | --- | --- |
| | ROE2 | ROESLAU | SBC | 80 | --- | --- | --- | --- |

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| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| GERMANY | SEE30 | SEEBACH | SBC | 80 | -- | -- | -- | -- |
| | TRA1 | TRAUNBACH | SBC | 80 | 26 | -0.41 | 0.341 | -4.85 |
| | VOR32 | VORDERER_SCHACHTENBACH | SBC | 80 | -- | -- | -- | -- |
| | ZIN2 | ZINSE | SBC | 80 | -- | -- | -- | -- |
| | DIC | DICKE_BRAMKE | SBC | 90 | 17 | -1.68 | 0.046 | -24.24 |
| | DUE6 | DUERREYCHBACH | SBC | 90 | 18 | 2.18 | 0.013 | 4.53 |
| | EGE1 | EGER | SBC | 90 | -- | -- | -- | -- |
| | GRA5 | GRAFENBACH | SBC | 90 | -- | -- | -- | -- |
| | GROS2 | GROSSE_SOESE | SBC | 90 | 20 | -0.72 | 0.240 | -6.53 |
| | HIN31 | HINTERER_SCHACTENBACH | SBC | 90 | -- | -- | -- | -- |
| | NIE5 | NIESTE | SBC | 90 | 18 | 0.00 | 0.500 | -0.68 |
| | ROE2 | ROESLAU | SBC | 90 | -- | -- | -- | -- |
| | ROM2 | ROMBACH | SBC | 90 | 20 | 2.08 | 0.018 | 25.96 |
| | SEE30 | SEEBACH | SBC | 90 | -- | -- | -- | -- |
| | TRA1 | TRAUNBACH | SBC | 90 | 20 | 0.86 | 0.200 | 9.97 |
| | VOR32 | VORDERER_SCHACHTENBACH | SBC | 90 | -- | -- | -- | -- |
| | ZIN2 | ZINSE | SBC | 90 | -- | -- | -- | -- |
| ITALY | ITA1 | PELLINO | SBC | 80 | 20 | 2.33 | 0.009 | 8.48 |
| | ITA2 | CANNOBINO | SBC | 80 | 40 | -0.22 | 0.412 | -0.71 |
| | ITA1 | PELLINO | SBC | 90 | 24 | -1.97 | 0.024 | -6.54 |
| | ITA2 | CANNOBINO | SBC | 90 | 24 | -0.85 | 0.201 | -6.25 |
| NETHERLANDS | GER | GERRITSFLES | SBC | 80 | 23 | 0.20 | 0.422 | 0.86 |
| | GER | GERRITSFLES | SBC | 90 | 19 | 0.39 | 0.349 | 1.91 |
| NORWAY | BIE01 | BIRKENES | SBC | 80 | 60 | -4.49 | 0.000 | -2.86 |
| | DALELV | DALELVA | SBC | 80 | 62 | -6.06 | 0.000 | -2.18 |
| | LAE01 | LANGTJERN | SBC | 80 | 62 | -6.06 | 0.000 | -2.18 |
| | OVE329 | VIKEDALSELVA | SBC | 80 | 22 | -0.74 | 0.232 | -0.59 |
| | OVE573 | GAULAR | SBC | 80 | 24 | -2.16 | 0.015 | -1.26 |
| | OVE901 | AURDØLA | SBC | 80 | 16 | -0.85 | 0.200 | -1.60 |
| | OVEL71 | TOVDALSELVA | SBC | 80 | 40 | -1.03 | 0.152 | -0.41 |
| | BIE01 | BIRKENES | SBC | 90 | 25 | -0.09 | 0.465 | -0.10 |
| | DALELV | DALELVA | SBC | 90 | 24 | 1.78 | 0.037 | 4.21 |
| | LAE01 | LANGTJERN | SBC | 90 | 24 | -0.66 | 0.257 | -0.41 |
| | OVE329 | VIKEDALSELVA | SBC | 90 | 25 | 2.73 | 0.003 | 2.14 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| NORWAY | OVE573 | GAULAR | SBC | 90 | 24 | 1.60 | 0.055 | 0.51 |
| | OVE901 | AURDOELA | SBC | 90 | 25 | 2.90 | 0.001 | 2.74 |
| | OVEL71 | TOVDALSELVA | SBC | 90 | 24 | 3.10 | 0.001 | 4.12 |
| SWEDEN | S01 | DALAANGERSAAN | SBC | 80 | 16 | -2.55 | 0.004 | -7.67 |
| | S04 | ANRAASEN | SBC | 80 | 15 | 0.00 | 0.500 | -0.63 |
| | S05 | TVAERINGEN | SBC | 80 | 18 | -1.47 | 0.071 | -3.64 |
| | S06 | STENSJOEN | SBC | 80 | 12 | 0.00 | 0.500 | -0.27 |
| | S08 | BRUNNSJOEN | SBC | 80 | 20 | -3.06 | 0.001 | -27.18 |
| | S09 | FIOLEN | SBC | 80 | 17 | -3.57 | 0.000 | -12.64 |
| | S10 | STORASJOEN | SBC | 80 | 14 | -1.20 | 0.117 | -8.07 |
| | S11 | FRAECKSJOEN | SBC | 80 | 17 | -2.80 | 0.002 | -14.61 |
| | S12 | HAERSVATN | SBC | 80 | 15 | -1.35 | 0.089 | -5.98 |
| | S01 | DALAANGERSAAN | SBC | 90 | 20 | -0.37 | 0.358 | -0.99 |
| | S04 | ANRAASEN | SBC | 90 | 20 | -2.33 | 0.009 | -3.68 |
| | S05 | TVAERINGEN | SBC | 90 | 24 | -3.48 | 0.000 | -10.33 |
| | S06 | STENSJOEN | SBC | 90 | 23 | -2.18 | 0.014 | -1.92 |
| | S08 | BRUNNSJOEN | SBC | 90 | 23 | -1.59 | 0.056 | -20.08 |
| | S09 | FIOLEN | SBC | 90 | 23 | 0.00 | 0.500 | 0.17 |
| | S10 | STORASJOEN | SBC | 90 | 23 | 0.00 | 0.500 | 0.00 |
| | S11 | FRAECKSJOEN | SBC | 90 | 23 | 0.40 | 0.347 | 1.14 |
| | S12 | HAERSVATN | SBC | 90 | 21 | -1.13 | 0.131 | -4.68 |
| U.K. | UK01 | LOCH_COIRE | SBC | 90 | 21 | 0.00 | 0.500 | -0.08 |
| | UK04 | LOCHNAGER | SBC | 90 | 20 | -1.84 | 0.032 | -2.25 |
| | UK07 | ROUND | SBC | 90 | 21 | 0.45 | 0.328 | 2.48 |
| | UK10 | SCOAT_TARN | SBC | 90 | 20 | -0.95 | 0.172 | -0.68 |
| | UK15 | LLYN_LLAGI | SBC | 90 | 20 | -0.95 | 0.172 | -3.83 |
| | UK21 | BLUE_LOCH | SBC | 90 | 17 | -1.07 | 0.144 | -3.60 |
| U.S.A. | 1A1-017S | CONSTABLE_POND_OUTLET | SBC | 80 | 31 | 0.26 | 0.400 | 0.70 |
| | 1A1-052O | ARBUTUS_LAKE | SBC | 80 | 28 | 4.88 | 0.000 | 7.52 |
| | 1A1-102O | HEART_LAKE | SBC | 80 | 31 | 1.55 | 0.061 | 1.11 |
| | 1A1-106O | DART_LAKE | SBC | 80 | 31 | 1.68 | 0.047 | 0.90 |
| | 1A1-109O | MOSS_LAKE | SBC | 80 | 31 | 2.06 | 0.020 | 1.88 |
| | 1A1-110O | LAKE_RONDAXE | SBC | 80 | 31 | 0.52 | 0.303 | 1.31 |
| | 1A2-078S | OTTER_LAKE_OUTLET | SBC | 80 | 31 | -0.52 | 0.303 | -0.82 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---------|------|-----------|----------|--------|---|---|---|-------|
| U.S.A. | 1C1-101E | BRSO4H | SBC | 80 | 24 | 0.46 | 0.322 | 0.52 |
| | 1E1-132E | LITTLE_LONG_POND | SBC | 80 | 14 | 1.25 | 0.108 | 1.23 |
| | 1E1-133E | TILDEN_POND | SBC | 80 | 14 | 2.62 | 0.003 | 2.78 |
| | 2A2-063E | CRUISER | SBC | 80 | 13 | 1.89 | 0.028 | 1.50 |
| | 2B1-047E | JOHNSON | SBC | 80 | 13 | 1.20 | 0.117 | 1.07 |
| | 2B2-102E | BUCKEYE | SBC | 80 | 11 | 0.00 | 0.500 | -0.10 |
| | 2B2-106E | NEVINS | SBC | 80 | 13 | -3.29 | 0.000 | -15.60 |
| | 2B3-082E | ANDRUS | SBC | 80 | 14 | 0.00 | 0.560 | 0.60 |
| | 2C1-068E | SAND | SBC | 80 | 13 | 0.66 | 0.259 | 1.33 |
| | 2C1-069E | NICHOLS | SBC | 80 | 13 | 0.51 | 0.307 | 1.85 |
| | 2C2-062E | LUNA | SBC | 80 | 14 | 2.62 | 0.003 | 3.76 |
| | 1A1-017S | CONSTABLE_POND_OUTLET | SBC | 90 | 20 | -3.06 | 0.001 | -6.64 |
| | 1A1-052O | ARBUTUS_LAKE | SBC | 90 | 20 | -2.33 | 0.009 | -5.10 |
| | 1A1-102O | HEART_LAKE | SBC | 90 | 20 | -1.84 | 0.032 | -3.23 |
| | 1A1-106O | DART_LAKE | SBC | 90 | 20 | -2.08 | 0.018 | -3.53 |
| | 1A1-109O | MOSS_LAKE | SBC | 90 | 20 | -0.61 | 0.272 | -3.29 |
| | 1A1-110O | LAKE_RONDAXE | SBC | 90 | 20 | -1.35 | 0.090 | -4.50 |
| | 1A2-078S | OTTER_LAKE_OUTLET | SBC | 90 | 20 | -3.31 | 0.000 | -7.19 |
| | 1C1-101E | BRSO4H | SBC | 90 | 15 | -0.52 | 0.303 | -0.88 |
| | 1E1-132E | LITTLE_LONG_POND | SBC | 90 | 11 | 0.61 | 0.275 | 1.28 |
| | 1E1-133E | TILDEN_POND | SBC | 90 | 11 | 0.61 | 0.275 | 1.43 |
| | 2A2-063E | CRUISER | SBC | 90 | 14 | 0.15 | 0.440 | 0.98 |
| | 2B1-047E | JOHNSON | SBC | 90 | 14 | 0.00 | 0.560 | -0.19 |
| | 2B2-102E | BUCKEYE | SBC | 90 | 14 | -1.70 | 0.044 | -3.39 |
| | 2B2-106E | NEVINS | SBC | 90 | 14 | -2.62 | 0.003 | -6.71 |
| | 2B3-082E | ANDRUS | SBC | 90 | 14 | -1.70 | 0.044 | -2.01 |
| | 2C1-068E | SAND | SBC | 90 | 15 | -2.97 | 0.001 | -5.20 |
| | 2C1-069E | NICHOLS | SBC | 90 | 15 | -1.27 | 0.103 | -1.41 |
| | 2C2-062E | LUNA | SBC | 90 | 15 | -2.97 | 0.001 | -4.38 |

## $H^+$: Results of the Nonparametric Seasonal Kendall Test (SKT) for the Single ICP Waters Sites

Table 38. $H^+$ concentrations.

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| CANADA | C01 | BATCHAWANA | $H^+$ | 80 | 37 | -1.40 | 0.081 | -0.03 |
| | C02 | WISHART | $H^+$ | 80 | 40 | -2.64 | 0.004 | -0.02 |
| | C03 | LITLE_TURKEY | $H^+$ | 80 | 39 | -1.48 | 0.069 | -0.01 |
| | C04 | TURKEY | $H^+$ | 80 | 40 | -1.57 | 0.059 | -0.01 |
| | C05 | LAC_VEILLE | $H^+$ | 80 | 15 | 0.00 | 0.500 | 0.01 |
| | C06 | LAC_JOSSELI | $H^+$ | 80 | 13 | -0.45 | 0.329 | -0.14 |
| | C07 | BONNEVILLE | $H^+$ | 80 | 28 | 2.36 | 0.009 | 0.77 |
| | C08 | LAFLAMME | $H^+$ | 80 | 32 | -3.90 | 0.000 | -0.06 |
| | C09 | LAC_MACLEO | $H^+$ | 80 | 15 | 0.00 | 0.500 | 0.00 |
| | C10 | MOUNT_TOM_LAKE | $H^+$ | 80 | 15 | -0.67 | 0.252 | 0.00 |
| | C12 | RED_LAKE | $H^+$ | 80 | 15 | -1.02 | 0.154 | -0.82 |
| | C13 | KEJUMKUJIK_LAKE | $H^+$ | 80 | 36 | -2.09 | 0.018 | -0.25 |
| | C01 | BATCHAWANA | $H^+$ | 90 | 20 | -0.86 | 0.198 | -0.05 |
| | C02 | WISHART | $H^+$ | 90 | 20 | -0.86 | 0.198 | -0.01 |
| | C03 | LITLE_TURKEY | $H^+$ | 90 | 20 | -0.37 | 0.358 | 0.00 |
| | C04 | TURKEY | $H^+$ | 90 | 20 | -0.12 | 0.452 | 0.00 |
| | C07 | BONNEVILLE | $H^+$ | 90 | 20 | -2.22 | 0.012 | -1.41 |
| | C08 | LAFLAMME | $H^+$ | 90 | 20 | 0.12 | 0.452 | 0.00 |
| DENMARK | SK110B | SKAERBAEK_B | $H^+$ | 90 | 21 | -1.13 | 0.131 | -0.01 |
| | SK038F | SKAERBAEK_F | $H^+$ | 90 | 21 | 0.00 | 0.500 | -0.01 |
| FINLAND | SF01 | HIRVILAMPI | $H^+$ | 80 | 18 | 0.30 | 0.385 | 0.00 |
| | SF01 | HIRVILAMPI | $H^+$ | 90 | 19 | 1.27 | 0.102 | 0.00 |
| | SF02 | VUORILAMPI | $H^+$ | 90 | 19 | -0.51 | 0.306 | -0.17 |
| | SF03 | MAEKILAMPI | $H^+$ | 90 | 19 | -0.51 | 0.306 | -0.17 |
| GERMANY | EGE1 | EGER | $H^+$ | 80 | 24 | -0.66 | 0.257 | -0.59 |
| | HIN31 | HINTERER_SCHACTENBACH | $H^+$ | 80 | 28 | 0.00 | 0.530 | 0.01 |
| | ROE2 | ROESLAU | $H^+$ | 80 | 24 | -0.19 | 0.426 | -0.38 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| GERMANY | SEE30 | SEEBACH | H⁺ | 80 | 24 | 0.28 | 0.390 | 0.02 |
| | TRA1 | TRAUNBACH | H⁺ | 80 | 27 | -2.13 | 0.016 | -8.25 |
| | VOR32 | VORDERER_SCHACHTENBACH | H⁺ | 80 | 24 | -0.09 | 0.463 | -0.01 |
| | ZIN2 | ZINSE | H⁺ | 80 | 24 | 0.00 | 0.537 | -0.09 |
| | DIC | DICKE_BRAMKE | H⁺ | 90 | 17 | -0.15 | 0.440 | 0.00 |
| | DUE6 | DUERREYCHBACH | H⁺ | 90 | 20 | -0.12 | 0.452 | -0.21 |
| | EGE1 | EGER | H⁺ | 90 | 20 | 2.08 | 0.018 | 0.24 |
| | GRA5 | GRAFENBACH | H⁺ | 90 | 20 | 1.48 | 0.070 | 6.26 |
| | GROS2 | GROSSE_SOESE | H⁺ | 90 | 20 | -0.95 | 0.172 | -0.05 |
| | HIN31 | HINTERER_SCHACHTENBACH | H⁺ | 90 | 20 | -0.37 | 0.358 | -0.03 |
| | NIE5 | NIESTE | H⁺ | 90 | 18 | -1.37 | 0.087 | -3.26 |
| | ROE2 | ROESLAU | H⁺ | 90 | 20 | 1.59 | 0.056 | 2.71 |
| | ROM2 | ROMBACH | H⁺ | 90 | 20 | -1.59 | 0.056 | -3.53 |
| | SEE30 | SEEBACH | H⁺ | 90 | 20 | -0.12 | 0.452 | -0.03 |
| | TRA1 | TRAUNBACH | H⁺ | 90 | 20 | 1.84 | 0.032 | 9.16 |
| | VOR32 | VORDERER_SCHACHTENBACH | H⁺ | 90 | 20 | 0.00 | 0.548 | 0.00 |
| | ZIN2 | ZINSE | H⁺ | 90 | 20 | 0.12 | 0.452 | 5.00 |
| ITALY | ITA1 | PELLINO | H⁺ | 80 | 23 | 0.00 | 0.500 | 0.00 |
| | ITA2 | CANNOBINO | H⁺ | 80 | 44 | -1.56 | 0.596 | 0.00 |
| | ITA1 | PELLINO | H⁺ | 90 | 24 | 0.09 | 0.463 | 0.00 |
| | ITA2 | CANNOBINO | H⁺ | 90 | 23 | 0.00 | 0.500 | 0.00 |
| NETHERLANDS | GER | GERRITSFLES | H⁺ | 80 | 23 | 2.52 | 0.005 | 6.29 |
| | GER | GERRITSFLES | H⁺ | 90 | 19 | -1.44 | 0.076 | -2.27 |
| NORWAY | BIE01 | BIRKENES | H⁺ | 80 | 62 | 1.01 | 0.156 | 0.17 |
| | DALELV | DALELVA | H⁺ | 80 | 62 | 3.01 | 0.001 | 0.32 |
| | LAE01 | LANGTJERN | H⁺ | 80 | 62 | 3.01 | 0.001 | 0.32 |
| | OVE329 | VIKEDALSELVA | H⁺ | 80 | 22 | 0.11 | 0.459 | 0.01 |
| | OVE573 | GAULAR | H⁺ | 80 | 24 | 0.28 | 0.390 | 0.10 |
| | OVE901 | AURDØLA | H⁺ | 80 | 16 | -0.17 | 0.434 | -0.02 |
| | OVEL71 | TOVDALSELVA | H⁺ | 80 | 40 | -0.04 | 0.482 | -0.02 |
| | BIE01 | BIRKENES | H⁺ | 90 | 25 | -0.79 | 0.216 | -0.60 |
| | DALELV | DALELVA | H⁺ | 90 | 24 | -2.91 | 0.001 | -0.07 |
| | LAE01 | LANGTJERN | H⁺ | 90 | 24 | -1.97 | 0.024 | -1.50 |
| | OVE329 | VIKEDALSELVA | H⁺ | 90 | 25 | -2.37 | 0.008 | -0.44 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| NORWAY | OVE573 | GAULAR | H$^+$ | 90 | 24 | -2.35 | 0.009 | -0.32 |
| | OVE901 | AURDOELA | H$^+$ | 90 | 25 | -0.44 | 0.332 | -0.02 |
| | OVEL71 | TOVDALSELVA | H$^+$ | 90 | 24 | -3.48 | 0.000 | -1.13 |
| SWEDEN | S01 | DALAANGERSAAN | H$^+$ | 80 | 16 | 2.21 | 0.012 | 0.01 |
| | S04 | ANRAASEN | H$^+$ | 80 | 15 | 0.00 | 0.500 | 0.77 |
| | S05 | TVAERINGEN | H$^+$ | 80 | 18 | 2.35 | 0.008 | 0.02 |
| | S06 | STENSJOEN | H$^+$ | 80 | 12 | 0.00 | 0.586 | 0.00 |
| | S08 | BRUNNSJOEN | H$^+$ | 80 | 20 | -0.86 | 0.198 | -0.20 |
| | S09 | FIOLEN | H$^+$ | 80 | 17 | 0.76 | 0.225 | 0.02 |
| | S10 | STORASJOEN | H$^+$ | 80 | 14 | -0.86 | 0.199 | -0.48 |
| | S11 | FRAECKSJOEN | H$^+$ | 80 | 16 | 0.14 | 0.446 | 0.00 |
| | S12 | HAERSVATN | H$^+$ | 80 | 15 | 1.54 | 0.061 | 1.60 |
| | S01 | DALAANGERSAAN | H$^+$ | 90 | 20 | -2.33 | 0.009 | -0.01 |
| | S04 | ANRAASEN | H$^+$ | 90 | 20 | -2.57 | 0.004 | -1.83 |
| | S05 | TVAERINGEN | H$^+$ | 90 | 24 | 1.13 | 0.130 | 0.02 |
| | S06 | STENSJOEN | H$^+$ | 90 | 23 | -0.60 | 0.278 | -0.01 |
| | S08 | BRUNNSJOEN | H$^+$ | 90 | 23 | 0.99 | 0.162 | 0.10 |
| | S09 | FIOLEN | H$^+$ | 90 | 23 | -3.37 | 0.000 | -0.06 |
| | S10 | STORASJOEN | H$^+$ | 90 | 23 | 0.30 | 0.384 | 0.10 |
| | S11 | FRAECKSJOEN | H$^+$ | 90 | 23 | -1.59 | 0.056 | -0.04 |
| | S12 | HAERSVATN | H$^+$ | 90 | 21 | -1.60 | 0.055 | -1.36 |
| U.K. | UK01 | LOCH_COIRE | H$^+$ | 90 | 21 | -0.23 | 0.412 | -0.02 |
| | UK04 | LOCHNAGER | H$^+$ | 90 | 20 | 0.25 | 0.404 | 0.10 |
| | UK07 | ROUND | H$^+$ | 90 | 21 | -0.23 | 0.411 | -0.06 |
| | UK10 | SCOAT_TARN | H$^+$ | 90 | 20 | -2.28 | 0.010 | -0.56 |
| | UK15 | LLYN_LLAGI | H$^+$ | 90 | 20 | -0.49 | 0.314 | -0.35 |
| | UK21 | BLUE_LOCH | H$^+$ | 90 | 18 | -1.45 | 0.074 | -0.21 |
| U.S.A. | 1A1-017S | CONSTABLE_POND_OUTLET | H$^+$ | 80 | 31 | 0.39 | 0.349 | 0.10 |
| | 1A1-052O | ARBUTUS_LAKE | H$^+$ | 80 | 28 | -2.78 | 0.002 | -0.04 |
| | 1A1-102O | HEART_LAKE | H$^+$ | 80 | 31 | 0.65 | 0.259 | 0.02 |
| | 1A1-106O | DART_LAKE | H$^+$ | 80 | 31 | -1.81 | 0.035 | -0.23 |
| | 1A1-109O | MOSS_LAKE | H$^+$ | 80 | 31 | 0.26 | 0.398 | 0.01 |
| | 1A1-110O | LAKE_RONDAXE | H$^+$ | 80 | 31 | -0.26 | 0.398 | -0.01 |
| | 1A2-078S | OTTER_LAKE_OUTLET | H$^+$ | 80 | 31 | 1.29 | 0.099 | 0.19 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---------|------|-----------|----------|--------|----|-------|-------|-------|
| U.S.A. | 1C1-101E | BRSO4H | $H^+$ | 80 | -- | -- | -- | -- |
| | 1E1-132E | LITTLE_LONG_POND | $H^+$ | 80 | -- | -- | -- | -- |
| | 1E1-133E | TILDEN_POND | $H^+$ | 80 | -- | -- | -- | -- |
| | 2A2-063E | CRUISER | $H^+$ | 80 | 14 | -0.96 | 0.173 | -0.02 |
| | 2B1-047E | JOHNSON | $H^+$ | 80 | 21 | -1.49 | 0.068 | -0.34 |
| | 2B2-102E | BUCKEYE | $H^+$ | 80 | 14 | -2.04 | 0.020 | -0.01 |
| | 2B2-106E | NEVINS | $H^+$ | 80 | 14 | 3.50 | 0.000 | 0.05 |
| | 2B3-082E | ANDRUS | $H^+$ | 80 | 14 | 0.53 | 0.301 | 0.14 |
| | 2C1-068E | SAND | $H^+$ | 80 | 14 | -1.38 | 0.085 | -0.54 |
| | 2C1-069E | NICHOLS | $H^+$ | 80 | 14 | -2.65 | 0.003 | -0.15 |
| | 2C2-062E | LUNA | $H^+$ | 80 | 14 | -1.18 | 0.121 | -0.09 |
| | 1A1-017S | CONSTABLE_POND_OUTLET | $H^+$ | 90 | 20 | -0.12 | 0.452 | -0.05 |
| | 1A1-052O | ARBUTUS_LAKE | $H^+$ | 90 | 20 | 0.00 | 0.548 | 0.00 |
| | 1A1-102O | HEART_LAKE | $H^+$ | 90 | 20 | 0.12 | 0.452 | 0.01 |
| | 1A1-106O | DART_LAKE | $H^+$ | 90 | 20 | -3.06 | 0.001 | -0.71 |
| | 1A1-109O | MOSS_LAKE | $H^+$ | 90 | 20 | -1.59 | 0.056 | -0.02 |
| | 1A1-110O | LAKE_RONDAXE | $H^+$ | 90 | 20 | -1.10 | 0.137 | -0.11 |
| | 1A2-078S | OTTER_LAKE_OUTLET | $H^+$ | 90 | 20 | -1.35 | 0.090 | -0.46 |
| | 1C1-101E | BRSO4H | $H^+$ | 90 | -- | -- | -- | -- |
| | 1E1-132E | LITTLE_LONG_POND | $H^+$ | 90 | 11 | 0.00 | 0.500 | 0.05 |
| | 1E1-133E | TILDEN_POND | $H^+$ | 90 | 11 | -1.25 | 0.108 | -0.07 |
| | 2A2-063E | CRUISER | $H^+$ | 90 | -- | -- | -- | -- |
| | 2B1-047E | JOHNSON | $H^+$ | 90 | -- | -- | -- | -- |
| | 2B2-102E | BUCKEYE | $H^+$ | 90 | -- | -- | -- | -- |
| | 2B2-106E | NEVINS | $H^+$ | 90 | -- | -- | -- | -- |
| | 2B3-082E | ANDRUS | $H^+$ | 90 | 11 | -0.61 | 0.275 | -0.08 |
| | 2C1-068E | SAND | $H^+$ | 90 | -- | -- | -- | -- |
| | 2C1-069E | NICHOLS | $H^+$ | 90 | -- | -- | -- | -- |
| | 2C2-062E | LUNA | $H^+$ | 90 | -- | -- | -- | -- |

NO$_3$: Results of the Nonparametric Seasonal Kendall Test (SKT) for the Single ICP Waters Sites

Table 39. NO$_3$ concentrations.

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| CANADA | C01 | BATCHAWANA | NO$_3$ | 80 | 37 | 0.90 | 0.184 | 0.14 |
| | C02 | WISHART | NO$_3$ | 80 | 40 | 0.50 | 0.311 | 0.08 |
| | C03 | LITLE_TURKEY | NO$_3$ | 80 | 39 | 0.28 | 0.391 | 0.11 |
| | C04 | TURKEY | NO$_3$ | 80 | 40 | -0.13 | 0.447 | -0.02 |
| | C05 | LAC_VEILLE | NO$_3$ | 80 | 15 | 0.38 | 0.352 | 0.13 |
| | C06 | LAC_JOSSELI | NO$_3$ | 80 | 13 | 0.00 | 0.500 | 0.00 |
| | C07 | BONNEVILLE | NO$_3$ | 80 | 28 | 1.48 | 0.070 | 0.08 |
| | C08 | LAFLAMME | NO$_3$ | 80 | 32 | -3.29 | 0.000 | -0.14 |
| | C09 | LAC_MACLEO | NO$_3$ | 80 | 15 | 0.73 | 0.234 | 0.00 |
| | C10 | MOUNT_TOM_LAKE | NO$_3$ | 80 | 15 | 0.00 | 0.500 | 0.00 |
| | C12 | RED_LAKE | NO$_3$ | 80 | 15 | 0.75 | 0.227 | 0.00 |
| | C13 | KEJUMKUJIK_LAKE | NO$_3$ | 80 | 36 | 0.00 | 0.500 | 0.00 |
| | C01 | BATCHAWANA | NO$_3$ | 90 | 20 | -0.37 | 0.358 | -0.28 |
| | C02 | WISHART | NO$_3$ | 90 | 20 | -1.10 | 0.137 | -0.73 |
| | C03 | LITLE_TURKEY | NO$_3$ | 90 | 20 | -1.10 | 0.137 | -0.49 |
| | C04 | TURKEY | NO$_3$ | 90 | 20 | 0.61 | 0.272 | 0.07 |
| | C07 | BONNEVILLE | NO$_3$ | 90 | 20 | 0.00 | 0.500 | 0.00 |
| | C08 | LAFLAMME | NO$_3$ | 90 | 20 | -1.73 | 0.042 | -0.41 |
| DENMARK | SK110B | SKAERBAEK_B | NO$_3$ | 90 | 21 | -1.58 | 0.057 | -4.25 |
| | SK038F | SKAERBAEK_F | NO$_3$ | 90 | 21 | -2.49 | 0.006 | -2.99 |
| FINLAND | SF01 | HIRVILAMPI | NO$_3$ | 80 | 18 | 0.20 | 0.422 | 0.02 |
| | SF01 | HIRVILAMPI | NO$_3$ | 90 | 19 | 0.31 | 0.381 | 0.06 |
| | SF02 | VUORILAMPI | NO$_3$ | 90 | 19 | -0.90 | 0.187 | -0.18 |
| | SF03 | MAEKILAMPI | NO$_3$ | 90 | 19 | 0.30 | 0.384 | 0.07 |
| GERMANY | EGE1 | EGER | NO$_3$ | 80 | 24 | 2.35 | 0.009 | 2.28 |
| | HIN31 | HINTERER_SCHACTENBACH | NO$_3$ | 80 | 28 | 2.48 | 0.006 | 2.77 |
| | ROE2 | ROESLAU | NO$_3$ | 80 | 24 | 0.85 | 0.201 | 1.45 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| GERMANY | SEE30 | SEEBACH | $NO_3$ | 80 | 24 | 1.03 | 0.152 | 1.23 |
| | TRA1 | TRAUNBACH | $NO_3$ | 80 | 27 | 1.50 | 0.068 | 1.30 |
| | VOR32 | VORDERER_SCHACHTENBACH | $NO_3$ | 80 | 24 | -0.47 | 0.321 | -0.50 |
| | ZIN2 | ZINSE | $NO_3$ | 80 | 24 | 2.83 | 0.002 | 6.16 |
| | DIC | DICKE_BRAMKE | $NO_3$ | 90 | 17 | -3.83 | 0.000 | -19.68 |
| | DUE6 | DUERREYCHBACH | $NO_3$ | 90 | 20 | --- | --- | --- |
| | EGE1 | EGER | $NO_3$ | 90 | 20 | 1.00 | 0.160 | 1.69 |
| | GRA5 | GRAFENBACH | $NO_3$ | 90 | 20 | -1.84 | 0.032 | -16.58 |
| | GROS2 | GROSSE_SOESE | $NO_3$ | 90 | 20 | -2.62 | 0.004 | -4.36 |
| | HIN31 | HINTERER_SCHACTENBACH | $NO_3$ | 90 | 20 | -1.48 | 0.070 | -2.26 |
| | NIE5 | NIESTE | $NO_3$ | 90 | 18 | -1.37 | 0.087 | -2.38 |
| | ROE2 | ROESLAU | $NO_3$ | 90 | 20 | 0.00 | 0.500 | 0.00 |
| | ROM2 | ROMBACH | $NO_3$ | 90 | 20 | 0.37 | 0.358 | 4.26 |
| | SEE30 | SEEBACH | $NO_3$ | 90 | 20 | -2.61 | 0.004 | -2.38 |
| | TRA1 | TRAUNBACH | $NO_3$ | 90 | 20 | -2.57 | 0.004 | -4.09 |
| | VOR32 | VORDERER_SCHACHTENBACH | $NO_3$ | 90 | 20 | -1.35 | 0.090 | -2.17 |
| | ZIN2 | ZINSE | $NO_3$ | 90 | 20 | -3.46 | 0.000 | -9.22 |
| ITALY | ITA1 | PELLINO | $NO_3$ | 80 | 23 | 1.19 | 0.118 | 3.45 |
| | ITA2 | CANNOBINO | $NO_3$ | 80 | 44 | 5.10 | 0.000 | 2.26 |
| | ITA1 | PELLINO | $NO_3$ | 90 | 24 | 0.00 | 0.500 | -0.42 |
| | ITA2 | CANNOBINO | $NO_3$ | 90 | 23 | -0.47 | 0.321 | -0.25 |
| NETHERLANDS | GER | GERRITSFLES | $NO_3$ | 80 | 23 | -2.05 | 0.019 | -0.54 |
| | GER | GERRITSFLES | $NO_3$ | 90 | 19 | -1.58 | 0.057 | -0.80 |
| NORWAY | BIE01 | BIRKENES | $NO_3$ | 80 | 62 | 4.31 | 0.000 | 0.29 |
| | DALELV | DALELVA | $NO_3$ | 80 | 62 | -0.21 | 0.416 | -0.01 |
| | LAE01 | LANGTJERN | $NO_3$ | 80 | 62 | -0.21 | 0.416 | -0.01 |
| | OVE329 | VIKEDALSELVA | $NO_3$ | 80 | 22 | 1.37 | 0.086 | 0.22 |
| | OVE573 | GAULAR | $NO_3$ | 80 | 24 | 0.85 | 0.201 | 0.13 |
| | OVE901 | AURDØLA | $NO_3$ | 80 | 16 | 0.51 | 0.308 | 0.03 |
| | OVEL71 | TOVDALSELVA | $NO_3$ | 80 | 40 | 1.83 | 0.033 | 0.22 |
| | BIE01 | BIRKENES | $NO_3$ | 90 | 25 | -2.02 | 0.021 | -0.53 |
| | DALELV | DALELVA | $NO_3$ | 90 | 24 | 1.78 | 0.037 | 0.06 |
| | LAE01 | LANGTJERN | $NO_3$ | 90 | 24 | 0.09 | 0.463 | 0.01 |
| | OVE329 | VIKEDALSELVA | $NO_3$ | 90 | 25 | -0.26 | 0.397 | -0.08 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| NORWAY | OVE573 | GAULAR | $NO_3$ | 90 | 24 | 2.35 | 0.009 | 0.35 |
| | OVE901 | AURDOELA | $NO_3$ | 90 | 25 | 1.49 | 0.068 | 0.08 |
| | OVEL71 | TOVDALSELVA | $NO_3$ | 90 | 24 | 1.03 | 0.152 | 0.16 |
| SWEDEN | S01 | DALAANGERSAAN | $NO_3$ | 80 | 16 | 2.21 | 0.012 | 1.15 |
| | S04 | ANRAASEN | $NO_3$ | 80 | 15 | 1.10 | 0.137 | 0.42 |
| | S05 | TVAERINGEN | $NO_3$ | 80 | 18 | -1.61 | 0.054 | -0.23 |
| | S06 | STENSJOEN | $NO_3$ | 80 | 12 | -1.31 | 0.097 | -0.27 |
| | S08 | BRUNNSJOEN | $NO_3$ | 80 | 20 | -2.57 | 0.004 | -0.71 |
| | S09 | FIOLEN | $NO_3$ | 80 | 17 | 2.04 | 0.020 | 1.41 |
| | S10 | STORASJOEN | $NO_3$ | 80 | 14 | 0.00 | 0.500 | 0.00 |
| | S11 | FRAECKSJOEN | $NO_3$ | 80 | 17 | 1.78 | 0.037 | 0.14 |
| | S12 | HAERSVATN | $NO_3$ | 80 | 15 | 1.20 | 0.117 | 0.43 |
| | S01 | DALAANGERSAAN | $NO_3$ | 90 | 20 | -1.84 | 0.032 | -0.29 |
| | S04 | ANRAASEN | $NO_3$ | 90 | 20 | -3.55 | 0.000 | -0.89 |
| | S05 | TVAERINGEN | $NO_3$ | 90 | 24 | 0.38 | 0.354 | 0.03 |
| | S06 | STENSJOEN | $NO_3$ | 90 | 23 | 0.00 | 0.539 | 0.00 |
| | S08 | BRUNNSJOEN | $NO_3$ | 90 | 23 | -0.40 | 0.347 | -0.13 |
| | S09 | FIOLEN | $NO_3$ | 90 | 23 | -1.59 | 0.056 | -0.89 |
| | S10 | STORASJOEN | $NO_3$ | 90 | 23 | 2.60 | 0.004 | 0.38 |
| | S11 | FRAECKSJOEN | $NO_3$ | 90 | 23 | -1.19 | 0.118 | -0.18 |
| | S12 | HAERSVATN | $NO_3$ | 90 | 21 | -2.62 | 0.004 | -0.91 |
| U.K. | UK01 | LOCH_COIRE | $NO_3$ | 90 | 21 | 0.26 | 0.398 | 0.00 |
| | UK04 | LOCHNAGER | $NO_3$ | 90 | 18 | 3.01 | 0.001 | 2.17 |
| | UK07 | ROUND | $NO_3$ | 90 | 21 | 1.04 | 0.152 | 0.33 |
| | UK10 | SCOAT_TARN | $NO_3$ | 90 | 20 | -1.91 | 0.027 | -1.55 |
| | UK15 | LLYN_LLAGI | $NO_3$ | 90 | 20 | 0.12 | 0.451 | 0.00 |
| | UK21 | BLUE_LOCH | $NO_3$ | 90 | 18 | 0.57 | 0.288 | 0.83 |
| U.S.A. | 1A1-017S | CONSTABLE_POND_OUTLET | $NO_3$ | 80 | 31 | 2.84 | 0.002 | 1.22 |
| | 1A1-052O | ARBUTUS_LAKE | $NO_3$ | 80 | 28 | 4.43 | 0.000 | 0.87 |
| | 1A1-102O | HEART_LAKE | $NO_3$ | 80 | 31 | 4.23 | 0.000 | 0.95 |
| | 1A1-106O | DART_LAKE | $NO_3$ | 80 | 31 | 1.55 | 0.061 | 0.55 |
| | 1A1-109O | MOSS_LAKE | $NO_3$ | 80 | 31 | 0.52 | 0.303 | 0.19 |
| | 1A1-110O | LAKE_RONDAXE | $NO_3$ | 80 | 31 | 1.55 | 0.061 | 0.38 |
| | 1A2-078S | OTTER_LAKE_OUTLET | $NO_3$ | 80 | 31 | 3.48 | 0.000 | 1.33 |

| Country | Code | Site Name | Variable | Decade | N | Z | p | Slope |
|---|---|---|---|---|---|---|---|---|
| U.S.A. | 1C1-101E | BRSO4H | $NO_3$ | 80 | 15 | 0.00 | 0.500 | 0.55 |
| | 1E1-132E | LITTLE_LONG_POND | $NO_3$ | 80 | 14 | 0.00 | 0.500 | 0.00 |
| | 1E1-133E | TILDEN_POND | $NO_3$ | 80 | 14 | 0.00 | 0.500 | 0.00 |
| | 2A2-063E | CRUISER | $NO_3$ | 80 | 15 | 2.23 | 0.012 | 0.35 |
| | 2B1-047E | JOHNSON | $NO_3$ | 80 | 16 | 1.29 | 0.099 | 0.15 |
| | 2B2-102E | BUCKEYE | $NO_3$ | 80 | 17 | 0.00 | 0.547 | 0.00 |
| | 2B2-106E | NEVINS | $NO_3$ | 80 | 17 | 0.75 | 0.226 | 0.04 |
| | 2B3-082E | ANDRUS | $NO_3$ | 80 | 17 | 0.61 | 0.273 | 0.03 |
| | 2C1-068E | SAND | $NO_3$ | 80 | 18 | -0.11 | 0.455 | 0.00 |
| | 2C1-069E | NICHOLS | $NO_3$ | 80 | 18 | 0.00 | 0.500 | 0.00 |
| | 2C2-062E | LUNA | $NO_3$ | 80 | 17 | 0.50 | 0.308 | 0.00 |
| | 1A1-017S | CONSTABLE_POND_OUTLET | $NO_3$ | 90 | 20 | -2.08 | 0.018 | -3.33 |
| | 1A1-052O | ARBUTUS_LAKE | $NO_3$ | 90 | 20 | -2.08 | 0.018 | -1.43 |
| | 1A1-102O | HEART_LAKE | $NO_3$ | 90 | 20 | -4.20 | 0.000 | -1.73 |
| | 1A1-106O | DART_LAKE | $NO_3$ | 90 | 20 | -2.57 | 0.004 | -3.98 |
| | 1A1-109O | MOSS_LAKE | $NO_3$ | 90 | 20 | -3.31 | 0.000 | -3.09 |
| | 1A1-110O | LAKE_RONDAXE | $NO_3$ | 90 | 20 | -3.06 | 0.001 | -3.34 |
| | 1A2-078S | OTTER_LAKE_OUTLET | $NO_3$ | 90 | 20 | -3.55 | 0.000 | -2.22 |
| | 1C1-101E | BRSO4H | $NO_3$ | 90 | 15 | -0.71 | 0.242 | -0.42 |
| | 1E1-132E | LITTLE_LONG_POND | $NO_3$ | 90 | 11 | 0.00 | 0.500 | 0.00 |
| | 1E1-133E | TILDEN_POND | $NO_3$ | 90 | 11 | 0.00 | 0.586 | 0.00 |
| | 2A2-063E | CRUISER | $NO_3$ | 90 | 14 | 0.99 | 0.166 | 0.05 |
| | 2B1-047E | JOHNSON | $NO_3$ | 90 | 14 | 0.00 | 0.562 | 0.00 |
| | 2B2-102E | BUCKEYE | $NO_3$ | 90 | 14 | -0.47 | 0.321 | -0.05 |
| | 2B2-106E | NEVINS | $NO_3$ | 90 | 14 | -0.16 | 0.437 | 0.00 |
| | 2B3-082E | ANDRUS | $NO_3$ | 90 | 14 | -1.60 | 0.055 | -0.10 |
| | 2C1-068E | SAND | $NO_3$ | 90 | 15 | 0.00 | 0.556 | 0.00 |
| | 2C1-069E | NICHOLS | $NO_3$ | 90 | 15 | 0.29 | 0.387 | 0.03 |
| | 2C2-062E | LUNA | $NO_3$ | 90 | 15 | 0.60 | 0.278 | 0.03 |