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The 12-year report:

Acidification of Surface Water in Europe and North America; Trends, biological recovery and heavy metals

Norwegian Institute for Water Research

REPORT

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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, evaluate dose/response relationships and long-term trends in aquatic chemistry and biota. The aim of the 12-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 years (1996-98) related to previous findings. The report includes an evaluation of the ICP Waters database and the representativeness of ICP Waters sites, long-term trends in water chemistry at the single ICP sites and on regional scales, characterisation of invertebrates on a regional scale and an assessment of heavy metals. The current ICP Waters database includes only active sites well suited to monitor changes in acidification in response to changes in acid deposition. In the 10-year periode 1989-1998, sulphate concentrations showed significant downward trends while nitrate concentrations showed no regional patterns of change. pH and ANC and also DOC have increased in most regions during the past 10 years. Biological response, reflecting improved water quality, is positive for sites in Scandinavia. In the most acidic sites in Central Europe improvements in water quality have not reached the level at which effects on biology can be detected. Concentrations of heavy metals are generally low in ICP Waters sites not influenced by local pollution sources.

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CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

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

The 12-year report: Acidification of Surface Water in Europe and North America; Trends, biological recovery and heavy metals

Prepared by the Programme Centre
Norwegian Institute for Water Research
Oslo, March 2000

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 it`s third session in Helsinki in July 1985. 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 led by Berit Kvæven, Norwegian State Pollution Control Authority.

The work plan of the Programme includes in-depth evaluations every third year. This 12-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, representativeness of the sites in the ICP Waters database, heavy metals and effects of acidification on biota. Project manager for the 12- year report has been Brit Lisa Skjelkvåle, NIVA. Richard F. Wright, NIVA, has been responsible for the discussion on the representativeness of the ICP Waters sites. John Stoddard, EPA (USA), Tom Andersen, NIVA, and Brit Lisa Skjelkvåle are responsible for the trends work, Gunnar Raddum, Godtfred Anker Halvorsen and Einar Heergaard, University of Bergen, for the biological section, and Brit Lisa Skjelkvåle for the section on heavy metals.

Many individuals and institutions have assisted in the preparation of this report. We thank all national focal centres for submitting the necessary data to the ICP Waters Programme Centre and for contributing with valuable comments at the 13th Programme Task Force meeting in Pallanza, Italy in October 1999, where the draft report was discussed. We also thank the National Focal Centres for submitting text for use in the report on the sites in the single countries: Canada - Dean Jeffries; Estonia - Ott Roots; Finland - Jaakko Mannio; Germany - Gerhard Burkl, Bruno Kifinger; Czech Republic - Josef Veselý, Jiri Kopácek; Ireland - Jim Bowman; Italy - Rosario Mosello; Latvia - Iraida Lyulko, Polina Berg; Sweden - Anders Wilander; UK - Don Monteith, Chris Evans. We also thank Jiri Kopácek, Rosario Mosello, Chris Evans and Don Monteith for their contribution to the chapter on trends.

The final report was reviewed by an evaluation panel nominated by the Programme Task Force; Dean Jeffries (Canada), Josef Veselý (Czech Republic) and Francois Guerold (France). Their comments and work with the report are appreciated.

At NIVA we thank Liv Bente Skancke for technical assistance, and Ann Kristin Buan for database management. Merete Johannessen Ulstein (NIVA) and Berit Kvæven (SFT) provided professional assistance.

Financial support to the work of the Programme Centre was provided by the Norwegian State Pollution Control Authority (SFT).

Oslo, March 2000

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Summary

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) is designed to assess the degree and geographical extent of acidification of surface waters, evaluate dose/response relationships and long-term trends in aquatic chemistry and biota. The aim of the 12-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 years (1996-98) related to previous findings. The report includes an evaluation of the ICP Waters database and the representativeness of ICP Waters sites, long-term trends in water chemistry at the single ICP sites and on regional scales, characterisation of invertebrates on a regional scale and an assessment of heavy metals.

The current ICP Waters database includes only active sites well suited to monitor changes in acidification in response to changes in acid deposition. In the 10-year periode 1989-1998, sulphate concentrations show significant downward trends while nitrate concentrations show no regional patterns of change. pH and ANC and also DOC have increased in most regions during the past 10 years. Biological response, reflecting improved water quality, is positive for sites in Scandinavia. In the most acidified sites in Central Europe, improvements in water quality have not reached the level at which effects on biology can be detected. Concentrations of heavy metals are generally low in ICP Waters sites not influenced by local pollution sources.

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) was established in July 1985 under the Executive Body of the Convention on Long-Range Transboundary Air Pollution (LRTAP). The Programme Centre is at the Norwegian Institute for Water Research, Oslo. The main aim of the Programme is to assess, on a regional basis, the degree and geographical extent of acidification of surface waters, evaluate dose/response relationships and long-term trends in aquatic chemistry and biota.

This report deals with programme activities during the 3-year period 1996-98. Altogether 23 countries have participated in one or more of the ICP Waters activities during this period. This report covers five topics: (1) An evaluation of the ICP Waters database (2) Representativeness of ICP Waters sites (3) Trends in water chemistry at the single ICP sites and on regional scales (1989-1998) (4) Effects of acidification on aquatic fauna (invertebrates) (5) An assessment of heavy metals

The ICP Waters database

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

Representativeness of the database

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

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

Trends in water chemistry

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

All of the regions had highly significant downward trends in SO_4^{2-}. The majority of the single sites (67 out of 98) showed a significant decrease in SO_4^{2-}. Nitrate, on the other hand, showed no regional patterns of change. Central Europe did show some significant decreasing NO_3^- trends, but the heterogeneity within the region was too large to identify a regional pattern. Decreasing trends in lake water NO_3^- occurred in the so called Black Triangle while southwest Germany and Italy (Southern Alps) exhibited no or increasing NO_3^- trends.

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

The sites were also grouped according to several characteristics. The low ANC sites showed the largest rates of recovery. Neither the high NO_3^- or low NO_3^- groups of sites exhibited significant trends in NO_3^- concentrations. Non-forested sites show clear and consistent signals of recovery in ANC and pH, and appropriate (relative to SO_4^{2-} trends) rates of base cation declines. Hence, the recovery we observe is, in fact, associated with declining SO_4^{2-}.

Biology

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

Heavy metals

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

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

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

Future perspectives

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

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

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

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

1. Introduction

1.1 Programme rationale and background

Over the past 30 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, cross national boundaries, and affect surface waters, groundwater and forest soils in other countries. Rethinking of air pollution control strategies was necessary, including long-term monitoring of affected receptors such as rivers and lakes. The Convention on Long-range Transboundary Air Pollution (CLRTAP) went into effect in 1983 and was the first step to enforce emission reduction measures in the international sphere aiming at controlling air pollutant emissions in Europe and North America. Its Working Group on Effects (WGE) has aided the Convention by developing science to support its Protocols. The WGE’s six International Cooperative Programmes (on Waters, Natural Vegetation and Crops, Forests, Materials and Cultural Heritage, Integrated Monitoring, and Mapping of Critical Loads and their Exceedances) and a Joint Task Force with WHO on Human Health, quantify effects on the environment through monitoring, modelling and scientific review.

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). 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 Programme Task Force has the mandate to plan and supervise activities undertaken within the ICP Waters 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. The aims and objectives of the programme are discussed and reviewed regularly at the yearly Task Force meetings.

The monitoring programme is designed to assess, on a regional basis, the degree and geographical extent of acidification of surface waters. The data collected 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). As reported to the fourth session of the Executive Body, the ICP Waters is based on existing programmes in participating countries, implemented by voluntary contributions. The operational part of the programme using standardised methodologies started in 1987. 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 programme aims and objectives were discussed and reviewed at the 15th Task Force meeting in Pallanza, Italy October, 1999. The reviewd aims and objectives are:

Aims:

- Assess the degree and geographic extent of the impact of atmospheric pollution, in particular acidification, on surface waters;
- Collect information to evaluate dose/response relationships;
- Describe and evaluate long-term trends and variation in aquatic chemistry and biota attributable to atmospheric pollution.

Objectives:

- Maintain and develop an international network of surface water monitoring sites;
- Promote international harmonisation of monitoring practices by:
    - maintaining and updating a manual for methods and operation;
    - conducting interlaboratory quality assurance tests;
    - compiling a centralised database with data quality control and assessment capabilities.
- Develop and/or recommend chemical and biological methods for monitoring purposes;
- Report on progress according to programme aims and short term objectives as defined in the annual work programme;
- Conduct workshops on topics of central interest to the Programme Task Force and the aquatic effects research community;
- Address water related questions in cooperation with other ICP's

These topics are addressed in depth by 3-year summary reports: 1987-1989 (Wathne 1991), 1990-1992 (Skjelkvåle et al. 1994), 1993-1995 (Lükewille et al. 1997), 1996-1998 (this report).

1.3 Major Programme Findings

1.3.1 The previous 3-year period 1993-95

Trends in Water Chemistry

Significantly decreasing SO_4^{2-} 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 upward trends in NO_3^- 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-}.

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 certain sites might have been influenced by earlier changes in deposition.

Dose / Response Relationships

In general, sensitive invertebrate species were associated with high ANC and conductivity and tolerant species with high Al, low pH and ANC. The chemical parameter most strongly related to fauna composition was pH. It was possible to predict pH from invertebrate assemblages at single sites. The effect of acidification on invertebrates varied from region to region, depending on Ca^{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.

Intercalibration Exercises

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

Implications for the Assessment of Critical Loads

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

1.3.2 The current 3-year period 1996-98

Trends in Surface Water Chemistry

Trends in water chemistry indicate that SO_4^{2-} 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. NO_3^- is of increasing importance as a strong-acid anion. In the Nordic Countries (Finland, Sweden, and Norway) alkalinity decreased in the 1980s (acidification), but then increased in the 1990s (recovery). At many European sites (Italy, Germany, Netherlands, Denmark) alkalinity also increased in the 1980s, and the rate accelerated in the 1990s. Other regions (e.g. Adirondacks/Quebec, Midwestern North America, UK) show either no recovery or further acidification.

Regions with declining SO_4^{2-} that fail to show recovery in alkalinity in the 1990s (Adirondacks/Quebec, Midwestern North America) are characterised by strongly declining concentrations of sum base cations (SBC= Σ Ca, Mg, K, Na). SBC concentrations are no longer declining in the Nordic Countries. All of the hydrogen ion (H^+) trends detectable at a regional level are consistent with the alkalinity trends observed within each region.

The 1980s were characterised by increases in NO_3^- in almost all regions. These increases have generally not continued in the 1990s. Regional-scale phenomena other than nitrogen deposition may be responsible for the development in the 1990s (e.g., changes in climate or climate extremes).

Nitrogen Leaching from ICP Waters Sites

SO_4^{2-} is the most important acidifying anion at ICP Waters Sites, but NO_3^- constitutes more than 10% of the non-marine acid anions at 63% of the sites. Exceptions are sites in Canada, Finland, Sweden and Russia. More than 50% of the sites have yearly-average NO_3^- concentrations > 10 µeq/L, and 27% are > 50 µeq/L. Dutch sites have ammonium values > 100 µeq/L.

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

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. Besides N-deposition, the overall N-status of ecosystems, changes in climate or climate extremes and hydrology can strongly influence leaching of excess NO_3^- (and ammonium) from a catchment.

Effects of Acidification on Aquatic Fauna

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

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

Intercalibration Exercises

The programme conducts yearly chemical and biological intercalibrations. The number of participating laboratories have increased from 9 in 1987 to 57 laboratories in 24 countries in the 13th chemical intercomparison in 1999. The investigated variables are pH, K25 (conductivity), HCO_3^- (alkalinity), NO_3^- $+NO_2^-$, Cl^-, SO_4^{2-}, Ca^{2+}, Mg^{2+}, Na^+, K^+, Al, reactive Al, non-labile Al, DOC and COD-Mn. The intercomparison confirm that the results are generally compatible between laboratories.

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

Implications for the Assessment of Critical Loads

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

1.4 Programme Status

Table 1 summarises the participation in the different parts of the programme in the 3-year period 1996-1998. Locations of ICP sites are presented in **Figure 1**.

Table 1. *Participation in the Programme during the 3-year period 1996-1998.*

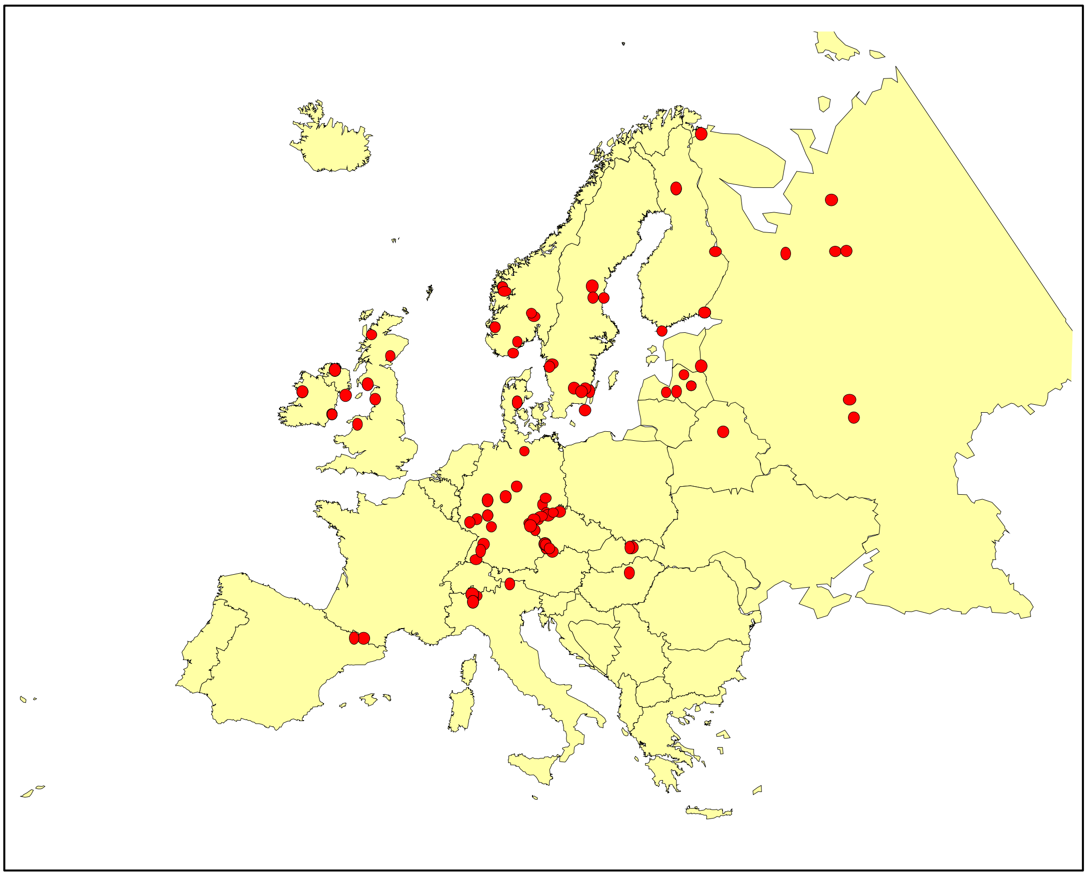
		Data contribution		Intercalibration	
	Partic. TF meetings	**Chemical data**	**Biological data**	**Chemical [1]**	**Biological**
Austria		*		*	
Belarus	*	*			
Canada	*	*		*	
Czech Rep.	*	*		*	
Croatia	*				
Denmark	*	*			
Estonia	*	*		*	
Finland	*	*		*	
France	*	*		*	
Germany	*	*	*	*	*
Hungary	*	*			
Italy	*	*		*	
Ireland	*	*	*	*	
Latvia	*	*	*	*	*
Norway	*	*	*	*	*
Poland	*	*		*	
Romania	*				
Russia	*	*		*	
Spain	*	*		*	
Sweden	*	*	*	*	*
Switzerland	*			*	
UK	*	*	*	*	
USA	*	*		*	
Total	**22**	**20**	**6**	**18**	**4**

[1] altogether 24 countries are participating in the chemical intercalibration organised by ICP Waters

1.5 Aim of the 12-year report

The aim of the 12-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 years (1996-98).

This report covers five topics:
1. An evaluation of the ICP Waters database (chapter 2)
2. Representativeness of ICP Waters sites (chapter 3)
3. Trends in water chemistry at the single ICP sites and on regional scales (chapter 4)
4. Effects of acidification on aquatic fauna (invertebrates) (chapter 5)
5. An assessment of heavy metals (chapter 6)



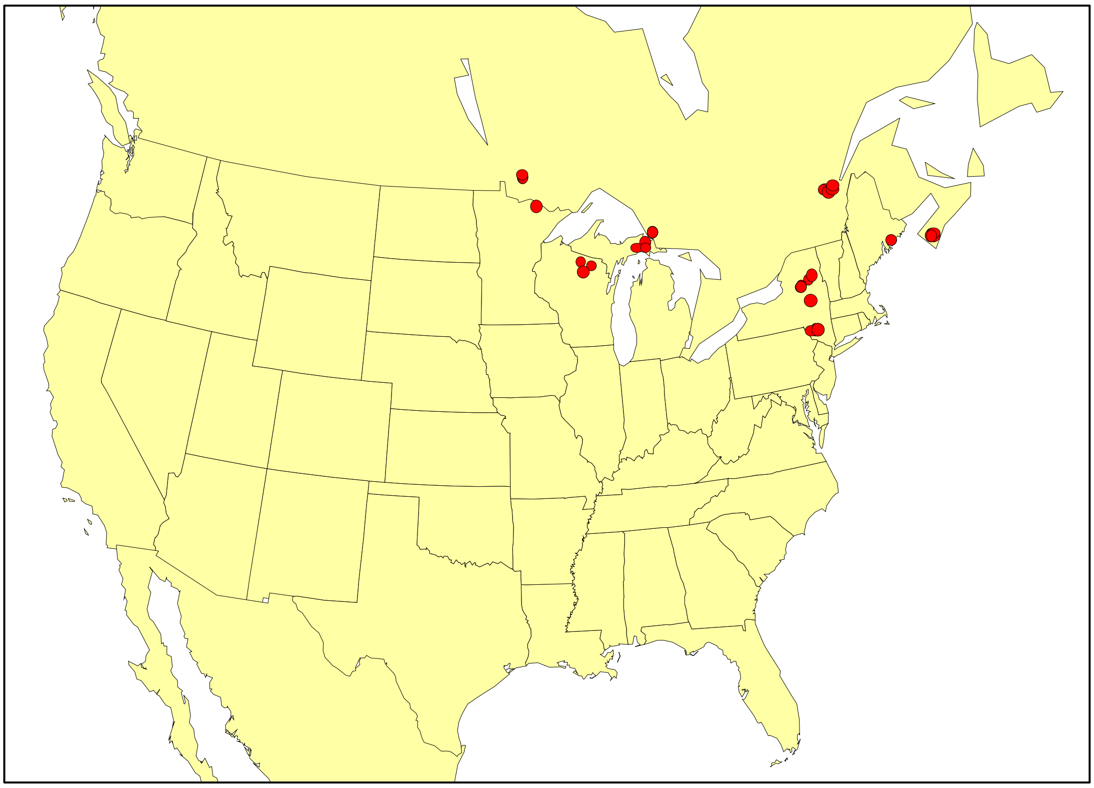


Figure 1. *Location of monitoring sites in Europe and North America as of October 1999.*

2. The ICP Waters database

The ICP Waters Programme Manual gives recommendations how to select sites with respect to sensitivity to acidic loading, regional coverage within a country, locations unaffected by local pollution etc.

The revised Programme Manual (Norwegian Institute for Water Research 1996) for the ICP Waters Programme was adopted at the 12th meeting of the Programme Task Force in 1996, Silkeborg, Denmark. The Task Force stressed the importance for all participating countries to follow the revised Programme Manual as closely as possible, especially for the selection of sites and the production and reporting of data. Experience with ICP Waters for 12 years has shown that participating countries usually deliver data from ongoing national monitoring programmes or research projects. As a result, not all countries have data that match with the requirements specified by the Programme Manual.

At the 13th meeting of the Programme Task Force in 1997, Pitlochry, Scotland, it was decided that the Programme Centre should carry out an assessment and evaluation of the database and the monitoring network with respect to the use of the data from future monitoring activities. At the 14th meeting in 1998 in Zakopane, Poland, it was decided that the work should continue and that it should focus both on the chemical and biological data.

The evaluation of the ICP Waters database and the monitoring network with respect to the use of the data from future monitoring activities is divided into two parts. The first part is the technical "cleaning" of the database and the results are presented in this chapter. The second part is a continuation of the recommendation from the report on critical loads for ICP Waters sites (Henriksen and Posch 1998). This topic is addressed in the next chapter.

2.1 Establishing a new Working database

As national monitoring programmes have changed over the years, it appears that there is a need for a refined database better suited for the work of the ICP Waters Programme. In the process of "cleaning" and streamlining the database, selected data were transferred from the old database to a new database ("Working database").

We used three criteria for selecting the sites and data to be included in the Working database:

1. Only active sites are included in the new database. By "active" we mean sites that are currently monitored and for which data from the period 1996-98 were reported to the programme centre.
2. Only the required or optional parameters (as specified in the Manual) are included. In addition we have included total organic carbon (TOC), reactive and non-labile aluminium and colorimetric analysed sulphate, when available.
3. For lakes where data for several depths have been reported, only the upper depth is included.

Data that are not transferred to the new database are still stored in the old database and can be accessed if necessary.

In the ICP Waters database, there are at present 142 sites (**Table 2**) with chemical data. In **Table 6** all the sites in the new working database are presented. Not all sites in the database have all the required background data. The missing data should, however, be available and can thus be collected.

Table 2. *Number of sites in each country*

Country	Number of sites	
	Chemistry	Biology (invertebrates)
Austria	1	*
Belarus	1	
Canada	17	
Czech Rep.	6	*
Denmark	2	
Estonia	1	
Finland	7	
France	1	*
Germany	33	33
Hungary	1	
Ireland	12	11
Italy	6	*
Latvia	5	3
Norway	9	63
Poland	2	*
Russia	10	*
Spain	1	*
Sweden	10	7
UK	6	6
USA	11	
Total	**142**	**123**

*) Sites with invertebrate available from other programmes.

2.2 Description of the sites

In this chapter we have expressed the deposition of S and N as kg or g S and N. The values should, however, only be used as indicative for the deposition level.

Austria

In Austria one site (AU03) report data to ICP Waters. This site (Schwarzsee ob Sölden) is a high mountain lake in the Tyrolian Alps, at 2799 m. The lake lies in gneiss/granitic terrain, is acid sensitive, and has a pH of about 5.6.

Belarus

The ICP Waters Programme in Belarus includes one site (BY01). It is located in the Berezinsky Biosphere Reserve in the upper part of the Berezina River. The river flows in the Verkhneberezinskaya (Upper-Berezina) depression along the western outskirts of the reserve. The catchment area of the Berezina from its head to the site BY01 is 1780 km^2. The length of the river is 55 km. The average annual precipitation is 705 mm, and average catchment runoff is 218 mm. The average long-term discharge of water in the site BY01 is 13.0 m^3/s.

In 1998 wet sulphate deposition in this area is 16 kg/ha/yr (1.6 g/m^2/yr), wet nitrate deposition is 10 kg/ha/yr (1.0 g/m^2/yr), and wet ammonium deposition is 6 kg/ha/yr (0.6 g/m^2/yr). The Verkhneberezinskaya depression is located in crystalline rocks overlain by sandstone and limestone rocks of the Upper-Proterozoic and Devonian periods. Present-day landforms include Quaternary deposits formed by Sozh (Ryss) glacier and by meltwaters from the Viurmsk glacier. Holocene deposits consist of bog, transition and fen types of peat, as well as alluvial deposits of clay and sand.

Canada

The 20 sites included in the ICP Waters program were selected from the more than 100 monitoring sites in southeastern Canada that have been used at various times to assess the aquatic effects of acidic deposition. All Canadian sites are lakes or lake outflows located in undeveloped, forested catchments. The sites are spatially distributed into 4 groups:

The 5 sites in southern Nova Scotia are within or near Kejimkujik National Park and are situated on slate-derived well-drained sandy soils, granitic tills, or peaty organic deposits. Mixed forest predominates throughout this part of Nova Scotia. The Nova Scotia sites experience a maritime climate and present wet sulphate deposition is ~5 kg/ha/yr (0.5 $g/m^2/yr$).

The 5 sites in southwestern Quebec are situated on a variety of igneous or metamorphic silicate bedrock types overlain by thin glacial deposits. The Quebec sites (like all of the remaining Canadian sites) experience a mid-continental climate and receive ~7 kg/ha/yr (0.7 $g/m^2/yr$) wet sulphate. Depending on site, the forests are either boreal or mixed types.

The 4 sites in central Ontario (Algoma region) are all located in the Turkey Lakes Watershed which has a metamorphic basalt ("greenstone") bedrock and generally thin, glacially-derived soils containing 0-2% $CaCO_3$. Hardwood forest dominates these sites and present wet sulphate deposition is ~6 kg/ha/yr (0.6 $g/m^2/yr$).

The 6 sites from northwestern Ontario are all associated with the Experimental Lakes Area (ELA) which has granodiorite bedrock overlain by patchy, thin, sandy soils. Boreal pine-spruce forest dominates the area, and wet sulphate deposition is ~1.5 kg/ha/yr (0.15 $g/m^2/yr$).

Discussion of some or all of these sites and/or the trends in water chemistry identified at them can be found in Clair et al. 1995, Jeffries et al. 1995, and Jeffries 1997. Monitoring was terminated at 2 Nova Scotia sites in 1996 and at 2 ELA sites in 1990. It will be possible to substitute new sites with approximately 20-year records for the discontinued Nova Scotia ICP sites, but the loss of ELA sites is permanent.

Czech Republic

All Czech sites are lakes located in spruce-forested catchments within or near Sumava (Bohemian Forest) National Park, 200 km or more south of major sources of emissions concentrated within the so-called Black Triangle. Present (1998) sulphate deposition is ~13 kg/ha/yr (1.3 $g/m^2/yr$). Lakes CZ03 and CZ06 are situated on granite, the rest on gneiss (mica schists), overlain by thin soils. Discussion of these sites and trends in water chemistry identified at them can be found in Veselý et al. 1998b.

Denmark

The two Danish sites (DK01 and DK02) are located in the same region in the Skaerbaek stream. The area is underlain by sand with podsolic soils (50 - 100 cm deep). The terrain surrounding station B, DK01, is covered 10% with coniferous (mainly spruce) forest, 80% with heather and grassland and 10% with wetlands and bogs. At station F, DK02, 30% of the vegetation is trees, 45% is heather and grassland and 5% is wetlands and bogs.

Estonia

The Estonian ICP Water site EE24 Ahja River (length 95 km, catchment area 1070 km^2) is located in the southeastern part of the country, in the drainage basin of Lake Peipsi. The river begins in the Otepää Upland. In the middle course it flows in the South-East Estonia Plain Region which has rolling surface topography and Devonian sandstone bedrock, covered with non-calcareous glacial till. The Ahja River has beautiful (partially protected) deep primeval valley with picturesque exposures of bedrock on the slopes. The catchment area of the Ahja River is comprised of fields 38%, forests 34%, bogs 12%, and meadows and swampy bushes

16%, respectively. The total discharge per year in the middle course of the river consists of ground water (51%), snowmelt water (27%), and rainwater (22%). In midsummer the water is cool (11-18° C) and weakly alkaline (pH 7.3-8.3). Sandy and gravely bottoms predominate in the riverbed. The river supports 30 fish species, among them brown trout (*Salmo trutta morpha fario*) and grayling (*Thymallus thymallus*).

The national hydrochemical monitoring of rivers is carried out at 58 river stations since the beginning of 1992. The biota is monitored every 5th year since 1994. Fifty-five stations in 24 rivers are sampled once per year.

Finland

The longest data record is from the three Finnish ICP sites (FI01-03) located in the southeastern corner of the country. One site is on the southwestern coast (FI07), one in inland southern Finland (FI09), one on the eastern border (FI08) and two in northern Finland (FI05-06 in central Lapland). All Finnish ICP sites are small, acid sensitive headwater lakes with thin soils in the catchment and granite/gneiss bedrock and no farmlands. All catchments are forest covered (mostly coniferous) with more open wetlands (FI05, 08) and exposed bedrock (FI01, 02,07) at some sites. Monitoring has terminated on the site FI04 and is less frequent on the site FI09, but the remaining 7 sites will be sampled 4-6 times per year.

The sulphur deposition ranges from 1.5 kg/ha/yr (0.15 $g/m^2/yr$) in Lapland to 6 kg/ha/yr (0.6 $g/m^2/yr$) (bulk S) on the southern coast. Nitrate and ammonia deposition both show similar pattern with values from < 1 kg/ha/yr (<0.1 $g/m^2/yr$) in north to 2-3 kg/ha/yr (0.2-0.3 $g/m^2/yr$) (bulk N) in south. Precipitation is on average 600-800 mm/yr in southern and central Finland and 500-600 in the north.

Site specific information of some of the lakes has been reported in, e.g., Tolonen et al. (1986) and Rask et al. (1995). Regional development of lake acidification in 171 lakes is discussed in Mannio and Vuorenmaa (1995).

France

Lake Aubé is located in southern France (Pyrénées mountains). Its elevation is 2091 m above sea level, and the main bedrock type is granite.

Germany

Germany has been involved in the ICP Waters programme from its inception in 1986. In the beginning Germany took part with 55 sampling sites within nine regions sensitive to acidification. In 1992 nine new sites in Saxony (in three regions sensitive to acidification) were added. Germany’s contribution now encompasses 33 sites (29 rivers, 2 lakes and 2 reservoirs). Most of them have been monitored since 1986, and for some sites data from as far back as 1953 are available.

The investigated areas are: Lauenburgische Seenplatte, Harz, Kaufunger Wald, Rothaargebirge, Taunus, Hunsrück, Odenwald, Schwarzwald, Ostbayerisches Grundgebirge (Bayerischer Wald, Oberpfälzer Wald, Fichtelgebirge), Erzgebirge, Elbsandsteingebirge, Sächsische Tieflandsbucht (Colditzer Forst, Dahlener Heide). The sites thus cover all main regions in Germany that are sensitive to acidification.

The sites are situated in typically acid-sensitive terrain; the predominant bedrock geology types are granite, gneiss, quartzite, phyllite and sandstone. The dominant land use is primarily coniferous forest and secondarily deciduous forest and grassland. Anthropogenic impacts are minimal in all of the watersheds. More detailed site-specific information can be found in Kifinger et al. (1998).

The German sites cover a wide range of sulphur depositon from < 20 kg/ha/yr (<2.0 g/m^2/yr) (catchment)/ < 10 kg/ha/yr (<1.0 g/m^2/yr) (open land) in some places in the Black Forest to > 100 kg/ha/yr (>10 g/m^2/yr) (catchment)/ < 20 kg/ha/yr (<2.0 g/m^2/yr) (open land) in some places in the Harz. The nitrate deposition varies between < 10 kg/ha/yr (1.0 g/m^2/yr) (catchment) (Black Forest) to < 5 kg/ha/yr(<0.5 g/m^2/yr) (open land) (Sächsische Tieflandsbucht) and > 60 kg/ha/yr (>6.0 g/m^2/yr) (catchment)/ > 10 kg/ha/yr (>1.0 g/m^2/yr) (open land) (Harz).

Over the years acidification levels have decreased in many German rivers and lakes. These reductions have occurred very slowly and not in all the monitored sites. pH levels have in fact decreased at 14% of the sampling sites. Sulphur concentrations have increased at 20% of the sites, and nitrogen concentrations at 19% of the sites.

Hungary

The elevation of Csórrét is 534 m above sea level. The catchment is 8.4 km^2. Average depth of the reservoir is 9.0 m (lake area = 0.1 km^2). Mean amount of rainfall is 650 mm/yr and runoff averages 205 mm/yr. Andesite is the predominant bedrock material, and soils are shallow (≈ 20 cm). Seventy-four percent of the catchment is covered with forest, of which more than 80% is deciduous. Fourteen percent of the area is exposed bedrock, while the rest consists of grassland, pasture, wetland and bogs.

Ireland

The three ICP Waters sampling sites in Ireland are small headwater lakes systems located in the principal acid sensitive regions of the country in Counties Donegal (Lough Veagh) and Galway (Lough Maumwee) along the western seaboard, and in County Wicklow (Glendalough Lake) on the east coast. The principal features of these lakes are listed in **Table 3**. The catchment of Lough Veagh, an uninhabited National Park, is comprised largely of heathland with small areas of deciduous forest. The lake contains brown trout/sea trout (*Salmo trutta*) and arctic char (*Salvelinus alpinus*). The uninhabited catchment of Lough Maumwee has sparse vegetation on its steep inclines; however, *Molinia* spp. and *Calluna* spp. are well established in the lower areas. Low intensity grazing by cattle and sheep is the main land use. The shallow lake contains brown trout, sea trout (*Salmo trutta*) and salmon (*Salmo salar*). The catchment of Glendalough Lake, also uninhabited National Park, contains a sizeable area of evergreen (*Picea sitchensis*) forest and mixed deciduous woodland; however the greater part of the catchment has exposed bedrock with sparse vegetation.

Typical Irish west coast deposition values for non-marine sulphur are in the range 2-5 kg/ha/yr (0.2-0.5 g/m^2/yr), nitrate < 2 kg/ha/yr (<0.2 g/m^2/yr) and ammonium 4 kg/ha/yr (0.4 g/m^2/yr). The corresponding east coast values for non-marine sulphur are 5-10 kg/ha/yr (0.5 - 1.0 g/m^2/yr) and nitrate < 4 kg/ha/yr (0.4 g/m^2/yr).

Table 3. *Some selected characteristics for the Irish ICP Waters sites.*

	Lough Veagh	Lough Maumwee	Glendalough Lake
Altitude of Lake m	43	48	133
Altitude of Lake Catchment m	43-650	48-525	133-700
Lake Surface Area km^2	2.3	0.27	0.38
Catchment Area km^2	35	4.3	18.7
Principal Rock Type	Granite	Quartz and Granite	Granite
Principal Soil Types	Podzols (75%)	Lithosols (70%)	Lithosols (70%)
	Lithosols (15%)	Blanket Peat (25%)	Blanket Peat (25%)
	Blanket Peat (10%)	Peaty Podzols (5%)	Peaty Podzols (5%)

Italy

The rivers and lakes chosen are all located in the Lake Maggiore watershed, in northwestern Italy, an area receiving high atmospheric load of pollutants; their main characteristics are reported in **Table 4**.

Lakes Paione Superiore and Inferiore are high mountain lakes situated above the timberline, in an area not affected by local disturbance or direct anthropogenic sources of pollutants. The only vegetation in their catchments is alpine meadow, limited to restricted areas; soils are absent or very thin and the geo-lithological composition is mainly acidic. As a result they are very sensitive to acidification (Mosello et al. 2000 in press). Their watershed receive from atmosphere average loads of 6, 6, 25 kg/ha/yr of N-NO3, N-NH4, S-SO4 respectively (0.6, 0.6 and 2.5 $g/m^2/yr$).

The other sites are located in the subalpine area, receiving average atmospheric loads of 14, 12, 16 kg/ha/yr of N-NO3, N-NH4, S-SO4 respectively (1.4, 1.2, 1.6 g/m2/yr). Their catchments are thinly populated and do not include any intensive industrial, stock-rearing or agricultural activity. The watershed of Lake Mergozzo is small compared to the lake surface; the northeastern part of the basin is characterised by metamorphic rocks, while the southwestern side is formed mainly by granites. About 69% of the catchment surface is covered with deciduous forest (Calderoni et al. 1978).

The land cover is quite similar in the catchments of the Rivers Pellino and Pellesino, comprised mainly of mixed deciduous forest, representing about 80% of the watershed area. Their catchments are made up only of granitic and granodioritic rocks. The River Cannobino watershed is subjected to a slight anthropogenic pressure near the villages along the shoreline of Lake Maggiore; vegetation occupies more of the watershed as the altitude rises, with coniferous forest covering about 43% of the total surface. Gneisses, micaschists and paragneiss are very common but in the upper part of the watershed basic rocks are also present (Boggero et al. 1996).

Table 4. *Some selected characteristics for the Italian ICP Waters sites.*

		Lake Paione Superiore	Lake Paione Inferiore	Lake Mergozzo	River Pellino	River Pellesino	River Cannobino
Altitude	m a.s.l.	2269	2002	194	290	290	193
Catchment area	km^2	0,50	1,26	10,43	17,5	3,4	110,4
Lake area	km^2	0,0086	0,0068	1,83	-	-	-
Mean depth	m	5,1	7,4	45,4	-	-	-
Max depth	m	11,5	13,5	73	-	-	-
Average precipitation	$mm\ y^{-1}$	1400	1450	1885	2000-2100	2000-2200	1600-2700
Renewal time	d	33	23	1716	-	-	-
Average catchment runoff	$l\ s^{-1}\ km^{-2}$	44	46	42	53	54	46
Yearly average flow	$m^3\ s^{-1}$	-	-	-	0,92	0,19	5,04

Latvia

The 5 sites included in the ICP Waters programme were selected from the more than 80 stations comprising the national monitoring network that cover water bodies having minimal local anthropogenic impact and locations in 3 major regions: Kurzeme, Zemgale and Vidzeme. Three sites are located on the rivers Barta, Liela Jugla and Tulija, one site on Lake Burtnieku, and one site represents a peat stream Zvirbuli. All the sites have long-term observation periods of 40 to 50 years. The main catchment data are listed in **Table 5**. The total S-deposition ranges from 7 kg/ha/yr (0.7 $g/m^2/yr$) in Kurzeme to 1.2 kg/ha/yr (0.12 $g/m^2/yr$) in Vidzeme; N-deposition ranges from 9 kg/ha/yr (0.9 $g/m^2/yr$) to 1.3 kg/ha/yr (0.13 $g/m^2/yr$) (EMEP data).

Table 5. *Some selected characteristics for the Latvian ICP Waters sites.*

Site data	Tulija (Zoseni)	Liela Jugla (Zaki)	Barta (Dukupji)	Zvirbuli Stream	Lake Burtnieku
Catchment area, km^2	33.4	663	1678	1.55	2220
Average precipitation, mm/yr	727	724	862	681	655
Lake area, km^2					40.2
Main type of bedrock	clay, dolomite, gypsum	clay, dolomite, gypsum	limestone, dolomite	peat	sand, rock, clay
Forest cover (total), %	44.3	51	40		51
Wetlands/Bogs, %	0	10	<5	100	
Elevation at site, m BS	176.7	12.50	10.7	8.38	39.9
Average runoff, mm.yr^{-1}	321	301	374	177	
Average depth, m	0.28-1.97	1.11-6.61	1.28-6.32	0.1-0.8	2.4

Norway

The 9 sites included in the ICP Waters program were selected from the Norwegian Monitoring Programme on Long-Range Transboundary Air Pollution (Johannessen 1995). The Norwegian monitoring programme includes 200 lakes in pristine areas sampled yearly, eight rivers sampled monthly, and 7 calibrated catchments sampled on weekly basis.

Four of the sites reporting data to ICP Waters are calibrated catchments. Birkenes, NO01, is located in the most polluted area in southern Norway with S-deposition of 5 kg/ha/yr (0.5 g/m^2/yr) and N-deposition of 15 kg/ha/yr (1.5 g/m^2/yr). The same deposition values for Storgama, NO10, located in the county of Telemark, north of Birkenes are 10 kg/ha/yr (1 g/m^2/yr) and 8 kg/ha/yr (0.8 g/m^2/yr). Langtjern, NO03, with S-deposition of 3 kg/ha/yr (0.3 g/m^2/yr) and N-deposition of 5 kg/ha/yr (0.5 g/m^2/yr), represents a typical lake in eastern Norwegian with TOC at about 10 mg C/L. Dalelva, NO04, having S-deposition of 1 kg/ha/yr (0.1 g/m^2/yr) and N-deposition of 0.5 kg/ha/yr (0.05 g/m^2/yr) is located in northern Norway on the border with Russia, and covers the effects of emissions from the Kola peninsula.

Four of the rivers are located in western Norway (Vikedalselva NO06, Gaula NO07, Nausta NO08 and Trodøla NO09). This is an area with very high precipitation and the ionic strength of the water is very low. Aurdøla NO05, is close to Langtjern and represents the rivers of eastern Norway.

Poland

Two Polish sites are lakes located in the southern part of the country within the Polish Tatra Mountains. This is one of the most sensitive aquatic ecosystems in Poland, which is a direct consequence of the specific bedrock lithology, soil type, land use and rainfall characteristics. The post-glacial lakes (about 40) of the Tatra Mts. are situated above the upper forest zone, on the level of mountain-pine and alps. The lakes Dlugi Staw (PL01) and Zielony Staw (PL02) lie close to each other and receive similar loads of acidity in precipitation. They differ in morphological and hydrological characteristics as well as in intensity of biological processes occurring in their catchments and waters. There is no forest in either catchment, only 1.5% of the catchment area of Dlugi Staw area and 6% of the catchment of Zielony Staw are covered with dwarf mountain-pine. The lake waters are ultraoligotrophic, and inhabited by phytoplankton dominated by smaller nanoplanctonic forms. Life forms in Dlugi Staw are much scarcer than in Zielony Staw; lower species diversity and lower invertebrate density have been observed. Also, there are no fish in Dlugi Staw whereas brook trout exists in Zielony Staw.

During the measurement period (1992-1998) similar deposition levels of sulphur (14 kg/ha/yr (1.4 g/m^2/yr)) and nitrogen (12 kg/ha/yr (1.2 g/m^2/yr)) were observed. Precipitation is on average 1600 mm/yr in the Tatra Mountains.

Russia

Two sets of sites are reported from Russia. Four sites (RU07-RU10) are located on the Kola peninsula, were there is heavy S-emission (and Cu and Ni) from the local smelter industry. The sites RU01-05 are slightly acidified lakes (pH 5.5.-6) situated in central parts of Russia.

Spain

There is only one ICP site in Spain, the high mountain lake Estany Redó (Lake Redó), located at 2240 m in the eastern Pyrenees Mountains. The lake is situated in granodiorite bedrock, is acid sensitive and has a pH of about 6.3. Acid deposition in this area is low. The lake has moderate concentrations of NO_3^-.

Sweden

At present ten Swedish sites are included in ICP Waters; seven lakes and three streams. They are geographically grouped with two sites in southwestern Sweden, five in the southeastern part of the country, and three in the central-eastern part. All are selected from various national monitoring programmes. These sites thus cover the range from high S-deposition (about 13 kg/ha/yr or 1.3 $g/m^2/yr$), moderate in the southwestern area (12 kg/ha/yr or 1.2 $g/m^2/yr$), and lowest in the central-eastern region (4 kg/ha/yr or 0.4 $g/m^2/yr$). pH-values range from 4.6 in Lake Härsvatten (SE12) to 6.9 in River Delångersån (SE01). The Swedish lakes are in general quite humic with a mean TOC of about 7 mg C/L. Lake Härsvatten (SE12) has the lowest concentration of the 7 lakes (< 3.0 mg C/L) while the highest concentration is found in Lake Brunnsjön (SE08). Calculations indicate that the critical loads for lakes in the southwest and the southeast are exceeded with respect to acid, while those in the central-eastern areas are not.

The streams are sampled monthly, while the lakes are sampled 4 or 8 times per year. All lakes are sampled for bottom fauna in the autumn.

UK

The six UK ICP Waters sites form part of the twenty-two site Acid Waters Monitoring Network, and have been sampled quarterly since 1988. Sites are located in Northern Scotland (UK01, UK04), Southwest Scotland (UK07), Northwest England (UK10), North Wales (UK15) and Northern Ireland (UK21). All are small glacial headwater lakes on acid-sensitive geology, with rough moorland catchments and peaty soils. Further details of all the UK Acid Waters Monitoring Network sites are available in Patrick et al. (1995).

USA

ICP Waters sites in the U.S. are located in the state of Maine in the extreme northeastern part of the country (2 lake sites), in the Adirondack and Catskill mountains of New York (7 lake and 2 stream sites, respectively), in the upper midwestern states of Wisconsin, Minnesota, and Michigan (7 lake sites), and in the mountains of Colorado in the western U.S. (4 lake sites). With the exception of the Colorado lakes, which lie above timberline (3000 m), all of the U.S. sites have watersheds that are completely forested; all of the watersheds are minimally disturbed. All of the sites lie in typically acid-sensitive terrain, with thin soils and base-poor bedrock (sandstone/silt/shale in the Catskill Mountains, granitic bedrock everywhere else). Rates of deposition in these areas range from more than 10 kg/ha/yr (1 $g/m^2/yr$) (wet S deposition) in the New York, to less than 1.5 kg/ha/yr (0.15 $g/m^2/yr$) in Colorado, with the midwestern and Maine sites falling in between these values. Characteristics and trends at the U.S. sites have previously been reported in numerous publications (Driscoll et al. 1995; Driscoll and van Dreason 1993; Murdoch and Stoddard 1993; Stoddard et al. 1998; Turk et al. 1993; Webster et al. 1993). Monitoring of the sites in Colorado and the Upper Midwest was terminated in 1995, and they will not continue as part of the ICP Waters network.

Table 6. *Sites in the ICP Waters pr. 1999 with water chemistry. Mean water chemistry are for for samples collected 1996-98. n indicates the number of observations in the 3-year period.*

Country	Dataset	ICP Site Number	Site Name	n	K25	pH	Ca	Mg	Na	K	Cl	SO_4	Alkal-inity	ANC	NH_4N	NO_3N	TOT-N	TOT-P	TOC	DOC
					mS/m		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µeq/L	µeq/L	µg N/L	µg N/L	µg N/L	µg P/L	mgC/L	mgC/L
Austria	IA-AT	AU03	Tirol, Schwarzsee ob Sölden	3	1.70	5.96	1.60	0.34	0.42	0.14	0.09	5.06	14	14	8	105		3		
Belarus	IA-BY	BY01	Berezinsky Biosphere Reserve	20	28.94	7.46	47.48	13.85	3.40	1.11	6.59	7.45	3014	3301	405	669	1087	49		
Canada	IA-CA	CA01	Ontario, Algoma Region, Batchawana Lake	148	2.15	5.98	2.18	0.36	0.43	0.19	0.31	4.57	52	38	66	278	568	5		4.37
	IA-CA	CA02	Ontario, Algoma Region, Wishart Lake	149	2.78	6.60	3.39	0.40	0.50	0.20	0.28	4.69	73	88	33	507	587	4		3.56
	IA-CA	CA03	Ontario, Algoma Region, Little Turkey Lake	147	3.22	6.76	4.18	0.43	0.53	0.20	0.31	4.84	96	130	46	465	558	4		3.54
	IA-CA	CA04	Ontario, Algoma Region, Turkey Lake	145	3.66	6.80	5.02	0.45	0.56	0.22	0.31	5.02	119	175	39	421	504	5		3.43
	IA-CA	CA05	Quebec, Lac Veilleux	2	1.17	6.05	1.18	0.22	0.38	0.15	0.15	2.10	46	46	15	40				2.45
	IA-CA	CA06	Quebec, Lac Josselin	2	1.21	5.97	1.22	0.22	0.40	0.14	0.15	2.30	45	44	15	40				3.40
	IA-CA	CA07	Quebec, Lac Bonneville	6	1.26	5.18	0.81	0.23	0.39	0.10	0.25	3.03	34	4	62	63				5.60
	IA-CA	CA08	Quebec, Lac Laflamme	29	2.11	6.39	2.21	0.52	1.01	0.23	0.34	3.10	81	124	15	72		10		5.20
	IA-CA	CA09	Quebec, Lac Macleod	2	1.06	5.11	0.66	0.17	0.30	0.08	0.15	2.30	30	7	11	40				5.15
	IA-CA	CA10	Nova Scotia, Mount Tom Lake	2	2.62	4.55	0.34	0.30	2.40	0.23	3.15	1.35	0	33		20	125			11.20
	IA-CA	CA11	Nova Scotia, Mountain Lake	5	2.15	5.40	0.47	0.28	2.48	0.20	3.42	1.68	3	26		20	82			4.00
	IA-CA	CA12	Nova Scotia, Little Red Lake	2	3.54	4.25	0.32	0.35	2.80	0.23	3.35	1.40	0	47		20	125			20.35
	IA-CA	CA13	Nova Scotia, Kejimkujik Lake	6	2.78	5.02	0.65	0.37	3.07	0.26	4.30	1.83	3	41		20	108			9.08
	IA-CA	CA14	Nova Scotia, Beaverskin Lake	25	2.05	5.48	0.31	0.30	2.48	0.22	3.63	1.85	2	11		20	64			2.33
	IA-CA	CA16	Ontario, Lake 224	8	2.04		1.86	0.47	0.73	0.36	0.19	2.93	47	107	8	1	156	2		3.24
	IA-CA	CA17	Ontario, Lake 239	5	3.20		3.06	0.93	1.23	0.60	0.42	3.72	85	209	8	5	229	3		7.26
	IA-CA	CA20	Ontario, Lake 373	8	2.51		3.09	0.72	0.87	0.44	0.24	2.55	90	203	8	2	212	3		3.86
Czech Republic	IA-CZ	CZ01	Bohemian Forest, erné	6	3.04	4.70	0.92	0.50	0.83	0.52	0.85	4.60	-55	-53	30	969				1.38
	IA-CZ	CZ02	Bohemian Forest, ertovo	6	3.71	4.41	0.57	0.37	0.67	0.36	0.66	5.42	-81	-81	75	662				2.56
	IA-CZ	CZ03	Bohemian Forest, Plešné	6	2.87	4.79	1.12	0.24	1.00	0.42	0.67	6.32	-53	-53	29	448				2.90
	IA-CZ	CZ04	Bohemian Forest, Prášilské	6	2.23	4.75	0.70	0.38	0.72	0.38	0.65	3.18	-17	-17	26	554				4.34
	IA-CZ	CZ05	Bohemian Forest, Laka	6	2.00	5.45	0.98	0.45	1.05	0.37	0.74	2.43	27	27	29	596			3.85	3.15
	IA-CZ	CZ06	Bohemian Forest, Zd´árské	6	3.27	6.05	2.30	0.56	1.90	0.52	0.92	6.55	88	88	20	92			12.45	10.80
Germany	IA-DE	DE01	Schwarzwald, Dürreychbach	23	3.39	5.47	2.08	0.79	0.87	1.18	1.84	3.66	80	20	23	1245		2		3.45
	IA-DE	DE02	Fichtelgebirge, Eger	36	6.10	5.90	2.63	1.25	5.00	0.95	9.22	4.81	51	59		789			2.41	
	IA-DE	DE03	Rothaargebirge, Elberndorfer Bach	59	6.78	7.12	3.70	3.17	2.90	0.87	2.83	13.09	175	162	45	1105				1.78

Country	Dataset	ICP Site Number	Site Name	n	K25 mS/m	pH	Ca mg/L	Mg mg/L	Na mg/L	K mg/L	Cl mg/L	SO_4 mg/L	Alkal-inity µeq/L	ANC µeq/L	NH_4N µg N/L	NO_3N µg N/L	TOT-N µg N/L	TOT-P µg P/L	TOC mgC/L	DOC mgC/L
	IA-DE	DE04	Sächsische Tieflandsbucht, Ettelsbach	19	47.65	4.62	47.20	11.11	6.66	2.75	11.64	172.68	41	-456	92	2295		14	10.41	9.58
	IA-DE	DE05	Schwarzwald, Goldersbach	23	2.71	6.63	2.61	0.74	1.29	0.35	0.75	3.46	165	122	27	583		2		4.20
	IA-DE	DE06	Hunsrück, Gräfenbach	28	9.63	4.33	5.96	2.95	3.47	0.87	5.46	25.11	33	-5	21	569		19		6.03
	IA-DE	DE07	Erzgebirge, Grosse Pyra	34	7.72	4.54	5.58	1.26	2.28	1.16	5.30	21.94	20	-175	200	1121			2.68	3.22
	IA-DE	DE08	Bayerischer Wald, Grosse Ohe	68	2.90	6.41	1.81	0.61	1.75	0.65	1.07	3.28	66	70	33	908		15	3.39	
	IA-DE	DE09	Sächsische Tieflandsbucht, Heidelbach	17	53.42	4.60	45.90	8.16	8.95	2.47	26.41	180.53	867	-1176	329	1225		26	6.20	5.31
	IA-DE	DE10	Bayerischer Wald, Hinterer Schachtenbach	29	2.47	6.28	1.85	0.52	1.67	0.58	0.97	3.12	33	38		1296				
	IA-DE	DE11	Schwarzwald, Kleine Kinzig	24	4.78	6.79	3.51	1.06	1.99	1.18	3.75	3.45	180	146	19	777		8		0.58
	IA-DE	DE12	Harz, Lange Bramke	134	5.21	6.29	3.61	1.82	1.85	0.66	2.85	11.81	66	66	76	490		50	1.06	
	IA-DE	DE13	Erzgebirge, Talsperre Neunzehnhain	2	19.05	6.25	12.90	5.50			7.20	44.20			400	2429		18		2.25
	IA-DE	DE14	Kaufunger Wald, Nieste 3	11	10.45	4.34	4.28	2.81	3.07	0.95	7.70	22.36	73	-117	87	514				
	IA-DE	DE15	Kaufunger Wald, Nieste 5	33	13.08	4.37	8.11	3.13	3.96	1.41	8.86	29.21	71	-47	83	836				
	IA-DE	DE16	Lauenburgische Seenplatte, Pinnsee	9	5.21	5.18	1.70	0.59	3.53	1.15	6.40	5.14	31	25	36	50		41	10.39	6.54
	IA-DE	DE17	Bayerischer Wald, Rachelsee	8	2.18	4.86	0.62	0.38	0.67	0.32	1.09	4.21	-67	-67	51	678		6	2.15	
	IA-DE	DE18	Fichtelgebirge, Röslau	34	4.88	5.30	2.53	0.65	3.10	0.53	1.76	11.50	35	0		541			4.69	
	IA-DE	DE19	Taunus, Rombach 2	31	12.14	4.57	4.58	2.58	6.62	1.60	12.60	16.00	70	-126	74	2883				
	IA-DE	DE20	Taunus, Rombach 3	34	7.96	6.32	6.56	2.55	3.29	0.56	8.12	10.88	215	103	73	1917				
	IA-DE	DE21	Erzgebirge, Rote Pockau	26	11.52	5.12	9.40	3.76	4.45	2.14	8.09	35.40	54	-43	3862	1469		31	4.88	5.80
	IA-DE	DE22	Odenwald, Schmerbach 3	24	10.89	4.06	3.04	2.11	1.87	1.91	6.45	16.71	69	-132	85	793				
	IA-DE	DE23	Bayerischer Wald, Seebach	29	2.32	6.38	1.49	0.61	1.53	0.53	1.17	2.95	63	56		764				
	IA-DE	DE24	Erzgebirge, Talsperre Sosa	43	9.04	4.95	5.66	1.84			4.22	23.27			42	725		23		2.67
	IA-DE	DE25	Elbsandsteingebirge, Taubenbach	16	18.39	6.78	25.61	1.87	1.80	1.68	4.08	56.56	294	145	20	1625				4.35
	IA-DE	DE26	Hunsrück, Traunbach 1	31	6.77	4.33	2.15	1.53	4.58	0.73	9.10	8.16	45	-9	14	476		20		6.62
	IA-DE	DE27	Bayerischer Wald, Vorderer Schachtenbach	29	2.74	6.51	2.14	0.61	2.07	0.58	1.15	3.69	91	96		789				
	IA-DE	DE28	Oberpfälzer Wald, Waldnaab 2	6	4.3	6.07	7.76	1.68	3.11	0.88	2.75	7.16	464	259	61	1449		24	4.28	4.13
	IA-DE	DE29	Oberpfälzer Wald, Waldnaab 8	6	6.78	4.83	7.63	1.36	3.82	0.87	2.82	17.05	994	181	46	924		348	9.38	8.15
	IA-DE	DE30	Erzgebirge, Wilde Weisseritz	37	9.98	6.30	8.81	1.93	2.26	1.47	3.84	29.16	117	-94	63	1574		91		4.94
	IA-DE	DE31	Erzgebirge, Wolfsbach	29	18.13	7.03	17.28	6.41	9.87	2.50	15.93	35.89	323	426	154	3641		134	4.04	3.99
	IA-DE	DE32	Rothaargebirge, Zinse	59	5.89	6.79	3.49	2.57	2.58	0.85	2.56	11.69	160	146	44	802				1.67
	IA-DE	DE33	Fichtelgebirge, Zinnbach	33	9.79	4.07	3.84	0.91	2.80	0.86	1.72	22.77	59	-185		1010			4.51	

Country	Dataset	ICP Site Number	Site Name	n	K25	pH	Ca	Mg	Na	K	Cl	SO_4	Alkal-inity	ANC	NH_4N	NO_3N	TOT-N	TOT-P	TOC	DOC
					mS/m		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µeq/L	µeq/L	µg N/L	µg N/L	µg N/L	µg P/L	mgC/L	mgC/L
Denmark	IA-DK	DK01	Sepstrup Sande, Skaerbaek, Station B	26	10.43	5.55	5.66	2.06	8.41	0.91	15.45	14.66	22	15	6	1189	1306	11		
	IA-DK	DK02	Sepstrup Sande, Skaerbaek, Station F	25	13.87	6.69	9.48	3.11	9.22	0.97	17.46	18.29	101	91	7	2668	2881	8		
Estonia	IA-EE	EE01	River Ahja, Kiidjärve	17	43.88	7.98	60.82	15.61	4.35	1.65	7.03	13.96	3993	3999	41	961	1423	54		
Spain	IA-ES	ES01	Lake Redó	12	1.02	6.38	1.46	0.09	0.29	0.04	0.26	1.27	16	50	30	150	255	3	1.16	
Finland	IA-FI	FI01	Hirvilampi	16	2.93	5.17	1.68	0.48	1.40	0.47	1.33	7.76	0	-6	36	45	258	6	2.53	
	IA-FI	FI02	Vuorilampi	16	3.16	5.54	1.99	0.55	1.74	0.71	1.56	7.23	5	40	105	70	469	7	5.36	
	IA-FI	FI03	Mäkilampi	15	2.64	5.57	2.18	0.40	0.98	0.47	1.08	6.17	0	35	42	37	333	8	4.73	
	IA-FI	FI05	Lapland, Suopalampi	11	1.30	6.13	0.82	0.31	1.12	0.16	0.52	1.30	15	75	12	30	290	10	6.07	
	IA-FI	FI06	Lapland, Vasikkajärvi	13	1.60	5.76	0.93	0.13	0.56	0.17	0.55	2.01	0	25	11	38	150	4	1.34	
	IA-FI	FI07	Vusimaa, Vitsjön	18	3.89	6.34	2.27	0.97	2.77	0.61	3.21	7.38	13	82	23	38	323	7	4.76	
	IA-FI	FI08	N-Karelia,Kakkisenlampi	22	1.08	5.10	0.32	0.10	0.36	0.18	0.30	1.92	0	-5	21	17	169	7	1.91	
France	IA-FR	FR01	Pyrenees, Lake Aubé	1	0.60	5.71	0.48	0.07	0.69	0.12	0.21	0.96	29	29	14	112				
Hungary	IA-HU	HU01	Matra Mountains, Csórrét Reservoir	7	23.74	7.43	22.44	7.50			26.14	26.17	743		90	2057				
Ireland	IA-IE	IE01	Wicklow, Glendalough, Lake Upper, Mid L.	6	4.63	5.95	1.45	0.67	4.05	0.31	6.18	3.44	3	53	23	169	273			
	IA-IE	IE02	Wicklow, Glendalough, Lake Upper, Inflow 1	6	5.44	6.76	2.48	0.77	4.03	0.35	5.25	4.47	16	119	12	166	275			
	IA-IE	IE03	Wicklow, Glendalough, Lake Upper, Inflow 2	6	6.93	7.11	3.91	1.29	4.63	0.32	6.65	6.17	21	171	10	334	538			
	IA-IE	IE04	Wicklow, Glendalough, Lake Upper, Inflow 3	6	5.43	5.24	0.93	0.71	5.37	0.25	8.25	3.57	2	23	11	201	332			
	IA-IE	IE05	Galway, Lough Maumwee, Mid Lake	7	7.39	6.31	1.75	1.11	8.61	0.51	13.79	3.13	3	105	14	94	99			
	IA-IE	IE06	Galway, Lough Maumwee, Inflow 1	7	7.03	6.44	2.01	1.05	7.95	0.68	11.74	3.01	5	152	13	51	59			
	IA-IE	IE07	Galway, Lough Maumwee, Inflow 2	7	7.46	6.38	1.76	1.09	8.74	0.40	13.89	3.21	4	104	11	59	94			
	IA-IE	IE08	Donegal, Lough Veagh, Mid Lake	6	6.64	6.54	1.55	1.15	7.97	0.63	12.28	2.77	3	124	24	80	76			
	IA-IE	IE09	Donegal, Lough Veagh, Inflow 1	5	7.12	6.48	1.51	1.25	9.09	0.53	13.56	2.58	6	148	12	39	32			
	IA-IE	IE10	Donegal, Lough Veagh, Inflow 2	6	5.98	5.54	1.20	0.98	7.47	0.46	10.58	2.34	0	128	12	22	25			
	IA-IE	IE11	Donegal, Lough Veagh, Inflow 3	6	6.28	6.27	1.54	1.04	7.67	0.55	11.37	2.48	7	135	10	34	46			
	IA-IE	IE12	Donegal, Lough Veagh, Inflow 4	6	7.56	6.73	2.06	1.33	8.99	0.53	13.48	3.04	11	168	9	73	89			
Italy	IA-IT	IT01	Piemonte, Lake Paione Inferiore	16	1.32	6.49	1.37	0.14	0.32	0.36	0.14	2.11	4	29	7	362		2		
	IA-IT	IT02	Piemonte, Lake di Mergozzo	3	5.56	6.84	5.40	1.37	1.99	0.93	1.29	9.04	199	222	4	644	791	4		
	IA-IT	IT03	Piemonte, Lake Paione Superiore	25	1.04	5.83	0.84	0.10	0.24	0.27	0.16	1.71	0	2	34	352		3		
	IA-IT	IT04	Piemonte, River Cannobino	36	4.57	7.36	4.11	1.43	1.67	0.71	0.91	5.53	196	225	7	665	784	6		
	IA-IT	IT05	Piemonte, River Pellino	35	5.29	7.27	4.47	1.11	3.28	0.50	1.82	4.68	178	209	8	1560	1827	13		
	IA-IT	IT06	Piemonte, River Pellesino	35	4.73	7.22	3.50	0.76	3.61	0.57	2.09	3.34	140	171	33	1542	1838	34		

Country	Dataset	ICP Site Number	Site Name	n	K25	pH	Ca	Mg	Na	K	Cl	SO_4	Alkal-inity	ANC	NH_4N	NO_3N	TOT-N	TOT-P	TOC	DOC
					mS/m		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µeq/L	µeq/L	µg N/L	µg N/L	µg N/L	µg P/L	mgC/L	mgC/L
Latvia	IA-LV	LV01	Burtnieku Lake, hydrosite	10	37.43	7.92	53.24	13.28	3.53	1.94	7.12	29.07	3044	3238	74	1023	1283	18		
	IA-LV	LV02	Barta, Dukupji	22	46.95	7.93	66.91	13.23	5.66	2.56	9.14	31.89	3062	3880	119	1064	2094	41		
	IA-LV	LV03	Liela Jugla, Zaki	20	45.89	8.06	68.43	15.82	3.91	2.36	7.37	53.95	3514	3732	82	1349	2496	39		
	IA-LV	LV04	Tulija, Zoseni	38	36.43	7.96	53.29	13.21	3.16	1.87	5.83	28.45	3139	3261	197	642	1785	23		
	IA-LV	LV05	Zvirbuli stream, hydrosite	11	6.22	4.34	2.42	0.65	1.38	0.83	4.59	7.15	0	41	306	95		22		
Norway	IA-NO	NO01	Aust-Agder, Birkenes	157	3.48	4.79	0.97	0.31	2.92	0.09	4.55	4.53	1	-27		96	293	5	5.62	
	IA-NO	NO03	Buskerud, Langtjern	137	1.58	4.98	1.09	0.17	0.60	0.10	0.51	2.14	3	37		19	288	5	11.22	
	IA-NO	NO04	Finnmark, Dalelv	157	3.81	6.22	1.56	0.89	3.66	0.27	6.06	4.87	39	43		22	130	2	3.20	
	IA-NO	NO05	Oppland, Aurdøla	39	1.41	6.31	1.34	0.21	0.75	0.27	0.73	2.18	42	54		43	231	3	3.75	
	IA-NO	NO06	Rogaland, Vikedalselva	41	2.02	5.78	0.74	0.33	1.90	0.20	3.30	1.95	11	6		161	230	3	1.14	
	IA-NO	NO07	Sogn og Fjordane, Gaula	37	1.17	5.80	0.44	0.17	1.01	0.22	1.71	1.03	11	9		90	172	4	1.29	
	IA-NO	NO08	Sogn og Fjordane, Nausta	58	1.39	5.95	0.54	0.23	1.29	0.28	2.26	1.05	18	18		70	156	6	1.58	
	IA-NO	NO09	Sogn og Fjordane, Trodøla	156	1.52	5.66	0.37	0.24	1.54	0.23	2.69	1.11	8	6		75	143		1.47	
	IA-NO	NO10	Telemark, Storgama	149	1.60	4.84	0.56	0.10	0.70	0.06	0.90	2.09	0	-7		80	321	4	5.37	
Poland	IA-PL	PL01	Tatra Mountains, Dlugi Staw Gasienicowy	29	2.05	5.76	1.91	0.13	0.34	0.12	0.25	3.07	18	5	36	676	743		0.31	
	IA-PL	PL02	Tatra Mountains, Zielony Staw Gasienicowy	28	2.34	6.61	2.64	0.20	0.36	0.20	0.29	3.08	76	67	60	427	600		0.45	
Russia	IA-RU	RU01	Shidmozero	1	1.60	5.50	2.10	0.70	1.60	0.30	1.30	8.30	22	28	12	30				4.10
	IA-RU	RU02	Yamozero	1	1.90	5.60	1.20	0.30	0.30	0.10	0.50	2.90	9	14	27	165				3.60
	IA-RU	RU03	Lum	1	2.20	5.70	2.70	0.40	0.90	0.50	1.20	7.70	18	24	0	16				3.30
	IA-RU	RU04	Soozero	1	3.10	6.20	1.10	0.20	0.60	0.20	0.50	2.40	20	37	0	16				1.90
	IA-RU	RU05	Dubovoye	1	1.30	5.50	2.00	0.70	1.00	0.40	0.90	6.60	15	46	13	29				4.20
	IA-RU	RU06	Ivan	1	2.30	5.70	1.50	0.90	1.30	0.30	2.50	5.90	27	20	11	0				3.20
	IA-RU	RU07	Chuna, Kola peninsula	2																
	IA-RU	RU08	Svetloe, Kola peninsula	1																
	IA-RU	RU09	Arkashkino, Kola peninsula	1																
	IA-RU	RU10	Glubokoe, Kola peninsula	1																
Sweden	IA-SE	SE01	Delångersån Iggesund	33	4.92	6.97	3.85	1.25	2.90	0.74	3.00	4.94	244	245	7	109	484	9	5.95	
	IA-SE	SE02	Alsterån Getebro	36	8.48	6.76	6.72	1.68	5.96	0.90	8.24	11.18	282	283	19	111	800	15	11.58	
	IA-SE	SE03	Alsterån Strömsborg	36	9.55	6.65	7.32	1.65	7.86	0.97	9.08	12.70	333	334	25	174	946	18		
	IA-SE	SE05	Tväringen	12	2.60	6.65	2.46	0.61	1.22	0.47	0.75	2.37	166	167	8	18	365	6	8.08	
	IA-SE	SE06	Stensjön	22	1.94	6.45	1.38	0.40	1.25	0.30	0.85	2.34	90	90	10	17	370	7	6.56	

Country	Dataset	ICP Site Number	Site Name	n	K25	pH	Ca	Mg	Na	K	Cl	SO_4	Alkal-inity	ANC	NH_4N	NO_3N	TOT-N	TOT-P	TOC	DOC
					mS/m		mg/L	mg/L	mg/L	mg/L	mg/L	mg/L	µeq/L	µeq/L	µg N/L	µg N/L	µg N/L	µg P/L	mgC/L	mgC/L
	IA-SE	SE08	Brunnsjön	24	6.41	5.44	3.98	1.46	4.54	0.78	5.71	10.85	139	139	27	147	779	12	21.04	
	IA-SE	SE09	Fiolen	24	6.26	6.50	3.63	1.25	4.30	1.17	7.37	10.18	78	78	13	45	506	10	5.79	
	IA-SE	SE10	Storasjö	12	3.64	5.58	1.66	0.67	3.01	0.41	3.98	4.39	73	73	33	36	594	11	11.78	
	IA-SE	SE11	Fräcksjön	24	7.51	6.43	4.01	1.31	6.93	0.73	11.26	8.60	126	126	14	73	569	9	9.10	
	IA-SE	SE12	Härsvatten	20	6.37	4.62	0.78	0.83	6.12	0.45	10.59	6.58	-60	-60	56	124	403	4	2.34	
United Kingdom	IA-UK	UK01	Scotland, Loch Coire nan Arr	9	3.94	6.34	0.87	0.73	5.19	0.30	9.01	1.88	41	41		44	156			2.90
	IA-UK	UK04	Scotland, Lochnagar	9	2.31	5.22	0.60	0.41	2.18	0.22	3.23	2.66	-4	-5		310	355			1.37
	IA-UK	UK07	Scotland, Round Loch of Glenhead	9	3.34	4.91	0.61	0.50	3.38	0.32	5.72	2.93	-10	-8		175	312			3.30
	IA-UK	UK10	England, Scoat Tarn	9	3.29	5.04	0.61	0.58	3.49	0.26	6.02	2.88	-23	-19		362	430			1.32
	IA-UK	UK15	Wales, Llyn Llagi	9	2.45	5.50	1.01	0.55	3.58	0.13	6.20	2.79	7	9		177	286			2.78
	IA-UK	UK21	N.Ireland, Blue Lough	9	5.59	4.74	0.74	0.77	6.24	0.59	10.28	4.38	-34	-32		521	726			4.06
United States	IA-US	US05	Maine, Little Long Pond	2	2.17	5.67	0.80	0.30	2.30	0.30	3.35	3.20	11	11		3				0.00
	IA-US	US06	Maine, Tilden Pond	2	2.22	6.06	1.10	0.35	2.25	0.30	3.10	2.65	46	46		11				0.00
	IA-US	US11	New York, Adirondack Mnt., Arbutus	24	2.59	6.42	2.76	0.48	0.68	0.29	0.35	5.60	79	79	6	129				5.04
	IA-US	US12	New York, Adirondack Mnt., Constable	24	2.49	5.11	1.69	0.30	0.54	0.33	0.34	5.38	-10	-10	9	414				4.81
	IA-US	US13	New York, Adirondack Mnt., Dart Lake	24	2.24	5.34	1.70	0.30	0.58	0.33	0.38	5.08	3	3	7	343				4.33
	IA-US	US14	New York, Adirondack Mnt., Heart Lake	24	1.86	6.32	2.07	0.26	0.50	0.10	0.26	4.03	47	48	8	142				2.68
	IA-US	US15	New York, Adirondack Mnt., Lake Rondaxe	24	2.30	6.03	2.04	0.37	0.70	0.34	0.38	4.98	35	35	8	311				3.85
	IA-US	US16	New York, Adirondack Mnt., Moss Lake	23	2.70	6.33	2.52	0.46	0.81	0.40	0.41	5.25	63	63	6	352				3.81
	IA-US	US17	New York, Adirondack Mnt., Otter Lake	24	2.13	5.44	1.48	0.33	0.64	0.20	0.36	5.42	-5	-5	4	226				2.37
	IA-US	US23	New York, Catskill Mnt., E. Branch Neversink, Headwaters	18	1.99	5.03	1.29	0.42	0.31	0.28	0.53	4.55	-6	-7		237				3.43
	IA-US	US24	New York, Catskill Mnt., Rondout Creek	15	2.08	4.70	0.95	0.41	0.25	0.23	0.43	4.17	-13	-14		187				5.33

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3. Representativeness of the ICP Waters sites

The aims of the ICP Waters programme are to assess the degree and geographic extent of acidification of surface waters (Norwegian Institute for Water Research 1996):

- collect information to evaluate dose/response relationships
- define long-term trends and variations in aquatic chemistry and biota attributable to atmospheric pollution

To fulfil these aims the ICP-Water programme has a 3-level hierarchy for data collection (Johannessen and Skjelkvåle 1997):

- small catchments with monthly or seasonal sampling (level I)
- lakes or streams sites with minimum annual sampling (level II)
- regional surveys of lakes and/or streams (level III)

Data from sites in the first 2 levels are held in the ICP Waters database (chemical data at NIVA, biological data at University of Bergen). These sampling localities comprise the ICP Waters sites (**Table 6**). The primary data from regional surveys are not held in the ICP Waters database, but the Programme Centre tries to maintain an accurate inventory of such surveys (**Table 7**).

The ultimate rationale for the ICP Waters programme is to monitor changes in acidification, in particular changes resulting from implementation of the Protocols under the Convention on Long-Range Transported Air Pollution. Thus ICP Waters should provide “ground truth” to document the acidification and the recovery from acidification of surface waters in areas impacted by long-range transported air pollutants.

For this reason many of the sites included in the ICP-Water programme are especially sensitive to acidification. They need not be representative of all the surface waters in a region, but rather represent the acid-sensitive surface waters.

This same reasoning is largely used in the setting of emission ceilings under the Protocols, in particular the Second Sulphur Protocol and the Multi-Pollutant Multi-Effect Protocol currently under negotiation. Both these are based on the critical loads concept. The goal is to protect 95% of all the ecosystems within each EMEP grid square. In practice this means that the target load for acid deposition within each EMEP square is set such that the critical load is exceeded for only 5% of the ecosystems. Henriksen and Posch (1998) have calculated the critical load for each of the European ICP-Water sites and compared this with the distribution of critical loads for the ecosystems within the EMEP grid (**Figure 2**). This comparison shows that while some of the sites are more sensitive than the 5th-percentile (i.e., have lower critical load), some of the sites are much less sensitive.

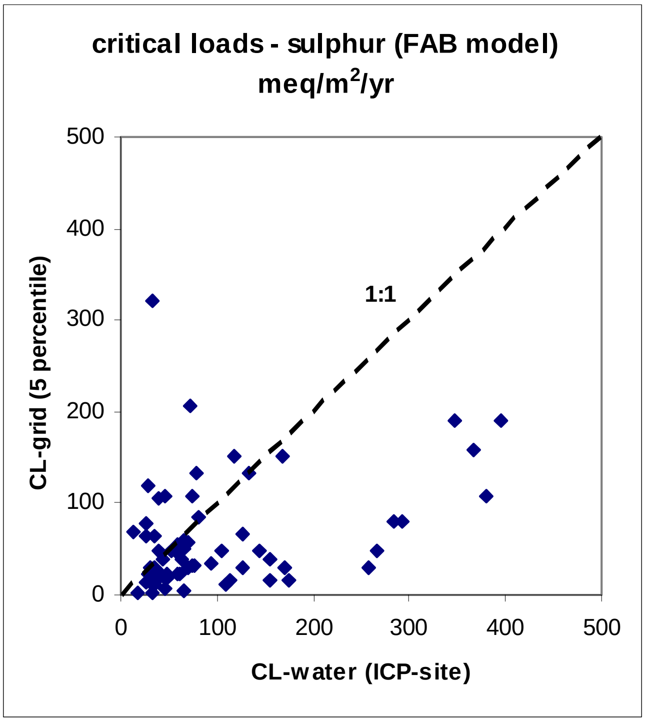


Figure 2. *Comparison of critical loads of sulphur (meq/m2/yr) for the European ICP Water sites with the 5 percentile contiguous grid values for the EMEP grid (data from Henriksen and Posch 1998).*

This comparison, however, cannot be used to evaluate the representativeness of the ICP Water sites, because the EMEP grid critical loads are for all types of ecosystems within each grid square. The ecosystems include forests and grasslands as well as freshwaters. In most countries in Europe it is the terrestrial ecosystems that comprise most of the ecosystems for which critical loads are used for determining the 5th-percentile (de Smet and Posch 1999). Thus the ICP Waters sites might be representative of the surface water ecosystems within the grid square, even though the 5th percentile critical load for all ecosystems in the square is much different. The exceptions to this are the three Scandinavian countries, Norway, Sweden and Finland, where surface waters comprise the majority of the ecosystems used to determine the 5th-percentile (de Smet and Posch 1999).

The North American countries, Canada and USA, have conducted extensive regional surveys of surface water sensitivity and acidification, especially in areas subject to acid deposition. In Canada these data have been used to estimate critical loads, albeit using a somewhat different approach than that used in Europe (Jeffries 1997, Jeffries et al. 1999). The USA has not taken the critical loads approach at all.

An assessment of the representativeness of the ICP Waters sites requires information on the sensitivity of surface waters in the region around each site. Such information is generally obtained by synoptic surveys, such as those listed in **Table 7**. Ideally a statistically selected sample of waters is made, such that the frequency distributions sample of key parameters such as pH and ANC in the statistical sample can be related to the distribution in the whole lake population.

3.1 Comparison of ICP Water sites with regional surveys

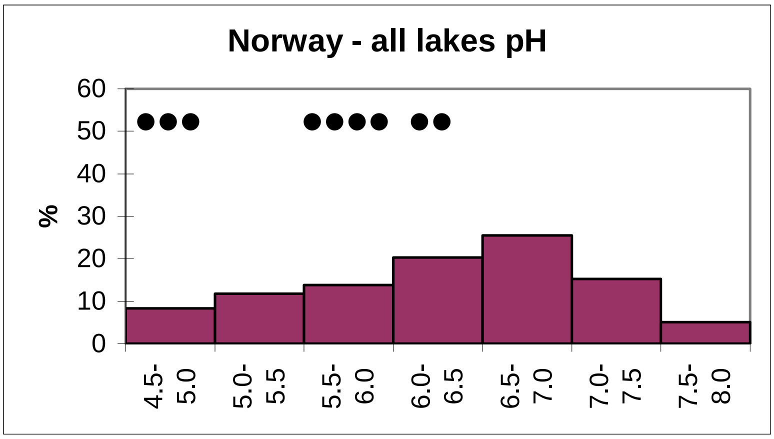
For those regions for which synoptic surveys of water quality have been carried out (**Table 7**) the ICP Waters sites can be compared with the general acidification situation for the entire region. This comparison is possible for Norway, Sweden, Finland, UK (Scotland and Wales), Germany (Bavaria), the southern Alps (Austria, Italy) and the Pyrenees (Catalonia, Spain) in Europe; and the Atlantic region (Labrador, Newfoundland, Nova Scotia and New Brunswick), Quebec, and Ontario in Canada; and the Adirondack Mountains, NY, and the Catskills, NY, in the USA.

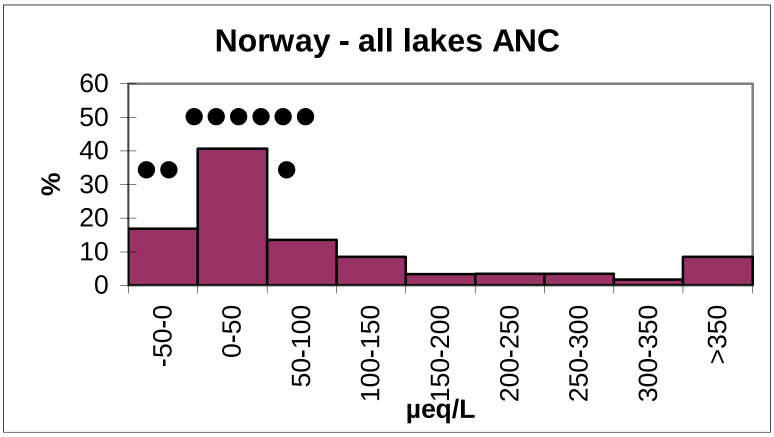
Table 7. *Overview of regional surveys of water quality and acidification*

Country	Year	Type	Number of sites	Notes	Reference
Norway	1974-75	lakes	154	small lakes only	Wright and Henriksen 1978
	1986	lakes	1006	small lakes only	Henriksen et al. 1988
	1995	lakes	1006	statistical sample	Skjelkvåle et al. 1996
Sweden	1985	lakes	6900	statistical sample	Naturvårdsverket 1986
	1990	lakes	4017	statistical sample	Naturvårdsverket 1990
	1995	lakes	3075	statistical sample	Henriksen et al. 1997; Henriksen et al. 1998
Finland	1987	lakes	987	statistical sample	Forsius et al. 1990; Kämäri et al. 1991
	1995	lakes	873	statistical sample	Henriksen et al. 1997; Henriksen et al. 1998
UK Scotland	1984-85	lakes	83	sensitive lakes	Maitland et al. 1987
	1995	lakes	136	statistical sample	Henriksen et al. 1997; Henriksen et al. 1998
UK Wales	1983-84	streams	141		Edwards et al. 1990
	1995	streams	102		Stevens et al. 1997
	1995	lakes	52		Henriksen et al. 1997; Henriksen et al. 1998
Denmark	1995	lakes	19	sensitive lakes	Henriksen et al. 1997; Henriksen et al. 1998
Germany	1983-96	rivers, lakes, reservoirs	36	sensitive waters	Bayerisches Landesamt für Wasserwirtschaft 1999
Czech Republic	1984-95	streams and lakes	12988	unpolluted waters	Veselý and Majer 1996; Veselý and Majer 1998
Slovakia, Tatra Mountains	1984-90	lakes	53	sensitive lakes	Kopácek and Stuchlík 1994
Poland, Tatra Mountains	1995	lakes and streams	11	sensitive lakes	Lydersen et al. 1996
Spain, Pyrenees, Catalonia	1987	lakes	102	small high mountain lakes	Catalan et al. 1993
Switzerland, southern Alps	1990-95	lakes	65	small high mountain lakes	Boggero et al. 1998
Italy, southern Alps	1981	lakes	320	small high mountain lakes	Mosello 1984

Country	**Year**	**Type**	**Number of sites**	**Notes**	**Reference**
	1988-91	lakes	295	small high mountain lakes	Mosello et al. 1993
Austria, Tyrolian and Carinthian Alps	1983-86	lakes	70	small high mountain lakes	Honsig-Erlenburg and Psenner 1986; Psenner 1988; Psenner 1989; Mosello et al. 1993
	1995	lakes	73	small high mountain lakes	Psenner and Sommaruga-Wögrath 1998
Canada Atlantic region	1985-1996	lakes	416	sensitive lakes	Jeffries 1997
Quebec	1985-1996	lakes	1377	sensitive lakes	Jeffries 1997
Ontario	1985-1996	lakes	1035	sensitive lakes	Jeffries 1997
USA Adirondack Mountains, NY	1991-1994	lakes	220	sensitive lakes	
Catskills, NY	1993-1994	streams	131	statistical sample	
Upper Midwest	1984	lakes	592	statistical sample	Eilers et al. 1988
Northeast	1984	lakes	768	statistical sample	Brakke et al. 1988
Southeast	1984	lakes	252	statistical sample	Eilers et al. 1988

For Norway the 9 ICP sites are all highly acid-sensitive, as they all have critical loads below 100 meq/m^2/yr (sulphur using the SSWC model) (**Figure 3**). They span a range of ANC and pH, but all fall into the lower part of the range for all the lakes in the country. Because the sites are located over the entire country the amount of acid deposition varies widely from highly impacted sites in the south to nearly pristine reference sites in the central and northern parts of the country. The sites are well suited for monitoring changes in acidification in response to changes in acid deposition.





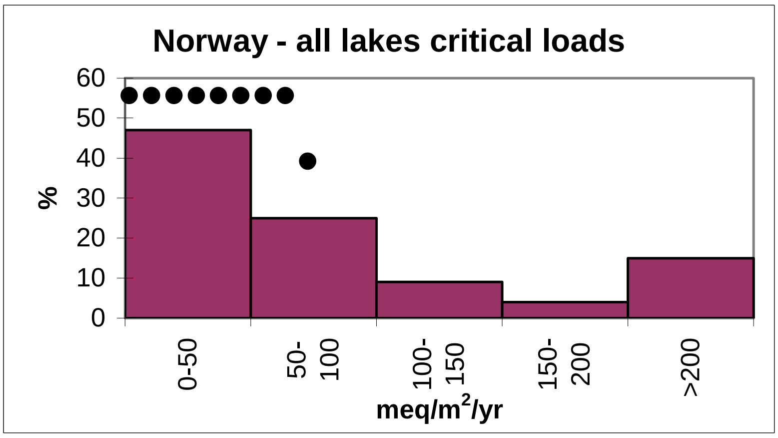


Figure 3. *Frequency distributions of pH, ANC and critical loads (sulphur using SSWC model) for all lakes in Norway derived from the 1995 synoptic survey (Henriksen et al. 1997; Henriksen et al. 1998) (columns) and values for the 9 Norwegian ICP Waters sites (means of all samples for years 1996-98) (points).*

For Sweden the 10 ICP-Water sites are acid-sensitive also, with critical loads below 150 meq/m^2/yr (**Figure 4**). They fall within the lower part of the range of pH and ANC relative to the entire lake population of Sweden. As in Norway the Swedish sites are located over the entire country, and thus receive a wide range of acid deposition. The sites are well suited for monitoring changes in acidification in response to changes in acid deposition.

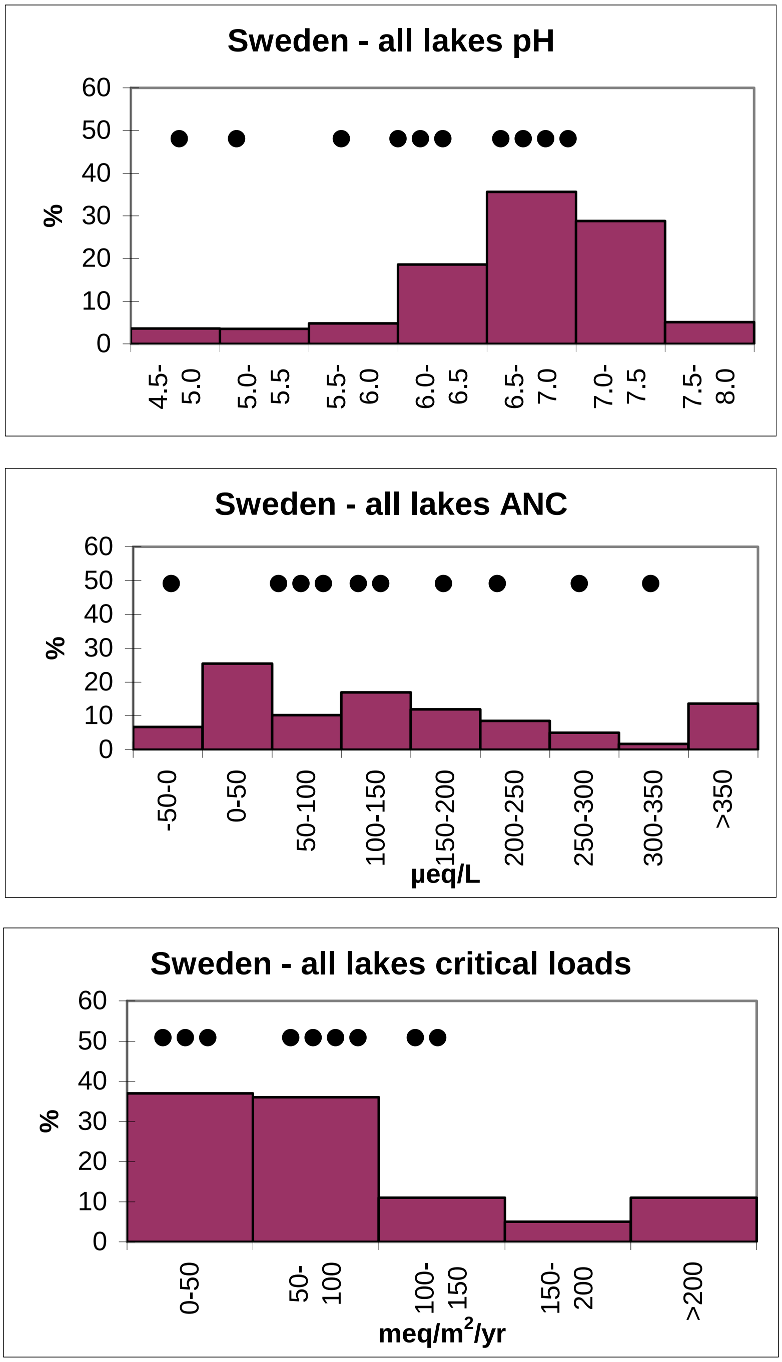


Figure 4. *Frequency distributions of pH, ANC and critical loads (sulphur using SSWC model) for all lakes in Sweden derived from the 1995 synoptic survey (Henriksen et al. 1997; Henriksen et al. 1998) (columns) and values for the 10 Swedish ICP Waters sites (means of all samples for years 1996-98) (points).*

For Finland the 7 ICP Waters sites are acid-sensitive also, with critical loads below 100 meq/m^2/yr (**Figure 5**). They fall within the lower part of the range of pH and ANC relative to the entire lake population. As in Norway and Sweden, the Finnish sites are located over the entire country, and thus receive a wide range of acid deposition. The sites are well suited for monitoring changes in acidification in response to changes in acid deposition.

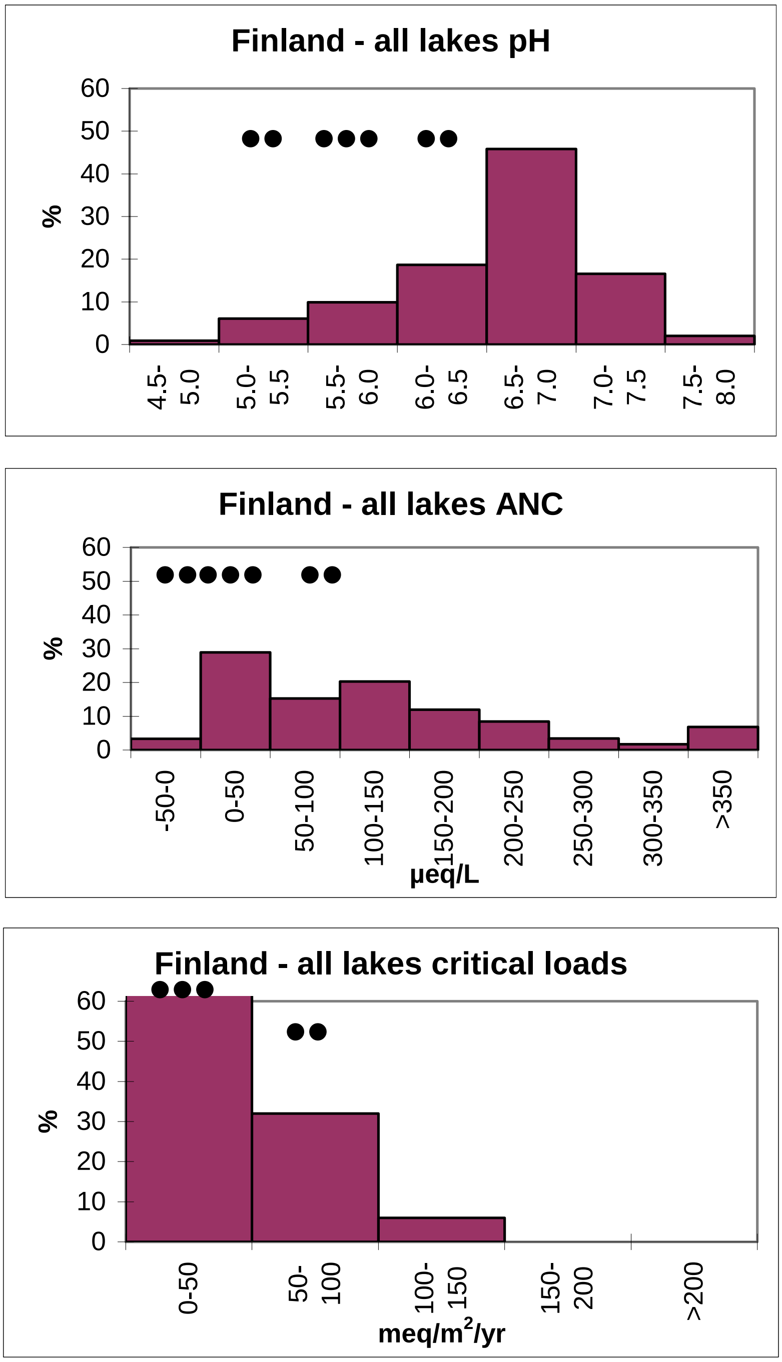


Figure 5. *Frequency distributions of pH, ANC and critical loads (sulphur using SSWC model) for all lakes in Finland derived from the 1995 synoptic survey (Henriksen et al. 1997; Henriksen et al. 1998) (columns) and values for the 7 (5 for critical loads) Finnish ICP Waters sites (means of all samples for years 1996-98) (points).*

For the UK there are synoptic survey data available only for Scotland and Wales. Of the 6 UK ICP sites, 3 are in Scotland (UK01 Loch Coire nan Arr, 04 Lochnagar, 07 Round Loch of Glenhead) and 1 is in Wales (UK15 Llyn Llagi) while the remaining 2 are in the English Lake District (UK10 Scoat Tarn) and Northern Ireland (UK21 Blue Loch), respectively. The Lake District site can be regarded are sufficiently similar to the Scottish survey sites, whereas the Northern Ireland site falls in a region whose general surface water characteristics is insufficiently known. The 5 ICP Waters sites are acid-sensitive, with critical loads below 100 meq/m^2/yr (**Figure 6**). They fall within the lower part of the range of pH and ANC relative to the entire lake population. As in Norway, Sweden and Finland, these UK sites receive a wide range of acid deposition. The sites are well suited for monitoring changes in acidification in response to changes in acid deposition.

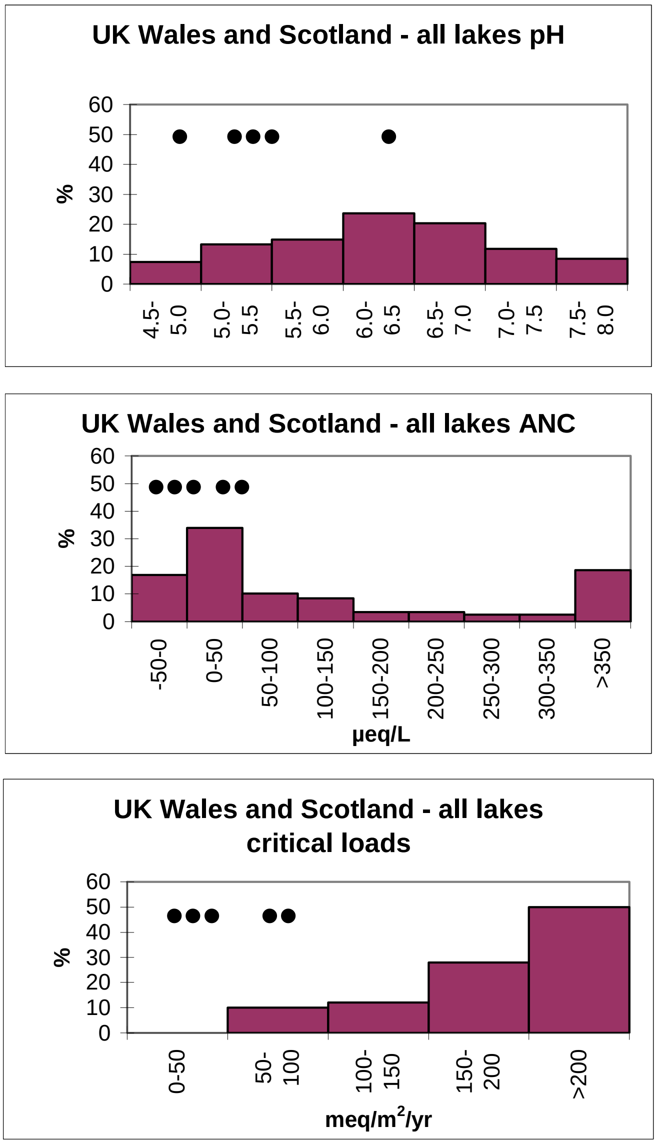


Figure 6. *Frequency distributions of pH, ANC and critical loads (sulphur using SSWC model) for all lakes in Wales and Scotland derived from the 1995 synoptic survey (Henriksen et al. 1997; Henriksen et al. 1998) (columns) and values for 5 British (Wales, England and Scotland) ICP Waters sites (means of all samples for years 1996-98) (points). The ICP site in the English Lake District (UK 10) is included, as it is similar to lakes in nearby Scotland included in the survey. The site in N. Ireland (UK21) is not included as it falls in another region.*

For Bavaria (southern Germany) there are data from 36 rivers, lakes and reservoirs (including 5 ICP sites) (Bayerisches Landesamt für Wasserwirtschaft 1999). These sites are chosen to be in acid-sensitive geological settings. With respect to pH three of the ICP sites (DE17, DE18, DE33) fall into the acid part of the range (pH4-5) while two of the sites (DE02, DE08) fall in the middle of the range (pH 5.5-6.0) (**Figure 7**). The sites are well suited for monitoring changes in acidification in response to changes in acid deposition.

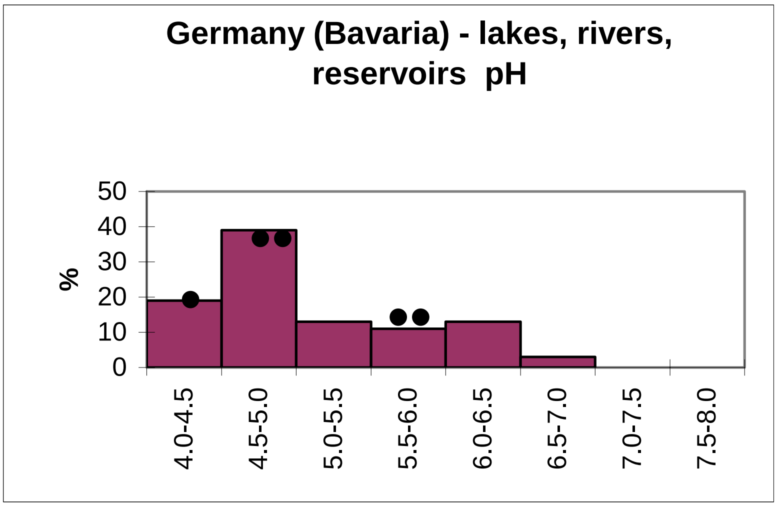


Figure 7. *Frequency distribution of pH for 36 rivers, lakes and reservoirs in Bavaria (Spessart, Frankenwald, Fichtelgebirge, Bayerischer Wald) surveyed in 1983-96 (columns), and values for the 5 ICP sites in this region (points). Data from Bayerisches Landesamt für Wasserwirtschaft (1999).*

For the Alps there are survey data from 413 high elevation lakes in sensitive regions in southern Switzerland (Ticino) (Boggero et al. 1998), northern Italy (Mosello 1984; Mosello et al. 1999), and Austria (Tyrolian and Carinthian Alps) (Honsig-Erlenburg and Psenner 1986; Psenner 1988; Psenner 1989; Psenner and Sommaruga-Wögrath 1998) (**Table 7**). In total there are synoptic data from 413 lakes, and these are summarised by Marchetto et al. (1994) and Mosello et al. (1993). These lakes generally have low ANC and low critical loads, features that confirm their general sensitivity to acidification (**Figure 8**). Only 3 ICP Waters sites fall in this geographic region and also are high-elevation. These are 2 Italian sites (IT01 Lake Paione inferiore, IT03 Lake Paione Superiore) and the one Austrian site (AU01 Schwarzsee ob Sölden). These three sites have low critical loads, low ANC and moderate pH (**Figure 8**), and appear well suited for monitoring the response to changes in acid deposition in this region. The 4 other Italian sites, on the other hand, are low lying streams and are much less acid sensitive, with high critical loads and high pH and ANC. These are less well suited as monitoring objects for acidification.

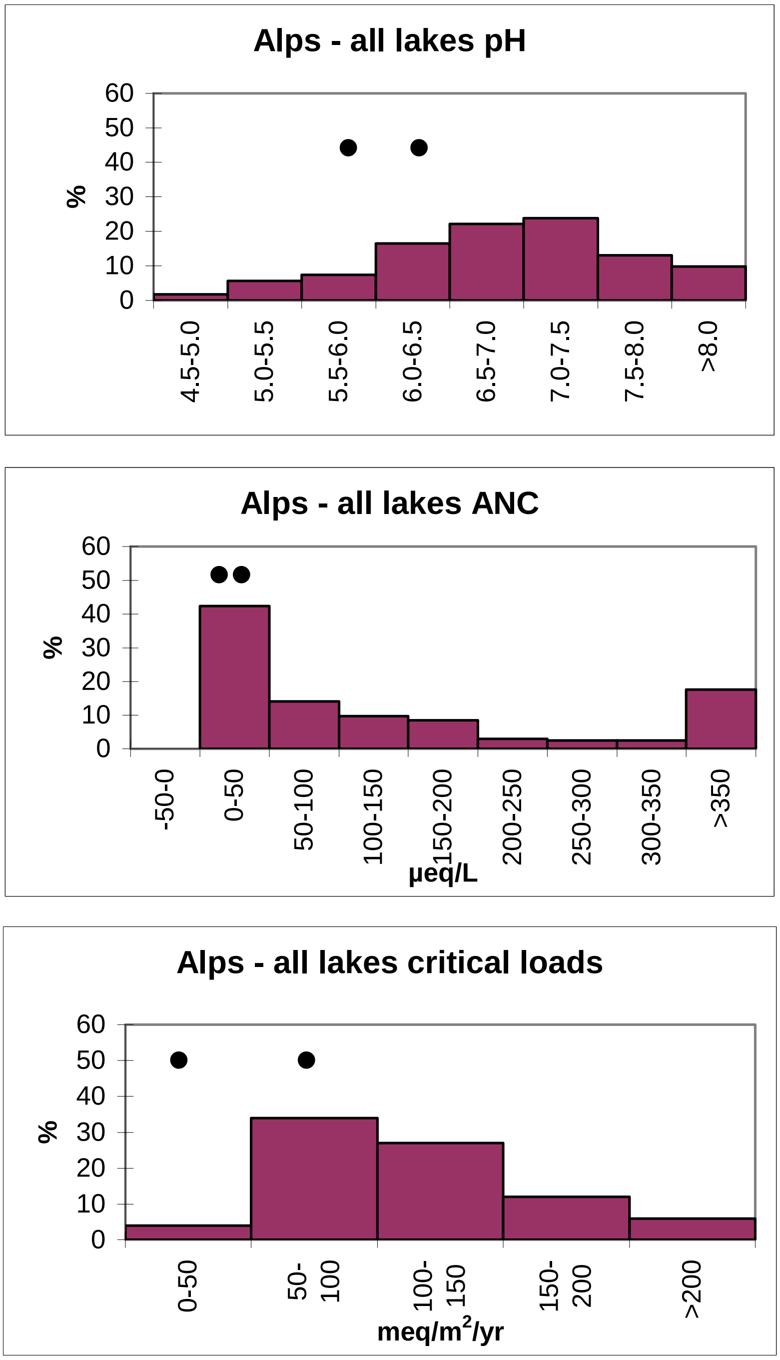


Figure 8. *Frequency distributions of pH, ANC and critical loads (sulphur using SSWC model) for 413 lakes in the southern Alps (Italy, Switzerland and Austria) surveyed in 1989-91 (Mosello et al. 1993; Marchetto et al. 1994) (columns) and values for the 2 high-elevation Italian ICP Waters sites (IT01, IT03) and one Austrian site (AU01) (means of all samples for years 1996-98) (points).*

For the Pyrenees in Catalonia, Spain, there are synoptic survey data for 102 high elevation lakes (Catalan et al. 1993). This region receives very low levels of acid deposition and thus serves as a reference area for much of the rest Europe. The lakes are generally acid-sensitive with low to moderate ANC and moderate pH (**Figure 9**). The Spanish ICP site (ES01 Redó) is located in this region, and has both pH and ANC near the lower part of the range for the region (**Figure 9**). It is thus well suited to monitor changes due to change in acid deposition or other environmental driving variables. Critical loads have not been calculated for these lakes.

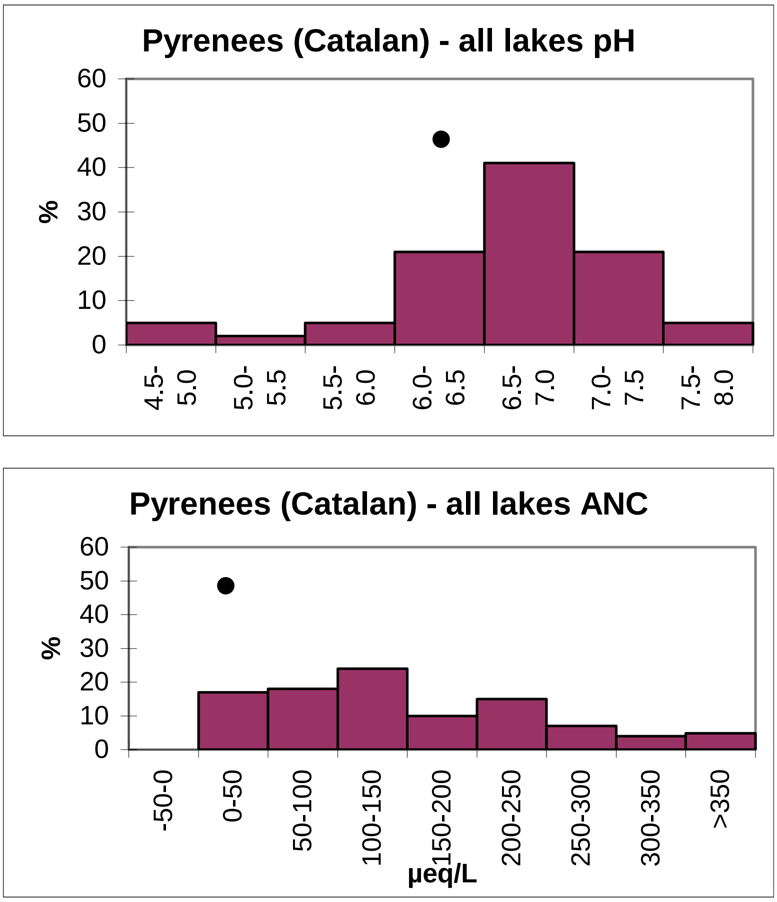


Figure 9. *Frequency distributions of pH and ANC (critical loads not reported) for 102 lakes in the Pyrenees Mountains of Catalonia, Spain surveyed in 1989-91 (Catalan et al. 1993) (columns) and values for the high-elevation Spanish ICP Waters site (ES01) (means of all samples for years 1996-98) (point).*

For a number of the ICP-Water sites in Europe there are apparently no relevant synoptic survey data with which the representativeness of these sites can be evaluated. These sites include most of the sites in eastern Europe (**Table 8**).

Table 8. *ICP Waters sites with no regional chemistry data. The representativeness of these sites cannot be assessed due to the lack of appropriate regional data.*

Site	Area
UK21	N. Ireland
IR01-03	Ireland
EE01	Estonia
LV01-04	Latvia
P01-02	Poland (Tatras)
HU01	Hungary
BY01	Belarus
RU01-06	Russia

For the 3 regions in eastern Canada there are extensive regional surveys of lake chemistry with which the ICP-data can be compared. The 1997 national assessment (Jeffries 1997) summarises results from surveys of several thousands of lakes, many of which are located in acid-sensitive terrain in eastern Canada. The 5 ICP sites in the Atlantic region (C10-C14) are all located Nova Scotia, and have low pH and low ANC relative to the entire set of 416 lakes surveys (**Figure 10**). The 5 ICP sites in Quebec (C05-C09) also have low pH and ANC relative to the set of 1377 surveyed lakes in the province (**Figure 10**). The 4 ICP sites in Ontario (C01-C04) are all located in the Turkey Lakes area of Algoma National Park, and have intermediate pH and high ANC relative to the set of surveyed lakes in the province (**Figure 10**). The Canadian ICP sites in both Quebec and Ontario appear not to represent acid-sensitive lakes, or alternatively they have not been acidified to the same extent as more sensitive sites in the regions.

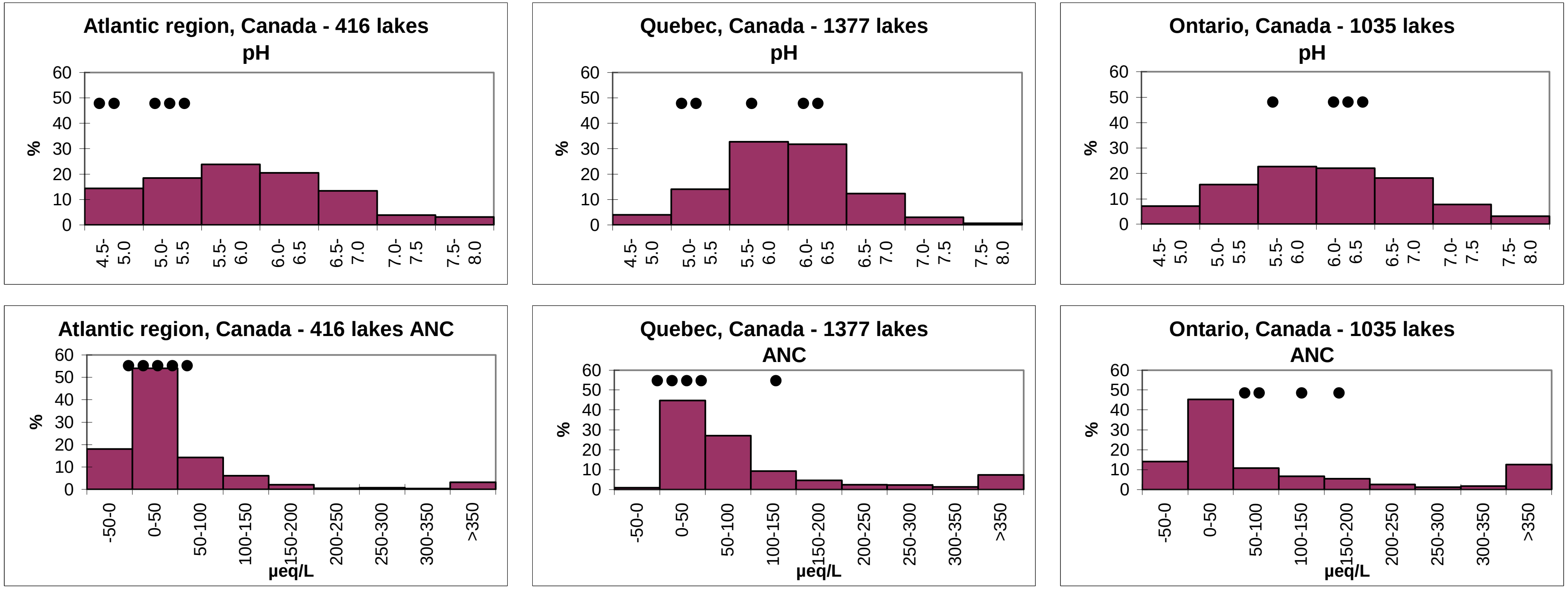


Figure 10. *Frequency distributions of pH and ANC for lakes in 3 regions of eastern Canada (Jeffries 1997) (columns) and the values for the ICP sites in each region (points). Critical loads not reported.*

For the 2 regions in the USA, there are extensive regional surveys of lakes and steams with which the ICP site data can be compared. Several synoptic surveys of lakes in the Adirondack Mountains, New York, have been conducted (**Table 7**). Data from 220 generally acid-sensitive lakes conducted in 1991-94 show many lakes that are acidified with low pH and low ANC (**Figure 11**). Seven ICP sites are located in the Adirondacks, and these generally fall into the more sensitive range of the entire distribution. For the Catskill region of NY there are data from a survey of 131 streams conducted in 1993-94 (**Table 7**). These data show that a few of the steams are acidified with low pH and low ANC. The 2 ICP sites located in the Catskills fall into this end of the distribution (**Figure 11**).

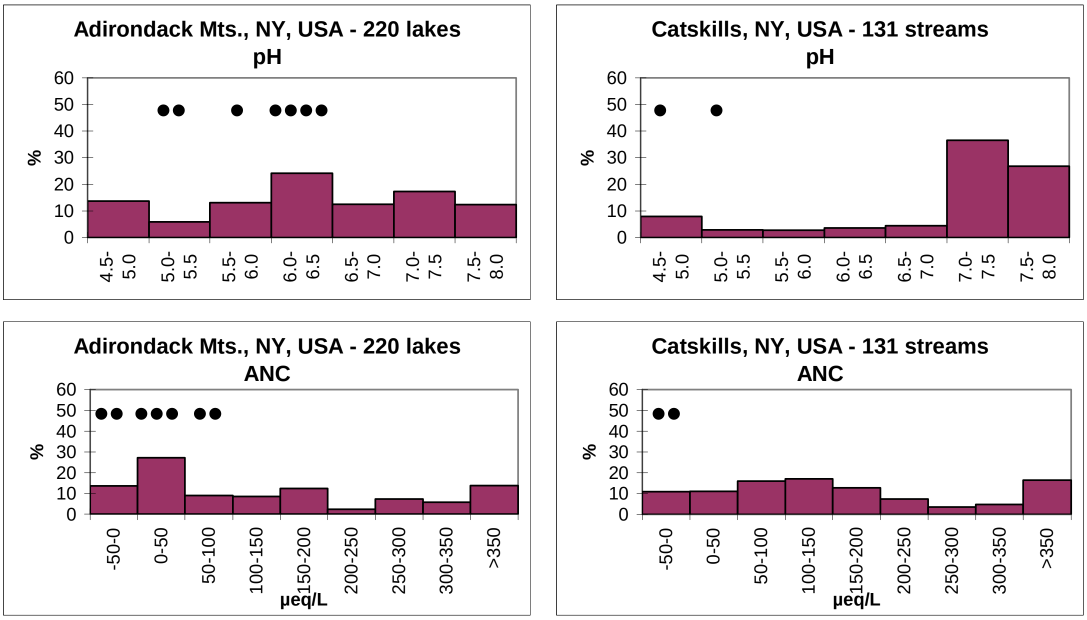


Figure 11. *Frequency distributions of pH and ANC (critical loads not reported) for waters in 3 regions in the USA (columns) and the values for the ICP sites in each region (points).*

In summary for those ICP sites for which regional synoptic survey data are available, most of the sites appear to be well suited to monitor changes in acidification in response to changes in acid deposition. The sites are generally representative of the lower ANC, lower critical load levels of the distributions for all the waters surveyed in the region.

3.2 Assessment of the total coverage of the ICP Water sites and regional surveys

Surface water acidification can be expected in acid sensitive areas where acid deposition is high. Acid sensitive areas are generally characterised by bedrock and overburden poor in readily-weatherable minerals. Small high-altitude lakes and streams are often more sensitive than low-lying large lakes and rivers.

Based on these considerations, maps of areas in Europe with waters sensitive to acidification have been compiled (Merilehto et al. 1988; Skjelkvåle and Wright 1991) (**Figure 12**). This map can be overlain with maps of S and N deposition (**Figure 13**, **Figure 14**) to identify areas that are suitable for monitoring response of changes in acid deposition.

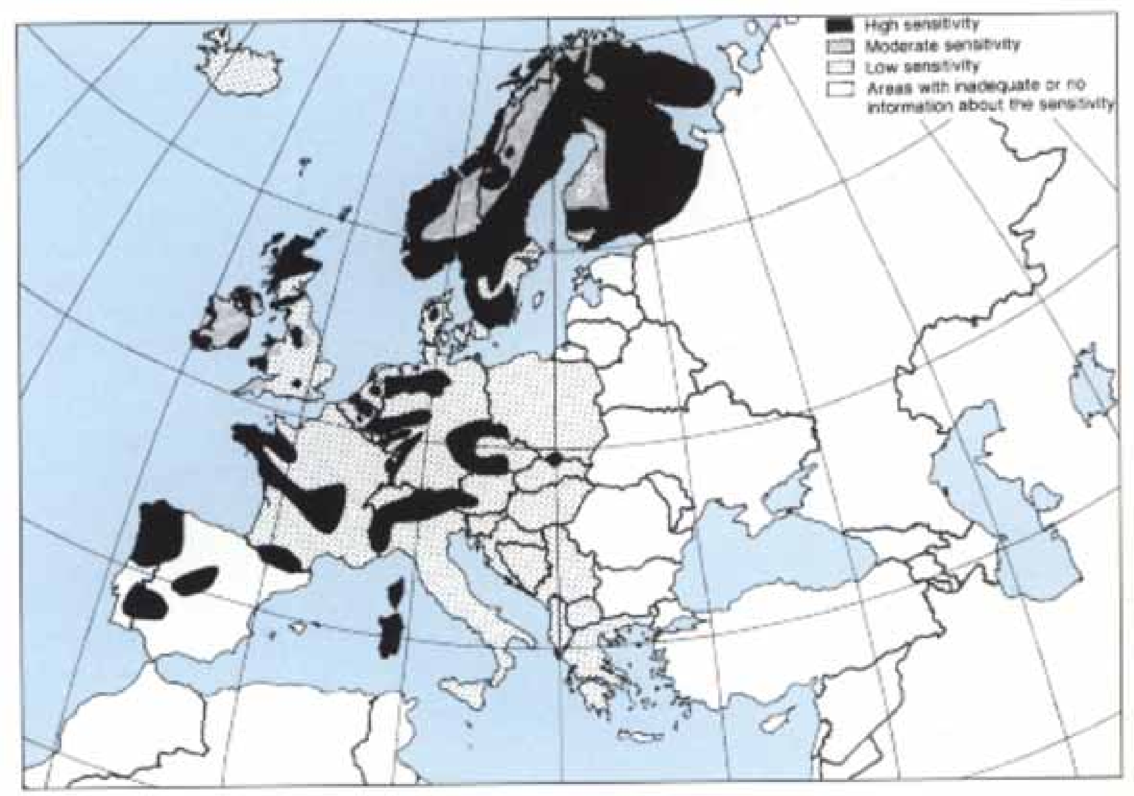


Figure 12. *Areas of Europe sensitive to surface water acidification (from Skjelkvåle and Wright 1991).*

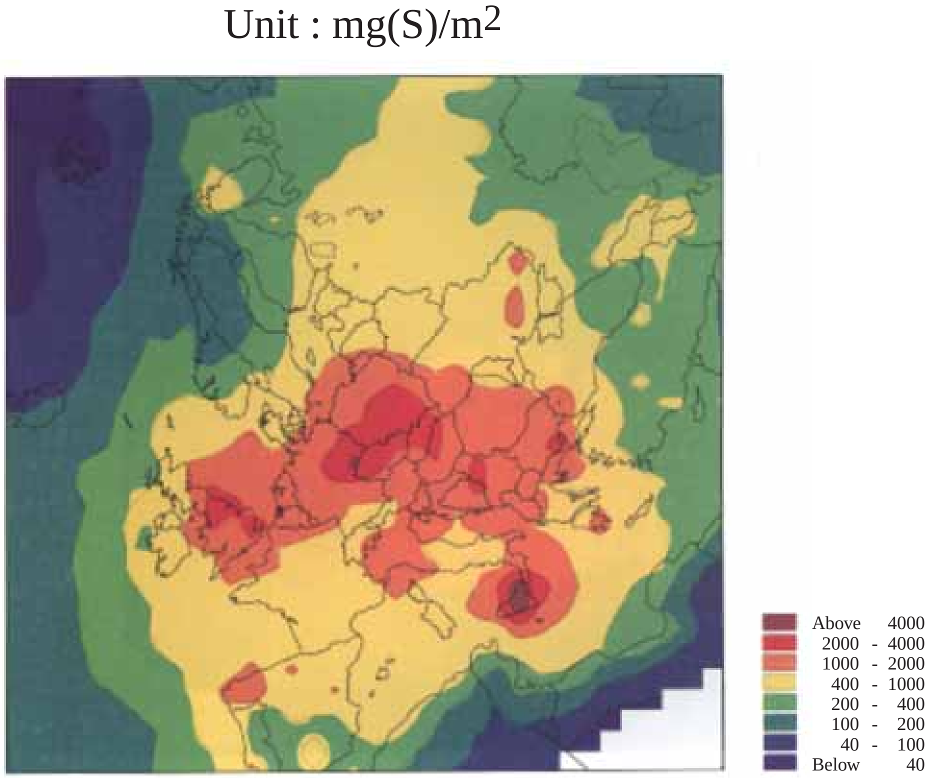


Figure 13. *Total deposition of oxidised sulphur in 1996 in Europe (from Meteorological Synthesizing Centre - West 1998).*

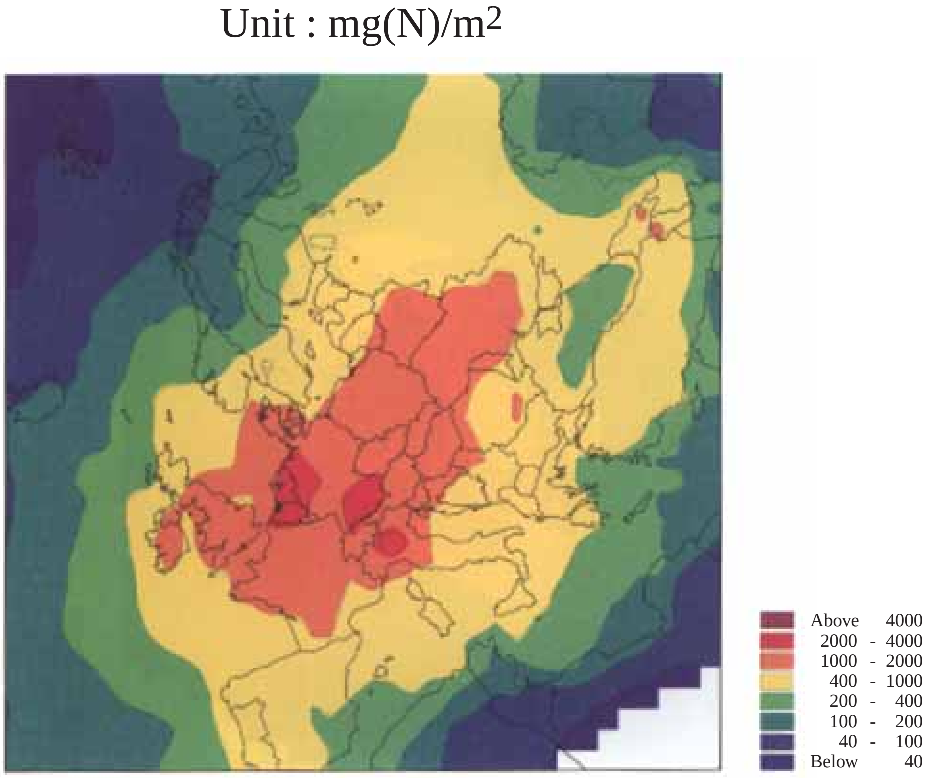


Figure 14. *Total deposition of oxidised nitrogen in 1996 in Europe (from Meteorological Synthesizing Centre - West 1998).*

A similar sensitivity map has been compiled for Canada and overlain by isopleths for sulphur deposition (RMCC 1990) (**Figure 15**). Note that in the Canadian map, sensitivity is expressed as the "potential of soils and bedrock to reduce the acidity of atmospheric deposition". Hence the most sensitive map area are classified as "low". It is clear that acidified waters can be expected in southeastern Canada, and that all the Canadian ICP sites lie in areas receiving intermediate to high levels of acid deposition. However, the catchment sensitivity of ICP lakes varies greatly from region to region, i.e. most sensitive for the Nova Scotia sites, less sensitive for the Quebec sites, and least sensitive for the Ontario sites. Because of the scale of the Canadian map, these differences in sensitivity are not readily apparent.

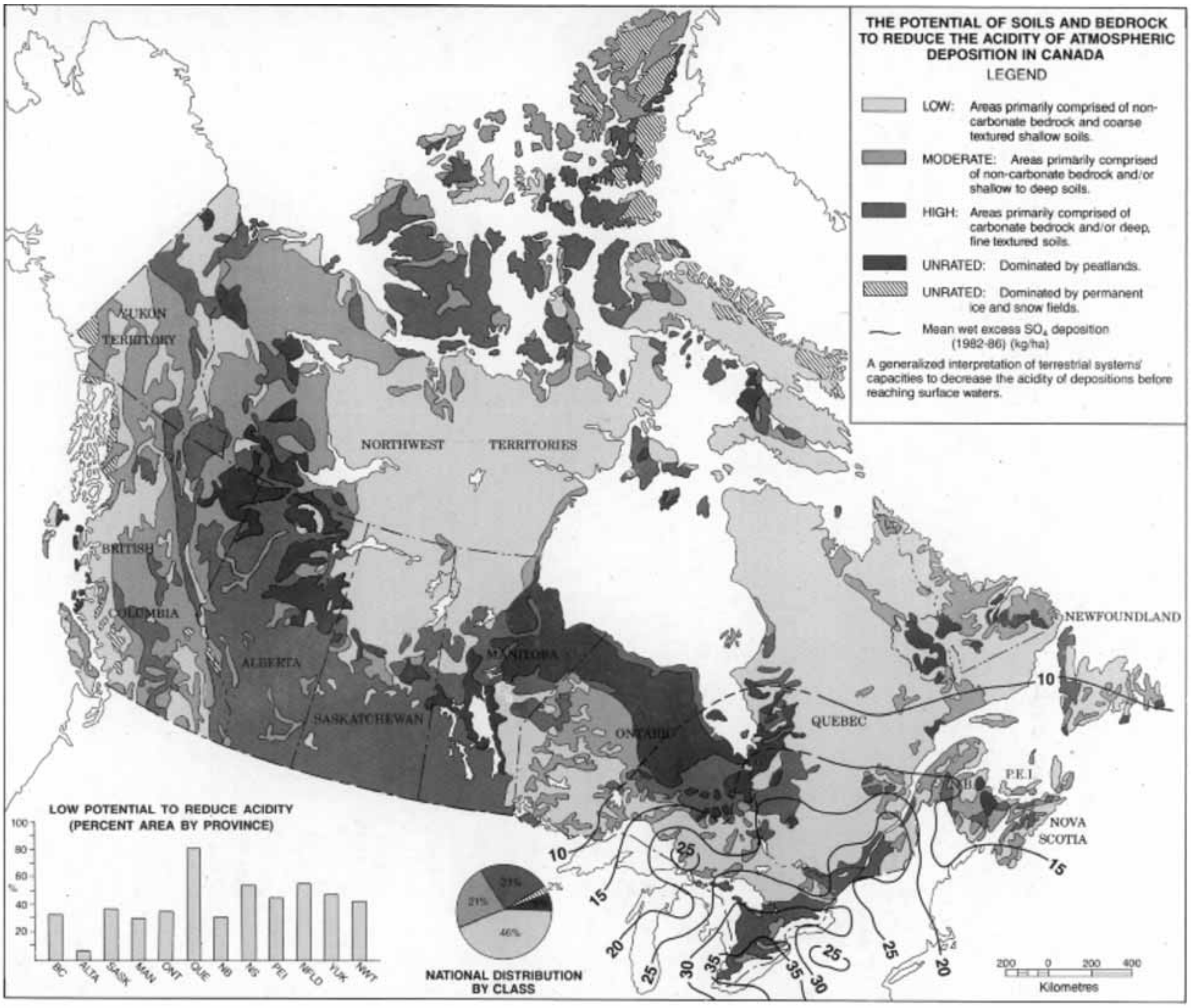


Figure 15. *Map of Canada showing areas sensitive to surface water acidification (from RMCC 1990).*

The ICP Waters sites cover most of the acid-sensitive areas in Europe that receive significant acid deposition. There are no ICP sites, however, in regions that have been or are potentially affected, such as northwestern Spain, southern Switzerland, the Vosges Mountains of France, Belgium, the Netherlands, Slovakia (Tatra Mountains) and northwestern Russia (Karelia, Kola Peninsula) (**Table 9**). Furthermore there are insufficient data from many regions in eastern Europe to adequately assess both the risk of acidification and the sensitivity of surface waters.

For North America the ICP sites cover several but not all of the acid-sensitive regions that receive significant acid deposition, and that can be expected to show changes in response to changing levels of deposition in the future. ICP sites cover important areas in New York (Adirondacks, Catskills), but there are currently no ICP sites in other impacted areas in the US such as New England, the southern Appalachians (Maryland, West Virginia, Virginia, Tennessee, Kentucky, North Carolina). Several areas of the western US are also potentially of interest (the Front Range in Colorado, mountains in southern California) and currently not included in the ICP site network.

In Canada there are ICP sites in several of the acid-sensitive and impacted regions. These include lakes in southern Quebec and Nova Scotia, but only one region of Ontario that receives moderate acid deposition. The highly impacted areas of Ontario near Sudbury and the Haliburton-Muskoka region are not represented. There are no ICP sites in north-central Alberta, a region subject to acid deposition and with potential increases in deposition in the future.

Table 9. *Sensitive areas with no ICP sites, potentially impacted by acidification.*

Europe

France (Vosges Mountains)
Switzerland (southern Alps, Ticino)
Slovakia (Tatra Mountains)
Belgium
Netherlands
Russia (Karelia)

North America

USA, southern Appalachian Mountains
USA, Front Range, Colorado
USA, San Bernadine Mountains, California
Canada, Ontario, Haliburton-Muskoka region
Canada, Ontario, Sudbury region
Canada, Alberta, Fort McMurray region

4. Long-term trends in surface water chemistry

4.1 General Overview of Trend Analysis

One of the most valuable uses to which data from the ICP Waters program can be put is an evaluation of long-term trends. Trend analyses have been conducted previously on ICP Waters data and have provided important indications of the geographic extent of acidification and recovery of lakes and streams (Lükewille et al. 1997; Newell and Skjelkvåle 1997; Skjelkvåle et al. 1994; Stoddard et al. 1999 in press). In this 12-year report, we report on trends from 98 ICP Waters monitoring sites, for the 10-year period 1989-1998. This decadal period is one in which all of the regions covered by the ICP monitoring sites have experienced substantial reductions in sulphur deposition, and this can be expected to be reflected in trends in surface water SO_4^{2-} concentrations and acidity (pH and alkalinity). We include in the current analysis two variables that are the direct result of atmospheric deposition (seasalt-corrected SO_4^{2-} and NO_3^-), one that will respond indirectly to changes in SO_4^{2-} and NO_3^- (sum of base cations – for the purposes of this report we use the sum of seasalt-corrected Ca^{2+} and seasalt-corrected Mg^{2+} as a surrogate for the complete list of base cations), and two variables indicative of recovery or acidification (hydrogen ion concentration and calculated acid neutralising capacity [ANC]). We calculate ANC as Σ(Base Cations) - Σ(Acid Anions), equal to $\Sigma(Ca^{2+} + Mg^{2+} + Na^+ + K^+) - \Sigma(SO_4^{2-} + NO_3^- + Cl^-)$, where the units are µeq/L.

As in the 9-year report, we again present the trends for each individual site, as well as aggregated trends organised by regions. While it is important to know how individual sites in various countries are responding to decreased atmospheric deposition, the strongest evidence that emissions control programs are having their intended effect comes from a consistent pattern of recovery (decreasing SO_4^{2-} and increasing pH and ANC) across a large number of sites; the regional trend analysis is intended to test for these large-scale patterns.

4.2 ICP Sites Chosen for Trend Analysis

Sites in the ICP Waters database exhibit a wide range of sampling frequencies, completeness of chemistry, and length of record. In order to make meaningful comparisons of trends among these sites, it is necessary to impose a minimum set of requirements for all of these data characteristics. We chose to focus the current analysis on sites sampled during at least 2 periods per year, and where data are available for at least 7 out of the 10 years. Further, we included only sites that had all of the variables that would have a direct response to changes in atmospheric deposition (i.e., sulphate, nitrate and base cations), and at least one of the two indicators of recovery (i.e., pH and ANC). Sites considered insensitive to acidification (ANC > 200 µeq/L) were excluded, as were any sites with documented watershed disturbances (e.g., agriculture) or trends clearly driven by climatic fluctuations (Psenner and Schmidt 1992). The results of this selection process are summarised in Appendix A. Ninety-eight sites had sufficient data for trend analysis.

4.3 Statistical Methods Used for Trend Analyses

4.3.1 Nonparametric Seasonal Kendall tau Test (SKT)

At each site, trends in ANC, hydrogen ion concentration, non-marine base cations ($\Sigma(Ca^{2+} + Mg^{2+})$), sulphate ($SO_4^{2-}$), nitrate ($NO_3^-$), and dissolved organic carbon (DOC) were analysed using the nonparametric Seasonal Kendall test (SKT) (Hirsch et al. 1982). The SKT has become a standard method for detecting site-specific trends in water quality data, largely because it can accommodate the non-normality, missing and censored data, and seasonality that are common in data of this type, but it is nevertheless a powerful (in a statistical sense) trend test (Loftis and Taylor 1989). Because the SKT is sensitive to serial correlation, we use a modified form of the test described by Hirsch and Slack (1984), which uses a covariance term to correct the seasonal Z scores for serial dependence. One limitation of the SKT is that it detects only monotonic trends; trends need not be linear, but they must proceed in only one direction (increasing or decreasing) to be detectable.

In brief, 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. The signs (indicating whether the second observation in each pair-wise comparison is higher or lower than the first) for all pair-wise comparisons within each block are summed, and a Z statistic calculated as the ratio of the sum of signs divided by the standard deviation in the signs. Z statistics are produced for each season, and if the trends are homogenous among the seasons, the seasonal Z statistics can be combined to produce an overall trend result. The SKT does not estimate the slopes of trends, but it has become customary to associate slopes calculated according to the method of Sen (1968), which estimates the slope by calculating 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.

4.3.2 Regional Meta-Analysis

In order to infer regional trends from the separate trend tests performed by SKT, we employ a variation of meta-analysis, 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 (1932), who recognised 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 sites and seasons 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 determining whether the variance among the Z statistics (in this case, from 98 individual sites) is sufficiently small to warrant their being combined into a single test for trend. In practice, this is accomplished by submitting the Z statistics to an analysis of variance (ANOVA), and testing the resulting sums of squares against a chi-square distribution (Mattson et al. 1997; van Belle and Hughes 1984). This procedure has been termed Analysis of Chi-Squares (ANOCHIS) by Mattson et al. (1997).

There is no established rule for determining when heterogeneity among trends (from individual sites) is sufficient to invalidate combining trend tests from individual sites. van Belle and Hughes (1984) propose setting a relatively high significance level (e.g., $p = 0.01$), provided that all of the Z statistics are of similar sign and magnitude. We find that in some cases where there is little seasonal variability in Z scores (e.g., for lake SO_4^{2-} concentrations) all of the Z statistics may be very similar (all of the same sign, and within a factor of 3 of one another), yet between-site differences are estimated to be highly significant ($p < 0.01$). For this reason we use the threshold suggested by van Belle and Hughes (1984) as a “red flag,” requiring more in-depth analysis of the Z scores, rather than as an absolute limit. For each case where homogeneity tests yield p values less than 0.01, we assess the consistency in the magnitude and signs of Z values, and determine qualitatively whether the homogeneity is ecologically significant (e.g., whether the trend is the overwhelmingly dominant pattern in the data, despite some heterogeneity in absolute Z values).

4.4 Results of trend analysis

4.4.1 General trends

The results of the SKT test for the single sites and the calculated slopes are presented in Appendix B. The summarised results presented in **Figure 16** show the general trends in the data.

Sixty-seven out of the 98 sites show significant decreasing trends in non-marine SO_4^{2-} while only one site (FI5, Suopalampi in Finish Lapland) shows a significant increasing trend. Of the sites not showing significant trends, 22 show a tendency to decrease while 6 show a tendency to increase. The majority of the significant decreasing trends show slopes in the range from -1 to -8 µeq/L/yr (**Figure 16** bottom panel). The decreasing trends in SO_4^{2-} over the last 10 years are a major characteristic for the ICP Waters sites.

The distribution of Z-scores for NO_3^- show a bell-shaped curve with about the same number of decreasing trends and increasing trends, and the majority of the sites (85 out of 98) show no significant trends. The decreasing trends in NO_3^- have in general a steeper slope than the increasing trends (**Figure 16** bottom panel). These results confirm the conclusion from the 9-year report, that there is no clear picture with regard to trends in NO_3^-.

The majority of the of the trends for base cations (SBC*) (36 out of 98 sites) show significant decreasing trends for base cations (SBC*), while 4 sites show significant increasing trends. Of the sites not showing a significant trend, 40 sites show a tendency to decrease while 12 show a tendency to increase. As for SO_4^{2-}, the decrease in base cations is a pronounced pattern in the ICP Waters sites. The slopes of the base cation trends (**Figure 16** bottom panel), show that a majority of the slopes are in the range from -2 to -8 µeq/L.

The Z-scores for the trends in H^+ show a bell-shaped curve, with the mid-point on the negative side. Nineteen sites show a significant negative trend, while only 2 sites show a significant positive trend. A majority of the decreasing trends show slopes in the range < 0.5 µeq/L/yr. Also for H^+ there is a general pattern in the ICP Waters sites towards decreasing H^+ (increasing pH).

Twenty-one sites show significant positive trends in ANC, while 6 sites show significant negative trends. The slopes of the ANC trends (**Figure 16** bottom panel), show that the trends are either positive in the range from 2 to 10 µeq/L, or negative in the range form -2 to -10 µeq/L. This is a very different pattern from H^+ were most of the trend slopes cluster around zero, and also different from SO_4^{2-} and base cations, were there is a strong tendency towards only negative slopes.

At some stations we have data for DOC, for some we have TOC, and for some station we have data for both or even no reported values. In the results presented in **Figure 16**, we have presented DOC, and if DOC does not exist, we have used TOC. Altogether 92 sites have trend analysis for DOC and/or TOC. The curve presented in **Figure 16** (upper panel) is bell shaped with the mid-point on the positive side. All significant trends are positive. The trend slopes for these sites are in the range 0.1-0.3 mg C/L/yr. Of the non-significant trends only 16 show negative trends. Also for DOC/TOC there is a strong pattern in the ICP Waters sites towards increasing trends.

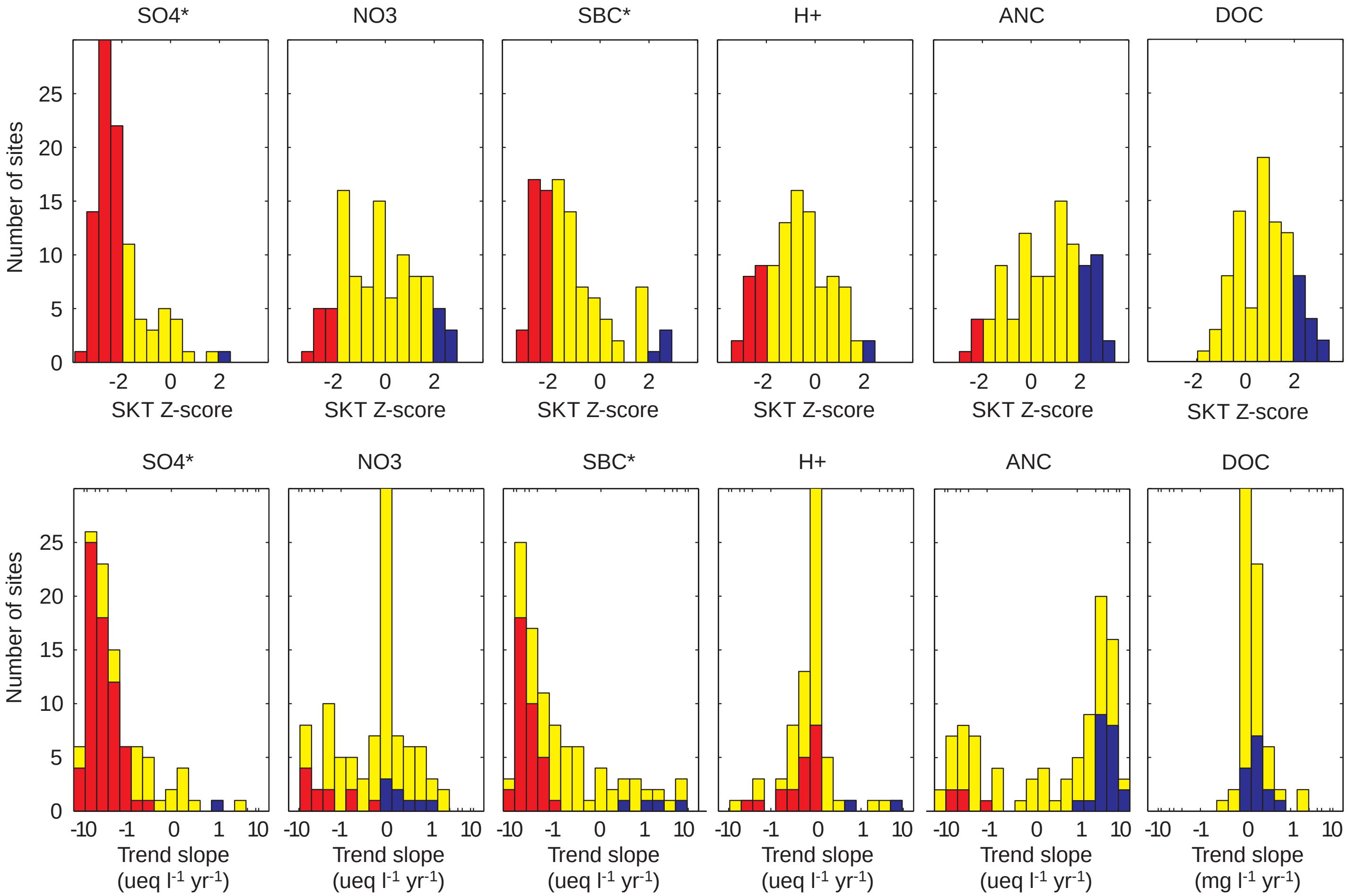


Figure 16. *SKT Z-scores and Theil trend slope for H^+, SO4*, NO_3, SBC, ANC and TOC for all the analysed sites. Negative scores indicate decreasing trends, while positive scores indicate increasing trends. Scores > 1.96 or < -1.96 are significantly different from 0 at the 0.005 significance level. The positive slopes are blue and the negative are red. Note that the axis on the bottom panel is arctan transformed.*

4.4.2 Regional trends

If the large-scale emission reductions that have occurred in Europe and North America over the past decade have had their intended effect, then equally large-scale patterns of recovery should be observable in acid sensitive surface waters. The goal of a regional trend analysis is to test whether a consistent pattern (or trend) is present in a large number of sites. In practice, we do this by examining the individual trends at all available sites within a large region, and testing whether the variability is sufficiently low to warrant their compilation into a single regional trend estimate.

For this report, we have divided the 98 available ICP Waters sites into 5 regions, on the basis of their geologic similarity, geographic proximity, and similarity of deposition. The 5 regions are: (1) the northern portions of the Nordic Countries (all sites north of 61° latitude); (2) the remaining portions of the Nordic Countries plus the United Kingdom; (3) Central Europe (all sites on the European mainland); (4) Midwestern North America (northwestern Ontario, Michigan, Wisconsin and Minnesota); and (5) Eastern North America (Central Ontario, Quebec, Atlantic Canada, and New York). These regions represent some compromises made in order to have sufficient sample sizes in each. Including the UK with the Nordic Countries is not an ideal arrangement, for example, but there are not sufficient sites to treat the UK as a separate region, and the sites are more similar to the Nordic Countries than to Central Europe. Trends for the UK do differ significantly from those for the Nordic Countries in some cases, however, and the UK situation is therefore given further consideration below. Similarly, the Central Europe region contains sites as dissimilar as several alpine lake sites in Italy and Poland, forested sites with thick soils in Germany, and stream sites in Denmark. In most cases these disparate sites have similar responses to reductions in rates of deposition; in a sense, any conclusions that we can make regarding the recovery of regional populations of lakes and streams are strengthened when the sites' dissimilarities are considered.

Results of the regional trend analysis are shown in **Table 10**. Evidence for regional declines in SO_4^{2-} are very strong. All of the regions had highly significant downward trends in SO_4^{2-}, with rates of decline ranging from -1.3 µeq/L/yr in the region with the lowest rates of deposition (Northern Nordic Countries) to -2.9 µeq/L/yr in Central Europe, where rates of deposition are the highest. Of the 98 sites included in this analysis, only 3 had Z scores higher than 0, and only one was significant. Similarly strong downward regional trends in SO_4^{2-} were reported in the 9-year report (Lükewille et al. 1997) and by Stoddard et al. (1999 in press), and it is now very clear that emissions reductions programs are having an important direct effect on surface water chemistry by reducing the concentrations of the most significant acidifying anion.

Nitrate, on the other hand, shows no regional patterns of change. Only one region (Central Europe) exhibited a significant regional trend in NO_3^- (downward), but the variability within this region was too high to make a valid regional trend estimate possible. Nitrogen deposition has decreased across much of Europe in the past decade (Tarrason 1998), but has remained largely unchanged in North America for the past 2 decades (Lynch et al. 1996). Because nitrogen is very biologically active, it is much more difficult to describe relationships between deposition and runoff chemistry for nitrogen than for sulphur. Climatic fluctuations, differences in histories of land use and insect outbreaks, and hydrologic differences can all create substantial site-to-site variability in how nitrogen is processed within catchments, leading to considerable differences in nitrogen retention and nitrogen leaching. This concept of variability is strongly supported by the trend results in **Table 10**.

Regional declines in SO_4^{2-} concentrations create an expectation of widespread recovery in acidified lakes and streams. This recovery is most clearly shown in the Nordic Countries/UK region, where ANC has increased at a rate of +2.4 µeq/L/yr in the past decade, and hydrogen ion concentrations have declined by -0.17 µeq/L/yr. This widespread recovery has also been indicated by the results of the Northern European Lake Survey (Henriksen et al. 1998), where lakes in southern Norway, for example, increased in ANC by 11 µeq/L between 1986 and 1995 (Skjelkvåle et al. 1998). Some recovery is also evident in the Northern Nordic Countries, where hydrogen ion has decreased by -0.04 µeq/L/yr, and in Midwestern North America, where ANC has increased by +1.6 µeq/L/yr. The lack of a significant ANC trend in the Northern Nordic Countries most likely results from the relatively small decreases in SO_4^{2-} that have occurred in this region, while the lack of a hydrogen ion trend in Midwestern North America results from a lack of data for this variable (only 2 of 9 sites in this region had complete pH data for the decade).

Two important regions failed to show significant recovery. In Central Europe, there was a regional tendency toward increasing ANC (+1.1 µeq/L/yr), but significant heterogeneity; ten out of 28 sites in this region showed no significant change in ANC. In Eastern North America there was no regional pattern for ANC or pH. The lack of recovery in this region has been reported previously (Driscoll et al. 1998; Stoddard et al. 1998; Stoddard et al. 1999 in press), and attributed to strongly declining base cation concentrations. In both Central Europe and Eastern North America there is strong evidence that base cation behaviour is an important factor controlling recovery (**Table 10**). In both cases, sites with declining ANC values showed the largest declines in base cation concentrations.

Finally, although the UK could not be treated as a separate region due to data limitations, it is clear from individual site analyses that significant differences in trends exist between the UK and the Nordic Countries. The regional decline in SO_4^{2-} concentrations, for example, is only significant at one of the six UK sites, and none show significant recovery in ANC. This lack of consistent UK recovery during the last decade has also been observed in the UK Acid Waters Monitoring Network (Monteith and Evans 2000), and has been attributed to (a) the absence of significant sulphur deposition reductions in western areas, and (b) the impact of natural climatic variations on water chemistry. Of particular importance appear to be inter-annual fluctuations in sea-salt deposition, which showed a large peak in the early 1990s. The resulting displacement of acidity from soil exchange sites by marine cations, and the apparent temporary retention of marine SO_4^{2-}, have significantly impacted surface water chemistry at many near-coastal sites. Longer monitoring periods may therefore be necessary for the detection of recovery trends in these areas.

Declining concentrations of acid anions should be expected to produce declining concentrations of base cations in lakes and streams, where some proportion of cations result from mobilisation by anions passing through catchment soils (Galloway et al. 1983). These declines are evident in all of the regions shown in **Table 10**. In all cases where base cation declines are smaller than measured decreases in SO_4^{2-} there are signs of recovery. In the one region (Eastern North America) where base cations are declining more steeply than SO_4^{2-} there are no signs of recovery. Thus the regional trends results reinforce the conclusions made by Stoddard et al. (1999 in press) (**Figure 17**) that base cation behaviour plays an important role in allowing or preventing recovery from acidification. It should be pointed out that there is no currently accepted explanation for why (or how) base cations could decline so steeply at many sites, although numerous hypotheses have been proposed (Driscoll et al. 1989; Lawrence et al. 1999 in press; Likens et al. 1996).

Table 10. *Results of trend analyses for the 10-year period 1989-1998 for 98 ICP sites, grouped by regions (unit: μeq/L/yr). Regions with no significant heterogeneity and trends significant at p <0.05 are in bold.*

Region	N	SO_4^{2-} Trend	NO_3^- Trend	ANC Trend	Hydrogen Trend	Base Cation Trend	DOC Trend
		μeq/L/yr	*μeq/L/yr*	*μeq/L/yr*	*μeq/L/yr*	*μeq/L/yr*	*mg C/L/yr*
Northern Nordic Countries	6	**-1.3** ***	-0.0	-0.7	**-0.04** ***	-1.0	+0.06
Nordic Countries/United Kingdom	24	**-2.1** ***	+0.1 †	**+2.4** ***	**-0.17** ***	**-1.0** ***	**+0.04** ***
Central Europe	34	**-2.9** ***	-0.9 ***†	+1.1 **†	+0.00	**-2.1** ***	+0.02
Eastern North America	22	**-2.4** ***	-0.1 †	+0.1 †	+0.00	**-2.5** ***	**+0.03** **
Midwestern North America	9	**-2.6** ***	0.0	**+1.6** *	-0.00	**-1.2** **	**+0.10** ***

* = p <0.05

** = p<0.01

*** = p<0.001

† = heterogeneity significant; this regional trend should be interpreted with caution.

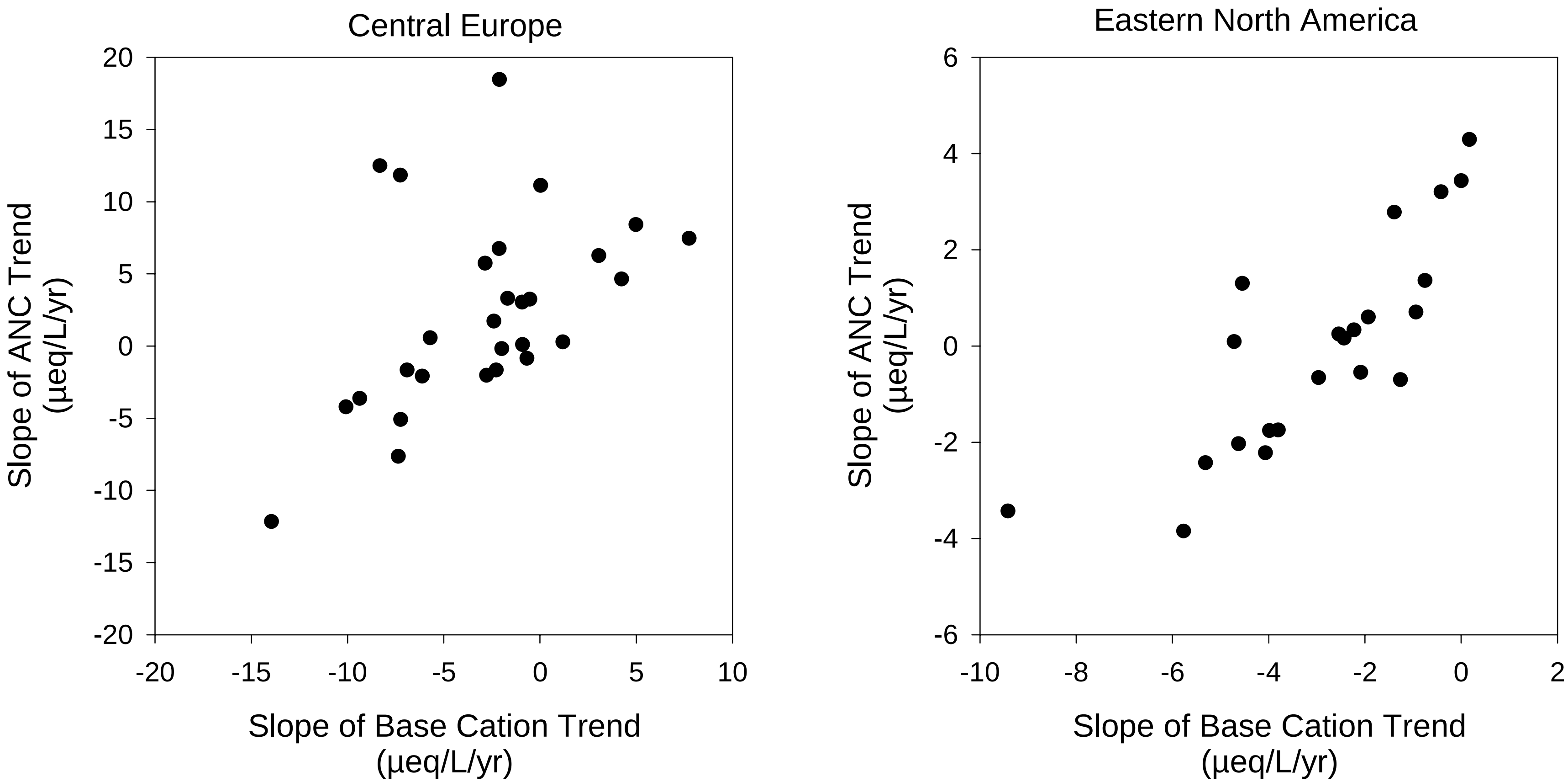


Figure 17. *Relationships between slopes of ANC trends and Base Cation trends in two regions not showing recovery. In both cases, strongly downward base cation trends are present at all of the sites with downward ANC trends.*

We also present in **Table 10** for the first time regional trends in dissolved organic carbon (DOC) concentrations at ICP Waters sites. DOC values are important indicators of the potential for lakes and streams to be organically acidified, or at least for organic acids to contribute to the overall acidity of these sites. All of the regions showed tendencies toward increasing DOC, with three of the regions showing significant increases. The mechanism causing these changes is currently unknown, although both climate and the changing chemistry of watershed soils (particularly in reaction to lowered rates of S deposition) can be suspected. One common technique for assessing the importance of organic acids is to apply a charge density of 5 µeq per mg of carbon in DOC, and to calculate the potential equivalents of charge that they represent. Using this general rule, the increases in acidity (decrease in ANC) that could be estimated to have occurred due to increases in organic acids during the decade range from 1 to 5 µeq/L (0.1 to 0.5 µeq/L/yr).

4.4.3 Central European mountain lakes

The trend analysis in this report used only sites sampled at least twice per year, which excludes most sites from the Tatra Mountains in the Slovak Republic. Because of this selection criterion, we are missing sites that show decreasing NO_3^- trends in some Central European lakes. Decreasing NO_3^- has been observed in territory close to the so called Black Triangle. Mixing these trend results with those from southwest Germany and Italy where no or increasing NO_3^- trends were observed must result in a large heterogeneity in the regional trend as it is shown in this report.

This section briefly summarises what is known up to now about the trends in S and N compounds in the atmosphere and water in Central Europe. More detailed evaluation and explanation of these trends will be completed next year within both national and European projects.

Atmospheric deposition of inorganic nitrogen (TIN = NO_3-N+NH_4-N) and sulphur (S) compounds in the former Czechoslovakia originates predominantly from emissions in Germany, Poland, Czech Republic and Slovakia (up to 85% of SO_4^{2-}, 65 % of NO_3^- and 80% of NH_4^+) (Barrett et al. 1995). Extremely high emissions of TIN and S pollutants in this region dropped by ~30% and ~55%, respectively, during the decade from 1985-1995 due to restructuring of industry and farming in post-communist countries and controlling of sulphur and NO_x emissions in Central Europe (Kopácek et al. 1997). Consequently, a decrease in deposition rates of SO_4^{2-}, NO_3^- and NH_4^+ in the Czech Republic and Slovakia was parallel to the emission decline in Central Europe. The reduction in the atmospheric input of S and N compounds to mountain ecosystems has resulted in a rapid change in the hydrochemistry of acidified lakes in the Bohemian Forest (the Šumava Mts.) and Tatra Mts. (Kopácek et al. 1998).

In 1997, lake water concentrations of SO_4^{2-} were lower by 36 and 46 µeq/L and NO_3^- concentrations by 24 and 32 µeq /L in Černé and Čertovo Lakes, respectively, compared to their 1986-1990 levels. The decline in strong acid anions was mostly compensated by declines in concentrations of Al, Ca and Mg. The decreases of nitrate concentrations in Černé and Čertovo lakes were undoubtedly connected with the decline in the N-deposition around 1990 and suggested a reversibility of N-saturation. However, the reliability between the rate of the changes in N-deposition and nitrates in lake water is complex, indicating a greater biological and chemical reactivity of N-compounds (Veselý et al. 1998a). For example elevated nitrate leaching from terrestrial sources after forest reduction (gales and insects) caused a slight increase of NO_3^- in the first half of the 1990s in the PráÜilské Lake, during the time when NO_3^- was decreasing in the remaining lakes.

Concentrations of Al and other metals have shown decreasing trends in all the Bohemian Forest lakes in parallel with increasing pH (Veselý et al. 1998b). First signs of biological recovery have been observed in erné Lake (a return of *Ceriodaphnia quadrangula*; unpublished data).

In the early 1980s, lake water pH of the Tatra Mountains lakes ranged from 4.0 to 7.4; 86% of lakes had ANC <100 µeq/L and 42% were < 25 µeq/L (Stuchlík et al. 1985). The estimated average loss of ANC (~100 µeq/L) due to atmospheric acidification was equal to the mean increase in SO_4^{2-} (70 µeq/L) and NO_3^- (30 µeq/L) concentrations between 1937 and the late 1980s (Fott et al. 1992; Kopácek and Stuchlík 1994). Concentrations of SO_4^{2-} and NO_3^- reached their maxima in the late 1980s and have been decreasing since 1989 due the reduction in S and TIN emissions (Kopácek et al. 1998). The most pronounced decrease in NO_3^- concentrations has been observed in the lakes with sparse vegetation in the watersheds (rocks or sparse alpine meadows) which are the most N-saturated among the Tatra lakes (Kopácek et al. 1995). The recent decline of concentrations of strong acid anions in lake water has been compensated predominantly by a decline in base cations; pH and ANC have been slightly increasing but are still low for biological recovery (Kopácek et al. 1998; Stuchlík and Kopácek, unpublished data).

4.4.4 The Southern Alps

The Alps represents another European environment where historical acidification has driven both hydrochemical and biological changes. The Southern Alps receive sulphur deposition of between 3 and 16 kg/ha/yr, while ammonium and nitrate deposition are in the range 1.4 to 12 and 1.6 to 10 kg N/ha/yr, respectively (Mosello and Marchetto 1996). Sulphate deposition declined by about 50% form 1984 to 1998, while nitrate increased from 1984 to 1992 and then it started to decrease.

An important feature of this region is that nitrogen deposition is higher than sulphur deposition, an occurrence expected to extend to other European areas as a consequence of decreasing sulphur emissions and deposition. Ammonium counteracts the acidity of atmospheric deposition, but it increases the potential acidity. When it reaches the soil it can be taken up by vegetation thereby releasing a mole of H^+ for each mole of ammonium, or it can be oxidised through bacterial processes with production of two moles of H^+ for every mole of ammonium oxidised (de Vries et al. 1994). Peculiar to Northern Italy and the Southern Alps are also the high concentrations of ammonium in precipitation. Episodic deposition of alkaline Saharan dust is a further important feature of atmospheric deposition in southern Europe, including Italy and the Alps (de Angelis and Gauidichet 1991; Rodà et al. 1993).

Although the atmospheric deposition of pollutants is quite high, most Italian lakes and rivers are buffered by relatively high values of alkalinity because of the geo-lithological characteristics of their watersheds. However a number of high altitude lakes in the Alps are sensitive to acid deposition because of the presence of acidic rocks, thin or absent soil and scarce vegetation in their catchments (Boggero et al. 1993). In two small, acid sensitive, alpine lakes (Lakes Paione), pH increased during the period 1984 to 1998 from 5.3-5.8 to around 6.0 and from ca. 6.0 to 6.4, respectively. A similar upward trend for alkalinity was also observed. Sulphate and NO3- did not present such clear variations, though SO_4^{2-} concentrations appear to be slightly decreasing.

The major problem affecting surface water in Italy is the atmospheric deposition of nitrogen compounds, which has led to increasing NO_3^- concentration in most Italian lakes (Mosello and Giussani 1997). In oligotrophic Lake Mergozzo from 1970 to 1978 alkalinity decreased from 0.22 to 0.18 meq/L and then increased to 0.23 meq/L in recent years. From 1984 to 1998, SO_4^{2-} decreased from 10.7 to 8.7 mg/L. In contrast, NO_3^- increased from 0.40 mg N/L in 1971 to 0.52 in 1984 and 0.67 in 1997, in good agreement with deposition trends.

High NO_3^- concentrations are also found in small rivers draining forested catchments in the Southern Alps (ca 1.5 mg N/L for Rivers Pellino and Pellesino and 0.52 mg N/L for River Cannobino), an indication that atmospheric input of nitrogen exceeds the uptake capacity of vegetation and soils. Nitrate increased by about 25% from the 1970s to the 1990s, while SO_4^{2-} decreased (Mosello et al. 2000 in press).

4.4.5 Trends – sites grouped by chemistry and site characteristics

The regional groupings of sites shown in **Table 10** are only one technique for looking for patterns in the trend data. Other regional groupings are possible, as are groupings along non-geographic lines. Several of these are shown in **Table 11**. Trends for all of the sites on the two continents (Europe and North America) are shown primarily because they highlight the difference in responses between the two major regions covered by ICP Waters. Sites on both continents exhibit remarkably similar (and very significant) decreases in SO_4^{2-} concentrations, and a similar lack of trends in NO_3^-. Despite the comparable rates of change in acidic anions, however, sites on the two continents show very different levels of recovery. There is a strong tendency (indicated by the very significant trend statistics) for ANC to increase and for H^+ to decrease (increase in pH) at sites across all of Europe, although the heterogeneity across such a large area makes definitive conclusions impossible. North American sites, however, show no regional trends in either ANC or pH. As was the case with the individual regions discussed above, the behaviour of base cations appears to play an important role in creating these continental-scale differences. The steeply declining base cation concentrations exhibited by North American sites leave little room for recovery, while the base cation trends in Europe are substantially smaller than those for SO_4^{2-} , and recovery is the predictable outcome. Interestingly, both continents exhibit very significant, and similar, increases in DOC concentrations.

It is also possible to relate rates of recovery to the levels of acid sensitivity in the sites as a whole. We examined the trends in groups of sites in three acidity classes: Acidic sites (i.e., those with ANC < 0 µeq/L); Low ANC sites (those with 0 < ANC < 50 µeq/L); and Moderate ANC sites (those with ANC > 50 µeq/L). The largest rates of recovery are expected to occur in the most sensitive sites, and the trend statistics bear this out. The acidic class exhibited the greatest decreases in SO_4^{2-} concentrations, the largest increases in ANC, and the largest decreases in hydrogen ion concentrations (**Table 11**). For both ANC and pH, the pattern was for recovery to be greatest in the acidic sites and lowest in the sites with highest ANC. DOC concentrations increased in all three classes.

Sulphate declined sharply in brown water sites (defined here as those with DOC concentrations over 5 mg/L); this category exhibited the steepest SO_4^{2-} declines of any category or region examined for this report. This large decrease in SO_4^{2-} corresponds to a large increase in ANC observed in brown water sites. Not surprisingly, these sites also exhibited the largest increases in DOC.

Approximately 25% of the ICP Waters sites have NO_3^- concentrations high enough that they play a significant role in chronic acidification (Lükewille et al. 1997); at these sites, NO_3^- represents more than 25% of total acidic anions. Importantly, neither the high NO_3^- or low NO_3^- groups of sites exhibited significant trends in NO_3^- concentrations (both groups showed considerable trend variability). Both groups showed decreases in SO_4^{2-} (as did all of the groups and regions analysed for this report), and tendencies toward recovery.

Non-forested sites, particularly those at high elevations, are often considered to be excellent sites for measuring environmental change, because soil and forest mechanisms that can complicate the detection of chemical changes over time at forested sites are minimised. Our trend data support this concept (**Table 11**). Despite having lower rates of SO_4^{2-} decline than forested sites (due most likely to their location in areas of lower rates of deposition), non-forested sites show clear and consistent signals of recovery in ANC and pH, and appropriate (relative to SO_4^{2-} trends) rates of base cation declines. They also exhibit significant increases in DOC that are larger than those in the forested group; as mentioned earlier, the mechanisms that cause observed increases in DOC are largely unknown.

Finally, it is worthwhile to confirm that the recovery we observe is, in fact, associated with declining SO_4^{2-}. We examined the trends in sites where there is strong evidence of decreasing SO_4^{2-} (e.g., those with SO_4^{2-} Z scores less than -1) vs. those where SO_4^{2-} changes are more ambiguous. There was a clear tendency for sites with decreasing SO_4^{2-} to exhibit increasing ANC and decreasing hydrogen ion concentrations, while those with no SO_4^{2-} changes showed no pattern of recovery. While ICP Waters data are not sufficient to show cause and effect, the patterns shown in **Table 11** are consistent with recovery being the direct result of emissions reductions and resulting declines in surface water SO_4^{2-} concentrations.

Table 11. *Results of trend analyses for the 10-year period 1989-1998 for 98 ICP sites, grouped by continent and by several chemical criteria. Regions with no significant heterogeneity and trends significant at $p < 0.05$ are in bold.*

Category	N	SO_4^{2-} Trend	NO_3^- Trend	ANC Trend	Hydrogen Trend	Base Cation Trend	DOC Trend
		µeq/L/yr	*µeq/L/yr*	*µeq/L/yr*	*µeq/L/yr*	*µeq/L/yr*	*mg C/L/yr*
Europe	64	**-2.3** ***	-0.0 **†	+1.9 ***†	-0.03 ***†	**-1.5** ***	**+0.06** ***
North America	31	**-2.4** ***	-0.0 †	+0.3 †	-0.00	**-2.1** ***	**+0.04** ***
Acidic (ANC < 0 µeq/L)	27	**-3.7** ***	-0.6 ***†	**+3.0** ***	**-0.34** ***	**-2.1** ***	**+0.10** ***
Low ANC (0 < ANC <50 µeq/L)	30	**-2.0** ***	**+0.0** **	**+1.1** ***	**-0.04** ***	**-1.0** ***	**+0.10** ***
Moderate ANC (ANC > 50 µeq/L)	38	**-2.2** ***	-0.0 **†	-1.1 †	**-0.00** ***	-2.3 ***†	**+0.03** **
Brown water (DOC > 5 mg/L)	23	**-4.2** ***	-0.0	**+3.0**	-0.03 **†	**-2.1** ***	**+0.18** ***
Clear Water (DOC < 5 mg/L)	64	**-2.0** ***	-0.0 †	**+1.2**	**-0.01** ***	-1.7 ***†	**+0.04** ***
High Nitrate (NO_3^- > 25% of acid anions)	26	**-1.2** ***	+0.0 †	**+0.9** **	**-0.01** ***	-0.8 **†	+0.02
Low Nitrate (NO_3^- < 25% of acid anions)	69	**-2.9** ***	-0.0 †	+1.4 ***†	-0.02 ***†	-2.1 ***†	**+0.08** ***
Forested Watersheds (Forest cover > 15%)	80	**-2.5** ***	-0.0 *†	+1.6 ***†	-0.0 ***†	-2.1 ***†	**+0.04** ***
Unforested/Alpine Watersheds (Forest cover < 15%)	13	**-1.2** ***	+0.0 †	**+1.9** **	**-0.15** ***	**-0.9** ***	**+0.10** ***
Strong sulphate decreases	80	**-2.9** ***	-0.0 †	**+1.7** ***	-0.03 ***†	**-1.8** ***	**+0.05** ***
No significant change in Sulphate	15	+0.0	-0.0 †	-1.3	-0.00	-0.8 †	**+0.05** *

* = $p < 0.05$

** = $p < 0.01$

*** = $p < 0.001$

† = heterogeneity significant; this categorical trend should be interpreted with caution.

5. Effects of acidification on aquatic fauna: characteristics, geographical extent, dose/response, relationships, and long-term trends

5.1 General overview of the status

In Chapter 4 the focus is on changes in acidification of water that are related to changes in acid deposition. In areas of Europe with decreasing SO_4^{2-}, generally ANC has increased and hydrogen ion concentrations decreased. The question is whether the reductions in sulphur emissions, reductions in sulphur deposition and improvements in surface water chemistry are reflected in biological components of the aquatic ecosystems. Since one of the main aims of reducing S-emissions is to restore damaged ecosystems, documentation of recovery of biological components is especially important. The basis for this evaluation rests on the critical load concept which is defined as *"the highest deposition of a compound that will not cause chemical changes leading to long-term harmful effects on ecosystem structure and function"* (Nilsson 1986).

Here we have treated sites/regions for which at least 10 years of biological data exist. Two types of biological data have been used (1) material that officially has been delivered to the Programme Centre and (2) published biological data from ICP Water sites, sampled under other programs, such as AL:PE, AL:PE2 and MOLAR (Wathne and Rosseland 1999). This additional material supports the database and is important for evaluating the representativeness, geographical extent of acidification and the dose/response relationships.

The relationship between water chemistry and biology (mainly invertebrate fauna) is well-established and for ICP Water has previously been summarised by Wathne (1991), Skjelkvåle et al. (1994) and Lükewille et al. (1997). Different strategies have been developed for monitoring trends, geographical extent and dose/response relationships of acidification based on the aquatic fauna. The strategies range from simple acidification score systems, evaluated by Raddum (1999) and Wiederholm (1999), to more complex multivariate techniques (Heegaard 1999). The basis for biological monitoring in the ICP Water program has been an acidity index, based on presence/absence of sensitive species of invertebrates (Norwegian Institute for Water Research 1996; Raddum 1999). Through this method the degree and geographical extent of biological damages have been established as well as dose/response relationships and trends in acidification based on sensitive species (Raddum and Skjelkvåle 1995; Lien et al. 1996; Lükewille et al. 1997 and Raddum 1999).

Multivariate techniques have been used for evaluating the connection between invertebrates and water chemistry and the importance of different chemical compounds (Larsen et al. 1996). These techniques have the potential for giving better resolution than the acidity index, but require harmonised data with respect to quantity, level of species identification, habitat, etc. The requirements for these analyses are almost impossible to meet for all regions in a large monitoring program due to different practices among the different participants. Within a country or a specific region, however, the requirements can be met and when this is the case, multivariate techniques have been used.

5.2 Methods used for evaluations

Critical load and trends of acidification have been based on an acidity index for aquatic invertebrates, a basic level of analysis where all types of data can be used. To meet the critical load concept of Nilsson (1986) we have chosen the score of 1 for the acidity index as the limit for no harmful effects on ecosystem structure and function. This is a conservative estimate, as the index does not take into account sublethal effects of acidification on the invertebrate community. The acidity index score of 1 is compared with the ANC values. The evaluations are carried out for spring and autumn samples. Spring is defined as the ANC for March, April and May samples, while autumn is defined by the September, October and November measurements.

For trend analysis simple linear regression has been performed for the relation between year and the acidity index.

A second level of analysis is to use multivariate techniques to detect the importance of different ions in the surface water for the invertebrate community. A third level is to use multivariate techniques to investigate if changes over time in water chemistry are reflected in the invertebrate community. This analysis weights the importance of water chemistry in relation to other factors for the recorded trend development.

5.3 Results

5.3.1 Extent and critical limits of acidification

Since the ICP Waters sites in Norway, Ireland, and the UK are situated mostly on bedrock sensitive to acidification, buffer capacity is low and yearly mean calcium concentrations vary mostly between 0.3 to 1 mg/ L. High similarity is also found for other compounds. The Norwegian and UK sites are chemically as well as biologically quite similar. The critical limit in Norway is set at ANC = 20 µeq/L, the level at which there is 10% probability of damage for brown trout populations. We propose the same critical limit for the UK. In Posch et al. (1999) the critical limit for water in UK is set to ANC = 0 µeq/L. This value is selected for 50% probability for damage to brown trout populations. The discrepancy between our critical limit and that of Posch et al. (1999) is a result of the different damage level used for the two evaluations.

Extensive work has been done on critical loads of acid deposition for UK freshwaters and the regressions between biological status and ANC, critical ANC values to ensure biological status and the predictions for biological status for stipulated ANC values (Patrick et al. 1995). Changes in the biological communities occur along an ANC gradient from -100 to 200 µeq/L. To meet the critical load concept by Nilsson (1986), the critical limit should probably be ~50 µeq/L to prevent any impoverishment of invertebrate assemblages (Juggins et al. 1995). The score of 1 for the invertebrate acidity index provides a clear definition of the critical limit related to direct toxic effect of acid water on some of the most sensitive organisms. Beyond the score of 1, the sensitive species will be related to a large number of both abiotic and biotic factors. Productivity, competition, predation etc. will be the main factors determining the biological community. Productivity is usually connected with Ca concentration and consequently ANC. Many species have minimum requirements for Ca, and the number of species is correlated with Ca concentration. The acidity index does not evaluate this type of locality or biological community, as these are attributed to ecosystems with low susceptibility to acidification. The invertebrate acidity index provides the basis for a common evaluation and understanding of critical limits that can be used for large areas. Different countries can, of course, chose other damage levels for critical limits, but when comparing large regions a common level is necessary.

The Irish fauna consists of species typical for the littoral zone of lakes and running waters. Most of the sites containing very sensitive organisms score 1, indicating no or little acidification. **Table 12** shows the sites and acidity index for 1998, illustrating the good situation in Ireland. The locality GLE 13, however, contains no sensitive species and thus reflects a strongly acidified site. The situation recorded for 1998 has been more or less the same since 1987, but occasional presence of sensitive species has been noted in GLE 13 during the period. The sensitive organisms have, however, not been able to establish any permanent population at the locality.

Table 12. *Acidification status for Irish sites in 1998.*

Lake	ICP site	Date	Station	Station	Station	Station	Station
Glendalough Lake Upper	**IE01**		**GLE 11**	**GLE 13**	**Shore**		
		03.02.98	1	0	0		
		21.05.98	1	0	0		
		08.12.98	1	0	0		
Lough Maumwee	**IE05**		**MAU 10**	**MAU 11**	**MAU 12**		
		07.05.98	1	1	1		
		25.11.98	1	1	1		
Lough Veagh	**IE08**		**VEA 11**	**VEA 12**	**VEA 13**	**VEA 14**	**Shore**
		05.05.98	1	1	1	1	1
		25.11.98	1	1	1	1	1

The faunal composition of the UK sites is generally similar to the composition recorded in Ireland. Most of the UK sites, however, are more acidic, resulting in a lower number of sensitive individuals than found in the Irish sites. Also in the UK, a significant relationship between the species composition and water chemistry is recorded. A redundancy analysis including all of the UK sites for the whole 10-year period and the environmental variables pH, Ca, DOC, and ANC, gave a significant ordination ($p = 0.05$), explaining 26.7 % of the variance in the abundance data. All included environmental variables, except ANC, contributed significantly to the ordination. This is consistent with similar analyses and results from Norway (Larsen et al. 1996).

There are differences in the species composition between Norway, UK and Ireland due to the geographical distribution patterns. On the other hand, there are also considerable similarities, especially between the ICP sites. The reason for this is probably that the chosen sites have low buffer capacity, low calcium content etc., giving almost the same chemical environment (Raddum and Skjelkvåle 1995; Lükewille et al. 1997).

The relationship between ANC and the acidity indexes for the sites in UK are shown in **Figure 18**.

The number of observations for index 0 and 0.5 is 23 and 28 respectively, but only 6 for index 1. Since the ANC value for index 1 corresponds to the critical limit, large uncertainties are connected to this limit due to the low number of observations. However, the ANC values for the different acidity indexes for the UK sites, are similar to those found for water in Norway (Lien et al. 1996).

The Irish sites are biologically similar to the UK as well as the Western Norway sites. The biological material from Ireland is either from non-acidified or strongly acidified systems. Therefore a proper ANC gradient for evaluation of the critical limit for acidity index 1 is lacking. In the absense of more information, the critical limit for water in Ireland should be set similar to that proposed for UK and Norway.

For Sweden and central Europe ANC ≈50 µeq/L has been proposed for water (Raddum and Skjelkvåle 1995; Lükewille et al. 1997). This was based on available biological material from ICP Waters sites until 1995. Additional material up to 1998 for Sweden does not indicate that this level should be changed.

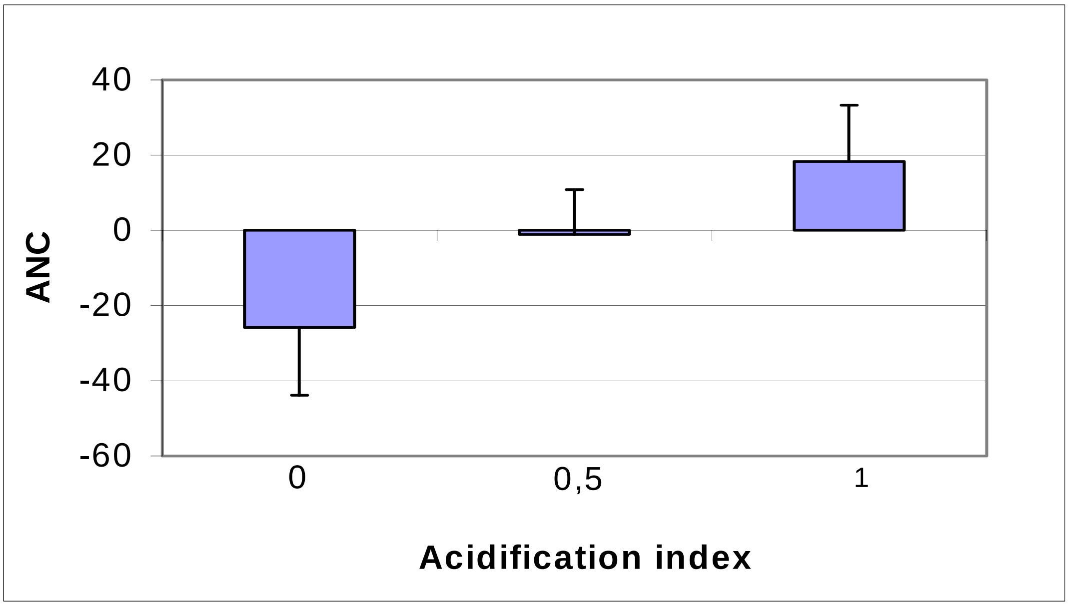


Figure 18. *Mean ANC for samples with the acidification score 0, 0.5 and 1 in ICP Waters sites in UK. Standard deviation is indicated.*

For Germany, as representative of central Europe, the newer material also supports the earlier proposed ANC level. Some of the sites in Germany are very acid, pH < 4, while others have pH > 7. They are also characterised by large fluctuations in the other chemical variables. This is reflected in the numbers of species present in the invertebrate communities, which range from almost none to very species-rich sites. In some of the most damaged sites only 2 - 5 taxa have been recorded, while in the less damaged sites > 40 taxa are found. This large variation in numbers of species is not recorded in the other countries. Recovery of biota in the seriously acidic German sites can therefore be more difficult, or take longer time, than recovery of the most acidic sites in other regions.

From France there is information on the extent of acidification in the Vosges Mountains (Guerold et al. 1999). This region is located in northeastern France close to the German border and can be regarded as a part of the central Europe. In this region, damages to the invertebrate fauna began at mean pH values below 6.5. At ANC values of 50 µeq/L, clear damages were seen in some streams. The results from the Vosges Mountains are similar to those from Germany both with respect to critical pH and ANC levels for sensitive invertebrates. The investigations by Guerold et al. (1999), therefore, support the proposed critical limit of ANC ≈50 µeq/L for central Europe.

Biological and chemical data also exist from other mountain regions of Europe. Some of the sites investigated in conjunction with other projects are also ICP Waters sites. From the Italian and Austrian Alps, data exist from the ICP sites Paione Superiore, Paione Inferiore and Schwarzee ob Sölden (Wathne and Rosseland 1999; Fjellheim et al. 2000 in press). From the Pyrenees we have the Spanish ICP site Lake Redó and the French site Lake Aubé, while in the Tatra Mountains, biological material exist from the ICP site Dlugi Staw. Analysis of the distribution of sensitive species from these regions show that the number of sensitive organisms is generally low in the European mountain lakes compared with non-acidified reference lakes in Norway (Fjellheim et al. 2000 in press). Lake Redó in the Pyrenees also hosted several acid-sensitive species, while areas in the Alps had reduced number of sensitive species. Lowest number of sensitive species was recorded in the Tatras.

Correlation analysis between pH and number of sensitive taxa indicated highest number of sensitive species at pH 6.5 (Fjellheim et al. 2000 in press). This is consistent with the results from the Vosges Mountains. Lake Redó and Paione Inferiore had acidity index 1 for all periods sampled and contained several taxa of the most sensitive organisms. The corresponding ANC values for Lake Redó for spring and fall in 1998 were 46 and 73 µeq/L, respectively. For Paione Inferiore, data exist from 1989 to 1998, showing ANC of 30 and 32 µeq/L during spring and autumn, respectively. In Paione Superiore, however, clear damages were observed on the most sensitive species. Here the mean ANC for was 1 µeq/L and 4 µeq/L during spring and autumn, respectively.

Altitude influences the distribution of species in these mountain areas (Fjellheim et al. 2000 in press). This complicates the evaluation of critical limits in mountain areas. The results so far from the ICP sites, however, indicate that the ANC in Lake Redó and Paione Inferiore is acceptable, while it is too low in Paione Superiore. We suggest that the critical limits should be close to 30 µeq/L in the high Alps.

Biological material from Latvia contains many sensitive species and indicates no acidification. Both pH and ANC are far above levels expected to be critical for invertebrates; ANC values are between 2000 and 4000 µeq/L. The sites are valuable as references for water with very high ionic strength. Compared with the other lowland regions, the sensitive mussels and snails are more frequent in the Latvian waters than in other regions. Also several sensitive insect larvae of mayflies are present, while stoneflies appear to be absent. Since stoneflies are indicators of unpolluted oligotrophic water, the Latvian sites seem to be affected by high loads of organic material. This is also indicated by the high abundance of oligochaets. Typical for productive water is also the high abundance of leeches recorded in the Latvian sites. The invertebrate composition in the localities indicates that the sites are more suitable for monitoring eutrophication or organic pollution. But they also provide important examples for the ICP Waters programme of well-buffered ecosystems far above the critical limit for acidity.

5.3.2 Trends in acidification based on the acidity index

The biological data from Ireland, UK, Norway, Sweden and Germany allow evaluation of trends. This is done by use of the acidity index and by multivariate techniques on material that fulfils the requirements for this type of analysis, see chapter 5.3.3.

Ireland

From Ireland we will focus on the development of *Baetis rhodani,* one of the most widespread and sensitive species. At the non-acidic site GLE11 (ICP site number IE01) the number of individuals recorded was < 200 during spring in 1987 and 1988, while the corresponding number in 1993 and 1995 was > 600 (**Figure 19)**. The average pH for the spring in 1989 was 5.8. Occasional pulses of water with increased or decreased acidity are to be expected in this region depending on precipitation associated with easterly or westerly air masses. *B. rhodani* can be exposed to sublethal stress at pH below 6 in clear water (Raddum and Fjellheim 1984). The increased abundance of *B. rhodani* at GLE11 is possibly due to an improvement in water quality, changing from sublethal to a quality above the critical limit of the species. There are, however, many other factors independent of changes in water chemistry that might explain the changes. It is therefore difficult to conclude that the improvement is due to decreased acidification.

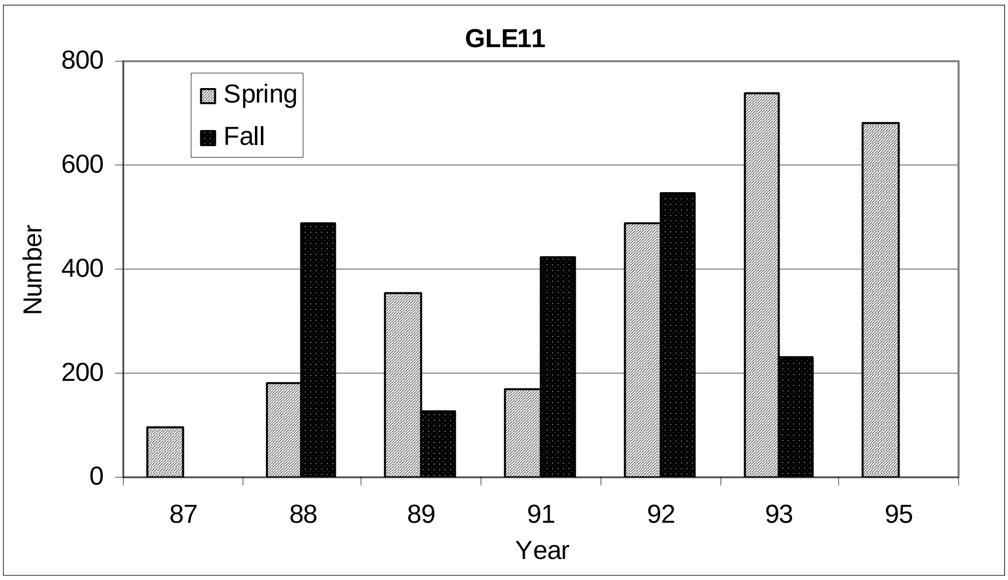
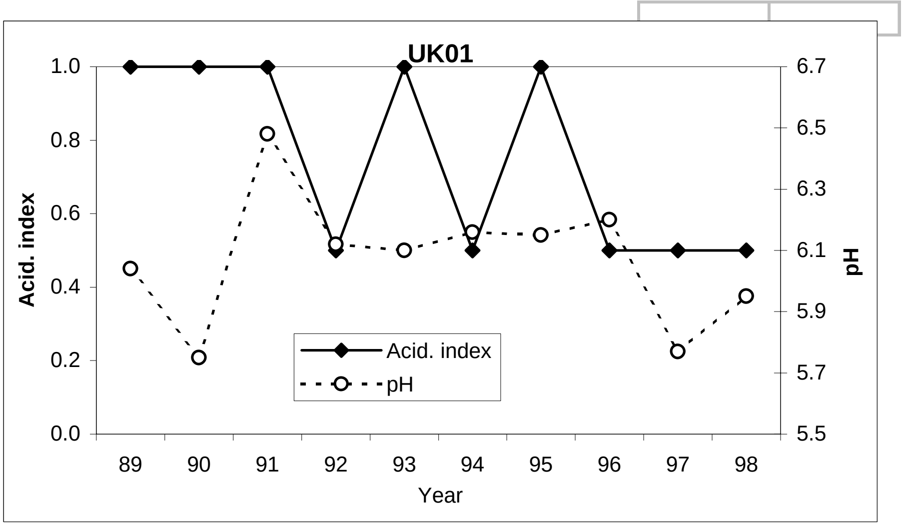
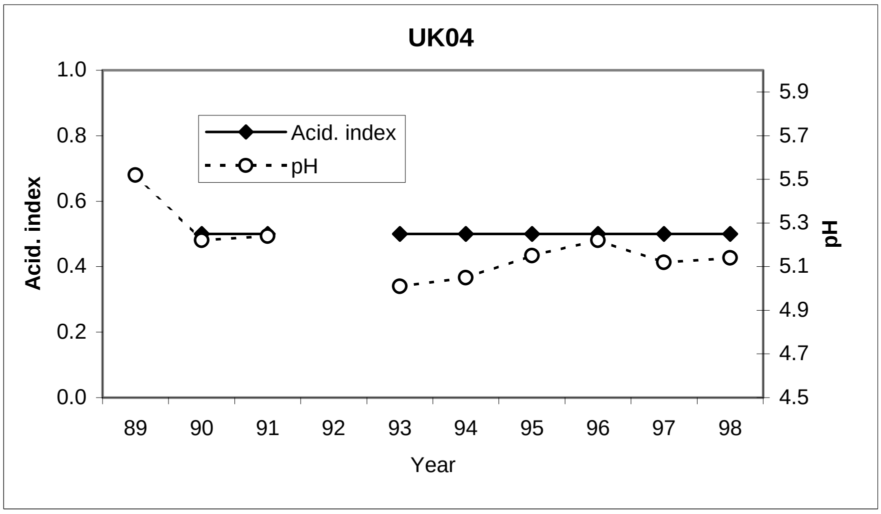


Figure 19. *Development of abundance of B. rhodani in GLE 11 (ICP site number IE01) during 1987-1995.*

UK

The acidity index and pH for the period 1989 to 1998 in the UK sites are shown in **Figure 20** and **Figure 21**. UK01 has the highest pH, varying between 5.7 and 6.5. The index was 1 from 1989 to 1991, varying between 1 and 0.5 from 1992 to 1995, while after 1995 the index decreased to 0.5, indicating a negative trend ($r^2 = 0.43$). No trend was recorded for pH. In the sites UK04 and UK15 the acidification indices have been 0.5 through the whole period. In the same period the mean pH has been within the range for this acidification level. In the other UK sites UK07, UK10 and UK21 the mean pH has stayed mostly below 5 with only minor changes. The acidity index has been 0 for all these sites throughout the whole period in accordance with the pH level of the sites. There have been changes in the invertebrate assemblage of the UK sites, but these variations have all been among species indicating the same acidification level. Factors other than acidity are therefore likely to be the reason for these changes. The main conclusion for UK so far is that the sensitive fauna do not show any significant change in acidification status.





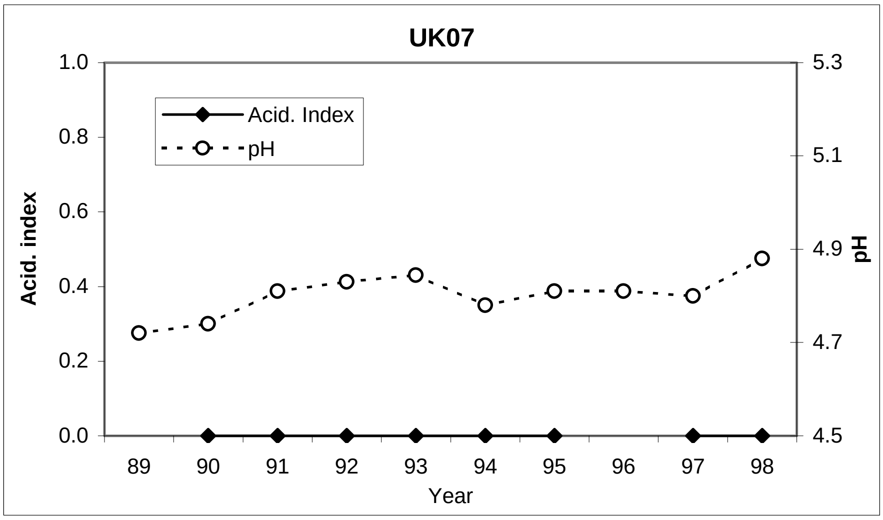
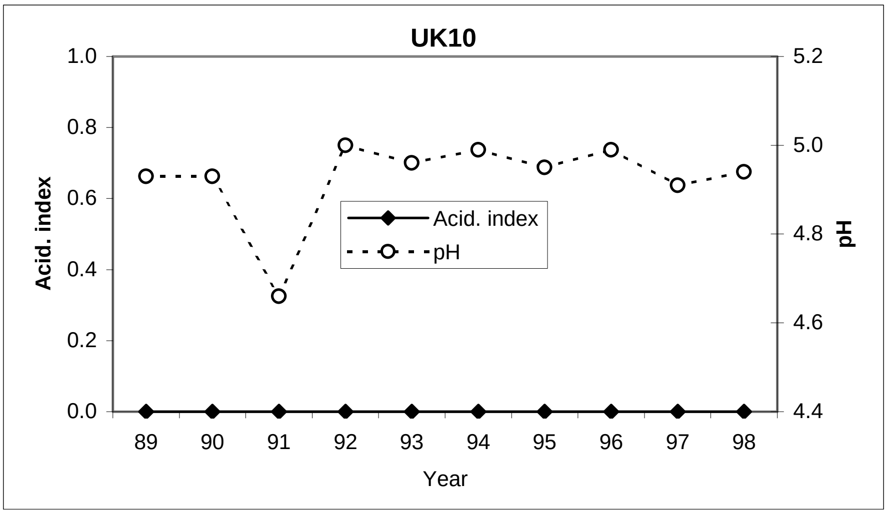
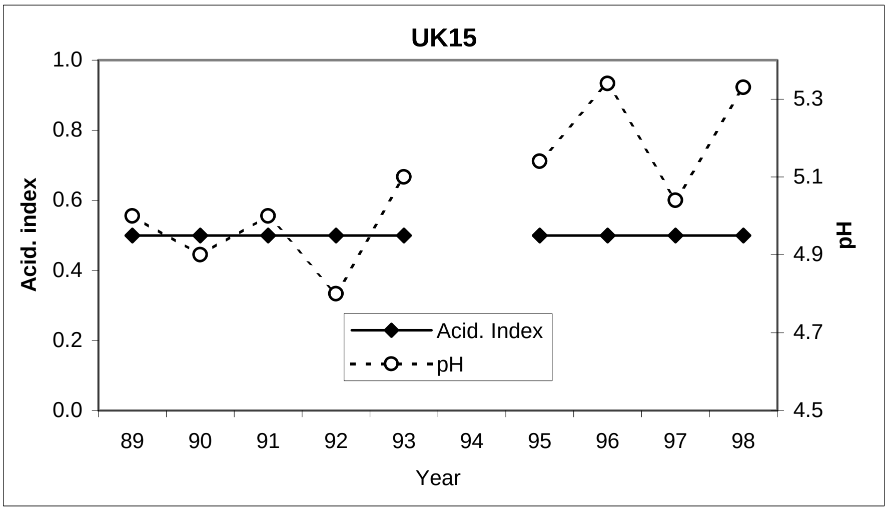


Figure 20. *Mean pH and acidity index for the period 1989 to 1998 in UK01, UK04 and UK07.*





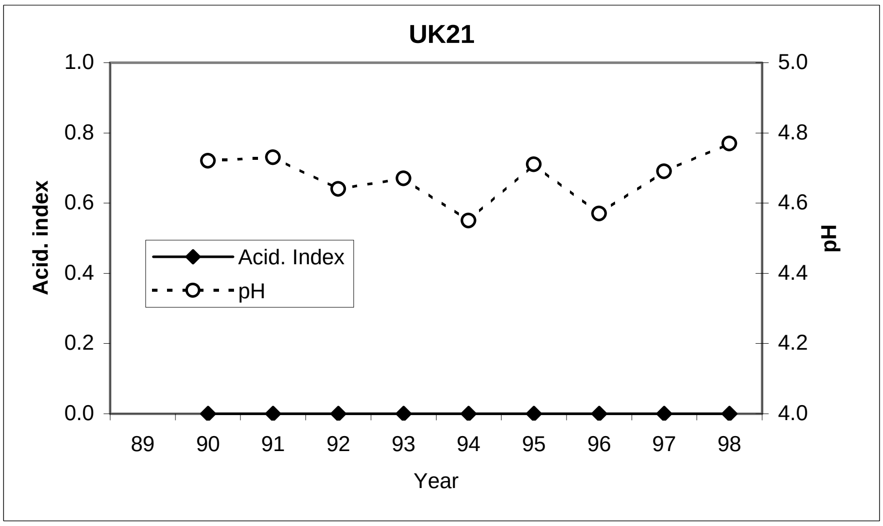


Figure 21. *Mean pH and acidity index for the period 1989 to 1998 in UK10, UK15 and UK21.*

Sweden
The mean acidity index for the Swedish sites has been estimated for the period 1985-1989, 1990-1995, 1997 and 1998 **Figure 22**. Mean pH for the different sites through the whole period is shown in **Figure 23.**

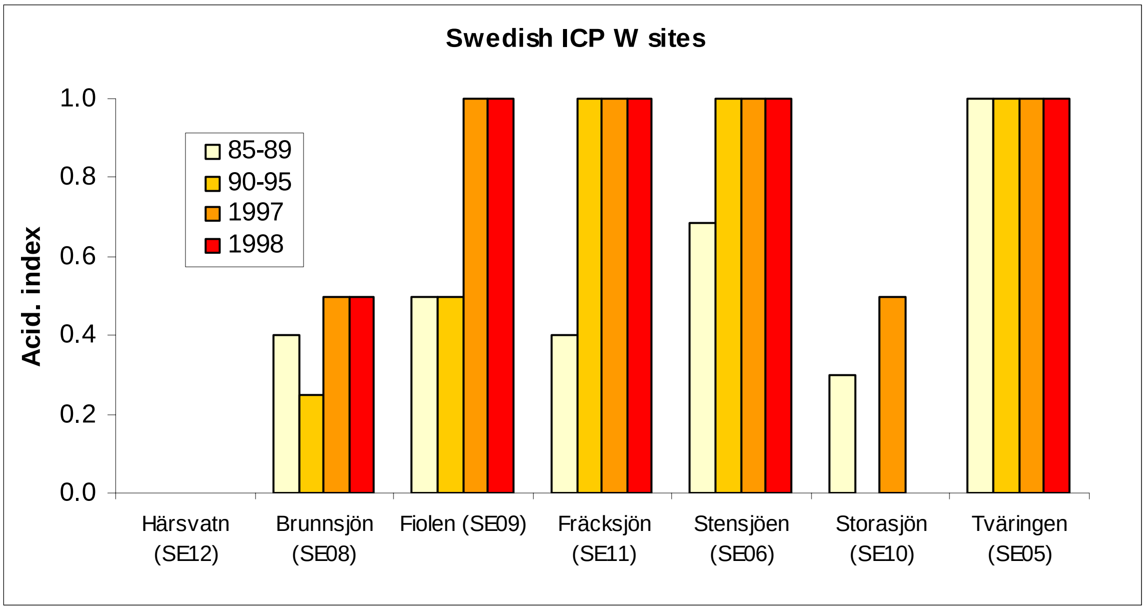


Figure 22. *Acidity index for different periods in Swedish ICP Waters sites.*

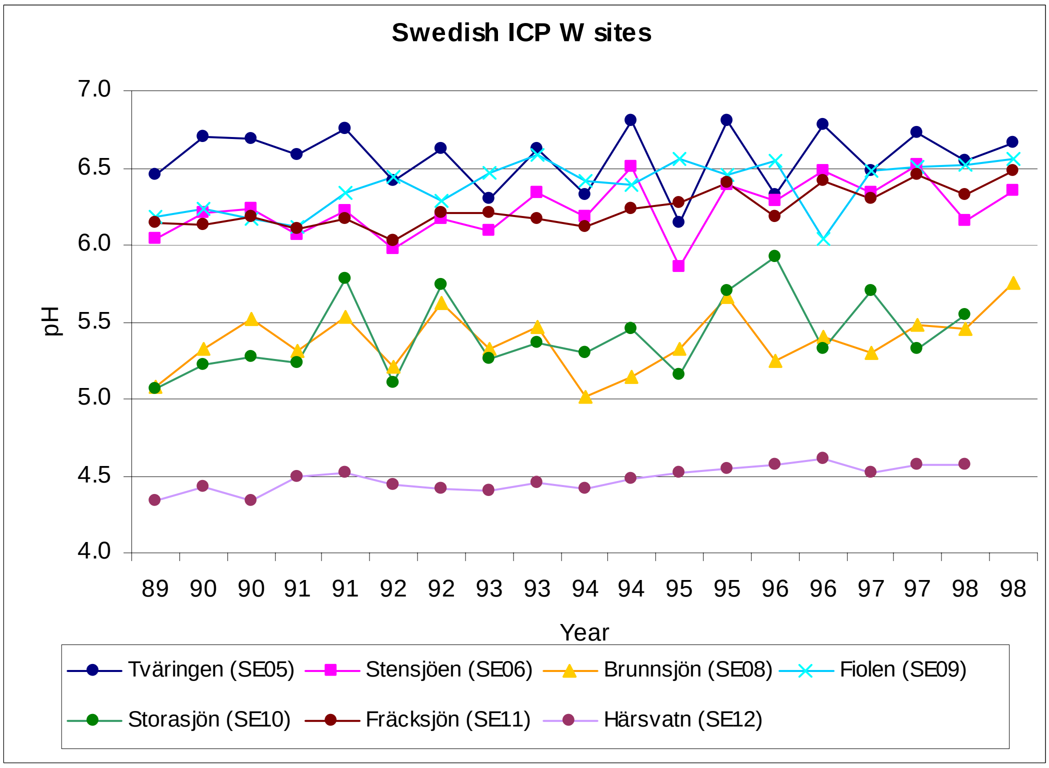


Figure 23. *Mean pH for the Swedish ICP Waters sites from 1989 to 1998.*

In Härsvatn (SE12) the acidity index has been 0 during the whole period. The mean pH has increased from 4.34 in 1989 to a maximum of 4.61 in 1996. The improvements are still within the score 0 range.

In Lake Brunnsjön (SE08) only small changes have occurred. At the start of the period the index value fluctuated between 0.25 and 0.5, while in 1997 and 1998 the index was 0.5. The mean pH shows no trend with time and varied within the range typical for index 0.5.

In Lake Fiolen (SE09) the acidity index increased from 0.5 during 1985-1995 to 1 in 1997 and 1998. The pH in the lake has been above the critical level for index 1 through the whole period. In the start the mean pH was 6.2 and at the end to 6.6. The positive trend with time has a $r^2 = 0.33$.

In Fräcksjön (SE11), Stensjön (SE06) and Tväringen (SE05) the acidity index has been 1 after 1990. The pH in Tväringen has been about 6.5 throughout the period without any trend. In Stensjön the lowest mean pH was 6.0 and 6.4 in 1989 and 1998, respectively. The trend analyses for mean pH over time gave a $r^2 = 0.62$ indicating an increase in pH of the lake. In Fräksjön the mean pH was 6.1 and 6.5 in 1989 and 1998, respectively. The trend analysis showed $r^2 = 0.62$. The improvements in the acidity index can therefor be explained with improvements in water quality.

Norway

In Norway biological material exists from the early 1980s in the Farsund (not an ICP site), Gaula (NO07) and Nausta (NO08) watersheds. The mean acidity index for Farsund is shown in **Figure 24** for spring and autumn.

During the past 10 years, significant improvements in water quality have been recorded in southern Norway as well as elsewhere in Scandinavia. We therefore divide the biological material into two periods, before and after 1989, respectively. For Farsund the acidity index in spring samples varied until 1989 and showed a weak negative trend ($r^2 = 0.20$). In autumn samples there was no trend for this period. From 1989 to 1998 the acidity index has increased both for spring and autumn concurrent with reduction in sulphur deposition and increased ANC. The situation during spring varies and events such as the seasalt episode during winter 1993 (Hindar et al. 1994) are reflected in the acidity index (**Figure 25**).

The trends both for spring and autumn are similar, but the significance of the trend differs, $r^2 = 0.63$ and 0.94 respectively. The results from Farsund indicate that the improvements of the invertebrate community started around 1990. Up to 1998 the improvements have led to stabilization of sensitive species indicating acidification score 0.5. It will probably take some time before the most sensitive species will be recorded in stable populations in the watershed.

In Nausta the acidity index increased over the period 1983 to 1998 (**Figure 26**). During spring the lowest values were recorded in 1983-1984 and 1989-1990, illustrating unstable conditions. Several seasalt events have been observed connected with the decreases in the acidity index. During autumn the lowest index was found in 1989. After that the index increased to 1 in 1994 and has stayed at 1 through 1997, indicating little or no damages to the invertebrate community.

For comparison with the development in water chemistry, the trend in the acidity index after 1989 is illustrated in **Figure 27**. For this period, the index shows an improvement ($r^2 = 0.69$) especially during spring. The index mostly indicated no or low acidification for autumn samples. Improvements beyond this level will not be recorded which explains why no trend exists after 1994.

For Gaular watershed, the trend development is similar to Nausta (**Figure 28)**. In this watershed we have about the same improvements for both spring and autumn. The highest acidification score in Gaular was 0.88 in autumn 1998, indicating still acidified sites in the watershed.

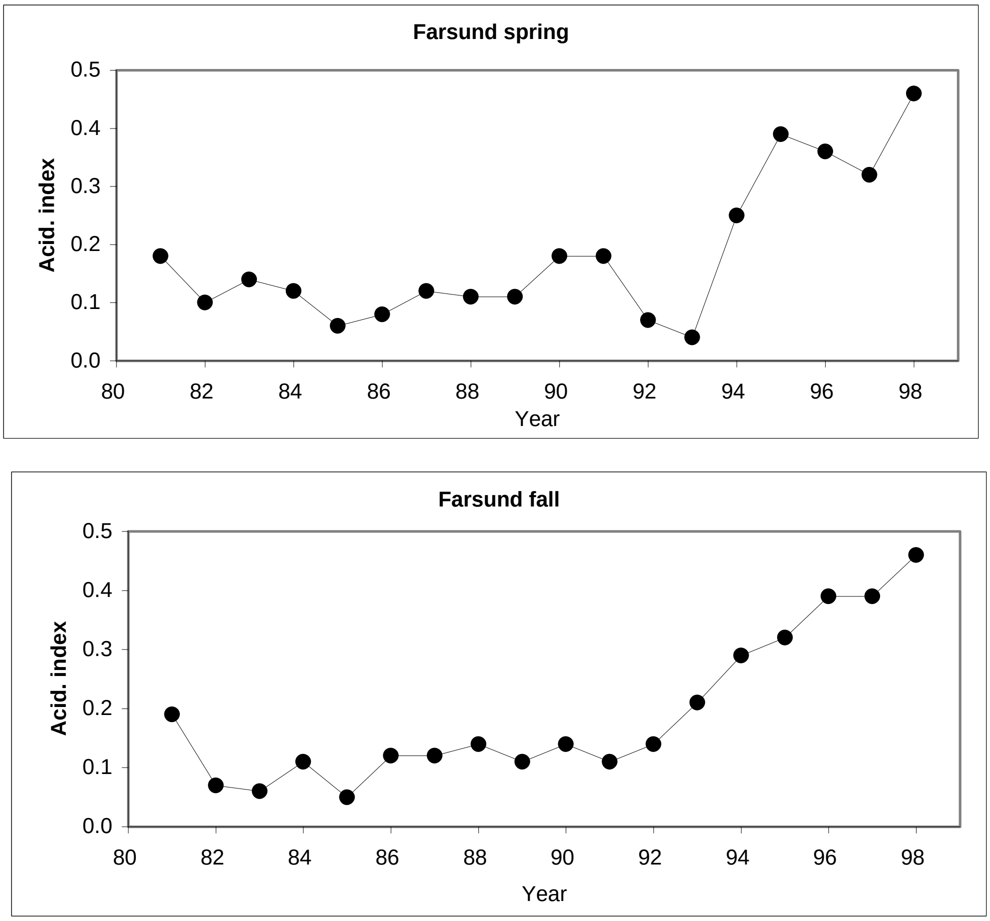


Figure 24. *Development of the acidity index during spring (upper panel) and autumn (bottom panel) in Farsund.*

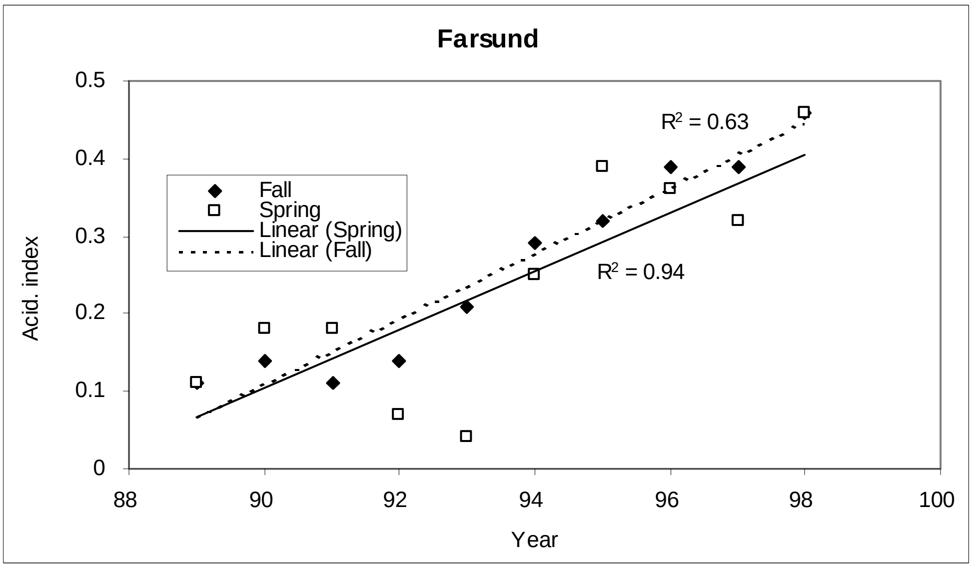
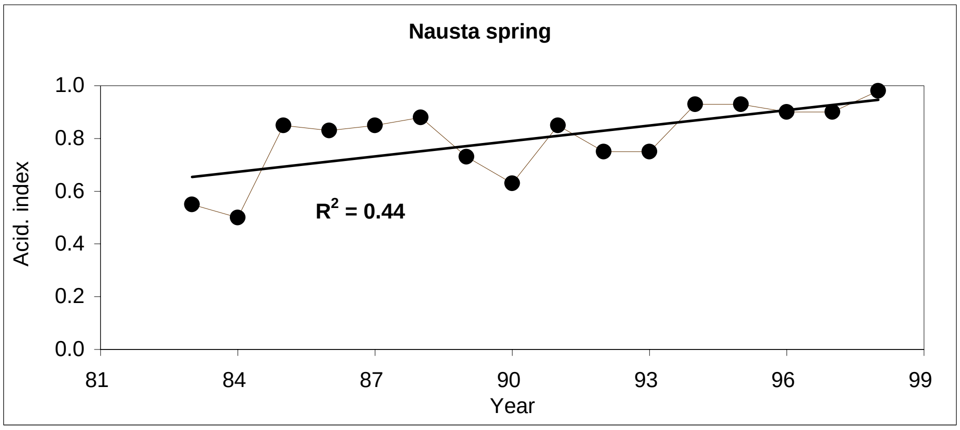


Figure 25. *Correlation between the acidity index and time for spring and autumn in the Farsund watershed.*



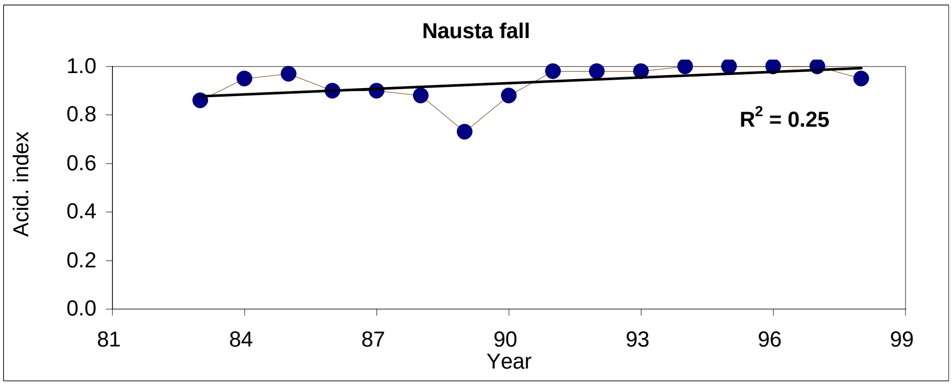


Figure 26. *Development of the acidity index in Nausta during spring and autumn.*

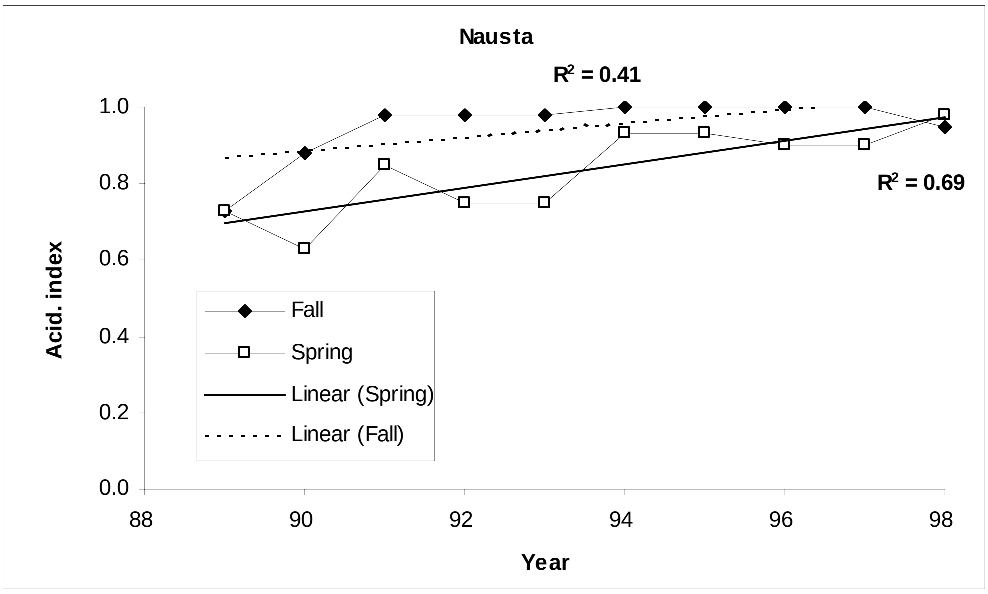


Figure 27. *Trend in the acidity index after 1989 in Nausta.*

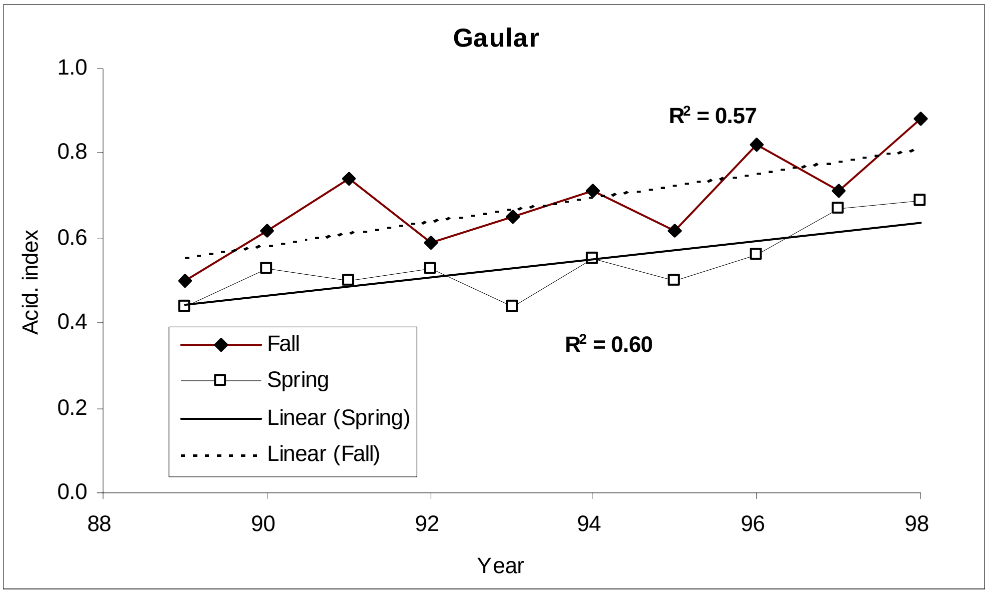


Figure 28. *Trend in the acidity index after 1989 in Gaular.*

Germany

Trends have been analysed for the mean acidity index of sites in the Black Forest and East Bavaria (**Figure 29**). The number of sites has varied through the period and this could influence the trends, but no sites have shown any stable improvements. In the Black Forest no general trend is seen, while a slightly positive trend (r^2 = 0.15) has occured in East Bavaria. The acidity index from Eger, Röslau and Zinnbach shows an index of 0.5 (stable) for Eger, 0.25 - 0.5 (varying) for Röslau, and 0 (stable) for Zinnbach (**Figure 29**).

The situation in Harz for Grosse Schacht, Grosse Bode and Grosse Söse is illustrated in **Figure 30** and for Rothaargebirge, Elbendorfer Bach, in **Figure 31**. Here the acidification class of Kifinger et al. (1998) is used for the trend analyses. The sites in Harz all show a small positive trend similar to that recorded for East Bavaria. In most cases the acidification class was 2 or 3. During the first half of the monitored period, class 4 (strong acidification) occurred several times, while in the second half of the period the class 1 (low or no acidification) was observed. However, $r^2 \approx 0.15$ indicates only little or no significant improvement. In Elbendorfer Bach, however, r^2 = 0.39, which is the highest significant positive trend among the German sites.

By using the acidification class method, Kifinger et al. (1998) found 15 sites with no improvement and 17 with small or some improvements. It is difficult, however, to point to significant and stabilised positive trends in the German biological data. The improvments observed are weak and occur in acid-damaged communities. This corresponds with the results from the water chemical analysis. One reason for the lack of stabilised improvements could be that many of the sites have water chemistry varying around the limit where biological changes can occur. This makes it difficult for sensitive organisms to develop stable populations. Permanent improvements in the invertebrate community can not happen before water chemistry reaches stable levels above the critical limits for the various invertebrate species. When this happens, a more permanent development of sensitive species can be expected.

The Program Centre has received data through 1998 from Germany. So far these latest data do not appear to indicate major changes in the acidification status in Germany.

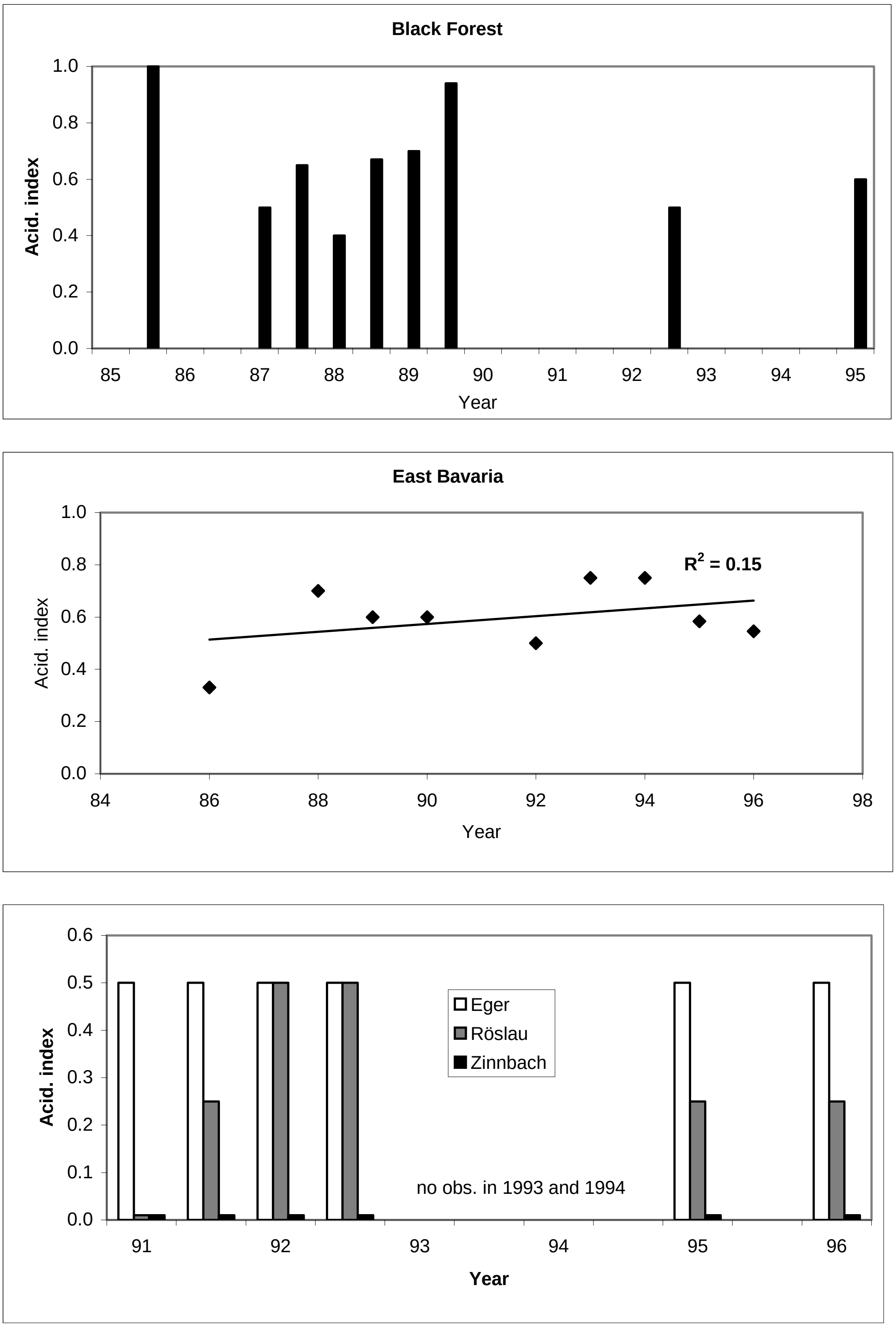


Figure 29. *Trends in the acidity index for different regions in Germany. At the bottom panel, Zinnbach has an acidity index = 0, and it is therfore not seen at the figure.*

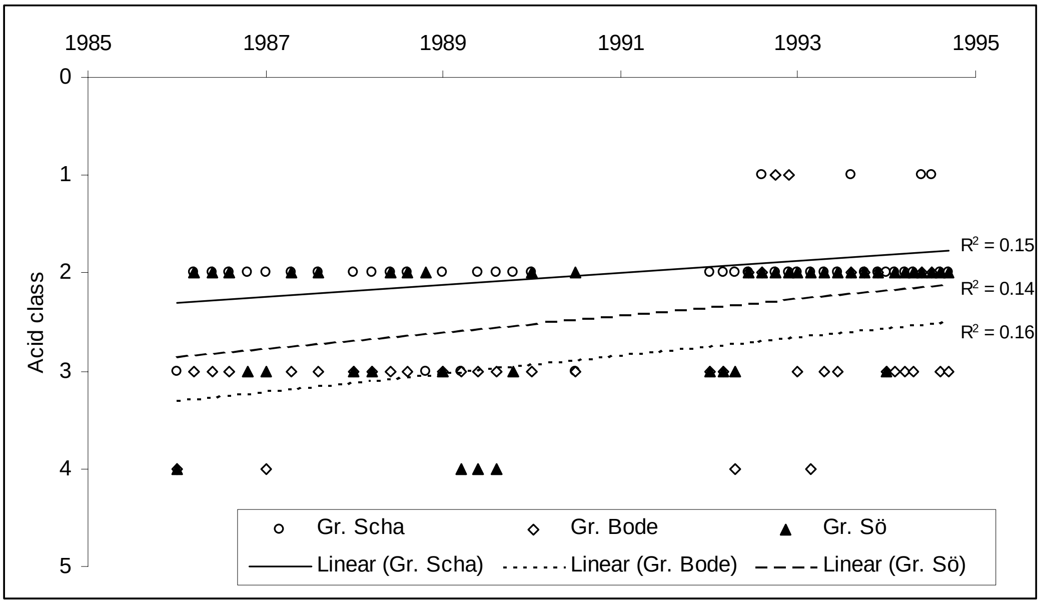


Figure 30. *Trends in the acidification class for different sites in Harz.*

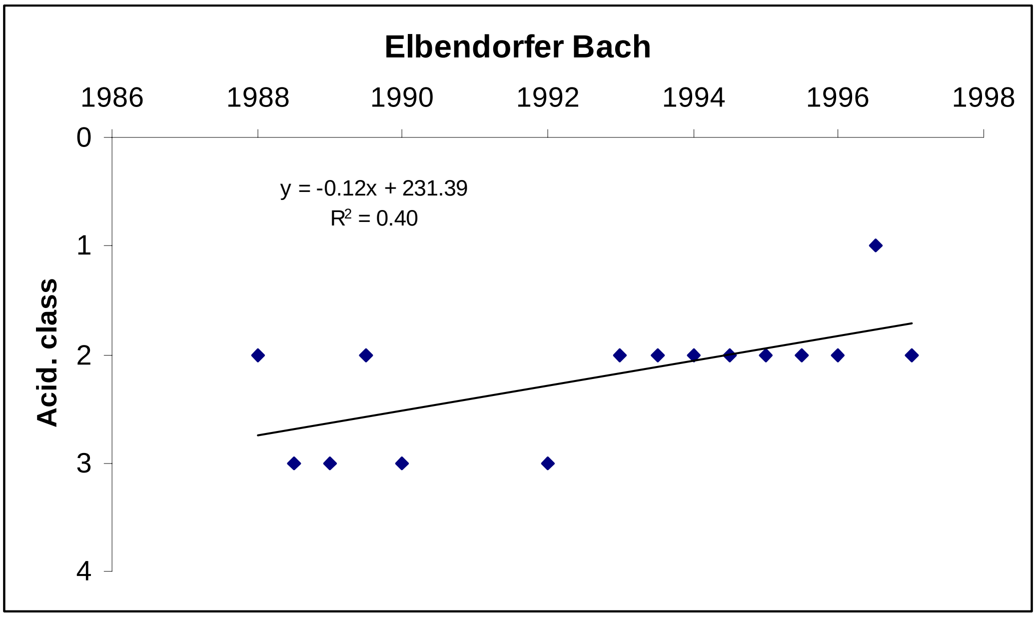


Figure 31. *Trends in the acidification class for Elberndorfer Bach in Rothaargebirge.*

5.3.3 Multivariate analysis of the total benthic community

The preceding sections have documented the response of individual species of benthic invertebrates to the chemical recovery in some of the ICP Waters sites, based on the *acidity index*. The next question is whether the recovery could be found in some of these watersheds when the whole benthic community was analysed.

The analysis was made on datasets that had both biological and chemical samples from the whole 10-year period. Two western Norwegian watersheds (Gaular - NO07 and Nausta - NO08) and six ICP sites (lakes) in the UK (Loch Coire nan Arr - UK01, Lochnagar - UK04, Round Loch of Glenhead – UK07, Scoat Tarn – UK10, Llyn Llagi - UK15, and Blue Lough – UK21) were chosen. The same method of analysis was used for all eight sites, as described below for the Norwegian watershed Nausta.

Norway
The Nausta watershed
The biological data from the Nausta watershed (NO08) consist of kick samples from 18 riffles in the main river and some of its tributaries. These localities were sampled with the same methodology every spring and autumn from 1989 to 1998. The species data analysed are the pooled data from all of the localities in the watershed from each spring and autumn sampling. A total of 75 species/taxa were included in the analysis.

The water chemistry used is based on the ICP Waters database. The spring data are the average concentrations from the samples collected in the months March, April and May, while the autumn data are the average for September, October and November.

An initial detrended correspondence analysis (DCA) using CANOCO 4 (ter Braak and Smilauer 1998) was first performed in order to identify the length of the underlying environmental axis in the abundance data. The abundance data were log-transformed to reduce the skewness and heteroscedasticity in the data. Rare species were not downweighted. This was done in order to record and give weight to the first few appearing individuals of a sensitive organism in the analysis of a potentially recovering system. The length of the gradient along the first ordination axis was shorter than 2 standard deviations, which means that a linear model best fits the data (Birks 1995). Consequently, a direct gradient analysis, redundancy analysis (RDA), was used (ter Braak 1994; ter Braak and Smilauer 1998).

In the RDA analyses the following water chemistry variables were included: pH, calcium concentration, acid neutralising capacity (ANC), total organic carbon concentration (TOC) and labile aluminium (LAl) concentration. All variables except pH were log-transformed. The time component was also included as a variable in some of the analyses. A linear variable called ‘Time’ was coded successively as 0 to 19 from the spring of 1989 to the autumn of 1998, and a nominal variable called ‘Spring’ was coded as 1 for the spring samples and as 0 for the autumn samples. The two time-related variables were included to compare a possible long-term trend with seasonality.

The first aim was to see whether the changes in water chemistry have influenced the benthic community. This was done by running a RDA with the five water chemistry variables included. The scaling focused on interspecies correlations, and the species scores were divided by the standard deviation (ter Braak 1994). The species data were log-transformed, the samples centred and standardised, and the species centred by species. Both the first ordination axis and all the axes together were tested with a Monte-Carlo permutation test with 999 permutations under a reduced model. Finally a permutation test restricted to time series was performed.

With this setup the total variation in the species data was set to 1. The RDA gave a significant ordination (p = 0.04 unrestricted permutation), with the sum of all canonical eigenvalues ($\Sigma \lambda$) equal to 0.333. When the analysis was tested with the restriction of being a time series, the result was not significant (**Table 13**). This means that about 33 % of the variation in the species data is accounted for by the variation in the five chemical variables included. The results from the RDA with the water chemistry variables included are shown in **Figure 32**.

Table 13. *Results from the RDA analyses of the spring and fall abundances from the Nausta watershed 1989 – 1998.* $\Sigma \lambda$ *is the sum of all canonical eigenvalues.*

Variables	Co-variable	$\Sigma \lambda$	**P unrestricted**	**P restricted**
Water chemistry		0.333	0.040	0.666
Water chemistry	Time	0.294		
Water chemistry + Time		0.447		
Time		0.154	0.001	
Water chemistry	Spring	0.220		
Water chemistry + Spring		0.410	0.010	0.810
Spring		0.191	0.005	1.000

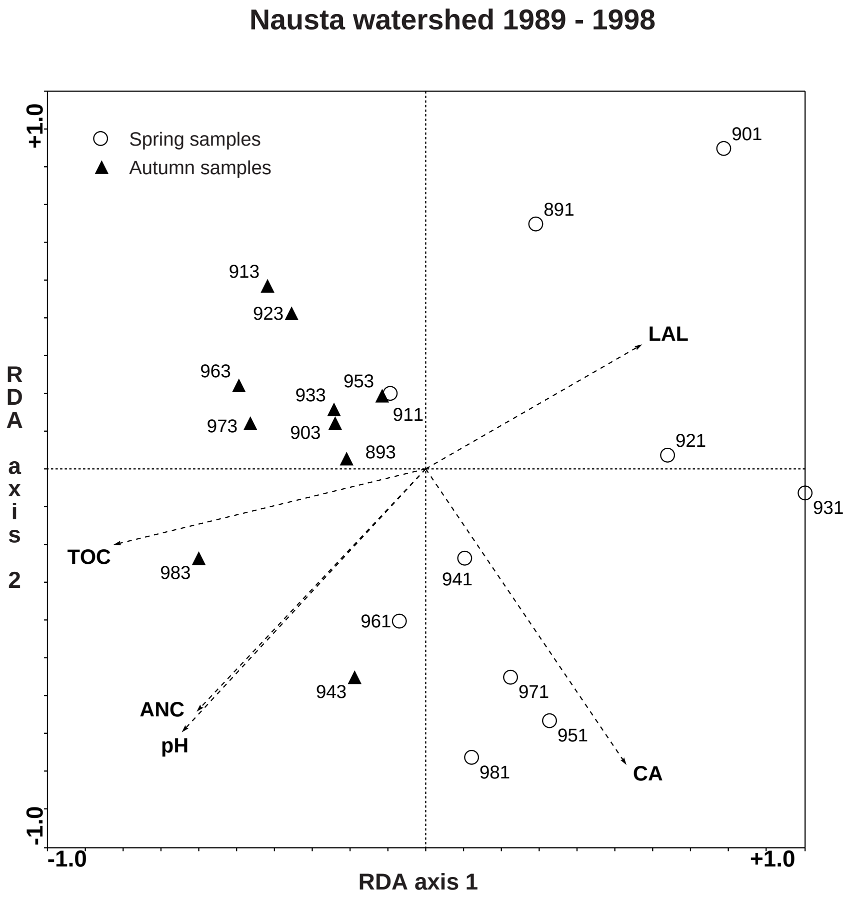


Figure 32. *RDA biplot of sites and water chemistry variables from the Nausta watershed 1989 – 1998. The sites are the pooled abundance data from 18 riffle localities in the river Nausta and its tributaries. The two first digits of the site names indicate the year sampled and the third digit (1 or 3) indicates spring or autumn samples.*

The next aim was to discover how much of this variation (33.3 %, **Table 13**) in the species data explained by the included chemical variables could be assigned to a linear change of the water chemistry through time, i.e., due to a possible natural recovery of an acidified watershed. Thus the linear variable 'Time' was run as a co-variable in a second run of the RDA. This removes the variation caused by the linear component of time. The sum of the constrained eigenvalues in this analysis is 0.294 (**Table 13**), indicating that only a small portion of the variation in the species data can be explained by a linear change in the water chemistry variables.

Since the RDA does not have a direct method for estimating the proportion and significance of the individual environmental variables, an indirect approach has to be used (Borcard et al. 1992; Legendre 1993; Borcard and Legendre 1994). The procedure is exemplified in **Figure 33**. The axis represents the total variation in the species data expressed in percent. The amount 'a' is the variation in the species data that can be explained by the water chemistry variables. This figure is taken from the sum of all constrained eigenvalues in the RDA with only the water chemistry variables included. 'b' is the amount of variation explained by linear time, and 'c' is the variation when both the water chemistry variables and linear time is included in the RDA. Of interest is the overlap between the amount explained by water chemistry and linear time. This overlap represents the possible amount of response by the benthic community to a recovery in water chemistry. The overlap then equals to ((a + b) – c). From **Table 13** we get **Figure 34**. The overlap between water chemistry and linear time is small. Only 4 % of the variation in the species data from the Nausta Watershed can be explained as a response to the amelioration due to the reductions in sulphur emissions during the last decade.

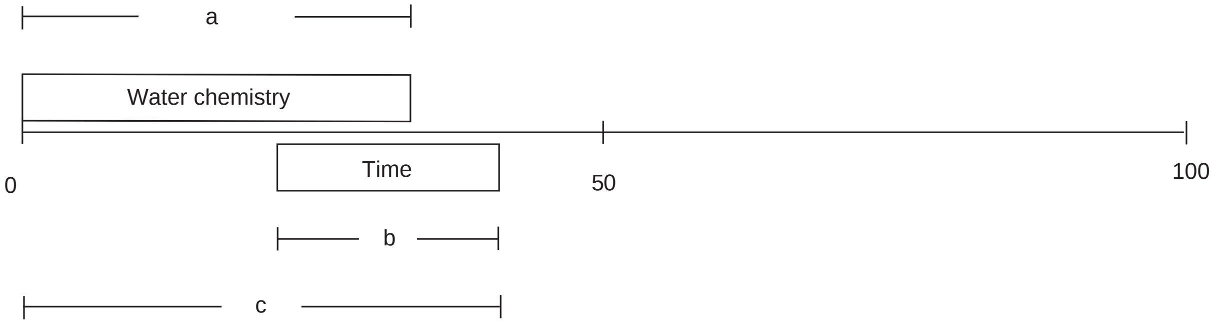


Figure 33. *Example of plot illustrating the explained variation in the species data that can be accounted for changes in water chemistry through the linear component of time. See text for further explanation.*

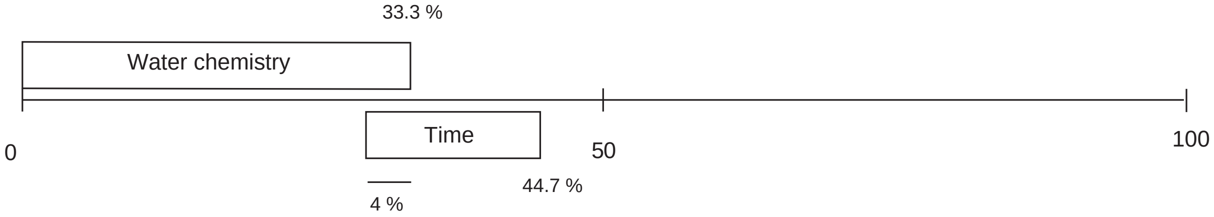


Figure 34. *The amount of variation in the species data in the Nausta watershed from 1989 to 1998 that can be explained by changes in the water chemistry variables and linear time. The overlap indicates the amount of variation that may be explained by linear changes in water chemistry through time.*

The spring and autumn samples from the same year are very different (**Figure 32**), a characteristic that is also shown by the *acidity index*. Acid episodes connected with the snowmelt and possible sea-salt episodes during the winter are the likely explanation to this. In the winter of 1993, a well-documented sea-salt episode took place in Western Norway (Hindar et al. 1994; Barlaup and Åtland 1996). This can easily be seen in the biplot, where the samples from spring 1993 (site 931 in **Figure 32**) are located at the extreme right in the ordination diagram.

Episodic events may confound or even eradicate an ongoing response in the community due to a chemical amelioration in the watershed. A test of this was carried out using the same procedure as above, but with the nominal variable 'Spring' substituting the linear time component. The RDA with Spring as a co-variable and with the water chemistry variables included, indicated that a larger part of the variation in the species data could be explained by these episodic events. The sum of all constrained eigenvalues is lower when the variation caused by the variable 'Spring' is removed, than when the variation caused by the linear component of time (variable 'Time') is run as a co-variable. **Figure 35** shows the amount of overlap between the variation explained by the water chemistry variables and the variable 'Spring'. This is larger than the proportion explained by the linear time component. This indicates that the signals of a beginning recovery in the benthic community caused by the reductions in sulphur emissions and deposition of sulphate during this 10-year period, may be set back or confounded by these episodic events.

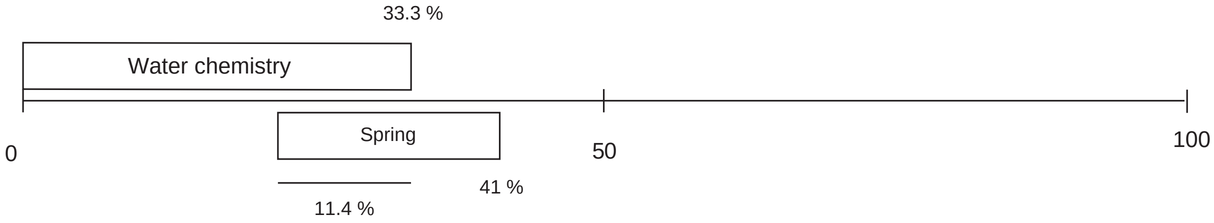


Figure 35. *The amount of variation in the species data in the Nausta watershed from 1989 to 1998 that can be explained by changes in the water chemistry variables and episodic events related to snowmelt and/or sea-salt episodes (the nominal variable' Spring'). The overlap indicates the amount of variation that may be explained by these episodic changes in water chemistry through time.*

A Spearman rank correlation test between the water chemistry variables and the two components of time is shown in **Table 14**. Linear time is significantly correlated with an increase in pH, ANC and a decrease in labile aluminium (LAl), which indicates an improvement of the water quality with respect to acidification. The nominal component of time (variable 'Spring') is significantly correlated with an increase in the calcium concentration during spring, which may be due to increased release of calcium in the catchment area during snowmelt and sea-salt episodes. Spring is also correlated with a decrease in TOC, perhaps due to dilution effects of snowmelt.

Table 14. *Spearman Rank Correlation matrix between the environmental variables from the Nausta watershed from 1989 – 1998 used in the RDA's.*

	Ca	**ANC**	**TOC**	**LAl**	**Time**	**Spring**
pH	0.347	0.749 **	0.624 **	- 0.704 **	0.743 **	- 0.226
Ca		0.344	- 0.015	- 0.212	0.358	0.566 **
ANC			0.690 **	- 0.670 **	0.588 **	- 0.295
TOC				- 0.449 *	0.321	- 0.504 *
LAl					- 0.679 **	0.313
Time						- 0.087

Two-tailed tests. * * significant at the 0.01 level. * significant at the 0.05 level.

In summary, the total benthic community of the Nausta watershed shows changes from 1989 to 1998 that may be interpreted as a trend of recovery due to reductions in sulphur deposition. The trend is weak, meaning that the recovery may be in its beginning, or that it is confounded by acid episodes caused by snowmelt and/or sea-salt episodes during the winter. The trend is, however, consistent with the results found with the *acidity index*.

Analysis of the spring and autumn samples separately with the water chemistry data did not give any significant ordinations. This may be due to too few samples in the analysis.

The Gaular watershed

The same procedure was followed at the Gaular watershed (ICP site NO07). The species data consisted of the pooled abundances from kick-samples in 17 riffles. The initial DCA gave a similar short gradient along the first ordination axis as for the Nausta watershed, but the RDA with the same water chemistry variables as above included did not give any significant ordination. The spring and autumn samples diverged in a similar way as in Nausta (**Figure 36**), but on a smaller scale. No trend in water chemistry could be traced to the total benthic community of the Gaular watershed.

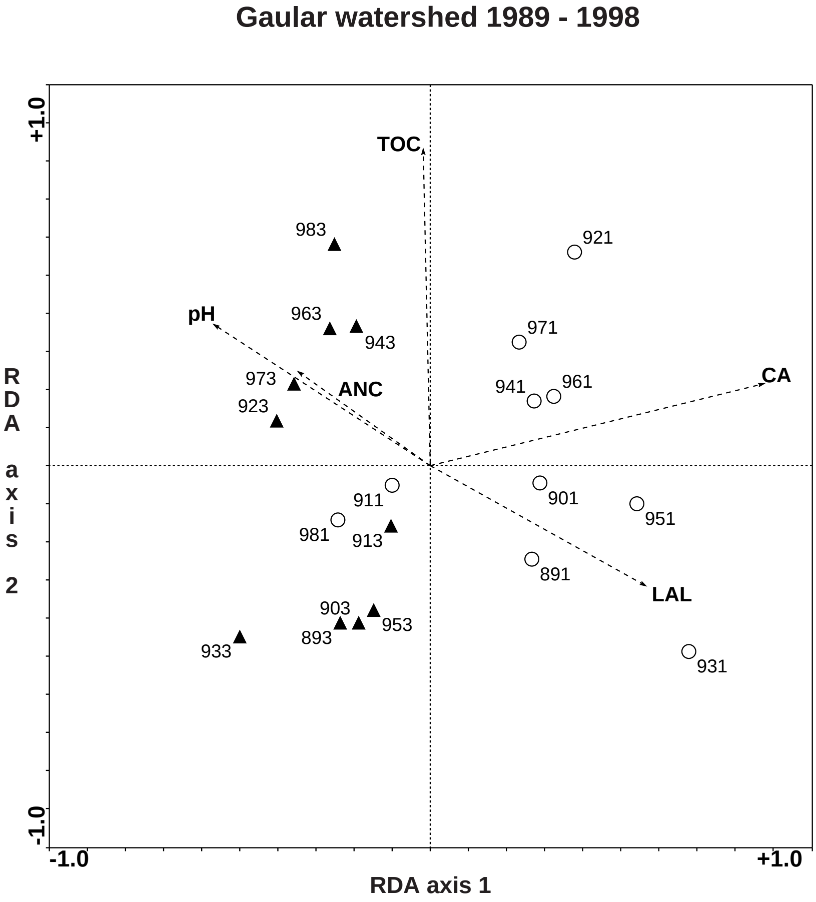


Figure 36. *RDA biplot of sites and water chemistry variables from the Gaular watershed 1989 – 1998. The sites are the pooled abundance data from 17 riffle localities in the river Gaular and in its tributaries. The two first digits of the site names indicate the year sampled and the third digit (1 or 3) indicates spring or autumn samples.*

UK

The species data from the six UK sites consisted of a varying number of kick-samples taken in the littoral of the lakes each spring. No autumn samples were taken. For the DCA and RDA, the average number of the different species/taxa were from each locality and sampling date were used. The localities and sampling methods are described in Juggins et al. (1992) and Patrick et al. (1995).

The initial DCA for all of the six sites showed a length of the gradient along the first ordination axis shorter than 2 standard deviation units, so a RDA was used in the following analyses as for the Norwegian sites. The water chemistry variables included in the analyses were pH, Ca concentration, dissolved organic carbon, and acid neutralising capacity (ANC). The ANC values were lacking for some of the sampling dates in Lochnagar (UK04), so this variable was not included in the analysis of this site.

Testing all ordination axes in the RDA's of the species data with the water chemistry variables included did not give significant results for any of the sites. There were no trends identified in the material. This is consistent with both the analyses of the water chemistry and with the results of the *acidity index*.

6. Heavy metals in surface waters

In June 1998 the protocol on heavy metals was signed in Aarhus, Denmark (UN/ECE 1998) (see text box below). As a result of this, work on heavy metals within all the ICPs under WGE has received a stronger focus. Heavy metals (Pb, Cd, Zn, Cu and Ni) are all optional variables in the ICP Waters Programme manual, and have until now received very little attention. At the 14th Task Force meeting in Zakopane, Poland a short survey of heavy metals in The ICP Waters database was presented. The survey showed that there is not as many data on heavy metals as there is on major chemistry in the database, and that many countries do not submit data on heavy metals at all. It was concluded that the work with the overview of heavy metals in the database should be expanded and documented more thoroughly. In this work a questionnaire should be sent out to all the participating countries to see if there existed more heavy metal data from monitoring of surface water that were not yet reported to the Programme centre.

The Aarhus Protocol on Heavy Metals.

The Aarhus Protocol on Heavy Metals (UN/ECE 1998) aims at regulation and reduction of emissons of particularly Cd, Pb and Hg to the atmosphere.

> ***Press release on the 1998 Aarhus Protocol on Heavy Metals***
> *The Executive Body adopted the Protocol on Heavy Metals on 24 June 1998 in Aarhus (Denmark). It targets three particularly harmful metals: cadmium, lead and mercury. According to one of the basic obligations, Parties will have to reduce their emissions for these three metals below their levels in 1990 (or an alternative year between 1985 and 1995). The Protocol aims to cut emissions from industrial sources (iron and steel industry, non-ferrous metal industry), combustion processes (power generation, road transport) and waste incineration. It lays down stringent limit values for emissions from stationary sources and suggests best available techniques (BAT) for these sources, such as special filters or scrubbers for combustion sources or mercury-free processes. The Protocol requires Parties to phase out leaded petrol. It also introduces measures to lower heavy metal emissions from other products, such as mercury in batteries, and proposes the introduction of management measures for other mercury-containing products, such as electrical components (thermostats, switches), measuring devices (thermometers, manometers, barometers), fluorescent lamps, dental amalgam, pesticides and paint.*

6.1 Heavy metals in the ICP Waters database

Nine countries have reported data on heavy metals to the Programme centre the last 3 years (1996-98). Of these, 8 have submitted data on Pb and Cd, 9 on Zn and Cu, and 5 on Ni.

Mean values for Pb, Cd, Zn, Cu and Ni for the period 1996-1998 are listed in **Table 15**. In calculating mean values, all values under detection limit are given the value of the detection limit. This means that mean value listed in the table is high for some of the stations. In some cases where the detection limit changed (to a lower value) within the 3-year period, only the latest observations are used.

Detection limits for the data in **Table 15** are listed in **Table 16**. This shows that the detection limit varies from very low values in Sweden and Finland to very high values in Hungary. In Germany, the laboratories analysing heavy metals are operating with different detection limits.

Table 15. *Mean values of heavy metals for the period 1996-1998 (units µg/L).*

	no. of observations in the period 1996-98					mean value for the period 1996-98				
	Pb	Cd	Zn	Cu	Ni	Pb	Cd	Zn	Cu	Ni
Site								µg/L		
BY01	7	6	18	18	18		2.17	13	4.9	4.6
CA01			146					31		
CA02			106					10		
CA03			105					10		
CA04			102					10		
CA08			29					10		
CA10	2	2	2	2		<2	<1	<10	<2	
CA11	4	4	4	4		<2	<1	<10	6.5	
CA12	2	2	2	2		<2	<1	<10	<2	
CA13	4	4	4	4		<2	<1	<10	6.5	
CA14	2	3	1	3		<2	<1	<10	<2	
CZ01	6	6	6	6		0.40	0.25	19	1.4	
CZ02	6	6	6	6		0.58	0.23	19	1.3	
CZ03	6	6	6	6		0.47	0.21	18	0.2	
CZ04	6	6	6	6		0.51	0.13	12	0.9	
CZ05	6	6	6	6		0.45	0.06	10	1.1	
CZ06	6	6	6	6		0.42	0.04	10	0.2	
DE01	12	12	12	12	12	0.87	0.11	14	0.5	1.3
DE02			1					<10		
DE03	56	57	55			0.82	0.15	22		
DE04	19	19	19	8	19	2.18	1.24	88	2.4	33.0
DE05	11	11	11	11	11	0.40	0.05	8	0.4	0.5
DE06	25	25	25	25	25	<3	0.50	39	<2	5.7
DE07	22	33	33	20	14	1.38	0.54	24	1.5	5.1
DE08	16	16	16	16	16	<1	<0.1	10	<1	<1
DE09	5	13	13	4	14	1.02	0.38	48	6.7	9.7
DE11	12	12	12	12	12	0.15	0.03	8	0.3	0.5
DE14	11	11	11	11	11	<6	0.52	26	<6	<6
DE15	32	32	32	32	32	<6	0.39	<20	<6	9.3
DE16	8	8	8	8	7	0.43	0.03	4	0.6	<0.5
DE18			1					10		
DE19	31	31	31	31	31	<6	0.92	66	<6	7.5
DE20	34	34	34	34	34	<6	<0.3	<20	<6	<6
DE21	17	24	25	22	19	1.45	0.79	22	7.9	4.4
DE22	24	24	24	24	24	<6	0.39	<20	<6	<6
DE25		9	7	1	12		0.41	32	14.1	6.3
DE26	31	31	31	31	31	<3	0.20	29	<2	<5
DE28	6	6	6	6	6	0.62	<0.07	<10	1.3	<20
DE29	6	6	6	6	6	0.68	0.08	14	1.2	<20
DE30	16	20	20	6	20	3.01	0.60	33	4.5	2.8
DE31	1		13	11	22	1.50		10	2.8	4.1
DE32	55	57	55			0.89	0.18	21		
DE33			1					10		
EE01	1	1	1	1		<1	<0.1	<10	25	
FI01	7	7	7	7	6	0.25	0.06	11	0.1	0.4
FI07	1	1	1	1	1	0.18	<0.03	4	0.5	0.4
FI08	6	6	5	6	6	0.26	0.03	5	0.2	0.2
LV02	17	18	18	18		0.1	0.02	3.9	1	
LV03	17	17	18	18		0.16	0.02	3.7	1	
LV04	16	17	16	17		0.12	0.02	4.1	1.2	
HU01	6	6	6	6	6	<15	<0.2	11	<10	<15
RU07	3	2	3	3	3	<0.3	0.06	0.6	0.5	0.5
RU08	1	1	1	1	1	<0.3		2.3	0.6	1.2
RU09	1	1	1	1	1	<0.3		0.3	0.7	1.2
RU10	1	1	1	1	1	<0.3		0.9	0.3	1
SE01	24	12	33	33	24	0.11	0.01	2	0.7	0.6
SE03	24	12	35	36	24	0.32	0.01	5	1.7	0.8
UK01			2					38		
UK04			2					45		
UK21			1	1				25	25.0	

Table 16. *Detection limits for heavy metals in surface waters for the countries submitting data to the programme centre (units: µg/L). For Germany the data come from several laboratories which operate with different detection limits.*

	Pb	Cd	Zn	Cu	Ni
Canada	2	1	5/10	2	
Czech R.	0.4	0.04	10	0.2	
Estonia	1	0.1	10	25	
Finland	0.03	0.03	0.3	0.07	0.04
Germany	0.2-6	0.02-4	1-20	0.5-20	0.2-6
Hungary	15	0.20	10	10	15
Latvia	0.07	0.01	0.21	0.18	0.13
Norway	0.03	0.02	0.3	0.2	0.1
Russia	0.3	0.05	0.02	0.1	0.2
Sweden	0.02	0.003	0.2	0.04	0.05
UK			10	10	

6.2 Results from the questionnaire

As a result of the short survey of heavy metals in the ICP Waters database, it was decided that a questionnaire should be sent out to all Focal Centres. The goals for the questionnaire were to obtain an overview of existing national databases on heavy metals in surface waters and to assess the interest for heavy metals in surface waters.

Belarus In Belarus the work on heavy metals in surface waters has been carried out for a long period of time (over 30 years) at many stations. But only one station in the Berezinsky reserve on the Berezina River submits data to the ICP Waters Programme. The laboratory analysing heavy metals participates in national intercalibrations.

Canada Most of the Canadian ICP sites report Zn, and a few report Cu. However, in Canada all the required heavy metals are probably measured at 100 sites with weekly-to-yearly sampling frequency. Some of these sites have time series as long as 25 years. The data may be accessible for ICP Waters. The laboratories participate in intercalibrations.

The Czech Republic Heavy metals (Pb, Zn, Cd and Cu) have been monitored at 6 Czech lakes since 1984 by the same analytical methods. Concentrations of Pb, Zn, Cd (and Mn, Be) significantly decreased in acidified lakes, Cu and Fe did not change (Veselý et al. 1998b). The laboratory analysing heavy metals participates in intercalibrations.

Estonia In Estonia, Hg, Cd, Zn, Cu and Pb are measured once per year in the main rivers in the country primarily for calculating fluxes to the Baltic Sea. The levels of heavy metals are in general low. The rivers are influenced by local pollution in addition to long-range air pollution.

Finland In Finland, both lakes and rivers are monitored for heavy metals, the rivers primarily for calculating fluxes to the Baltic Sea. Six small lakes are monitored for heavy metals under the Convention on LRTAP. Two report quarterly data to ICP Waters and four sites monthly data to ICP Integrated Monitoring. The laboratories analysing heavy metals participate and arrange intercalibration.

Germany In the ICP Waters database, there are heavy metals data for the last three years at more than 20 sites. Fifteen sites of these sites have been selected for continued monitoring of Pb, Cd, Zn, Ni and Cu. The German laboratories participate in intercalibration.

Hungary Heavy metals are monitored at a dense network of all the large rivers in Hungary. In addition heavy metals are monitored at the Hungarian ICP site HU01. However, the detection limits are relatively high for all the elements. The monitoring network for the large rivers is not directly suitable to monitor the effect of heavy metals air pollution on water quality.

Ireland Monitoring for heavy metals at Irish ICP sites will commence in December 1999. Heavy metals are measured at the lowermost freshwater sampling points on 17 Irish Rivers for the purpose of calculating loading as part of the PARCOM programme; however, these sampling sites are not suitable for measuring transboundary air pollution.

Italy There are no heavy-metal data from ongoing monitoring programmes in Italy. The Focal Centre in Italy presumes that there is interest for heavy metals in Italy, but funding is lacking.

Latvia In Latvia, heavy metal measurements started in 1972. The Latvian Hydrometeorological Agency has long experience in monitoring heavy metals in air, precipitation and surface- and ground water. The data are reported to EMEP, ICP IM, ICP Waters and to HELCOM for calculations of heavy metal fluxes to the Baltic Sea.

Norway Heavy metals in surface waters are not a part of the Norwegian monitoring programme for long-range transported air pollutants. However, heavy metals are measured at the mouths of 155 Norwegian rivers as a part of PARCOM (Paris Convention). The rivers at these stations are influenced by the total anthropogenic activity in the catchment and are thus not suitable for monitoring the effects of transboundary air pollutants. In 1995, a national survey in 1000 lakes was carried out to map the level of heavy metals in Norwegian surface waters.

Poland In Poland, national monitoring of surface waters has been carried out. Heavy metals, however, are determined only in waters contaminated with industrial effluents (mainly rivers). According to the Polish Focal Centre, no systematic analysis of heavy metals content in surface waters without effluents has been carried out.

Russia The Russian contact person at INEP in Apatity at the Kola Peninsula answered the questionnaire. In this region, pollution of air transported heavy metals is a huge problem, and the interest for work with heavy metals are consequently also high. INEP holds heavy metals data that can be accessible for ICP Waters.

Sweden In Sweden >25 stations are monitored for heavy metals. Two of these sites presently report data to ICP Waters. The laboratories analysing heavy metals participate in intercalibrations.

UK In the UK, heavy metals are monitored through the Environmental Change Network (ECN). Twenty-six rivers are monitored monthly and 17 lakes are monitored quarterly. Some of these data may be accessible for ICP Waters. In addition there are other long-term heavy metals monitoring at single sites. The interest for heavy metals is described as increasing by the Focal centre, due to forthcoming EU legislation.

6.3 Critical limits of heavy metals in surface waters

There is extensive work underway at the international level (Umweltbundesamt 1998) to determine toxic levels of heavy metals for aquatic and terrestrial biota. There are many factors that influence the concentration level at which harmful effects can be expected. Usually, only the free or inorganic bound form of the element is regarded as potentially toxic to aquatic biota, and complexation significantly reduces the concentration of the free and thus toxic metal ion. Organic matter is a strong complexing agent in natural waters, and in the presence of organic matter, higher concentrations of total metal can be tolerated without adverse effects on organisms. This complexation is particularly strong for Cu and Pb, and decreases in importance for Ni, Zn and Cd. Further there are other factors such as pH and hardness that affect concentration of free metal ions and thus reduce toxicity.

There are at present no internationally-agreed limits for toxic levels of heavy metals in surface waters. Critical limits for heavy metals in surface waters are listed (**Table 17**) for some of the participating countries in the Programme. The data are mostly taken from a compilation made for the "Workshop on critical limits and effect based approaches for heavy metals and persistent organic pollutants" (Umweltbundesamt 1998). The Swedish limits are generally the most stringent.

In Norway and Sweden, the "critical limits" for each of the elements are set according to present knowledge of adverse effects of heavy metal pollution. In the background literature for the Swedish classification systems (Alm et al. 1999), it is stressed that the limits must be used with caution, as knowledge about adverse effects is limited. Most dose-effect relationships between heavy metals and effects on biota have been made in laboratory, and the applications of laboratory results to field conditions are usually not straightforward.

Table 17. *Critical limit of heavy metals in surface waters for a number of countries within the ICP Waters programme. The data are taken from a compilation published in Umweltbundesamt (1998).*

	Pb	Cd	Zn	Cu	Ni
Belarus	100	5	10	1	1
Canada	1-7	0.2-1.8	30	2-4	25-150
Czech R.	50	5	50	50	50
Denmark	3.2	5	110	12	10
Germany	11	0.34	6.6	1.1	1.8
Norway	2.5	0.2	50	3	10
Russia	100	5	10	1	1
Sweden	1	0.1	20	3	5
UK	4-25		8-50	0.5-12	8-40

In general, the concentrations of heavy metals in ICP Waters sites not influenced by local pollution sources, is not very high. Based on the guidelines given by e.g., Sweden, a number of the ICP sites have a concentration level of heavy metals above the critical limit. To be able to give a good picture of the general level of heavy metals in surface waters throughout Europe and North America, heavy metal data for more sites with a larger geographical cover is needed.

6.4 Long-term trends in heavy metals

The methods for analysing heavy metals have changed and improved over the last years. In recent years the ICP-MS method has increasingly become the standard and has resulted in major improvement in detection limits for many metals. Both the change in method and the change in detection limit make it difficult to identify time trends in heavy metals for many sites. In addition, monitoring of heavy metals in many countries is a relatively new activity.

In the ICP Waters database, only the sites from Germany and the Czech Republic have both sufficiently long time-series and high quality data to allow for evaluation of trends. Trends for two sites in Germany and the Czech Republic are shown (**Figure 37**). The German site (DE03) illustrates the changes in decrease in detection limit. The detection limit for Pb changes from 2 µg/L prior to 1992 to 0.2 µg/L through 1998. The detection limit for Cd changes from 0.3 µg/L prior to 1991, to 0.2

µg/L through 1992, and then to 0.04 µg/L subsequently. The Czech site shows a clear decline in Pb and Cd over the last 10 years, while the German site shows no trend.

Due to the limited number of sites with heavy metals data and because of the shortcomings in the data (short series, changes in analytical methods), it is presently not possible to do an analysis of long-term trends in heavy metals on a regional scale, in a manner similar as for acidification (chapter 4).

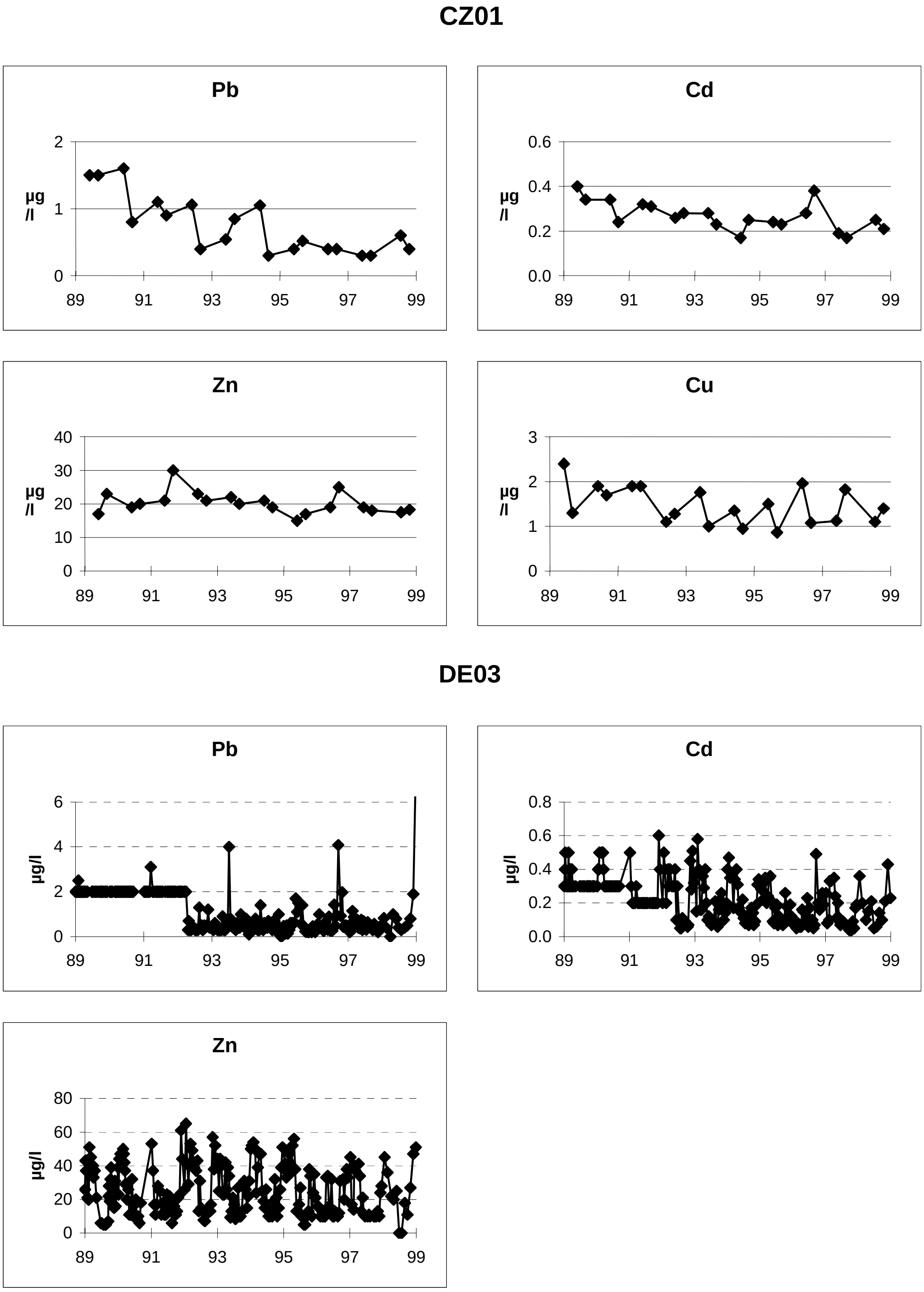


Figure 37. Trends in Pb, Cd, Zn and Cu for Lake Černé (CZ01) in Czech Republic and Pb, Cd and Zn for Elbendorfer Bach (DE03) in Germany. Units in µg/L.

6.5 Heavy metals from regional lake surveys

Several countries have carried out synoptic surveys of heavy metals in surface waters (**Table 18**). As the analytical methods and hence detection limits have changed over time, only the results from the most recent surveys are relevant for comparison. In **Table 19** mean concentrations for a number of recent lake surveys are listed. Both in Finland and Sweden, the ICP sites reporting heavy metals in general have higher values than the median value for the total lake population, while in the Czech Republic the 6 monitoring station have levels of heavy metals around the median value.

Table 18. *Overview of regional surveys of heavy metals in surface waters.*

Country	**Year**	**Type**	**Number of sites**	**Notes**	**Reference**
Norway	1974-75	lakes	154	small lakes only	Wright and Henriksen 1978 Steinnes and Henriksen 1993
	1995	lakes	985	statistical sample	Skjelkvåle et al. 1996; Skjelkvåle et al. 1999
Sweden	1995	lakes	1036	statistical sample	Wilander 1998; Skjelkvåle et al. 1999
Finland	1987	lakes	256	headwater lakes	Verta et al. 1990; Mannio et al. 1993
	1990-92	streams and lakes	1165 (1990) 154 (1992)	headwater	Mannio and Vuorenmaa 1995; Tarvainen et al. 1997
	1995	lakes	464	statistical sample	Skjelkvåle et al. 1999
Denmark	1995	lakes	19	sensitive lakes	Skjelkvåle et al. 1999
Czech Republic	1984-95	streams and lakes	10 680	unpolluted waters	Veselý and Majer 1996; Veselý and Majer 1998
Canada Atlantic region		lakes	435	sensitive lakes	unpublished

Table 19. *Median values for Pb, Cd, Zn, Cu and Ni for regional lake surveys in several countries. Unit: µg/L. Data from Finland, Norway, Sweden, Denmark and Russian Kola are published in Skjelkvåle et al 1999. Data from the Czech Republic are from Veselý and Majer (1998) and Veselý and Majer (1996). Data from Atlantic Canada are unpublished data from G.E.M. Hall, Geological Survey Canada, cited in Veselý and Majer (1998).*

	Finland	Norway	Sweden	Denmark	Russian Kola	The Czech republic	Atlantic Canada
n	464	985	1036	19	460	10 680	
Pb	0.17	0.14	0.16	0.38		<0.39	0.1
Cd	0.011	0.012	0.010	0.050		<0.04	
Zn	2.2	1.1	1.5	7.4	1.4	<10	1
Cu	0.42	0.33	0.36	0.6	0.73	0.8	0.4
Ni	0.37	0.24	0.35	1.0	0.33		

Results from the Nordic lakes survey in 1995 (Skjelkvåle et al. 1999) indicated that direct and indirect influence of long-range transported air pollution is the most important factor explaining the distribution of Pb, Cd and Zn in lakes on a regional scale, while bedrock geology and overburden is the major controlling factor for Cu and Ni with exception of areas around the smelters at Kola (Russia). The results further indicate that heavy metal pollution in lakes is a minor ecological problem on a regional scale in the Nordic Countries. Certain areas, however, in particular in southern Norway and Sweden, are affected by long-range transport leading to Pb, Cd and Zn concentrations in lakes above limits set by the national environmental authorities. There are indications that especially in small streams there may be generally higher levels of heavy metals than in lakes, and thus the ecological problem of heavy metals may be larger for running waters than for lakes.

Results of a Czech freshwater survey (Veselý and Majer 1996; Veselý and Majer 1998) showed that acid deposition mobilised Mn, Be, Cd and Zn from overburden and bedrock. Direct atmospheric deposition was important for Pb, F, As and Li concentrations in freshwaters. Natural enhanced concentrations were observed for Cu, As and Be. Local pollution of freshwater was observed for all heavy metals.

7. The ICP Waters sites as monitors of future changes: Reversibility, recovery and delay times

7.1 Work plan for 1999/2000

The workplan for 1999/2000 for ICP Waters was agreed at the 15th Task Force Meeting in Pallanza, October 1999:

a) Finalisation of the ICP Waters 12-year report (deadline 15 April 2000);
b) Finalisation of the report on trends in intercalibration results;
c) Participation in preparing a substantive report on occurrence and effects of heavy metals (joint effort of ICPs and the Task Force on the Health Aspects of Air Pollution);
d) Preparing a report on nitrogen including calculating critical loads at ICP sites using the First-order Acidity Balance (FAB) model) with progress report in 2000;
e) Collection and processing of 1999 data (deadline for submission of data 1 July 2000), and continuing management of the ICP Waters database, with more emphasis on biological data (following the 1998 workshop on biological monitoring);
f) Chemical intercalibration, including heavy metals;
g) Biological intercalibration;
h) Amendment of the ICP Waters manual dealing with heavy metals;
i) Update of the ICP Waters Internet home page;
j) Consideration of the importance of short-term climatic fluctuations (sea-salt episodes) and their influence on trends in water quality. A note will be prepared with the assistance of the UK and will be presented to the TF meeting in 2000.

7.2 Future perspectives

The ICP Waters programme is well positioned to monitor changes over the next 10+ years as the new reductions in emissions of S and N compounds come into effect. The 2nd Sulphur Protocol (Oslo protocol of 1994, UN/ECE 1994) and the new Multi-Pollutant, Multi-effect Protocol (Gothenburg protocol of 1999, UN/ECE 1999), if implemented as planned, entail dramatic reductions in both S and N emissions in Europe relative to present-day levels. The trends already documented in water chemistry and biology in the ICP Waters data are thus likely to continue in the years to come.

The number and geographical distribution of sites, as well as the sampling frequency and parameter set at existing sites in the ICP Waters Programme can be, of course, improved to better follow the expected changes in the future.

The results from the ICP Waters Programme, as reported here in the 12-year report and elsewhere, clearly show that surface waters respond to changes in atmospheric deposition. Indeed waters are much more responsive than either soils or terrestrial vegetation to changes in long-range transported acid pollutants. Lakes and rivers also have the advantage that they integrate the effects over the entire catchment area. The ICP site network is geographically extensive (with currently nearly 150 active sites) and includes long-term data series (< 15 years) for many sites. The network is thus well poised to document changes that result from implementation of the protocols.

The ICP Waters programme is well developed with respect to major-ion chemistry and chemical parameters associated with acidification. Addition of new biological parameters and expansion of sampling programmes at individual sites to include existing biological parameters could substantially strengthen the network. As reported here in the 12-year report, the invertebrate fauna responds to changes in water chemistry, and is a suitable group of organism for monitoring purposes. As the acidification of surface waters continues to diminish in the future, we expect the recovery of damaged groups of organisms.

The ICP Waters programme presently includes only a minor amount of data for heavy metal concentrations. An analysis of these data presented here indicates that ecological effects of long-range transported heavy metals are probably minor. An intercalibration of heavy metal analysis methods should be conducted. In the event that the Working Group on Effects (WGE) deems it prudent to include heavy metals in its activities, the ICP Waters sampling programme and database provides a ready vehicle for sampling, quality control and assessment of heavy metals in surface waters.

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Appendix A. Results of trend site selection process

Country	**Dataset**	**ICP Site Number**	**Site Name**	**Used for Trends**	**Reason for exclusion**						
					Incomplete Chemistry	Insufficient or Inconsistent sampling schedule	Multiple locations per site	Insensitive to Acidification	Disturbed Watershed	Other Documented Causes for Trends	Record too Short
Austria	IA-AT	AU03	Tirol, Schwarzsee ob Sölden	No						X	
Belarus	IA-BY	BY01	Berezinsky Biosphere Reserve	No					X		
Canada	IA-CA	CA01	Ontario, Algoma Region, Batchawana Lake	Yes							
	IA-CA	CA02	Ontario, Algoma Region, Wishart Lake	Yes							
	IA-CA	CA03	Ontario, Algoma Region, Little Turkey Lake	Yes							
	IA-CA	CA04	Ontario, Algoma Region, Turkey Lake	Yes							
	IA-CA	CA05	Quebec, Lac Veilleux	Yes							
	IA-CA	CA06	Quebec, Lac Josselin	Yes							
	IA-CA	CA07	Quebec, Lac Bonneville	Yes							
	IA-CA	CA08	Quebec, Lac Laflamme	Yes							
	IA-CA	CA09	Quebec, Lac Macleod	Yes							
	IA-CA	CA10	Nova Scotia, Mount Tom Lake	No		X					
	IA-CA	CA11	Nova Scotia, Mountain Lake	Yes							
	IA-CA	CA12	Nova Scotia, Little Red Lake	No		X					
	IA-CA	CA13	Nova Scotia, Kejimkujik Lake	Yes							
	IA-CA	CA14	Nova Scotia, Beaverskin Lake	Yes							
	IA-CA	CA16	Ontario, Lake 224	Yes							
	IA-CA	CA17	Ontario, Lake 239	Yes							
	IA-CA	CA19	Ontario, Lake 305	No							X
	IA-CA	CA20	Ontario, Lake 373	No		X					
Czech Republic	IA-CZ	CZ01	Bohemian Forest, erné	Yes							
	IA-CZ	CZ02	Bohemian Forest, ertovo	Yes							
	IA-CZ	CZ03	Bohemian Forest, Plešné	Yes							
	IA-CZ	CZ04	Bohemian Forest, Prášilské	Yes							
	IA-CZ	CZ05	Bohemian Forest, Laka	Yes							
	IA-CZ	CZ06	Bohemian Forest, Zd´árské	Yes							
Germany	IA-DE	DE01	Schwarzwald, Dürreychbach	Yes							
	IA-DE	DE02	Fichtelgebirge, Eger	Yes							
	IA-DE	DE03	Rothaargebirge, Elberndorfer Bach	Yes							

Country	**Dataset**	**ICP Site Number**	**Site Name**	**Used for Trends**	**Reason for exclusion**						
					Incomplete Chemistry	Insufficient or Inconsistent sampling schedule	Multiple locations per site	Insensitive to Acidification	Disturbed Watershed	Other Documented Causes for Trends	Record too Short
	IA-DE	DE04	Sächsische Tieflandsbucht, Ettelsbach	No							x
	IA-DE	DE05	Schwarzwald, Goldersbach	Yes							
	IA-DE	DE06	Hunsrück, Gräfenbach	Yes							
	IA-DE	DE07	Erzgebirge, Grosse Pyra	Yes							
	IA-DE	DE08	Bayerischer Wald, Grosse Ohe	Yes							
	IA-DE	DE09	Sächsische Tieflandsbucht, Heidelbach	No							X
	IA-DE	DE10	Bayerischer Wald, Hinterer Schachtenbach	Yes							
	IA-DE	DE11	Schwarzwald, Kleine Kinzig	Yes							
	IA-DE	DE12	Harz, Lange Bramke	Yes							
	IA-DE	DE13	Erzgebirge, Talsperre Neunzehnhain	No	X						
	IA-DE	DE14	Kaufunger Wald, Nieste 3	No			X				
	IA-DE	DE15	Kaufunger Wald, Nieste 5	Yes							
	IA-DE	DE16	Lauenburgische Seenplatte, Pinnsee	No							X
	IA-DE	DE17	Bayerischer Wald, Rachelsee	Yes							
	IA-DE	DE18	Fichtelgebirge, Röslau	Yes							
	IA-DE	DE19	Taunus, Rombach 2	No			X				
	IA-DE	DE20	Taunus, Rombach 3	Yes							
	IA-DE	DE21	Erzgebirge, Rote Pockau	Yes							
	IA-DE	DE22	Odenwald, Schmerbach 3	Yes							
	IA-DE	DE23	Bayerischer Wald, Seebach	Yes							
	IA-DE	DE24	Erzgebirge, Talsperre Sosa	No	x						
	IA-DE	DE25	Elbsandsteingebirge, Taubenbach	No							X
	IA-DE	DE26	Hunsrück, Traunbach 1	Yes							
	IA-DE	DE27	Bayerischer Wald, Vorderer Schachtenbach	Yes							
	IA-DE	DE28	Oberpfälzer Wald, Waldnaab 2	No		X					
	IA-DE	DE29	Oberpfälzer Wald, Waldnaab 8	No		X					
	IA-DE	DE30	Erzgebirge, Wilde Weisseritz	No	x						
	IA-DE	DE31	Erzgebirge, Wolfsbach	Yes							
	IA-DE	DE32	Rothaargebirge, Zinse	Yes							
	IA-DE	DE33	Fichtelgebirge, Zinnbach	Yes							
Denmark	IA-DK	DK01	Sepstrup Sande, Skaerbaek, Station B	Yes							
	IA-DK	DK02	Sepstrup Sande, Skaerbaek, Station F	Yes							
Estonia	IA-EE	EE01	River Ahja, Kiidjärve	No				X			
Spain	IA-ES	ES01	Lake Redó	No							X

Country	**Dataset**	**ICP Site Number**	**Site Name**	**Used for Trends**	**Reason for exclusion**						
					Incomplete Chemistry	Insufficient or Inconsistent sampling schedule	Multiple locations per site	Insensitive to Acidification	Disturbed Watershed	Other Documented Causes for Trends	Record too Short
Finland	IA-FI	FI01	Hirvilampi	Yes							
	IA-FI	FI02	Vuorilampi	Yes							
	IA-FI	FI03	Mäkilampi	Yes							
	IA-FI	FI05	Lapland, Suopalampi	Yes							
	IA-FI	FI06	Lapland, Vasikkajärvi	Yes							
	IA-FI	FI07	Vusimaa, Vitsjön	No							X
	IA-FI	FI08	N-Karelia,Kakkisenlampi	Yes							
France	IA-FR	FR01	Pyrenees, Lake Aubé	No							X
Hungary	IA-HU	HU01	Matra Mountains, Csórrét Reservoir	No				X			
Ireland	IA-IE	IE01	Wicklow, Glendalough, Lake Upper, Mid Lake	No		X					
	IA-IE	IE02	Wicklow, Glendalough, Lake Upper, Inflow 1	No		X					
	IA-IE	IE03	Wicklow, Glendalough, Lake Upper, Inflow 2	No		X					
	IA-IE	IE04	Wicklow, Glendalough, Lake Upper, Inflow 3	No		X					
	IA-IE	IE05	Galway, Lough Maumwee, Mid Lake	No		X					
	IA-IE	IE06	Galway, Lough Maumwee, Inflow 1	No		X					
	IA-IE	IE07	Galway, Lough Maumwee, Inflow 2	No		X					
	IA-IE	IE08	Donegal, Lough Veagh, Mid Lake	No		X					
	IA-IE	IE09	Donegal, Lough Veagh, Inflow 1	No		X					
	IA-IE	IE10	Donegal, Lough Veagh, Inflow 2	No		X					
	IA-IE	IE11	Donegal, Lough Veagh, Inflow 3	No		X					
	IA-IE	IE12	Donegal, Lough Veagh, Inflow 4	No		X					
Italy	IA-IT	IT01	Piemonte, Lake Paione Inferiore	Yes							
	IA-IT	IT02	Piemonte, Lake di Mergozzo	No		X					
	IA-IT	IT03	Piemonte, Lake Paione Superiore	Yes							
	IA-IT	IT04	Piemonte, River Cannobino	No				X			
	IA-IT	IT05	Piemonte, River Pellino	Yes							
	IA-IT	IT06	Piemonte, River Pellesino	Yes							
Latvia	IA-LV	LV01	Burtnieku Lake, hydrosite	No				X			
	IA-LV	LV02	Barta, Dukupji	No				X			
	IA-LV	LV03	Liela Jugla, Zaki	No				X			
	IA-LV	LV04	Tulija, Zoseni	No				X			
	IA-LV	LV05	Zvirbuli stream, hydrosite	No				X			

Country	Dataset	ICP Site Number	Site Name	Used for Trends	Reason for exclusion						
					Incomplete Chemistry	Insufficient or Inconsistent sampling schedule	Multiple locations per site	Insensitive to Acidification	Disturbed Watershed	Other Documented Causes for Trends	Record too Short
Norway	IA-NO	NO01	Aust-Agder, Birkenes	Yes							
	IA-NO	NO03	Buskerud, Langtjern	Yes							
	IA-NO	NO04	Finnmark, Dalelv	Yes							
	IA-NO	NO05	Oppland, Aurdøla	Yes							
	IA-NO	NO06	Rogaland, Vikedalselva	Yes							
	IA-NO	NO07	Sogn og Fjordane, Gaula	Yes							
	IA-NO	NO08	Sogn og Fjordane, Nausta	Yes							
	IA-NO	NO09	Sogn og Fjordane, Trodøla	Yes							
	IA-NO	NO10	Telemark, Storgama	Yes							
Poland	IA-PL	PL01	Tatra Mountains, Dlugi Staw Gasienicowy	Yes							
	IA-PL	PL02	Tatra Mountains, Zielony Staw Gasienicowy	Yes							
Russia	IA-RU	RU01	Shidmozero	No							X
	IA-RU	RU02	Yamozero	No							X
	IA-RU	RU03	Lum	No							X
	IA-RU	RU04	Soozero	No							X
	IA-RU	RU05	Dubovoye	No							X
	IA-RU	RU06	Ivan	No							X
	IA-RU	RU07	Chuna	No							X
	IA-RU	RU08	Svetloe	No							X
	IA-RU	RU09	Arkashkino	No							X
	IA-RU	RU10	Glubokoe	No							X
Sweden	IA-SE	SE01	Delångersån Iggesund	Yes							
	IA-SE	SE02	Alsterån Getebro	Yes							
	IA-SE	SE03	Alsterån Strömsborg	Yes							
	IA-SE	SE05	Tväringen	Yes							
	IA-SE	SE06	Stensjön	Yes							
	IA-SE	SE08	Brunnsjön	Yes							
	IA-SE	SE09	Fiolen	Yes							
	IA-SE	SE10	Storasjö	Yes							
	IA-SE	SE11	Fräcksjön	Yes							
	IA-SE	SE12	Härsvatten	Yes							
United Kingdom	IA-UK	UK01	Scotland, Loch Coire nan Arr	Yes							
	IA-UK	UK04	Scotland, Lochnagar	Yes							
	IA-UK	UK07	Scotland, Round Loch of Glenhead	Yes							

Country	**Dataset**	**ICP Site Number**	**Site Name**	**Used for Trends**	**Reason for exclusion**						
					Incomplete Chemistry	Insufficient or Inconsistent sampling schedule	Multiple locations per site	Insensitive to Acidification	Disturbed Watershed	Other Documented Causes for Trends	Record too Short
	IA-UK	UK10	England, Scoat Tarn	Yes							
	IA-UK	UK15	Wales, Llyn Llagi	Yes							
	IA-UK	UK21	N.Ireland, Blue Lough	Yes							
United States	IA-US	US01	Colorado, Seven Lakes	No							X
	IA-US	US02	Colorado, Summit Lake	No							X
	IA-US	US03	Colorado, Sunlight Lake	No							X
	IA-US	US04	Colorado, White Dome Lake	No							X
	IA-US	US05	Maine, Little Long Pond	No		X					
	IA-US	US06	Maine, Tilden Pond	No		X					
	IA-US	US07	Michigan, Andrus	Yes							
	IA-US	US08	Michigan, Buckeye	Yes							
	IA-US	US09	Michigan, Johnson	Yes							
	IA-US	US10	Minnesota, Cruiser	Yes							
	IA-US	US11	New York, Adirondack Mnt., Arbutus	Yes							
	IA-US	US12	New York, Adirondack Mnt., Constable	Yes							
	IA-US	US13	New York, Adirondack Mnt., Dart Lake	Yes							
	IA-US	US14	New York, Adirondack Mnt., Heart Lake	Yes							
	IA-US	US15	New York, Adirondack Mnt., Lake Rondaxe	Yes							
	IA-US	US16	New York, Adirondack Mnt., Moss Lake	Yes							
	IA-US	US17	New York, Adirondack Mnt., Otter Lake	Yes							
	IA-US	US18	New York, Catskill Mnt., E. Branch Neversink, Midreach	No							X
	IA-US	US19	Wisconsin, Luna	Yes							
	IA-US	US20	Wisconsin, Nichols	Yes							
	IA-US	US21	Wisconsin, Sand	Yes							
	IA-US	US22	New York, Catskill Mnt., High Falls Brook	Yes							
	IA-US	US23	New York, Catskill Mnt., E. Branch Neversink, Headwaters	Yes							
	IA-US	US24	New York, Catskill Mnt., Rondout Creek	Yes							

Appendix B. Z-scores and slopes for the single sites

SKT Z-scores and Theil trend slope for H^+, SO_4*, NO_3, SBC*, ANC, TOC and DOC. Negative scores indicate decreasing trends, while positive scores indicate increasing trends. Scores > 1.96 or < -1.96 are significantly different from 0 at the 0.005 significance level. These scores are outlined in bold and grey.

	SKT Z-scores							Theil trend slope						
Site	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC
								µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	mg C/L/yr	mg C/L/yr
CA01	-0.65	**-2.56**	0.84	**-2.93**	**-2.22**		1.31	-0.01	**-2.97**	0.08	**-4.63**	**-2.03**		0.05
CA02	-0.96	**-2.71**	1.13	**-2.60**	**-1.96**		0.23	0.00	**-3.03**	0.33	**-3.98**	**-1.76**		0.01
CA03	-0.48	**-2.71**	1.95	**-2.72**	-1.95		0.33	0.00	**-3.26**	0.61	**-3.80**	-1.75		0.01
CA04	-0.50	**-2.93**	**2.38**	**-2.82**	**-2.19**		1.57	0.00	**-3.05**	**0.38**	**-5.31**	**-2.43**		0.04
CA05	-0.93	**-2.95**	0.77	0.00	**2.18**		-0.10	-0.04	**-2.85**	0.17	0.00	**3.43**		0.00
CA06	-1.95	**-2.85**	0.89	0.47	**2.56**		0.71	-0.10	**-3.74**	0.00	0.17	**4.29**		0.03
CA07	-0.78	**-2.76**	1.64	-1.04	**2.17**		1.53	-0.27	**-3.33**	0.17	-0.42	**3.20**		0.18
CA08	0.17	**-2.97**	-1.81	-1.64	1.36		1.27	0.00	**-3.16**	-0.23	-1.39	2.78		0.12
CA09	1.13	**-2.20**	**2.49**	-1.59	0.88		0.86	0.35	**-2.15**	**0.27**	-1.93	0.60		0.17
CA11	0.33	**-2.45**	1.27	-1.68	1.06		1.92	0.00	**-1.86**	0.00	-0.75	1.36		0.12
CA13	**2.00**	**-2.92**	**2.48**	-1.87	-0.57		1.16	0.65	**-1.95**	**0.10**	-1.26	-0.70		0.36
CA14	1.33	**-2.85**	**2.33**	**-2.29**	1.08		1.57	0.17	**-1.79**	**0.09**	**-0.94**	0.70		0.13
CA16	0.00	**-2.37**	0.00	-0.45	1.15		1.91	0.00	**-1.66**	0.00	-0.35	1.36		0.08
CA17	-1.33	**-2.59**	1.18	-0.89	1.16		1.95	-0.05	**-4.20**	0.00	-1.18	1.64		0.17
CZ01	-1.55	**-3.47**	-1.64	**-2.52**	1.59		0.67	-0.45	**-4.38**	-1.30	**-1.68**	3.30		0.14
CZ02	-1.39	**-2.94**	-1.67	**-2.51**	**3.20**		1.33	-0.44	**-7.22**	-1.38	**-2.12**	**6.75**		0.50
CZ03	**-1.98**	**-2.37**	-0.74	**-2.46**	**2.53**		0.67	**-0.70**	**-7.78**	-0.90	**-2.85**	**5.72**		0.20
CZ04	-0.83	**-2.47**	0.98	-0.85	**2.86**		0.59	-0.46	**-3.90**	0.68	-0.53	**3.23**		0.16
CZ05	1.46	0.18	0.20	-1.30	**-2.08**	0.00	0.00	0.17	0.04	0.33	-0.68	**-0.86**	0.24	
CZ06	0.66	-1.51	-0.08	**-2.60**	-1.44	0.00	0.00	0.02	-2.35	0.00	**-2.27**	-1.67	0.63	
DE01	-1.46	-0.77	1.66	**2.56**	**2.36**		-0.76	-0.25	-0.38	0.51	**7.75**	**7.46**		-0.05
DE02	0.92	-0.72	1.71	1.82	0.00	**-1.27**	0.00	0.04	-0.60	0.59	1.50		**-0.05**	

	SKT Z-scores							Theil trend slope						
Site	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC	H^+ µekv/L/yr	SO_4* µekv/L/yr	NO_3 µekv/L/yr	SBC* µekv/L/yr	ANC µekv/L/yr	TOC mg C/L/yr	DOC mg C/L/yr
DE03	-0.56	**-2.28**	1.67	**-2.23**	-0.85		0.92	-0.01	**-4.92**	1.22	**-6.12**	-2.10		0.02
DE05	-0.48	**-2.59**	-0.35	1.76	1.69		-0.43	-0.01	**-3.24**	0.00	4.98	8.39		-0.02
DE06	1.51	**-2.78**	**-1.98**	-0.43	**2.72**	0.00	**2.15**	2.60	**-11.10**	**-6.46**	-2.11	**18.45**		**0.18**
DE07	0.05	-1.68	**-2.56**	-1.30	1.42	0.67	1.16	0.00	-55.16	**-7.10**	-19.65	41.89	3.05	0.20
DE08	**-2.05**	**-2.16**	-0.73	-1.23	1.42	-0.38	-1.32	**-0.05**	**-0.99**	-0.23	-0.92	3.04	0.00	-0.17
DE10	-1.06	**-2.02**	0.43	-0.09	0.00			-0.02	**-1.23**	0.60	-0.08	-2.75		
DE11	-1.20	-1.61	-1.80	1.93	1.77		-1.45	0.00	-0.53	-1.01	4.24	4.63		-0.07
DE12	0.34	0.17	**-2.63**	**-2.88**	-1.91	-0.83		0.00	0.18	**-1.58**	**-7.24**	-5.09	-0.07	
DE15	1.23	-1.76	-1.50	**-2.30**	1.21		0.89	1.77	-13.16	-1.32	**-7.26**	11.84		0.27
DE17	-0.83	**-3.04**	-0.76	**-3.14**	1.12	0.63		0.00	**-5.23**	-0.83	**-2.40**	1.72	0.01	
DE18	0.36	**-2.50**	-0.36	-1.58	0.00	1.68	0.00	0.16	**-3.66**	0.00	-1.14		0.08	
DE20	1.41	-1.87	0.95	0.91	0.90		-0.86	0.05	-2.70	0.70	3.05	6.26		-0.26
DE21	-1.68	-1.22	-1.93	-0.94	-0.11	0.75	1.68	-1.71	-30.29	-7.14	-9.37	-3.63	5.53	0.50
DE22	-0.14	**-2.12**	-1.56	**-2.46**	1.35		1.88	-0.17	**-13.09**	-3.84	**-8.31**	12.49		1.88
DE23	-1.35	**-2.47**	**-2.87**	**-2.58**	0.00			-0.03	**-1.63**	**-1.79**	**-1.44**	-22.70		
DE26	**2.28**	**-2.29**	-1.69	0.04	**2.57**	0.00	-1.45	6.81	**-6.22**	-1.72	0.04	**11.11**		-0.07
DE27	-0.21	**-2.10**	-1.63	-0.63	0.00			0.00	**-1.48**	-1.30	-0.49	-56.80		
DE31	-0.27	**-2.43**	-1.46	**-2.25**	**2.46**	0.67	-0.98	0.00	**-67.01**	-11.37	**-45.07**	**84.83**	4.20	-0.39
DE32	0.00	**-2.15**	-0.51	**-2.29**	-1.15		-1.50	0.00	**-5.83**	-0.25	**-10.07**	-4.22		-0.02
DE33	-0.66	-1.69	**-3.03**	**-2.49**	0.00	0.22	0.00	-1.87	-3.09	**-5.35**	**-3.63**		0.01	
DK01	0.45	0.00	-1.41	-1.05	-1.37		1.10	0.01	0.21	-1.65	-7.35	-7.65		0.05
DK02	-1.01	-0.20	**-2.42**	-1.39	-1.47		-0.44	0.00	-0.83	**-4.69**	-13.95	-12.17		-0.04
FI01	1.00	**-3.13**	1.28	**-3.36**	1.91	0.00		0.00	**-4.23**	0.16	**-3.68**	3.14	0.00	
FI02	1.33	**-2.29**	0.92	**-1.96**	0.95	0.88		0.02	**-5.61**	0.05	**-3.29**	3.70	0.27	
FI03	-0.05	**-2.91**	-0.31	-0.69	0.39	-0.38		0.00	**-4.96**	0.00	-4.44	1.10	-0.05	
FI05	-0.70	**2.16**	-0.85	-0.35	-1.13	0.87		-0.03	1.15	0.00	-0.25	-1.68	1.43	
FI06	-1.46	**-2.67**	-1.15	**-2.26**	-1.76	0.83		-0.05	**-0.87**	0.00	**-1.78**	-1.45	0.10	
FI08	**-2.75**	**-3.71**	0.91	**-2.03**	2.96	2.66		-0.41	**-2.98**	0.09	**-0.40**	2.75	0.16	
IT01	-0.30	**-3.02**	-1.19	**-2.57**	0.00			0.00	**-1.57**	-0.63	**-1.99**	-0.19		

	SKT Z-scores							Theil trend slope						
Site	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC
								µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	mg C/L/yr	mg C/L/yr
IT03	**-1.99**	**-2.07**	0.08	-1.29	0.27			**-0.15**	**-1.15**	0.04	-0.91	0.09		
IT05	0.66	-1.17	1.89	0.58	0.14			0.00	-0.69	1.58	1.18	0.28		
IT06	0.58	**-2.79**	1.72	-1.95	-1.28			0.00	**-1.70**	1.49	-2.77	-2.03		
NO01	-0.92	-1.59	**-2.29**	**-2.79**	1.86	1.45		-0.58	-1.85	**-0.25**	**-1.79**	2.35	0.10	
NO03	**-2.00**	**-2.47**	1.19	-0.68	**2.72**	**2.98**		**-0.43**	**-2.51**	0.02	-0.59	**2.67**	**0.36**	
NO04	**-2.51**	**-3.37**	1.86	-1.78	1.31	0.86		**-0.04**	**-2.86**	0.03	-1.31	2.04	0.04	
NO05	**-2.75**	**-2.76**	**1.98**	1.77	**2.78**	**2.54**		**-0.04**	**-1.11**	**0.07**	0.78	**2.50**	**0.12**	
NO06	**-2.78**	-1.07	-0.35	**2.86**	**2.53**	**2.03**		**-0.34**	-0.38	-0.05	**1.08**	**2.06**	**0.02**	
NO07	**-2.90**	-1.62	0.84	1.84	**2.31**	1.37		**-0.28**	-0.42	0.13	0.72	**1.19**	0.03	
NO08	**-2.93**	-1.51	-0.26	1.77	**2.24**	1.23		**-0.13**	-0.28	-0.03	0.55	**1.44**	0.02	
NO09	**-2.79**	**-1.97**	**2.00**	**2.04**	1.76	**2.08**		**-0.23**	**-0.32**	**0.19**	**0.37**	0.89	**0.02**	
NO10	**-2.89**	**-3.06**	-0.44	-1.30	**3.23**	**3.00**		**-1.41**	**-3.55**	-0.05	-0.44	**3.34**	**0.22**	
PL01	-1.54	**-2.77**	**-2.65**	**-2.77**	0.76	-0.43		-0.15	**-3.82**	**-2.76**	**-5.70**	0.55	0.00	
PL02	-0.51	**-2.94**	-1.90	**-2.39**	-1.14	0.70		0.00	**-4.15**	-1.50	**-6.90**	-1.67	0.02	
SE01	**-2.62**	-1.70	-0.97	**2.59**	**2.38**	0.63		**0.00**	-1.48	-0.14	**1.83**	**3.02**	0.02	
SE02	**-1.97**	**-2.87**	0.75	-1.85	**2.89**	**2.27**		**-0.02**	**-18.65**	0.09	-5.64	**12.72**	**0.40**	
SE03	**-2.11**	**-2.11**	-0.97	0.11	**2.10**			**-0.03**	**-14.59**	-0.25	0.45	**13.24**		
SE05	-0.04	**-2.45**	-0.25	-1.87	-0.93	1.12		0.00	**-1.80**	-0.02	-2.53	-1.47	0.07	
SE06	-1.82	**-1.97**	-1.10	-1.08	0.09	0.08		-0.04	**-1.08**	-0.02	-0.60	0.12	0.01	
SE08	-0.96	**-2.57**	0.91	**-2.05**	**1.99**	**1.97**		-0.07	**-22.91**	0.34	**-20.05**	**3.82**	**0.65**	
SE09	**-2.96**	-1.85	**-2.25**	**-2.27**	1.24	-0.81		**-0.04**	-2.30	**-0.83**	**-1.86**	1.70	-0.05	
SE10	**-2.11**	**-2.36**	1.14	-1.69	1.57	1.60		**-0.27**	**-6.50**	0.06	-2.55	3.35	0.42	
SE11	**-2.29**	**-3.02**	-1.68	**-2.17**	1.94	1.75		**-0.03**	**-6.17**	-0.12	**-4.57**	4.60	0.17	
SE12	**-2.98**	**-3.41**	**-2.90**	**-2.79**	1.26	0.04		**-2.00**	**-5.70**	**-0.56**	**-3.72**	2.96	0.00	
UK01	-0.29	0.36	1.10	-0.54	0.80		**2.76**	0.00	0.12	0.00	-0.83	1.83		**0.20**
UK04	1.86	**-2.54**	**2.02**	-1.24	-0.48		1.36	0.21	**-0.63**	**1.25**	-0.32	-0.71		0.05
UK07	-0.75	0.04	**2.62**	0.15	1.27		**2.33**	-0.20	0.25	**0.75**	0.15	1.24		**0.11**
UK10	-1.76	0.66	0.33	-1.76	0.00		**2.54**	-0.26	0.32	0.34	-0.83	0.04		**0.10**
UK15	**-2.32**	-0.24	0.76	-1.49	0.94		1.76	**-0.56**	-0.56	0.27	-1.50	0.86		0.10

	SKT Z-scores							**Theil trend slope**						
Site	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC	H^+	SO_4*	NO_3	SBC*	ANC	TOC	DOC
								µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	µekv/L/yr	mg C/L/yr	mg C/L/yr
UK21	-1.74	-1.30	1.07	-1.37	0.61		**2.45**	-0.49	-1.81	1.00	-1.19	2.43		**0.20**
US07	0.00	**-2.15**	0.00	-1.25	**2.29**		**2.13**	-0.10	**-4.22**	0.00	-1.08	**3.09**		**0.27**
US08	0.00	-0.20	0.19	-1.14	-1.27		0.50	0.00	-0.56	0.00	-2.40	-2.13		0.04
US09	-1.18	**-2.49**	0.00	-1.83	1.76		1.60	-4.00	**-4.18**	0.00	-1.58	3.60		0.17
US10	-1.06	-0.21	-0.16	0.00	0.76		**2.19**	0.00	0.00	0.00	0.00	1.50		**0.10**
US11	0.92	**-2.95**	-1.79	**-3.17**	**-2.68**		0.49	0.00	**-2.69**	-0.44	**-5.77**	**-3.85**		0.03
US12	-1.19	**-2.12**	-0.35	-1.87	0.10		0.60	-0.45	**-2.42**	-0.33	-2.54	0.25		0.02
US13	-1.83	**-2.44**	-1.37	**-2.63**	0.29		1.29	-0.25	**-2.09**	-0.99	**-2.43**	0.16		0.07
US14	-0.78	**-2.94**	-1.07	**-2.20**	-0.74		0.00	0.00	**-1.98**	-0.35	**-2.09**	-0.55		0.00
US15	-1.35	**-2.81**	-1.80	**-2.28**	0.27		0.88	-0.04	**-2.39**	-1.01	**-2.23**	0.33		0.05
US16	-1.04	**-2.84**	-1.66	**-2.32**	-1.33		0.76	0.00	**-2.35**	-0.88	**-4.07**	-2.22		0.03
US17	-1.79	**-2.54**	-1.34	**-2.40**	-0.48		-0.91	-0.23	**-1.59**	-0.60	**-2.96**	-0.66		-0.01
US19	0.89	1.80	-0.46	1.77	**-2.30**		-0.83	0.07	2.10	0.00	0.43	**-4.46**		-0.07
US20	0.61	**-2.52**	0.40	-1.94	1.72		0.00	0.10	**-4.87**	0.00	-1.43	3.05		0.00
US21	0.27	**-3.02**	-0.10	**-2.35**	1.89		1.16	0.05	**-5.53**	0.00	**-3.33**	3.05		0.10
US22	-0.10	-0.73	-1.94	-1.37	-0.31		-0.58	0.00	-2.00	-5.55	-9.42	-3.43		-0.10
US23	0.70	**-2.14**	**-2.10**	-1.69	0.09		0.03	0.20	**-1.68**	**-2.54**	-4.72	0.09		0.00
US24	-0.58	**-2.02**	-1.82	**-2.13**	1.18		0.00	-0.50	**-1.87**	-1.94	**-4.55**	1.30		0.00

Appendix C. ICP Waters Publications

1. Manual for Chemical and Biological Monitoring. Programme Manual. 1987. Prepared by the Programme Centre, Norwegian Institute for Water Research. NIVA, Oslo.
2. Norwegian Institute for Water Research. 1987. Intercalibration 8701. pH, Ks, SO_4, Ca. Programme Centre, NIVA, Oslo.
3. Norwegian Institute for Water Research. 1988. Data Report 1987 and available Data from Previous Years. Programme Centre, NIVA, Oslo.
4. Norwegian Institute for Water Research. 1988. Intercalibration 8802. pH, K_{25}, HCO_3, NO_3, SO, Cl, Ca, Mg, Na, K. Programme Centre, NIVA, Oslo.
5. Proceedings of the Workshop on Assessment and Monitoring of Acidification in Rivers and Lakes, Espoo, Finland, 3rd to 5th October 1988. Prepared by the Finnish Acidification Research Project, HAPRO, Ministry of Environment, October 1988.
6. Norwegian Institute for Water Research. 1989. Intercalibration 8903: Dissolved organic carbon and aluminium fractions. Programme Centre, NIVA, Oslo. NIVA-Report SNO 2238-89. ISBN 82-577-1534-4.
7. Hovind, H. 1989. Note: Some reflections about the determination of pH and alkalinity. Programme Centre, Norwegian Institute for Water Research, NIVA, Oslo.
8. Hovind, H. 1990. Intercalibration 9004: pH and alkalinity. Programme Centre, NIVA, Oslo. NIVA-Report SNO 2465-90. ISBN 82-577-1776-2.

- Skjelkvåle, B.L. and Wright, R.F. 1990. Overview of areas sensitive to acidification: Europe. Programme Centre, NIVA, Oslo. Acid Rain Research Report 20/1990. NIVA-Report 2405-90. ISBN 82-577-1706-1.

9. Johannessen, M. 1990. Intercalibration in the framework of an international monitoring programme. Proceedings of the third annual Ecological Quality Assurance Workshop, Canada Centre for Inland Waters, Burlington Ontario. Programme Centre, NIVA, Oslo.
10. Norwegian Institute for Water Research. 1990. Data Report 1988. Programme Centre, NIVA, Oslo.
11. Norwegian Institute for Water Research. 1990. Data Report 1989. Programme Centre, NIVA, Oslo.
12. Proceedings for the 5th Meeting of the Programme Task Force Freiburg, Germany, October 17 -19, 1989. Prepared by the Umweltbundesamt, Berlin July 1990.
13. Hovind, H. 1991. Intercalibration 9105: pH, K_{25}, HCO_3, $NO_3 + NO_2$, Cl, SO_4, Ca, Mg, Na, K and TOC. Programme Centre, NIVA, Oslo. NIVA-Report 2591-91. ISBN 82-577-1931-5.
14. Norwegian Institute for Water Research. 1991. The Three Year Report. Summary and results 1987 – 1989: Results from the International Co-operative Programme on Assessment and Monitoring of Acidification in Rivers and Lakes. Programme Centre, NIVA, Oslo.
15. Norwegian Institute for Water Research. 1991. Summary of The Three Year Report 1987 – 1989. Programme Centre, NIVA, Oslo.
16. Scientific papers presented at the Sixth Task Force meeting in Sweden 23 - 24 October 1990. Swedish Environmental Protection Agency, Sweden, September 1991.

17. Seventh Task Force meeting of international Co-operative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes. Galway, Ireland. September 30 - October 3 1991. Proceedings.
18. Johannessen, M., Skjelkvåle, B.L. and Jeffries, D. 1992. International cooperative Programme on Assessment and Monitoring of Rivers and Lakes. In: Conference Abstracts, Intern. Conference on Acidic Deposition, Glasgow 16-21, sept. 1992, p. 449. Kluwer Academic Press.
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