REPORT SNO. 6218-2011

ICP Waters Report 106/2011
Trends in precipitation chemistry, surface water chemistry and aquatic biota in acidified areas in Europe and North America from 1990 to 2008

Langtjern (Photo: Olav Olsen/Aftenposten)

International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes

Convention on Long-Range Transboundary Air Pollution

Norwegian Institute for Water Research
– an institute in the Environmental Research Alliance of Norway

REPORT

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Title Trends in precipitation chemistry, surface water chemistry and aquatic biota in acidified areas in Europe and North America from 1990 to 2008.	Report No.. SNO 6218/11 ICP Waters report 106/2011	Date
	Project No. 23300	Pages: 126 Price:
Ed(s) Skjelkvåle, B.L. and de Wit, H.A.	Topic group Effects of air pollution	Distribution
	Geographical area Europe and North America	Printed NIVA

Client(s) The Norwegian Climate and Pollution Agency (Klif) United Nations Economic Commission for Europe (UNECE)	Client ref.

Abstract
Results from international monitoring programmes on precipitation chemistry and surface water chemistry and biota for the time period 1990 to 2008 are reported here. Concentrations of sulphate and nitrate in precipitation in large areas in Europe and North America have decreased due to reduction of emissions to the atmosphere. For sulphate in surface waters, the same pattern can be seen although the reduction is not as large as in precipitation. Nitrate, however, does not show uniform decreasing trends. The acidity of lakes and rivers has decreased and in many areas there are good conditions for recovery of aquatic biological communities. Several areas in Europe will, however, not achieve good (non-acidified) water quality with current legislation of emissions of acidifying components. Future reductions of both S and N deposition are necessary to achieve biological recovery. hjelpelinje på 4 pt som gir "luft" etter siste linje - skrives ikke ut - er "hidden"

4 keywords, Norwegian	4 keywords, English
1.	1. Air pollution
2.	2. Surface water chemistry
3.	3. Biological recovery
4.	4. Long-term trends

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ISBN 978-82-577-5953-7

CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON
ASSESSMENT AND MONITORING EFFECTS OF AIR
POLLUTION ON RIVERS AND LAKES

Trends in precipitation chemistry, surface water chemistry and aquatic biota in acidified areas in Europe and North America from 1990 to 2008.

Prepared at the ICP Waters Programme Centre
Norwegian Institute for Water Research
Oslo, October 2011

Preface

The international cooperative programme on assessment and monitoring of air pollution on rivers and lakes (ICP Waters) was established under the Executive Body of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) in July 1985. Since then ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. Numerous assessments, workshops, reports and publications covering the effects of long-range transported air pollution have been published over the years.

The ICP Waters Programme Centre is hosted by the Norwegian Institute for Water Research (NIVA), while the Norwegian Climate and Pollution Agency (Klif) leads the programme. The Programme Centre's work is supported financially by Klif and from the UNECE LRTAP Trust Fund.

The main aim of the ICP Waters Programme is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters. More than 20 countries in Europe and North America participate in the programme on a regular basis.

The results from the ICP Waters Programme clearly show that surface waters respond to changes in atmospheric deposition. Surface waters are far more responsive than either soils or terrestrial vegetation to changes in long-range transported acid deposition. Lakes and rivers also have the advantage that they integrate response over the entire catchment area. The ICP Waters site network is geographically extensive and includes long-term data series (> 20 years) for more than 100 sites. The network is thus well poised to document changes that result from implementation of the protocols.

Beginning in 1991, ICP Waters has evaluated trends in surface water chemistry and biology in Europe and North America due to decreases in emissions of S and N since 1980. This report is the 7th of these 3-year reports and comprises data for the period from 1990 to 2008.

Oslo, November 2011

Brit Lisa Skjelkvåle
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Summary

Under the Convention on Long-range Transboundary Air Pollution (CLRTAP), acidifying components in precipitation, surface water chemistry and biota are monitored in international programmes. Here, results and trends are reported for Europe and North America for the period 1990 to 2008 in precipitation chemistry (from EMEP, Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe) and surface water chemistry and biota (from ICP Waters, The International Cooperative Programme on Assessment and Monitoring of Air Pollution of Rivers and Lakes).

From 1990 to 2008 the concentrations of sulphate and nitrate in precipitation have decreased in large areas in Europe and North America due to emission reductions. The reductions were larger from 1990 to 1999 than from 1999 to 2008. The same pattern can also be seen for sulphate in surface water. Nitrate, in contrast to sulphate, does not show uniform decreasing trends despite the decrease in nitrogen deposition. The acidity of lakes and rivers has decreased due to the decrease in sulphate and many places there are good conditions for recovery of aquatic biological communities that have been damaged due to acidification.

Full biological recovery is not documented anywhere. A return to pre-industrial biodiversity is unlikely in most cases, because original species are extinct, new species have been introduced and biological processes are often non-reversible. Several areas in Europe will never achieve good (non-acidified) water quality with current legislation of emissions of acidifying components. Future reductions of both S and N deposition are necessary to achieve biological recovery.

Extended summary

Atmospheric drivers of surface water acidification are strongly reduced; precipitation is much less acidic

Trends in precipitation chemistry (SO_4, NO_3, NH_4, base cations and H^+) from 27 European sites (EMEP) and 34 North American sites (CAPMoN - Canadian Air and Precipitation Monitoring Network; NADP in US) for the period 1990-2008, as well as for the two 10-year periods 1990-1999 and 1999-2008 are reported here. There were no significant trends in precipitation amount. Trends in chemistry were therefore assumed to reflect trends in deposition.

- **SO_4:** All European sites and all North American sites, except one, showed decreasing trends in SO_4 for 1990-2008, with an average reduction of 56% and 37% respectively. The largest reductions took place before 2000.
- **NO_3:** North America had a more frequent and larger reduction (30%) in NO_3 than European stations (23%) for 1990-2008. In Europe, most of the reductions in NO_3 took place before 2000, while in North America the largest decline was observed after 2000. This was related to timing of implementation of emission reduction policy.
- **pH**: Trends in hydrogen ions (pH) were lower in Europe than in North America with reductions of 34% and 46%, respectively.

The trends in precipitation chemistry were consistent with reported trends in emissions of acidifying components in Europe and North America.

Lakes and rivers are less acidic; potential for biological recovery

Water chemical trend analyses have been performed in nearly 200 acid-sensitive monitoring sites in Europe and North America. The results show a consistent pattern of chemical recovery across a large number of these sites from 1990 to 2008. Trends were calculated also for 1990-1999 and 1999-2008. Key variables that describe surface water acidification status are sulphate, pH, alkalinity and acid neutralizing capacity (ANC). Trends in these variables in 12 regions in Europe and North America document chemical recovery of acidification.

- **Sulphate:** The decline in sulphate between 1990 and 2008 ranged between 20% and 58% in the 11 regions where significant reductions were found. About 2/3 of this was achieved before 2000. The declines in sulphate were largest before 2000 both in precipitation and in surface waters.
- **Nitrate:** The relative importance of nitrate for acidification of surface waters has increased in most regions because of the decreases in sulphate, but sulphate is still the far most important acidifying anion in most acidified surface waters. Nitrate, in contrast to sulphate, does not show uniform decreasing trends despite the decrease in nitrogen deposition.
- **Acidity: pH/alkalinity/ANC:** All regions except one showed positive regional trends in pH, and/or alkalinity and/or ANC in one or both periods, which shows recovery from acidification. Most recovery took place between 1990 and 1999, but chemical recovery continued after 2000. The largest and regionally most extensive reductions were found for sulphate, while trends in pH, alkalinity and ANC were less distinct both temporally and geographically.
- **Base cations:** The concentrations of base cations, particularly at low levels, are important for aquatic biota. Base cations can protect the biota i.e. damage caused by acidification. Base cations decline almost everywhere as a result of the decline in sulphate, but more slowly after 2000. The Alps are the exception to this rule with increases in base cations after 2000.
- **Organic carbon:** DOC has increased in many of the monitoring sites. Changes in DOC affect light penetration, primary production and oxygen concentrations. Binding by DOC reduces the toxicity of aluminium. Labile forms of aluminium, whose trends are highly dependent on

acidification but not discussed in this report, are toxic at high concentrations to aquatic biota. About 15 % of all sites showed significantly increasing concentrations of DOC in both periods.

The improvements in acidification of surface waters are due to lower acid deposition. The reductions of acidifying components in precipitation were larger and more rapid than the observed improvements in water chemistry. Water chemistry responses to changed precipitation chemistry are delayed by catchment processes. Increase in pH, alkalinity and ANC indicate that biological recovery can be expected.

Biological recovery is under way in Europe, but full recovery is still far ahead.

Six countries (Czech Republic, Finland, Germany, Norway, Sweden and Switzerland) reported on biological recovery from national monitoring programmes. Most contributions focused on recovery of zoobenthos (small organisms that live on the bottom of rivers and lakes such as aquatic insects, worms and snails), but status of fish populations, algae and macrophytes (water plants) were also given. Zoobenthos have a short life cycle and are therefore able to respond more quickly to improved water chemistry than fish, which makes these organisms suitable as early indicators of biological recovery.

Almost all contributions reported evidence of biological recovery which was attributed to improved water quality, although other factors such as climate also contributed to explaining temporal variations. Higher species diversity was observed while species composition in many places has become more similar to non-acidified communities.

Positive developments, however, must not be mistaken for full biological recovery. Comparison with reference sites suggests that species diversity in fully restored aquatic ecosystems could be much higher than is presently observed in aquatic systems that are under recovery from acidification. Additionally, extinction of original populations and recolonisation by other acid-sensitive species are two important reasons why recovery from acidification is unlikely to produce the pre-acidification biological community.

Do emission reduction policies have their intended effect – full biological recovery?

Emission reduction policies for sulphur and nitrogen have been effective in reducing the atmospheric deposition load to sensitive aquatic ecosystems. The relative large reduction of sulphate in deposition is not fully reflected in the reduction of sulphate in surface waters, an indication that catchment processes delay chemical recovery. Reductions in sulphate in surface waters have resulted in improved conditions for biology as documented by increased pH, alkalinity and ANC. These improvements are slower than changes in sulphate concentrations because they depend on chemical processes in the soil that can be slow. Soil base cations have been leached at increased rates from catchments since the onset of acidification and soil weathering processes are often too slow to replenish the soil base cation stores to pre-acidification sizes. Replenishment of soil base cation stores may take centuries.

Biological recovery has been documented in many regions in Europe, especially improvements in zoobenthos, small sediment-dwelling aquatic species with short life cycles. Data on fish populations are scarcer but also here improvements have been documented, in particular for fish that are not extremely sensitive to acidification. Still, full recovery has been not documented anywhere. A return to pre-industrial biodiversity is unlikely in most cases, because original species are extinct, new species have been introduced and biological processes are often non-reversible.

Several areas in Europe will not achieve good (non-acidified) water quality with current legislation of emissions of acidifying components. Future reductions of both S and N deposition are necessary to achieve biological recovery.

1. Introduction

1.1 The ICP Waters Programme

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 across national boundaries. Acidifying compounds thus affect surface waters, groundwaters and forest soils far beyond their country of origin. 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. The Working Group on Effects (WGE) has aided the Convention by developing science to support Protocols. The WGE’s six International Cooperative Programmes (Modelling and Mapping, Waters, Vegetation, Forests, Materials, Integrated Monitoring) and a Joint Task Force with the World Health Organisation (WHO) on Human Health quantify effects on the environment through monitoring, modelling and scientific review.

The International Cooperative Programme on Assessment and Monitoring of Air Pollution of Rivers and Lakes (ICP Waters) was established under the Executive Body of the Convention on LRTAP at its third session in Helsinki in July 1985. Canada was appointed as lead country for the first phase of the ICP Waters.

The monitoring programme is designed to assess, on a regional basis, the degree and geographical extent of acidification of surface waters. The collected data 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. The ICP Waters Programme is based on existing programmes in participating countries, implemented by voluntary contributions.

The programme aims and objectives (reviewed at the ICP Waters 15th Task Force meeting in Pallanza, Italy October, 1999) 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

1.2 The current trend report

The aim of this report is to assess and compare trends in acidifying air pollution, surface water chemistry and aquatic biota for the period 1990 through 2008 in Europe and North America. In contrast to earlier trend reports, the entire 20-year period is assessed for surface water chemistry which allowed for comparison of trends 1990-1999 and 1999-2008. Surface water chemistry trends have been analysed at the ICP Waters programme centre based data delivered to ICP Waters by national focal points. The assessment of biological recovery is based on contributions of relevant data from countries participating in the ICP Waters Programme and is synthesized by the ICP Waters subcentre.

A feature that was not present in earlier trend reports is the assessment on trends in precipitation chemistry in Europe and North America, the driver of change in surface water chemistry in acid-sensitive regions. This analysis was contributed by EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe), CAPMoN (Canadian Air and Precipitation Monitoring Network) and NADP (National Atmospheric Deposition Program in the US).

The report is structured as follows:

- **Precipitation chemistry** - trends in chemistry of precipitation in Europe and North America 1990-2008 (1990-1999 and 1999-2008) (Chapter 2)
- **Water chemistry -**Trends in surface water chemistry 1990-1999 and 1999-2008 (Chapter 3)
- **Biology –** Updated trend analysis on biological recovery (Chapter 3)

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2. Trends in chemistry of precipitation in Europe and North America in 1990-2008

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Atmospheric deposition of acidifying compounds drives surface water acidification, and biological responses to changed water chemistry, in acid-sensitive regions. Observed trends in surface water chemistry and biological recovery, based on ICP Waters data, are presented in the current report. Here, a similar analysis as for the surface water chemistry was conducted for atmospheric deposition.

2.1 Monitoring networks used for trend analysis

Data from the large scale deposition networks in Europe (EMEP) and North America (NADP/NTN and CAPMoN) have been used to assess the changes in contribution from wet deposition. We report trends from 27 European sites and 34 North American sites for the period 1990-2008, as well as for the two ten years period 1990-1999 and 1999-2009, to determine if the rate of deposition has changed. These networks are to a large extent comparable, they have all reference methods which are harmonised with the WMO Manual for precipitation sampling (WMO, 2004). The common siting criteria also ensures that the measurement locations are regionally representative, i.e., not affected by local sources of air pollution. Details of which sites have been used are found in Appendix A.

EMEP
On a regional European scale, investigation and mitigation of atmospheric pollution occur to a large extent under the framework of EMEP (Co-operative programme for monitoring and evaluation of the long-range transmission of air pollutants in Europe, www.emep.int) under the UNECE Convention on Long Range Transboundary Air Pollution (CLTRAP). The main objective of EMEP is to provide observational and modeling data on pollutant concentrations, deposition, emissions and transboundary fluxes on the regional scale and identify their trends in time. Furthermore, EMEP's aims are to identify the sources of the pollution concentrations and depositions and to assess the effects of changes in emissions. EMEP comprises almost 100 sites covering major ions in precipitation though only a selection of these have data for the whole time span 1990-2008. After the selection of suitable sites, 27 sites from 14 countries (Austria, Switzerland, Germany, Denmark, Finland, France, Great Britain, Hungary, Lithuania, Latvia, Norway, Poland, Sweden, and Serbia) have been used.

CAPMoN
The Canadian Air and Precipitation Monitoring Network (CAPMoN, http://www.ec.gc.ca/rs-mn) is Canada's national daily network for monitoring regional-scale air, ozone, nitrogen, particulate matter and precipitation quality. CAPMoN began operation in mid-1983 and the network is managed by the Science and Technology Branch of Environment Canada. As of 2008, there are 22 precipitation measurement sites in Canada (mostly in eastern Canada) and one in the USA. For the trend analysis representative for the ICP Water catchments, only sites located in the following provinces were chosen: Ontario (ON), New Brunswick (NB), Nova Scotia (NS), Québec (PQ) as well as one site in Pennsylvania USA (PA). In total 13 sites with sufficient data coverage were used.

NADP/NTN

In 1978, the National Atmospheric Deposition Program (NADP, http://nadp.sws.uiuc.edu/NADP/) was started in the USA under the leadership of the State Agricultural Experiment Stations to obtain data on atmospheric deposition and its effects on agricultural crops, forests, range lands, surface waters and other natural and cultural resources. In 1982, the NADP assumed responsibility for the National Trends Network (NTN), which was supported by the National Acid Precipitation Assessment Program. The Illinois State Water Survey is responsible for the analysis of the precipitation samples. NADP National Trends Network (NTN) currently has 250 sites distributed over the whole of USA. In this report we included only sites in northeastern USA with sufficient data coverage and representativity. In total 21 sites from the following states were used in the statistical calculations: Indiana (IN), Maine (ME), Maryland (MD), Massachusetts (MA), Michigan (MI), New Hampshire (NH), New York (NY), Ohio (OH), Pennsylvania (PA), Vermont (VT) and Virginia (VA).

2.2 Criteria for data selection and statistical method

The precipitation chemistry data for North America (both CAPMoN and NADP) were obtained from the Canadian National Atmospheric Chemistry Database and Analysis Facility operated by Environment Canada (NAtChem, http://www.on.ec.gc.ca/natchem). Although NADP data are downloadable from the NADP web site, the data were instead obtained from the NAtChem Database because this Database provides U.S. and Canadian data in a single format. The European data are from the EMEP database (ebas, http://ebas.nilu.no) hosted at the Norwegian Institute for Air Research (NILU)

To ensure that the trend analyses for the two different 10 year periods were comparable we used only sites that have data for both periods, and we have chosen sites where data were available for at least 18 out of 19 years (1990-2008). If 1990, 1999 or 2008 was the missing year, the site was excluded. A missing year is given an average value of the previous and following year, one site in North America and three sites in Europe. For the statistical analyses it is necessary to have at least ten years of data. The deposition networks in North America includes sites which are not necessarily regional representative, these sites are however flagged in the NAtChem database, and data which are coded with flag 3 and 2b were excluded here. Location of the sites is shown in ***Figure 1***.

For the present study, the non-parametric Mann-Kendall Test for detecting and estimating trends used annual volume-weighted means. The Sen slope estimator was used to quantify the scale of potential trends. The Mann-Kendall test and Sen’s slope estimator were calculated using the MAKESENS software (Salmi *et al.*, 2002).

2.3 Results of trend analysis

The results of the trend analysis for all the sites and a summary table for each network are given in Appendix A. In Table 1, a summary of the statistics for North America and Europe is presented. It shows the trends in concentration and not deposition. The inter-annual variations in deposition are larger than those in concentration, and concentration is a more robust parameter to assess trends in chemical parameters. Furthermore, the precipitation amount at the individual sites is not necessarily representative for the larger scale region. The data were investigated for a potential trend in precipitation amount to see check if wet deposition trends might be different from trends in precipitation chemistry. There were no clear trends in precipitation amount (***Table 1***), we therefore assume that the trends in concentrations reflect trends in wet deposition.

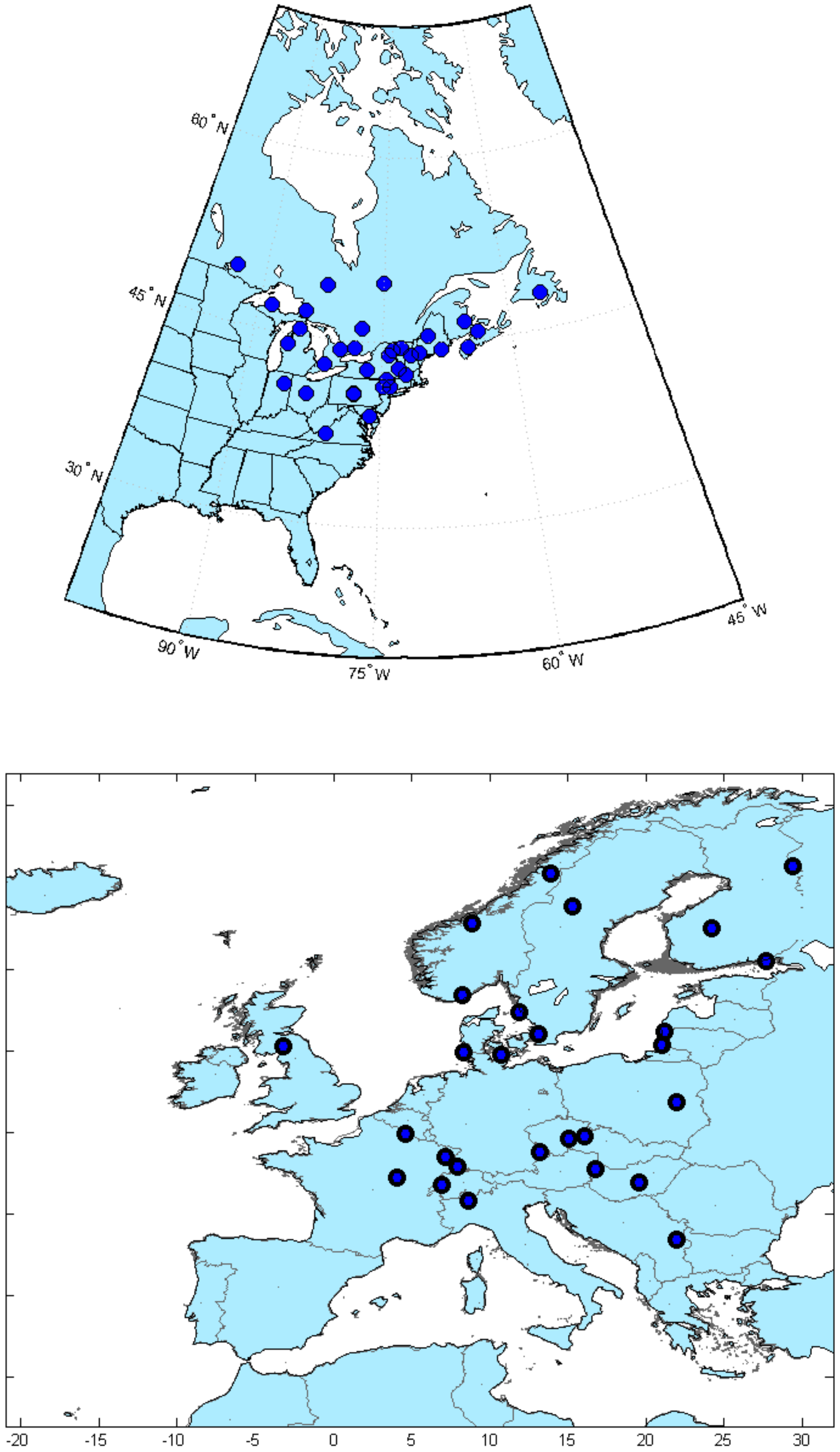

Figure 1. *Location of precipitation monitoring sites used for trends analyses in this report*

Table 1. *Mean trends in annual volume weighted mean and number of sites with significant positive or negative trend. No trend at significance level p<0.05. Mean change in % of average 1990-1992. Average for 1999 and 2008 based on 1998-2000, and 2006-2008, respectively.*

Comp	Network	Nr of sites	Period	Sites with sign. trend decrease	increase	Mean change
SO4	Europe	27	1990-2008	100 %	0 %	-56 %
			1990-1999	89 %	0 %	-40 %
			1999-2008	67 %	0 %	-16 %
	North America	34	1990-2008	97 %	0 %	-37 %
			1990-1999	50 %	0 %	-20 %
			1999-2008	41 %	0 %	-17 %
H	Europe	24	1990-2008	74 %	0 %	-34 %
			1990-1999	63 %	0 %	-29 %
			1999-2008	37 %	0 %	-4 %
	North America	34	1990-2008	100 %	0 %	-46 %
			1990-1999	47 %	0 %	-21 %
			1999-2008	71 %	0 %	-25 %
NO3	Europe	27	1990-2008	70 %	0 %	-23 %
			1990-1999	48 %	4 %	-17 %
			1999-2008	7 %	4 %	-6 %
	North America	34	1990-2008	94 %	0 %	-30 %
			1990-1999	12 %	0 %	-3 %
			1999-2008	85 %	0 %	-27 %
NH4	Europe	27	1990-2008	59 %	0 %	-29 %
			1990-1999	56 %	0 %	-25 %
			1999-2008	7 %	4 %	-4 %
	North America	34	1990-2008	0 %	9 %	2 %
			1990-1999	0 %	0 %	2 %
			1999-2008	9 %	0 %	0 %
Ca	Europe	27	1990-2008	37 %	0 %	-28 %
			1990-1999	33 %	0 %	-23 %
			1999-2008	7 %	19 %	-5 %
	North America	34	1990-2008	0 %	18 %	8 %
			1990-1999	6 %	6 %	9 %
			1999-2008	15 %	0 %	0 %
Total precip amount	Europe	27	1990-2008	4 %	11 %	13 %
			1990-1999	4 %	19 %	16 %
			1999-2008	4 %	0 %	-3 %
	North America	34	1990-2008	3 %	15 %	9 %
			1990-1999	3 %	3 %	-1 %
			1999-2008	0 %	18 %	10 %

Sulphate

The decrease in sulphur emissions in both Europe and North America have caused a substantial decrease in the atmospheric deposition the last decades, and this has been reported in several assessments and publications (i.e. EMEP, 2004; IJC 2008; Vestereng, 2007; Sickles, 2007). These reductions are naturally also reflected in precipitation chemistry. All the sites in Europe and 97% of the sites in northeastern North America show a decreasing trend from 1990-2008, with an average reduction of 56% and 37% respectively (***Figure 2***). The trends are not as strong for the two shorter ten year periods. Reductions in 1990-1999 were larger than in the period 1999-2008, most clearly in Europe.

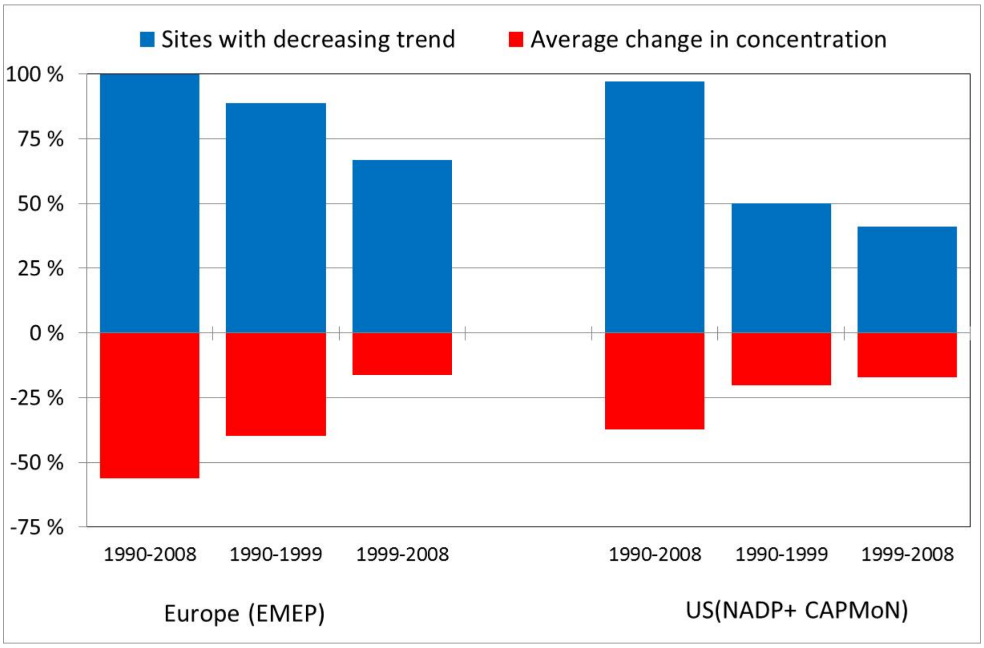

Figure 2. *Relative number of sites with decreasing trends in non-seasalt sulphur concentration in precipitation, and their average decrease.*

The emission reductions reported (***Table 2***) for Europe (EMEP 2009), Canada (Environmental Canada, 2010) and US (USEPA, 2009) reflect similar trends as seen in the observations of precipitation chemistry, where Europe as a whole has somewhat larger reduction than US and Canada, and the strongest reductions were in the first period. These are total emission reductions for the whole region and there are regional differences which may give different signals to the various sites.

Table 2. *Reductions in total sulphur emissions*

	US	Canada	Europe
1990-2008	50%	46%	61%
1990-1999[1)]	24%	28%	50%
1999-2008[1)]	35%	25%	24%

1) For Europe the trend is for the period 1990-2000 and 1999-2008.

Nitrate

Similar to sulphur, emissions of oxidised nitrogen have decreased in both North America and Europe due to regional and national abatement strategies of NO_x. However, reductions were smaller and started later than for sulphur (i.e. EMEP, 2004; IJC 2008; Vestereng, 2009; Sickles, 2007). These reductions are also reflected in the observed trends in the nitrate concentrations in precipitation in North America and Europe (***Figure 3***). The average reduction (30%) as well as the relative number of sites with significant decrease is slightly higher in North America compared to Europe, and the two regions show a very different development from 1990-2008. In Europe, most of the reductions were in the first period, 1990-1990, while in North America the reductions were implemented later and are better reflected in the last period, 1999-2008. The observed reductions in NO_x in precipitation are very much in line with the emission reductions reported by for Europe (EMEP 2009), Canada (Environmental Canada, 2010) and US (USEPA, 2010) (***Table** 3*).

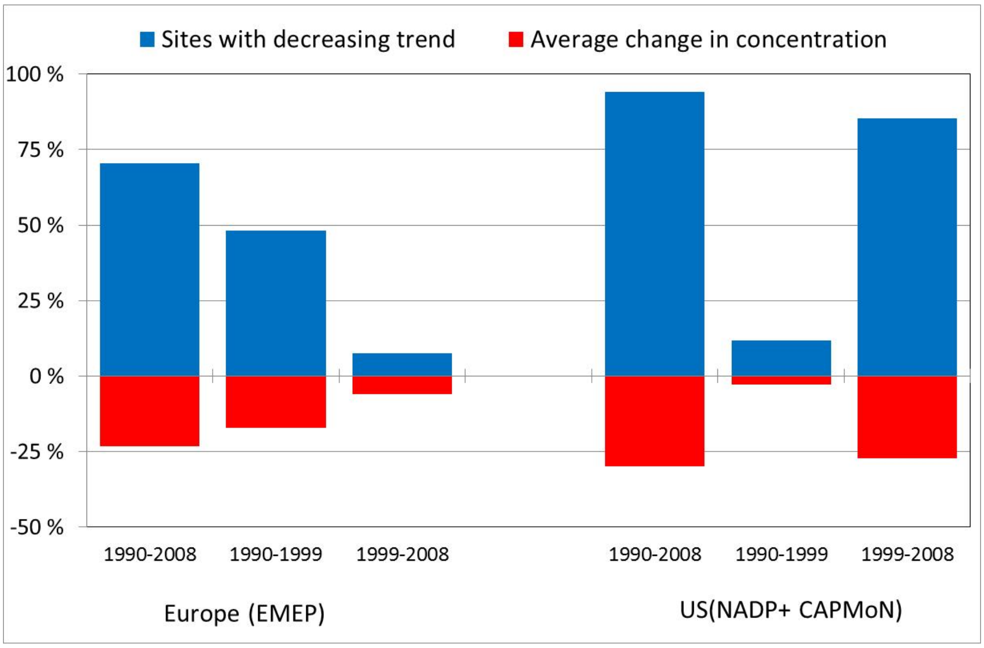

Figure 3. *Relative number of sites with decreasing trends in nitrate concentration in precipitation, and their average decrease.*

Table 3. *Reductions in total NOx emissions*

	US	Canada	Europe
1990-2008	36%	14%	25%
1990-1999[1)]	11%	-3%	24%
1999-2008[1)]	28%	13%	2%

1) For Europe the trend is for the period 1990-2000 and 1999-2008.

Ammonium, base cations and acidity

It is important to consider the deposition of ammonium and base cations when assessing exceedance of critical loads of acid deposition for surface waters. These are also important ions for assessing trends in pH (or hydrogen ions).

For ammonium there has been a clear reduction in Europe, both in the emissions (25%; EMEP, 2010) and in precipitation (27%; Table 1) since 1990, and most of the reductions were seen in the first period (1990-1999). In North America there has been little change in either period (Table 1).
The situation is similar for base cations, with significant reductions in the 1990-1999 period in Europe (Table A.5). This was a side effect of the implementation of effective abatement technologies for sulphur (EMEP, 2004). There seems to be an increase the last period at some sites which may be due to changes in the nearby surroundings and more influence of natural sources at some sites. In North America there is no clear signal, but scattered positive and negative trends at the individual sites.

To a large extent the trends in sulphur concentration are reflected in the trends in hydrogen ions (pH). A reduction of 54% and 55% occurred from 1990-2008 in Europe and North America respectively. The highest reductions came in the first period in Europe and in the second period in North America. This is probably due to the comparatively large reductions in nitrate in the second period in North America.

2.4 Conclusions

Trends in precipitation chemistry (SO_4, NO_3, NH_4, base cations and H^+) from 27 European sites and 34 North American sites for the period 1990-2008, as well as for the two ten years period 1990-1999 and 1999-2008 are reported. There was no general trend in precipitation amount. Trends in chemistry were therefore assumed to reflect trends in deposition.

- **SO_4:** All European sites and all North American sites, except one, showed decreasing trends in SO_4 for 1990-2008, with an average reduction of 56% and 37% respectively. The largest reductions took place before 2000.
- **NO_3:** North America had a more frequent and larger reduction (30%) in NO_3 than European stations (23%) for 1990-2008. In Europe, most of the reductions in NO_3 took place before 2000, while in North America the largest decline was observed after 2000. This was related to timing of implementation of emission reduction policy.
- **pH**: Trends in hydrogen ions (pH) were lower in Europe than in North America with reductions of 34% and 46%, respectively.

The trends in precipitation chemistry were consistent with reported trends in emissions of acidifying components in Europe and North America.

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3. Trends in surface water chemistry in Europe and North America 1990-2008

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3.1 Trend analysis of surface water chemistry

Data from ICP Waters are especially suitable for assessment of long-term trends in surface water chemistry. Previous trend analyses of ICP Waters data have provided important indications of the geographic extent of acidification and recovery of lakes and streams (Lükewille *et al.* 1997, Newell and Skjelkvåle 1997, Stoddard *et al.* 1999, Skjelkvåle *et al.* 1994, 2000, 2003, 2005, de Wit and Skjelkvåle 2007). Here, we report trends from 182 ICP Waters monitoring sites and 13 extra sites from Ontario for the period 1999-2008. We also compare these trends with trends for the period 1990-1999 to determine if the rate of recovery is changing. The latter interval was chosen to minimise the overlap between the time spans and meet the requirement of 10 years of data for calculation of time trends.

Previously, a significant decrease in surface water sulphate concentrations was reported for almost all regions covered by ICP Waters (de Wit and Skjelkvåle 2007). However, sulphate trends indicated slower rates of change in recent years. Widespread chemical recovery, i.e. increasing alkalinity, ANC and pH, was also observed, with European regions showing the largest changes. So-called confounding factors, including increased organic acidity, were found to delay recovery in some regions.

Our analysis of surface water response to changing deposition focuses on the variables that play major roles in acidification and recovery:

1) **SO_4^{2-}** and **NO_3^-**, the acid anions of acidic deposition. Trends in the concentrations of these anions reflect recent trends in deposition (especially SO_4^{2-}) and in ecosystem response to long-term deposition (e.g., NO_3^-).
2) **Base cations:** (Ca^{2+} + Mg^{2+}) are mobilised by weathering reactions and cation exchange that neutralise acids in watersheds. Base cations will respond indirectly to changes in SO_4^{2-} and NO_3^-.
3) **Acidity,** including **pH**, **measured (Gran) alkalinity** and **calculated ANC**, reflect the outcome of interactions between changing concentrations of acid anions and base cations.
4) Concentrations of **dissolved organic carbon (DOC)** or alternatively **total organic carbon (TOC)**. These are considered as surrogates for organic acids, mostly derived through degradation of natural organic matter in catchment soils.

Both SO_4^{2-} and base cation concentrations were sea-salt corrected (Skartveit *et al.* 1980) (denoted by an asterisk (SO_4^*, $(Ca+Mg)^*$), and pH was transformed to H^+ concentrations (assumed to be equal to activity) prior to statistical analysis.

Similar to the previous reports, we present the trends for each individual site, as well as aggregated trends 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 have 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.

3.2 ICP Waters sites chosen for trend analysis

Sites in the ICP Waters database exhibit a range of sampling frequencies, analytical programmes, and differences in lengths of record. In order to make a meaningful comparison of trends among these sites, it is necessary to impose a minimum set of requirements for inclusion of data. We chose to focus the current analysis on:

- sites where data were available for at least 7 out of 10 years (1990-1999 and/or 1999-2008).
- sites that are sensitive to acidification (ANC < 300 µeq/L and/or alkalinity < 300 µeq/L)
- sites with undisturbed catchments (i.e. no known point sources of pollution or agricultural influence)

An overview of mean chemical data for each site for the time period 2005-2008 is presented in Appendix B.

Sites that were excluded from analyses are summarised in Appendix C. Thirteen sites from Ontario, Canada, that are not routinely included in the ICP Waters database were added to this analysis to improve the data representativeness for this region. The number of sites that met the criteria was 189 and 195 for the time spans 1990-1999 and 1999-2008, respectively (***Table 4***). Sites that only met the criteria for one of the time spans were excluded from the regional analyses.

3.3 Quality assurance of data

Standardization of sample collection and analytical methodologies are addressed in the ICP Waters Programme Manual (ICP Waters Programme Centre 2011, http://www.icp-waters.no). Aspects of site selection, water chemistry/biological monitoring and data handling are also described in detail in the manual.

Three levels of quality control of water chemistry data are distinguished: in-laboratory controls in individual countries, between-laboratory controls and quality control of data reported to the National

Focal Points and to the Programme Centre at NIVA. The latter does not involve physical-chemical analysis of single parameters in the laboratory, but is a technical procedure including:

- looking for outliers
- evaluation of continuity in time series
- calculation of ionic balance

Table 4. *Number of sites in each country, and number of sites per country included in trend analysis.*

Europe				North America			
Country	no of sites in analysis		no of ICP Waters sites	Country	no of sites in analysis		no of ICPWaters sites
	90-99	99-08			90-99	99-08	
Belarus	0	0	1	USA	86	89	94
Czech Republic	8	8	8	Canada	17+13	17+13	18
Estonia	0	0	1				
Finland	8	8	8				
Germany	28	23	35				
Italy	6	6	6				
Latvia	0	0	8				
Norway	5	5	5				
Poland	2	2	4				
Sweden	10	9	10				
Switzerland	0	9	9				
United Kingdom	6	6	6				
	73	76	101		116	119	112

3.4 Statistical methods used for trend analysis

Only annual means were used in the statistical analyses. The pH was back calculated from arithmetic mean H^+. The frequency of observations per station varied from a single annual observation to weekly sampling, and the frequency of observations for some stations differed between years. For each site, a representative annual value was calculated for each variable by taking the arithmetic mean. Thus, seasonality in the data only influenced the value of the annual value and did not affect the power of the statistical tests.

Numerous statistical techniques are available to analyse trends in time series like those presented here. In previous ICP Waters reports on assessment of trends we have used the Mann Kendall test (MKT) (Hirsch and Slack 1984, Hirsch *et al.* 1982). This method is robust against outliers, missing data and does not require normal distribution of data. The method was used to determine monotonic trends based on the values of the test statistic (Z-score). Slopes were calculated using the Sen estimator (Sen 1968).

We compiled the results of the MKT for the individual sites and summed up the total number of "increasing", "decreasing" and "no trend" occurrences for each parameter. This procedure is not strictly valid because the risk of falsely rejecting null hypotheses increases when individual results are summed. However, if each test is regarded as a Bernoulli trial with a success probability of 0.05, the total number of false rejections can still be expected to be relatively low. We therefore believe that the compilation of MKT results gives a good overall picture of trends in the data.

While the significance of sums of individual MKT tests are questionable from a statistical viewpoint, the slopes calculated for multiple sites within a region represent a distribution of results, which can in turn be examined and analysed for patterns. The non-parametric Wilcoxon test was used to test for different distributions of slopes (calculated with the Sen slope estimator) between two time periods, i.e. 1990-1999 and 1999-2008. The sites were grouped in the regions used for the regional trend analysis (see below), and all sites that met the criteria (see above) for one or both criteria, were included in the analysis. The time intervals were chosen to minimise the overlap between the time spans.

Regional trends in the data were assessed using the Regional Kendall Test (Helsel and Frans 2006), which gives median slopes and a p value for the trend's significance. Only sites that met the criteria for both periods were included. The Regional Kendall Test has similar strengths as the MKT, and does not require normal distribution of the data.

3.5 Results of trend analysis 1990-1999 and 1999-2008

3.5.1 Trends at single sites

Patterns and qualitative results of the trend analyses are summarised in ***Table 5*** and described below. Results of trend analysis for each individual site are given in Appendix D and summarised in ***Figure 4***. Magnitude of slopes for sites and regions, and further statistical analyses are presented in the next section.

Table 5. *Result of trend analyses for ICP Waters sites (and 13 extra from Ontario) for the periods 1990-1999 and 1999-2008, for* SO_4^*, NO_3, *sum of base cations* (Ca^*+Mg^*), *alkalinity, Acid Neutralizing Capacity (ANC),* H^+, *and organic carbon (OC). Number of sites with significantly increasing or decreasing trends for given variables (note that data for all parameters are not available from all sites). No trend at significance level* $p<0.05$.

	SO_4*		NO_3		Ca*+Mg*		Alkalinity		ANC		H^+		OC	
	90-99	99-08	90-99	99-08	90-99	99-08	90-99	99-08	90-99	99-08	90-99	99-08	90-99	99-08
Europe														
Increasing	0	0	8	7	2	6	12	13	23	23	1	0	15	8
Insignificant	23	35	54	42	43	52	50	42	38	48	56	58	30	48
Decreasing	50	40	12	20	28	18	1	3	2	0	17	18	0	0
North America														
Increasing	1	0	8	7	1	3	22	14	16	18	1	6	8	16
Insignificant	33	25	94	101	45	52	89	91	80	82	99	104	92	89
Decreasing	82	94	14	9	59	49	3	4	9	4	15	8	1	8
Total no of sites	189	194	190	186	178	180	177	167	168	175	189	194	146	169
Total increasing	1	0	16	14	3	9	34	27	39	41	2	6	23	24
Total insignificant	56	60	148	143	88	104	139	133	118	130	155	162	122	137
Total decreasing	132	134	26	29	87	67	4	7	11	4	32	26	1	8
% increasing	0.5	0.0	8	8	2	5	19	16	23	23	1.1	3	16	14
% insignificant	30	31	78	77	49	58	79	80	70	74	82	84	84	81
% decreasing	70	69	14	16	49	37	2	4	7	2	17	13	0.7	5

Most sites show a significant decrease in non-marine sulphate. For nitrate, there are more sites that show significant decrease than increase, but a large majority (>75 %) show no significant trend. A more detailed description of the mixed tendencies in nitrate trends is given in the last ICP Waters trend assessment report (de Wit and Skjelkvåle, 2007). The lack of a uniform trend in nitrate concentrations indicates that nitrate leakage from catchment is affected by a variety of processes, in contrast to sulphate leakage from catchment which is largely controlled by sulphate deposition (past and present), sulphate adsorption and desorption in soils, and modifications sometimes imposed by climatic events such as drought (Dillon et al 2003). The two time spans are very similar with respect to the fraction of sites showing significant positive or negative trends for sulphate and nitrate.

About half of the sites show a significant decrease in $(Ca+Mg)^*$ (non-marine base cations) between 1990 and 1999, while the fraction is somewhat lower for the time span 1999-2008. Few sites show significant increase.

About 20 % of sites display significantly increasing trends in alkalinity and ANC, while the majority (70-80 %) show no significant trend. The fractions are similar for both time spans. Less than 5% of all sites appear to be undergoing further acidification between 1999 and 2008.

Only 13 % of the sites show significantly decreasing trends in H^+ activity (increase in pH) between 1999 and 2008. This is somewhat lower than the corresponding fraction for the time span 1990-1999. The fraction of sites showing no significant trend is higher than 80 % in both periods.

The concentration of dissolved organic carbon (DOC) is not measured at all sites. For such sites, we have used total organic carbon (TOC) if data were available. A majority of sites (> 80 %) shows no trend in DOC, while 14-16 % displays a significant increase.

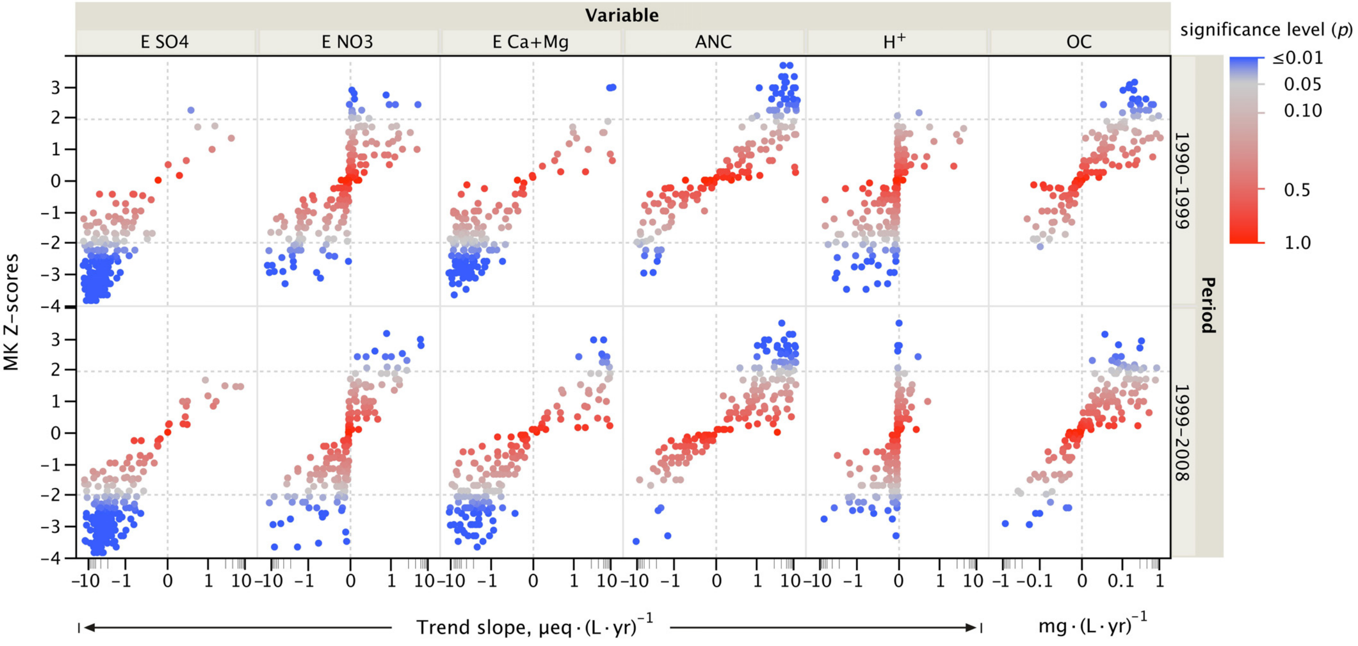


***Figure 4**. Mann Kendall (MK) Z-scores and trend slope for SO_4^*, NO_3, sum of base cations $(Ca+Mg)^*$, Acid Neutralizing Capacity (ANC), H^+, and organic carbon (OC) for all the analysed sites in the time spans 1990-1999 (upper row of plots), and 1999-2008 (lower row of plots). The letter E before the parameter name denotes that the concentrations are in equivalents. A MK Z-score > 1.96 or < -1.96 implies that the trend is significantly different from 0 at the 0.05 confidence level (blue colour). Note that the x-axes are arctan transformed.*

3.5.2 Regional trends

The strongest evidence that emissions control programs work is a consistent pattern of recovery (decreasing SO_4^{2-} and increasing pH, ANC and alkalinity) across a large number of sites. For this reason, we are again reporting trends for clusters of ICP sites (***Table 6***, ***Figure 5***). The sites are grouped into geographic regions based on similar acid-sensitivity (e.g., similar geology, soil characteristics) and rates of deposition. The regions, and sites they include, are almost identical to those used in the last trend assessment (de Wit and Skjelkvåle 2007). The main difference is that 13 lakes from south-central Ontario have been included in the "Ontario" region. The list of regions on which we report is based on scientific and pragmatic decisions resulting from availability of data.

Results of the trend analyses for different regions and time spans are shown in Figures 2-8. Results are also tabulated in Appendix E.

Table 6. *Regions in Europe and North America and number of sites for which trend analysis was undertaken in each region.*

Regions in Europe	Abbreviation	n	Regions in North America	Abbreviation	n
North Nordic	NoN	7	Maine and Atlantic Canada	Atl	20
South Nordic	SoN	15	Vermont and Quebec	Vt/Que	17
U.K.	UK	6	Adirondacks	Ads	50
West Central Europe	WCE	8	Appalachian Plateau	App	9
East Central Europe	ECE	25	Virginia Blue Ridge	Bri	3
Alps	Alps	15	Ontario	Ont	20
	Sum	76		Sum	119

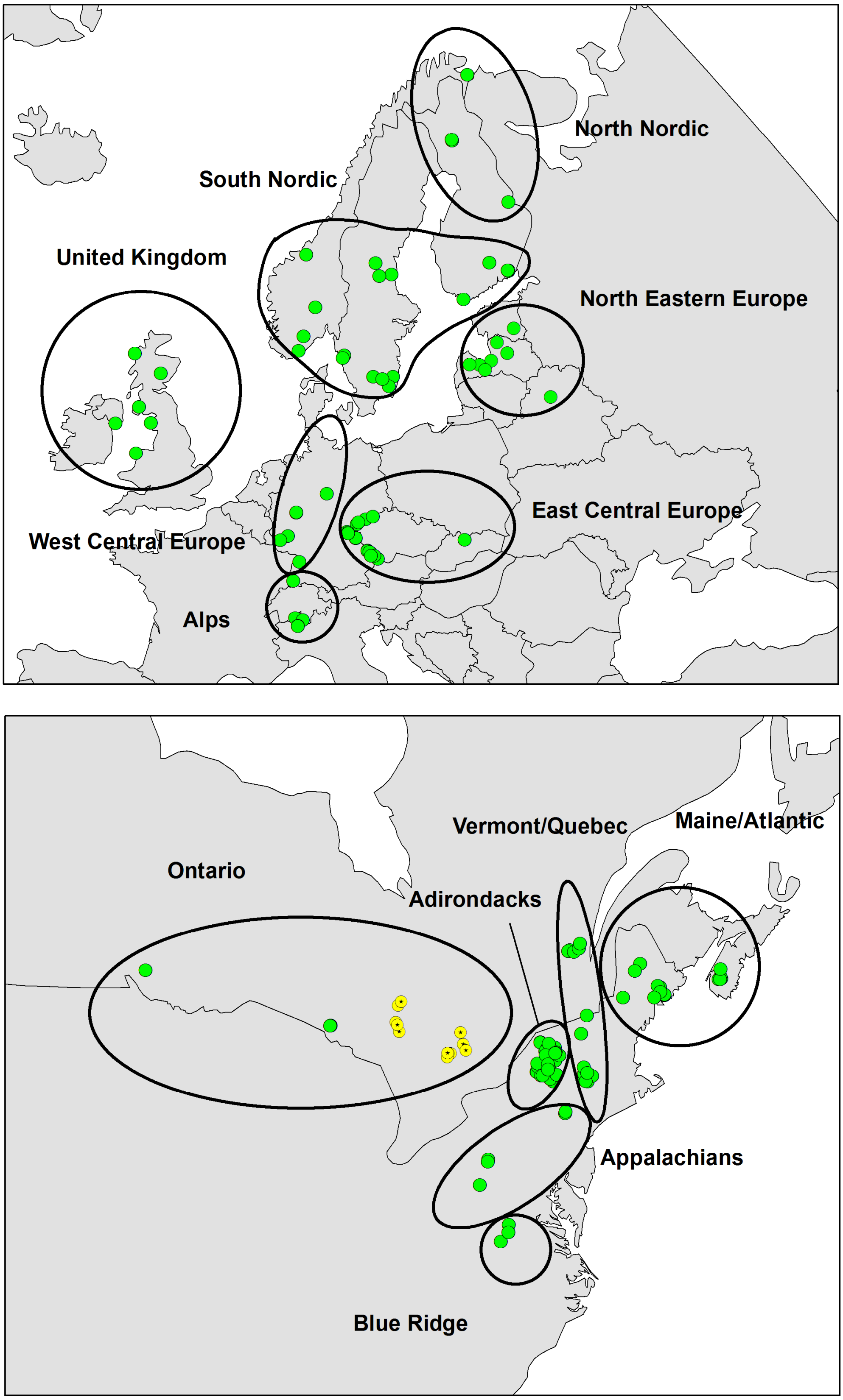


Figure 5. *Map showing outline of geographical regions and location of ICP Waters sites (green) and additional sites (yellow).*

Trends in sulphate by region

Decreasing concentrations of non-marine sulphate were found in all regions included in this analysis except Virginia Blue Ridge in North America (***Figure 6***). In this region, soil characteristics make a sulphate decrease unlikely (Church *et al.* 1990), and the three sites follow the pattern revealed by a more comprehensive assessment of streams in the region (Stoddard *et al.* 2003). Sulphur-adsorbing soils typical of the Southern Blue Ridge exert a strong control on atmospherically-deposited sulphate, and have driven small but significant increases in surface water SO_4^{2-}, even during a time of decreasing rates of acidic deposition. Relatively high concentrations of sulphate at some Alpine stations in recent years are possibly an effect of low precipitation in the catchments.

Other regions show median regional rates of SO_4 decline ranging from -1.1 µeq/L/yr (UK, Maine and Atlantic Canada) to -5.3 µeq/L/yr (Southern Nordic) between 1990 and 1999, and -0.8 µeq/L/yr (Alps) to -2.6 µeq/L/yr (Southern Nordic) between 1999 and 2008. Trends in most regions tend to have smaller slopes in the latter time span, but the difference is statistically significant only for Vermont/Quebec, and Ontario. However, if results are grouped into larger regions, the temporal difference in the distribution of slopes is significant ($p<0.01$) both for North America and Central Europe (WCE, ECE and Alps) (results not shown).

Mitchell *et al.* (2011) concluded that the non-marine S export relative to S input observed at many eastern North American watersheds indicates that previously deposited S stored in wetland and/or forest soils is now being released. This release affects the observed surface water SO_4 trends and acts to delay the pH or ANC recovery expected from reduced S deposition.

The largest rates of decline between 1990 and 1999 were observed in Europe. The regional differences are smaller for the time span 1999-2008, reflecting a substantial decrease of sulphur deposition especially in the areas that previously received the highest loads.

The trend analysis of sulphate in precipitation in Chapter 2 showed that concentrations of sulphate in precipitation declined with about 60 and 40% in the period 1990 to 2008 in Europe and North America, respectively, with the largest declines occurring in Europe between 1990 and 1999. According to an EMEP model (http://webdab.emep.int/Unified_Model_Results/AN/), deposition of sulphur in the European countries included in the present analysis declined on average with 60 % between 1990 and 1999, and about 20 % between 1999 and 2008 (relative to 1990 level, i.e. 80 % reduction in total). Corresponding relative changes in total deposition of sulphur in the eastern US are about 30 and 20 %, respectively (CASTNET, 2008), amounting to a total reduction of 50%. Thus, the modelled reductions of sulphate deposition for Europe are larger than those observed whereas the modelled estimates for in the eastern US are similar to what is measured.

The largest declines in sulphur deposition occurred during the 1990s. The same is found for sulphate in surface waters in 5 of 6 regions in Europe and in 3 of 6 regions in North America (***Figure 6*** and ***Table 7***). The relative changes of sulphate in in suface water are similar to those observed in precipitation (Chapter 2).

Table 7. *Percent change in concentrations of non-marine sulphate divided by regions, for the periods 1990-2008, 1990-1999 and 1999-2008. Estimates are based on averages of annual mean values for 3-year periods (1990-1992, 1998-2000 and 2006-2008. Changes are related to mean values for 1990-1992.*

Region		Number of sites	1990-2008	1990-1999	1999-2008
Europe	NoNordic	7	-39	-21	-18
	SoNordic	15	-58	-37	-21
	UK	6	-48	-33	-15
	WCEurope	7	-30	-18	-11
	ECEurope	23	-34	-35	1
	Alps	6	-25	-18	-7
North America	Maine_Atlantic	10	-38	-16	-22
	Vermont_Quebec	17	-39	-25	-14
	Adirondacks	50	-33	-16	-17
	Appalachians	9	-20	-13	-7
	Blue Ridge Mountains	3	-1	0	-1
	Ontario	20	-33	-27	-6

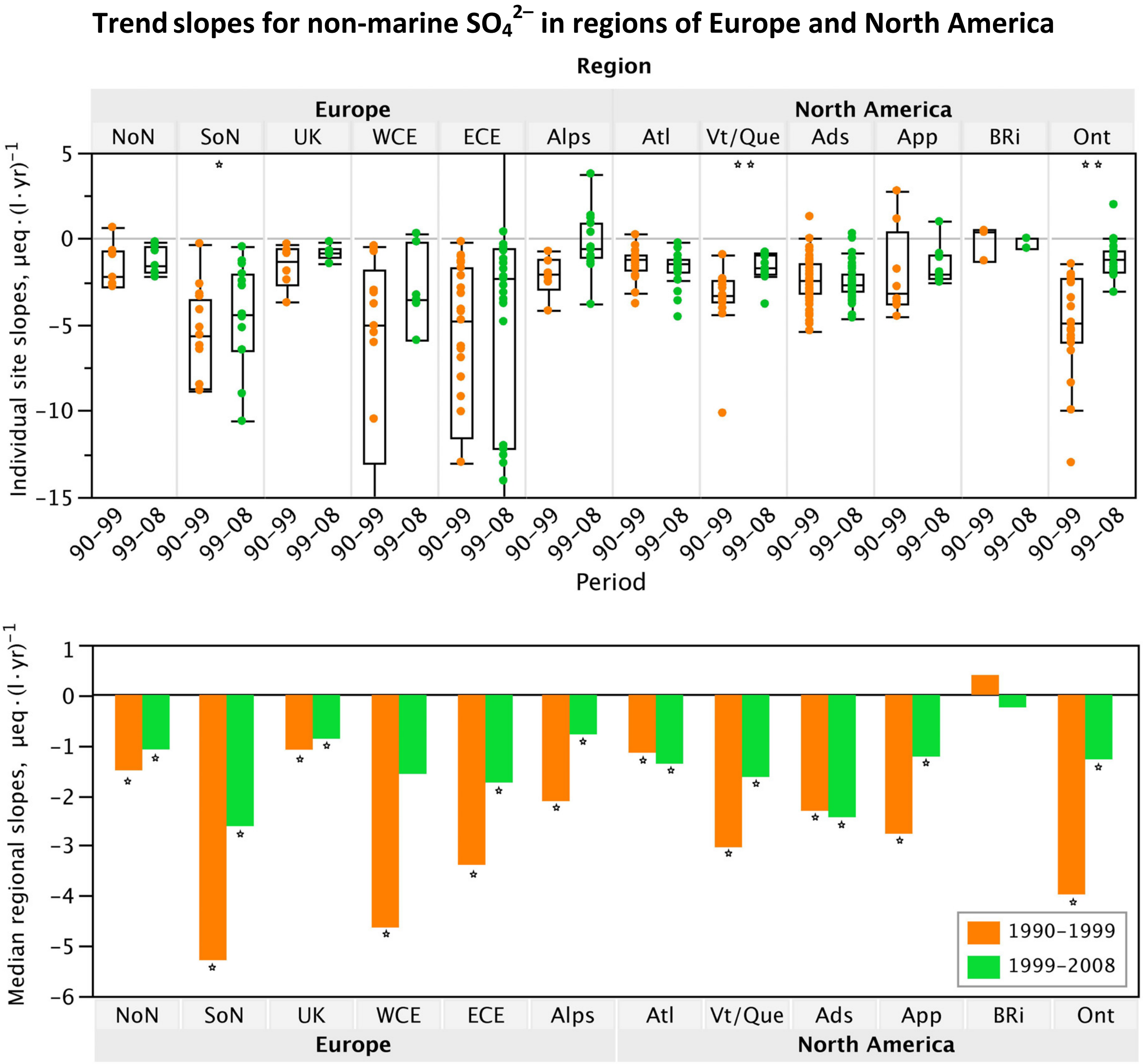


Figure 6. *Upper panel: Distribution of individual non-marine SO$_4^{2-}$ trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median is indicated with a line. Asterisks indicate the significance level (** $p<0.05$, * $p<0.10$) of a non-parametric test for difference between the distributions of slopes in the two time spans. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). An asterisk indicates that the slope is significantly different from zero ($p<0.05$), i.e. that there is a trend. Abbreviations are defined in Table 3. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

Trends in sulphate in sites not sensitive to acidification

Some of the ICP Waters sites are regarded as insensitive to acidification (ANC > 300 µeq/L). However, we wanted to see if the general decrease in sulphate that is observed in acid-sensitive sites is also found in the non-sensitive sites (***Table 8***). Only one of the seven sites shows a significantly decreasing trend, and the slope is large compared to most of the acid-sensitive sites. The sites might be influenced by local sources of sulphate in the catchment, either from natural conditions (geology) or anthropogenic input (agriculture).

Table 8. *Trend analysis for the time spans 1990-1999 and 1999-2008 of non-marine SO_4 in sites not sensitive to acidification. Values are median concentration (µEq/L), slope (µEq/L/yr) and significance level (Mann-Kendall test). n = nr of years. Significant slopes are indicated in bold.*

		1990-1999				1999-2008			
		Median concentration SO4 µEq/L	P	Slope	n	Median concentration SO4 µEq/L	P	Slope	n
Belarus	BY01					516	0.18	-8	10
Estonia	EE01	274	**0.01**	-11.0	8	273	0.93	-0.4	10
Latvia	LV01	571	0.80	-0.38	8				
Latvia	LV02	634	0.14	-9.7	8				
Latvia	LV03	1025	0.46	-16.2	8	878	0.21	55	9
Latvia	LV04	569	0.08	-9.9	8	460	0.14	-22	9
Latvia	LV05	153	0.62	0.89	8				

Trends in nitrate by region

The regional pattern of temporal trends for nitrate is more complex than for sulphate, and all regions except Virginia Blue Ridge include individual sites with increasing or decreasing trends (***Figure** 7*). Even so, the median trend of several regions is significant as determined by the Regional Kendall test. Between 1990 and 1999, nitrate concentrations decreased in West Central Europe, Vermont/Quebec, Adirondacks, and Virginia Blue Ridge but increased in Maine/Atlantic Canada and the Alps. The increase that we present for the latter region in this period, is entirely based on observations from Italian stations (***Appendix D***) and may not be representative for the whole alpine region. The pattern is very different between 1999 and 2008 with the Northern and Southern Nordic regions, Alps, and Ontario showing negative trends, while Vermont/Quebec and West Central Europe show a positive trend. However, a significant difference in the distribution of slopes between the two time spans was only found for the Northern Nordic region, the Alps, Maine/Atlantic Canada and Vermont/Quebec.

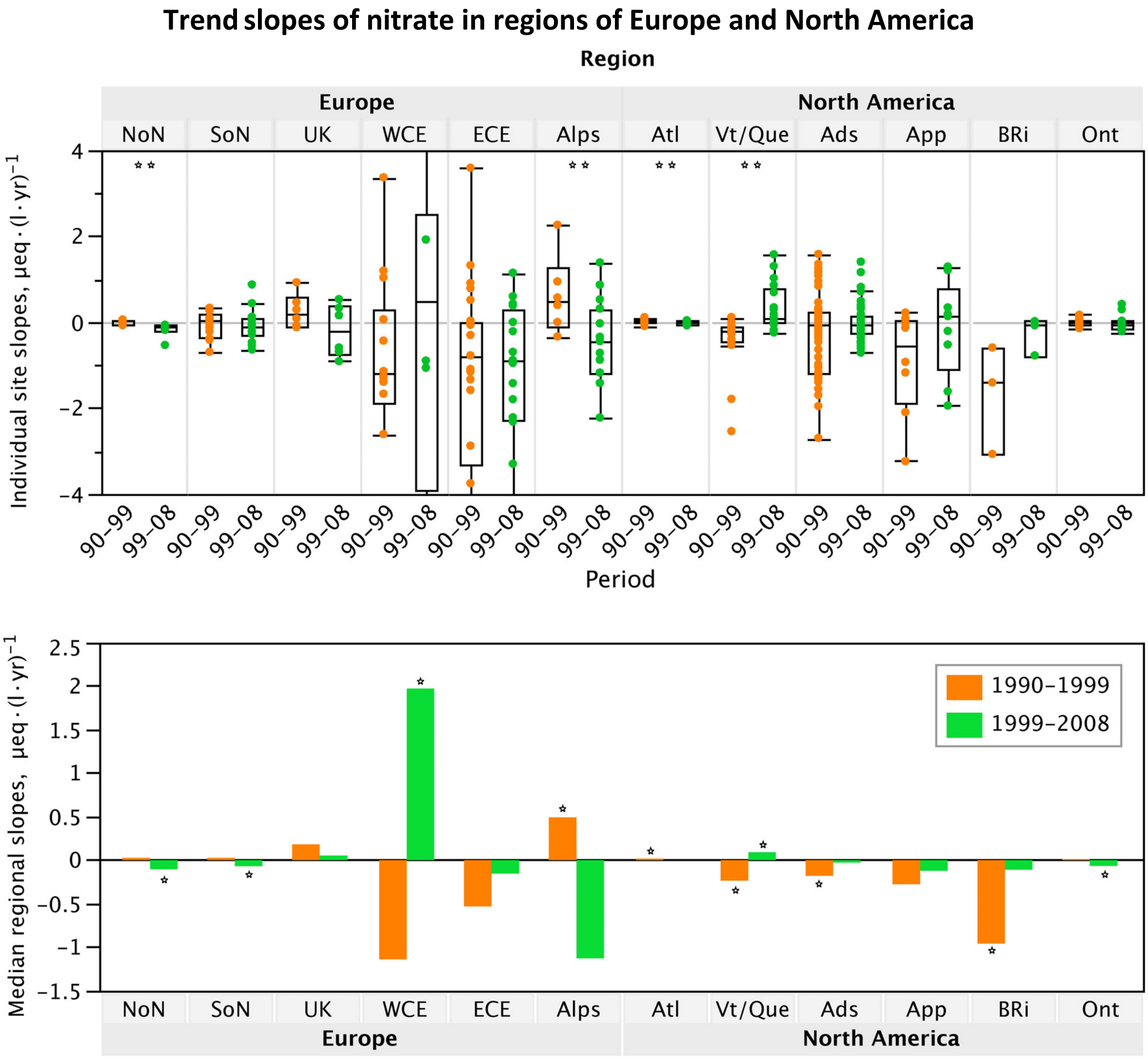

Trend slopes of nitrate in regions of Europe and North America

***Figure** 7. Upper panel: Distribution of individual nitrate trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in **Figure 6**. Note that some sites were* *excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

Regional decreases in SO_4 concentrations were much larger than the changes in NO_3 concentrations except in the regions Virginia Blue Ridge, the Alps and West Central Europe. This implies that the relative importance of NO_3 for acidification of surface waters has increased in most regions. The importance of NO_3^- in acidification relative to SO_4^{2-} in surface waters can be estimated by the concentration of NO_3^- divided by the sum of non-marine SO_4^{2-} and NO_3^-.

$$\text{N acidification ratio (NAR)} = \frac{NO_3^-}{\left(SO_4 * + NO_3^-\right)} \quad \text{(all concentrations in } \mu\text{eq L}^{-1}\text{)}$$

The ratio between the equivalent concentrations of nitrate and the sum of nitrate and non-marine sulphate (NAR-ratio) was less than 0.1 for 48 percent of sites and less than 0.5 for 95 percent of sites (calculated from data in ***Appendix B***), which means that sulphate is still the most important contributor to acidification. The highest NAR-ratios were in Europe (***Figure 8***), especially in central Europe and the Alps. Increasing NAR-ratios were observed in all regions except the Northern Nordic, Appalachians and the Blue Ridge Mountains (***Figure 9***).

While reduced deposition of sulphur is the main driver behind the almost universal decline of freshwater sulphate, the controls of nitrate concentration are complex. Factors that have been invoked to explain trends include changes in deposition of nitrogen and sulphur (e.g. Wright *et al.* 2001, Oulehle *et al.* 2008) as well as changes in the catchment (e.g. Burns *et al.* 2006, de Wit *et al.* 2007, Rogora 2007, Rogora *et al.* 2008, Baron *et al.* 2009, Brookshire *et al.* 2011). Furthermore, these processes are affected by climate and seasonal changes that increase the variability in nitrate concentrations. A more comprehensive discussion of nitrogen trends, relative importance of sulphate and nitrate for acidification of surface waters, and further references are given in the last trend assessment report (de Wit and Skjelkvåle 2007).

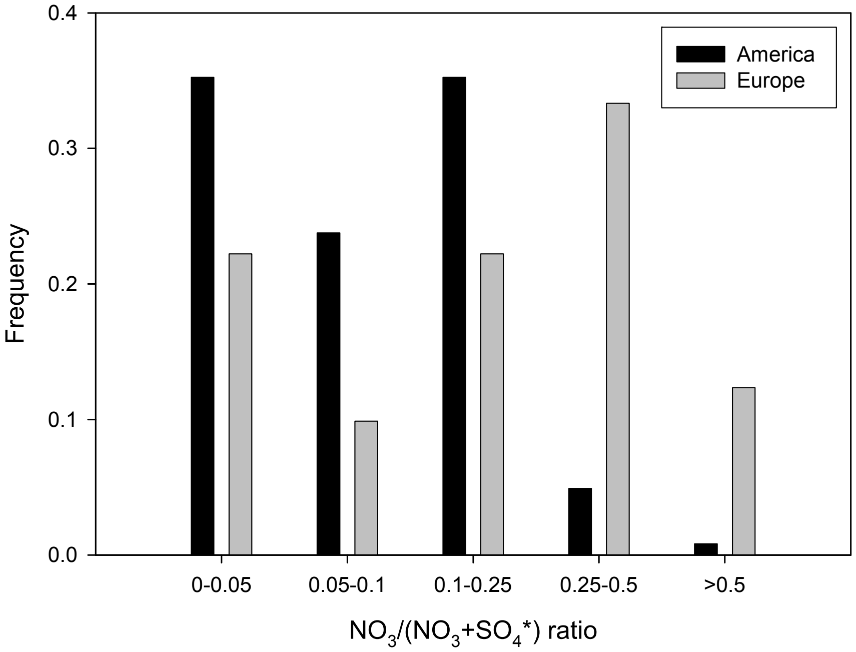


Figure 8. *Relative importance of nitrate in acidification at 122 American and 81 European sites. The ratios are calculated from mean equivalent concentrations at each site for the time span 2005-2008.*

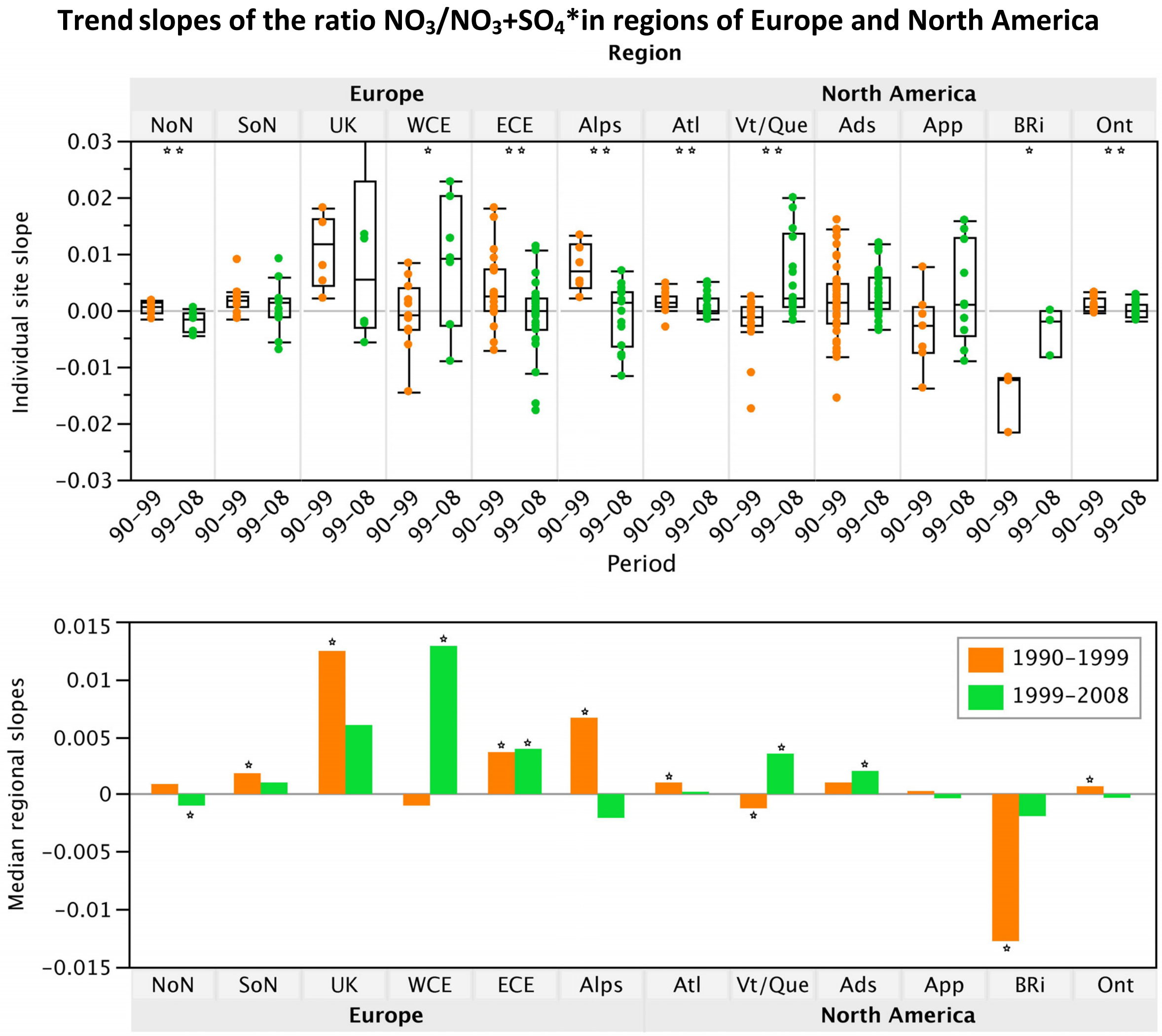


Figure 9. *Upper panel: Distribution of individual trend slopes of the ratio NO_3/NO_3+SO_4* in regions of Europe and North America for the time spans 1990-1999 and 1999-2008.Increasing trend means that nitrate is increasingly important for acidification of surface waters in the ICP Waters sites, while decreasing trends means that sulphate is increasingly important. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in* ***Error! Reference source not found.****. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

Trends in base cations by region

One of the expected responses of catchments to falling non-marine SO_4^{2-} inputs is the reduced leaching of non-marine base cations (Galloway *et al.* 1983). Lower concentrations of anions necessitate lower concentrations of one or more cations. In this report, we use the sum of the concentrations of non-marine calcium and magnesium $(Ca+Mg)^*$ as a surrogate for total base cation concentrations, because these cations are quantitatively most important at the majority of acid-sensitive monitoring sites.

All regions where data are available except West Central Europe and the Alps, showed decreasing concentrations of non-marine base cations in one or both periods (**Error! Reference source not found.**). However, the rate of decline appears to be decreasing, and many regions showed significantly smaller slopes in the latter time span. The Alps is the only region that shows increasing concentrations (between 1999 and 2008) of base cations. This is probably a consequence of the increasing SO_4^{2-} concentrations (***Figure 6***). Possible explanations for the observed recent increase in base cations in the Alps include hydrological effects (see section on sulphate), increased weathering rates caused by climate change (Rogora *et al.* 2003), and contribution from Saharan dust deposition episodes (Rogora *et al.* 2004)

For most regions the median decrease in base cations was lower than the equivalent decline in SO_4, but the difference appears to be larger in the European regions. If the decrease in SO_4 was entirely balanced by a decrease in base cations, no improvement of water chemistry (increase in pH, alkalinity and ANC) would be expected.

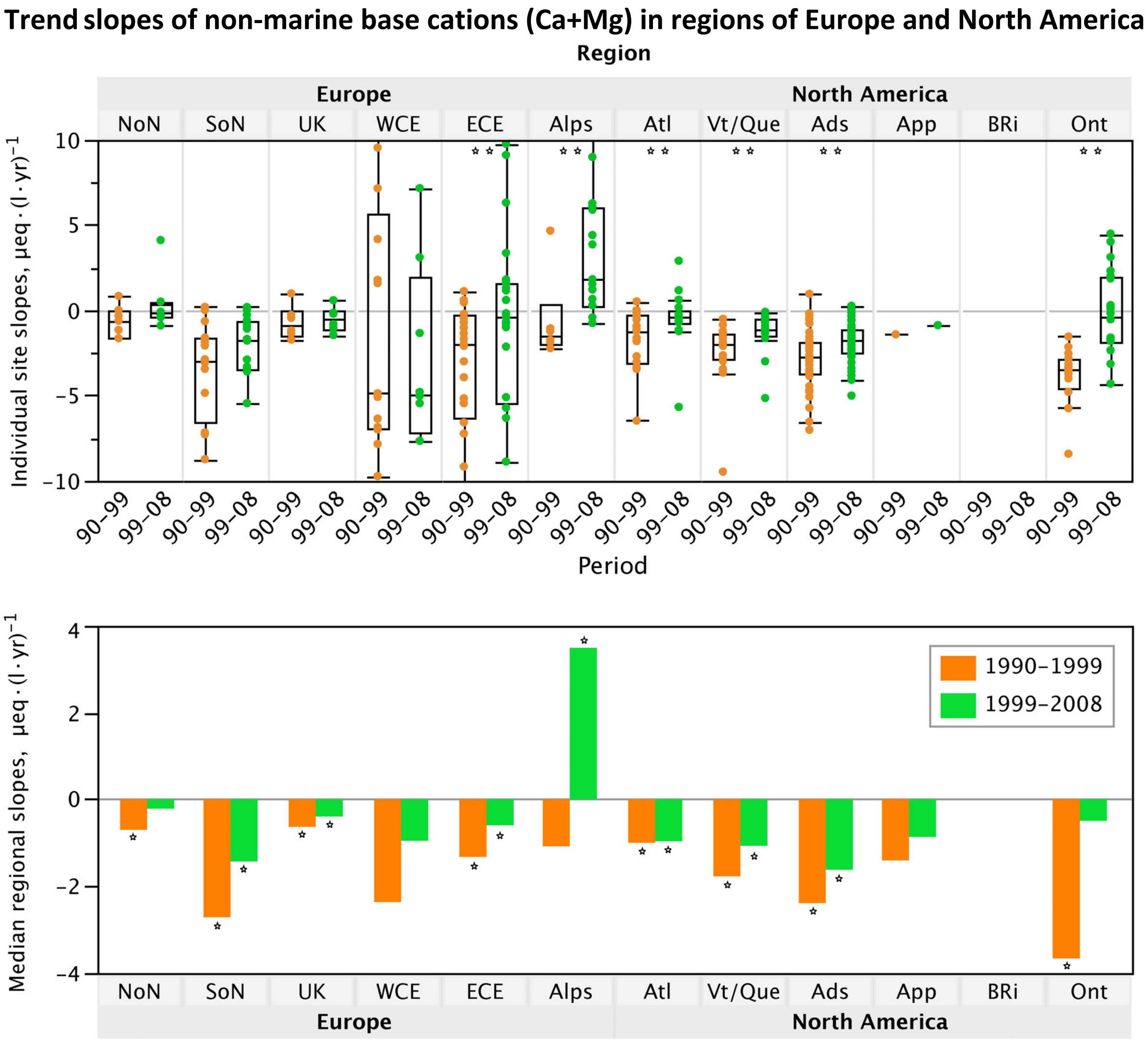


__Figure 10.__ Upper panel: Distribution of individual trend slopes for non-marine base cations (Ca+Mg) in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in __Error! Reference source not found.__. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.

Trends in alkalinity and ANC by region

Alkalinity is measured by titration and indicates the capacity of the water to buffer acidic inputs. Acid neutralising capacity (ANC), calculated from sum of base cations (Ca^{2+}+Mg^{2+}+Na^{+}+K^{+})minus the sum of acid anions (SO_4^{2-}+Cl^{-}+NO_3^{-}), is a surrogate for alkalinity. However, ANC can overestimate buffer capacity in waters rich in DOC where the concentration of strong organic acid anions is high compared to inorganic acid anions.

The steeper negative regional trends for SO_4 compared to those for base cations, combined with the regional signal of either no change or decreasing NO_3, imply increasing ANC and suggest that alkalinity is increasing. Indeed, all regions except UK, Maine/Atlantic Canada and Virginia Blue Ridge show positive regional trends in alkalinity and/or ANC in one or both periods (**Error! Reference source not found.** ***Figure 11*** and ***Figure 12***), indicating chemical recovery. The only region displaying a small but significant negative trend is ANC in Vermont/Quebec between 1999 and 2008.

Between 1990 and 1999, the largest regional improvements occurred in West Central Europe and the Appalachians, respectively, but progress later appears to have stagnated in these regions. The high rate of increase in ANC in the Southern Nordic region has also slowed down between 1999 and 2008. The largest improvements in alkalinity and ANC between 1999 and 2008 were observed in the Alps, consistent with the increase in base cations. In Ontario, it is the decrease in sulphate and nitrate that principally drives the increase in ANC between 1999 and 2008.

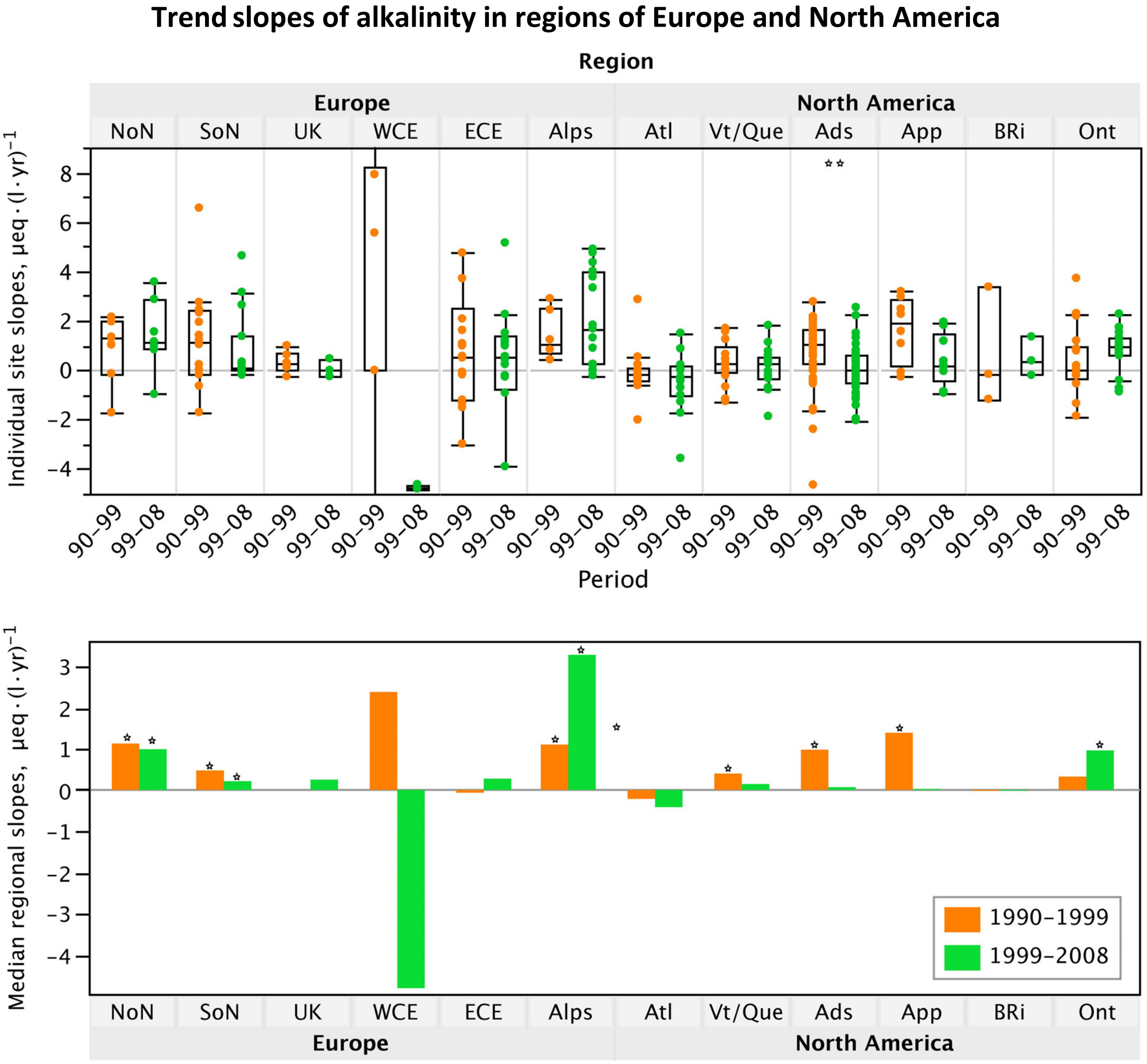


Figure 11. *Upper panel: Distribution of individual alkalinity trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in* ***Figure 6****. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

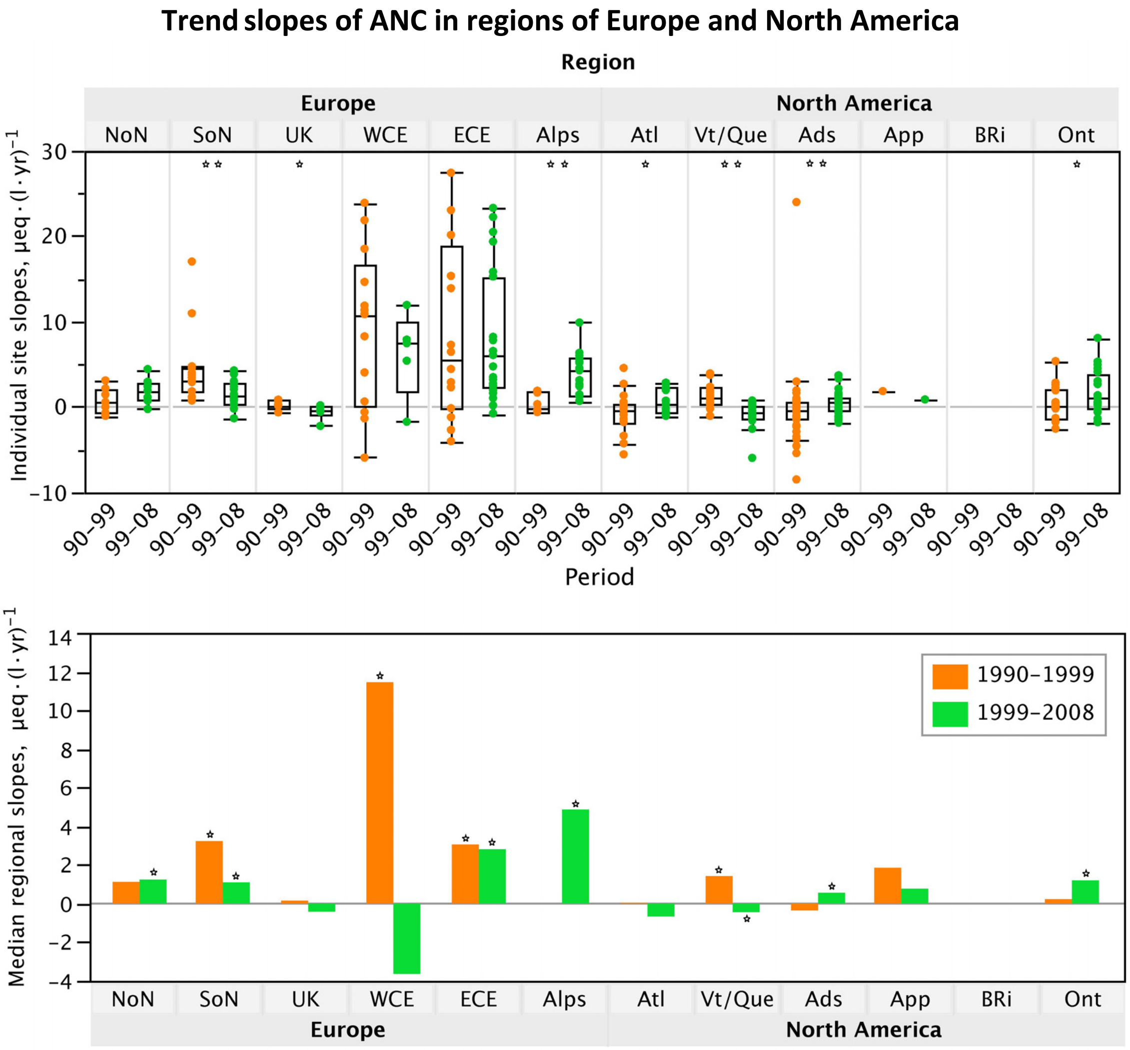

Trend slopes of ANC in regions of Europe and North America

__Figure 12.__ Upper panel: Distribution of individual ANC trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in__Error! Reference source not found.__. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.

Trends in DOC by region

Dissolved organic carbon (DOC) is a key component of aquatic chemistry, e.g. as an indicator of natural organic acidity (Hruška *et al.* 2003). In this respect, it has received considerable attention in recent years (see e.g. Erlandsson *et al.* 2011) because of rising levels in many regions. Possible drivers for rising DOC include factors related to climate change, but decreasing sulphur deposition appears to be an important determinant in the regions considered in this report (Monteith *et al.* 2007, de Wit and Skjelkvåle 2007). However, climatic factors are becoming more influential determinants of increasing DOC observed in some of the eastern Canadian sites (Clair *et al.* 2011, Couture *et al.* 2011). All regions (where DOC data are available) except West Central Europe and the Appalachians show increasing DOC concentrations in one or both periods with median rates varying between 0.01 and 0.23 mg/L/y (***Figure 13***). None of the regions show a significant difference in the distribution of slopes between the two periods, i.e. there are few indications that DOC concentrations are levelling off (there is a nearly significant difference in Vermont/Quebec). If the increase mainly has been driven by reduced acidification, a stabilisation of DOC levels is expected to occur in the near future, because further decrease in sulphate concentrations is expected to be low.

Increasing levels of DOC retard increases in pH and alkalinity (Driscoll *et al.* 2003, Evans *et al.* 2008, Erlandsson *et al.* 2010).

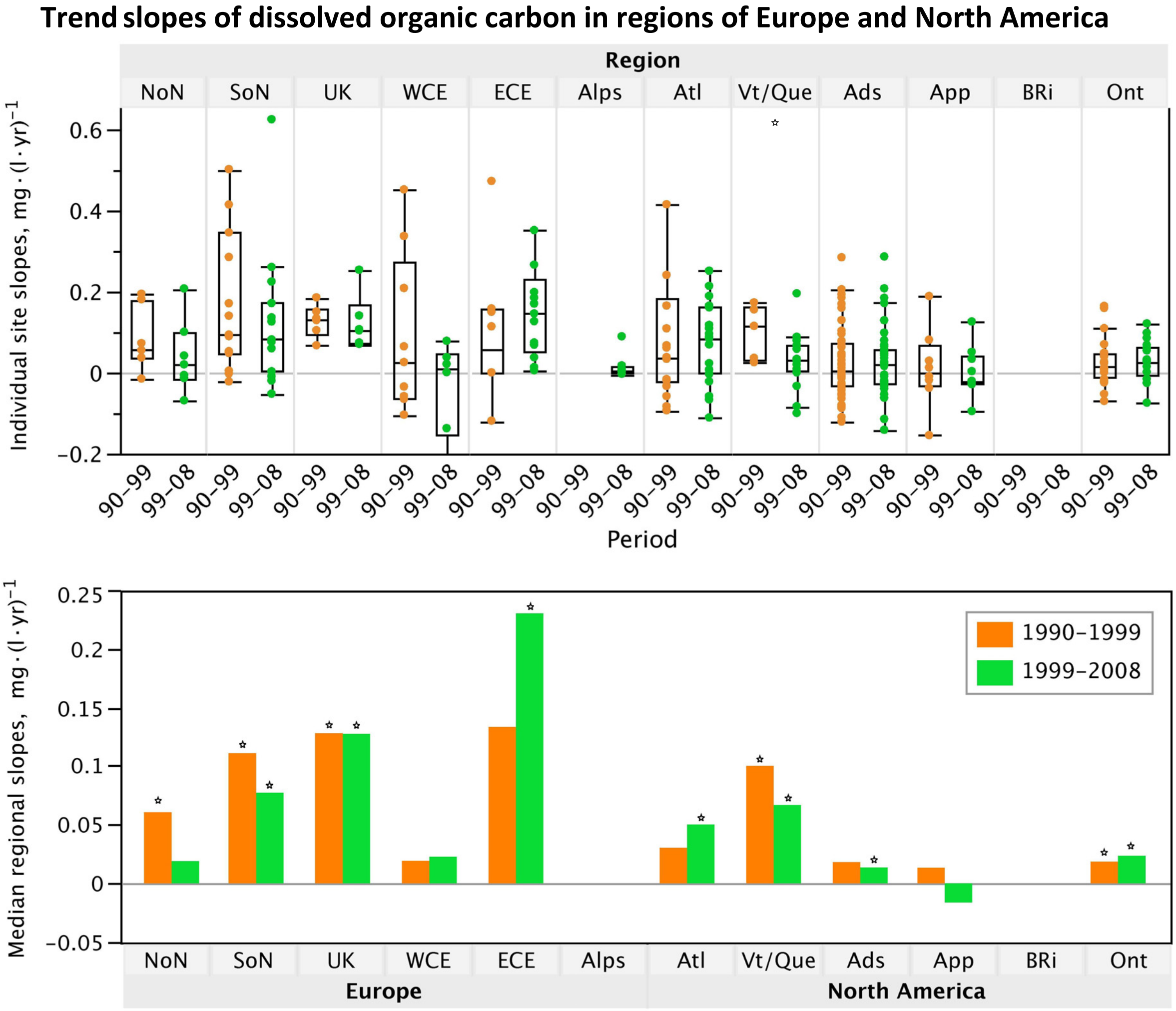


Figure 13. *Upper panel: Distribution of individual DOC trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in* ***Figure 6****. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

Trends in pH by region

Most processes in natural aqueous systems are pH dependent. Lake or stream pH can be used to assess the integrated effect on the rather delicate proton balance of all the reactions that acid deposition initiates. pH is also a very important parameter for understanding biological consequences of acid deposition. A decreasing pH is likely to give damage to the aquatic biota, while an increasing pH can indicate the possibility of a biological recovery. However, trends can be difficult to detect because of high measurement uncertainty in the low ionic strength water typical for most of the ICP Waters sites (Hovind 2010). Here we analyse trends in proton activity, calculated from pH measurements. An increase in pH is a decrease in proton activity.

All regions except the Appalachians and Virginia Blue Ridge show decreasing proton activity in one or both periods (***Figure 14***). None of the European regions show a significant difference in the distribution of slopes between the two periods, i.e. there is no evidence that trends in proton activities are levelling off. In the North American regions, the absolute values of median trend slopes are smaller than in most European regions, and tendencies are mixed: In the Adirondacks and Ontario regions, proton activities decreased between 1990 and 1999 and increased between 1999 and 2008, while the opposite may have happened in Maine/Atlantic Canada and Vermont/Quebec.

The trends in pH are consistent with those observed for other parameters on a regional scale. In the European regions, the median decreases in base cations have been appreciably smaller than the equivalent declines in acid anions in both periods. Consequently, ANC and alkalinity have increased and proton activity has decreased. Exceptions are West Central Europe where the variation in slopes for individual sites is large and the number of stations with adequate time series is small, and UK where no significant increase in ANC or alkalinity was observed in either of the periods. Possible explanations for the latter include long-term variability in sea-salt deposition (Evans *et al.* 2001a), relatively large increases in DOC, and compound analytical errors (Davies *et al.* 2005, Evans *et al.* 2001b,c). However, proton activity has decreased significantly.

In the North American regions, the rates of decrease of base cations are closer to those observed for acid anions (see also e.g. Stoddard *et al.* 2003, Driscoll *et al.* 2007). Changes in ANC/alkalinity are therefore relatively small. Moreover, increasing concentrations of DOC are found in most North American regions. North American pH trends are therefore more subtle than in Europe. The clearest examples are Adirondacks, Ontario and Vermont/Quebec between 1999 and 2008 where the respective trends for ANC and proton activity have the same sign (i.e., pointing in opposite directions as indicators of recovery).

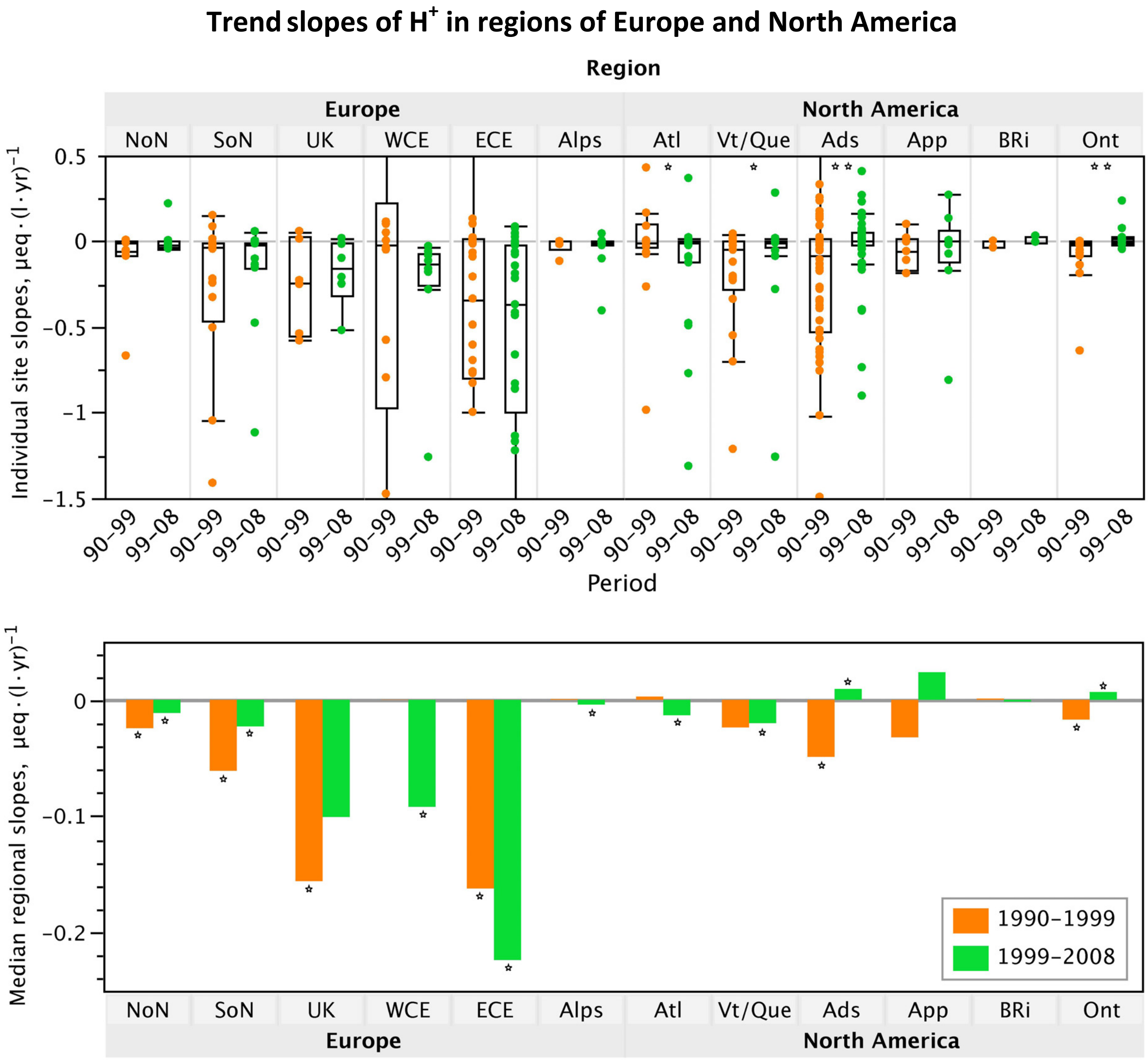
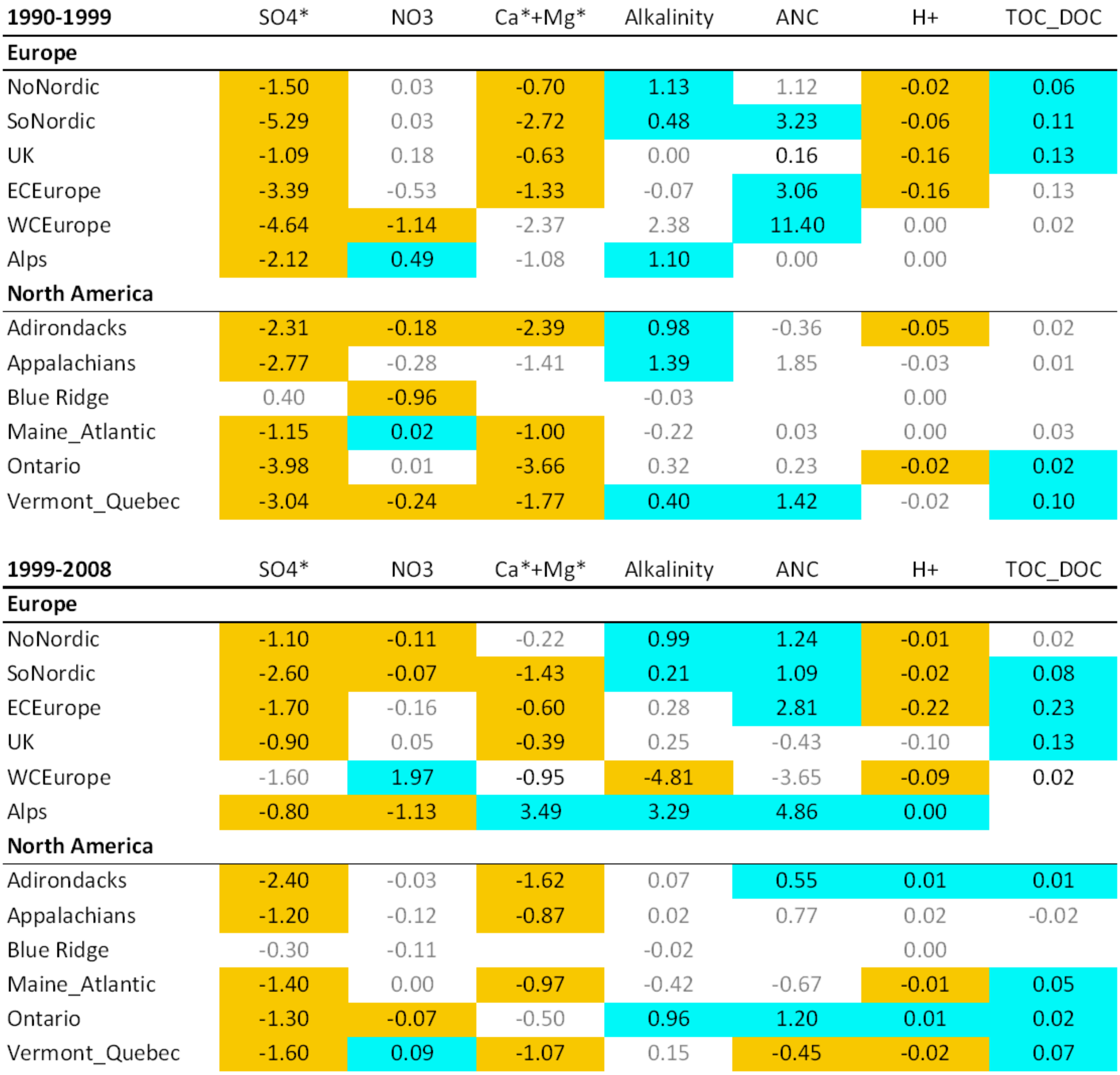


Figure 14. *Upper panel: Distribution of individual H$^+$ trend slopes in regions of Europe and North America for the time spans 1990-1999 and 1999-2008. Lower panel: Slopes of median regional trends assessed with the Regional Kendall test for the time span 1990-1999 (red) and 1999-2008 (green). Boxes, whiskers, asterisks and region names are defined as in* ***Figure 6****. Note that some sites were excluded in the Regional Kendall analysis, i.e. for some regions (WCE, ECE, Alps) the datasets presented in the upper and lower panels are not identical.*

3.6 Conclusions

In the trend analysis of nearly 200 acid-sensitive ICP Waters monitoring sites in Europe and North America distributed over 12 regions, we found a consistent pattern of chemical recovery in the time spans 1990 to 1999 and 1999 to 2008 as summarized in***Table 9***. The improved chemical status of acidified surface waters is related to the reduction of acid deposition. Because acid deposition is the consequence of emissions of acidifying compounds to the atmosphere (See Chapter 2), our results show that emission control programmes have their intended effect.

Table 9. *Summary of trend analysis. Median Mann Kendall trend slope for* SO_4^**,* NO_3*, sum of base cations* $(Ca+Mg)^*$*, Acid Neutralizing Capacity (ANC),* H^+*, and organic carbon (TOC_DOC) by region in the time spans 1990-1999 and 1999-2008. Significant upward trends in blue, significant downward trends in yellow, non-sigificant in grey. Units: µeq/L/yr for all except OC: mg C/L/yr*



1990-1999	SO4*	NO3	Ca*+Mg*	Alkalinity	ANC	H+	TOC_DOC
Europe							
NoNordic	-1.50	0.03	-0.70	1.13	1.12	-0.02	0.06
SoNordic	-5.29	0.03	-2.72	0.48	3.23	-0.06	0.11
UK	-1.09	0.18	-0.63	0.00	0.16	-0.16	0.13
ECEurope	-3.39	-0.53	-1.33	-0.07	3.06	-0.16	0.13
WCEurope	-4.64	-1.14	-2.37	2.38	11.40	0.00	0.02
Alps	-2.12	0.49	-1.08	1.10	0.00	0.00	
North America							
Adirondacks	-2.31	-0.18	-2.39	0.98	-0.36	-0.05	0.02
Appalachians	-2.77	-0.28	-1.41	1.39	1.85	-0.03	0.01
Blue Ridge	0.40	-0.96		-0.03		0.00	
Maine_Atlantic	-1.15	0.02	-1.00	-0.22	0.03	0.00	0.03
Ontario	-3.98	0.01	-3.66	0.32	0.23	-0.02	0.02
Vermont_Quebec	-3.04	-0.24	-1.77	0.40	1.42	-0.02	0.10

1999-2008	SO4*	NO3	Ca*+Mg*	Alkalinity	ANC	H+	TOC_DOC
Europe							
NoNordic	-1.10	-0.11	-0.22	0.99	1.24	-0.01	0.02
SoNordic	-2.60	-0.07	-1.43	0.21	1.09	-0.02	0.08
ECEurope	-1.70	-0.16	-0.60	0.28	2.81	-0.22	0.23
UK	-0.90	0.05	-0.39	0.25	-0.43	-0.10	0.13
WCEurope	-1.60	1.97	-0.95	-4.81	-3.65	-0.09	0.02
Alps	-0.80	-1.13	3.49	3.29	4.86	0.00	
North America							
Adirondacks	-2.40	-0.03	-1.62	0.07	0.55	0.01	0.01
Appalachians	-1.20	-0.12	-0.87	0.02	0.77	0.02	-0.02
Blue Ridge	-0.30	-0.11		-0.02		0.00	
Maine_Atlantic	-1.40	0.00	-0.97	-0.42	-0.67	-0.01	0.05
Ontario	-1.30	-0.07	-0.50	0.96	1.20	0.01	0.02
Vermont_Quebec	-1.60	0.09	-1.07	0.15	-0.45	-0.02	0.07

- About 70 % of nearly 200 sites, and 11 of 12 regions, show significant declines in non-marine sulphate in the periods 1990-1999 and 1999-2008. The total reductions for the whole period in the 11 regions ranged between 20 and 58%. About 2/3 of the reductions were achieved before 2000.
- Very few significant trends and regional patterns in nitrate concentration were found, probably because of multiple controls of nitrate leaching from catchments.
- The relative importance of nitrate for acidification of surface waters increased in most regions. Still, sulphate is by far the most important acidifying anion in most acidified surface waters.
- Base cations declined almost everywhere as a result of the decline in sulphate, but more slowly after 2000. The Alps were the exception to this rule with increases in base cations after 2000. Base cations are needed for chemical and biological recovery.
- About 20 % of all sites show increasing trends in alkalinity and/or ANC, and only few sites appear to be undergoing further acidification. On a regional level, all regions except UK, Maine/Atlantic Canada and Virginia Blue Ridge show positive trends in alkalinity and/or ANC in one or both periods.
- About 15 % of all sites show significant increases in pH and less than 3% show significant decreases. Trends in pH are more distinct and larger in Europe compared to North America.
- All, except 2, regions show increasing DOC concentrations in one or both periods

Trends in sulphate, pH and ANC data point to a consistent pattern of chemical recovery from acidification across a large number of sites. This chemical recovery is mostly due to the reduction in sulphur deposition.

For nitrate, however, no clear temporal or regional patterns were found. Nitrogen is an essential nutrient and most of deposited nitrogen is immobilized in catchments by biological processes. However, a small part of the nitrogen present in soils is mineralized and leaks as nitrate from soils to surface waters. Further enrichment of catchment soils by N deposition and climate change could reduce the N-retention of catchments and increase leaching of nitrate to surface waters.

The concentrations of base cations, particularly at low levels, are important for aquatic biota. Base cations can protect the biota i.e. damage caused by acidification. Pollutants such as POPs and heavy metals are also less toxic when the concentration of calcium (the most important base cation in the lakes and rivers included in ICP Waters) is high. Such effects are apparent at concentrations below 2 mg Ca/L and are large below 1 mg Ca/L. Low concentrations of base cations can affect biota even in the absence of external stressors.

DOC is increasing in many of the monitoring sites. Changes in DOC affect light penetration, primary production and oxygen concentrations. Binding by DOC reduces the toxicity of aluminium. Labile forms of aluminium, whose trends are highly dependent on acidification but not discussed in this report, are toxic at high concentrations to aquatic biota.

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4. Trends in biological recovery of acidified surface waters in Europe

4.1 Recovery of biota in acidified surface waters: a synthesis

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Recovery of biota in acidified surface waters
Acidification of freshwater habitats has received considerable attention as an international ecological problem over the past 40 years. The dynamics of natural recovery of aquatic biota from acidification in lakes and rivers is, however, due to the short time-scale of emission control, a relatively new issue. The ICP Waters programme monitors the effects of acid deposition on freshwater ecosystems with respect to water chemistry, biological and dose/response relationships.

Collection of data on biological recovery is done differently from country to country, in contrast to collection of surface water chemistry. Therefore, a unified approach to compare trends in regions across national boundaries was not feasible. Instead, each country supplied a report of relevant data that documented the status of biological recovery in acid-sensitive regions. The ICP Waters subcentre received contributions from the Czech Republic, Finland, Germany, Norway, Sweden and Switzerland. Here, the main focus is on recovery of zoobenthos (small organisms that live on the bottom of rivers and lakes such as aquatic insects, worms and snails), but data on recovery of fish populations and macrophytes (waterplants) are also given. Part of the data records presented in the current report are included in previous reports (Raddum *et al.* 2004, 2007) which focus on trends in biological recovery, evaluated against the background of trends in water chemistry.

Differences in biological responses during acidification and chemical recovery
There are fundamental differences in processes leading to biological damage under acidification, and the biological recovery processes after the critical load is no longer exceeded. The main driver for biological change during acidification is the toxic effect of water chemical components. The critical load is exceeded, resulting in toxic concentrations, which again lead to disappearance of acid-sensitive species (Raddum and Fjellheim, 2002, Posch *et al.* 2003). During the acidification process, the community structure is in general predictable at different acidic levels. Biotic interactions occur in relation to the disappearance of key sensitive organisms. An example is the effect of fish absence and the immediate increase of species sensitive to fish predation. In this case, invertebrate species become the new top predators and will structure the community. Biological responses to improved water quality during the recovery process are different because the former main driver of biological change during acidification, the toxicity, is of less importance. Instead, the main structuring drivers of a community during recovery are connected to the physical environment in the region, i.e. dispersal and colonisation ability of different species and their biological interactions, while water chemistry plays a less dominating role. The recovery of invertebrates may therefore not follow the reverse path of the changes in community structure during acidification. Due to this, predicting the process, the biological target, and the ‘end-point’ of the biological recovery is difficult. Recovery from acidification is unlikely to result in the pre-acidification biological community.

The development of the caddisfly *Hydropsyche siltalai* in rivers in the Farsund area in southwest Norway well illustrates the process of recovery (***Figure 15***). The species was missing from the samples in taken 1981-1991 and was first recorded in 1992, when the critical limits were no longer exceeded. This corresponds to timepoint t_5 of the schematic picture of acidification and recovery presented by Posch *et al.* (2003). During the first years the populations were small, as reflected by low densities. In 2000 - 2001, *H. siltalai* exceeded the critical abundance t6 (Posch *et al.* 2003) and has since then been found regularly in good numbers. The population of the species now has reached the stage 4 of the MIRACLE conceptual model (Raddum & Fjellheim 2002), population fluctuate natural. The time to reach stage 4 depends mostly on biotic factors (Raddum & Fjellheim 2002, Yan *et al.*, 2003).

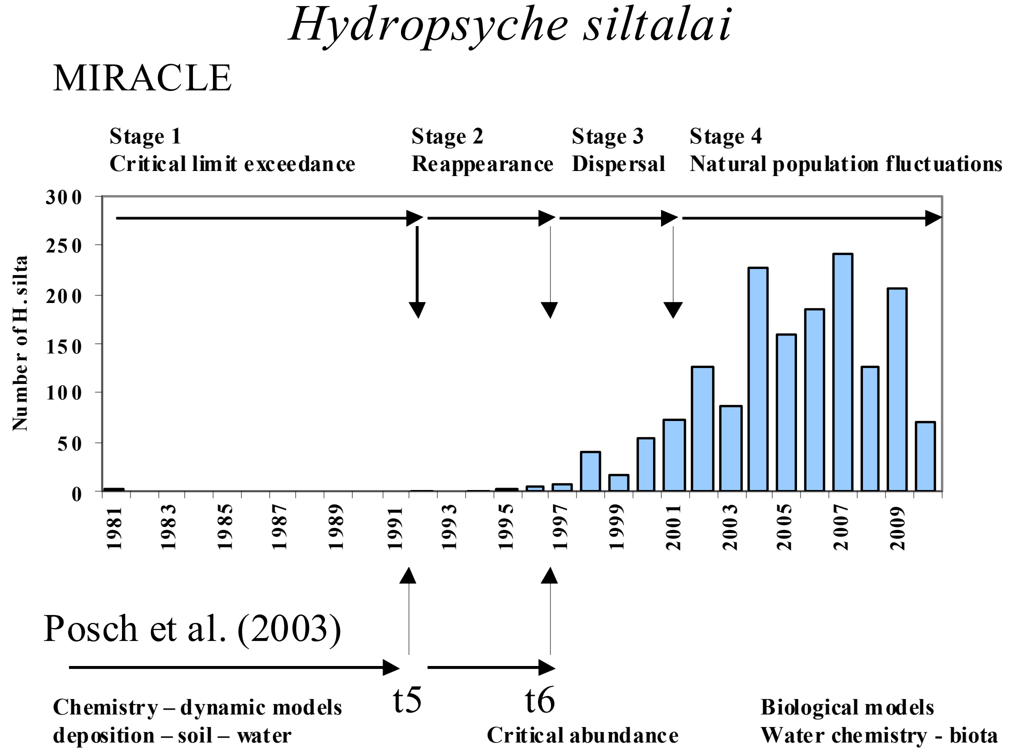

Figure 15. *Recolonisation of the caddisfly Hydropsyche siltalai in the Farsund area, within the frames of conceptual models suggested by Raddum and Fjellheim (2002) and Posch et al. (2003). Modified after Fjellheim and Raddum (2005). H. siltalai figure taken from Fjellheim and Halvorsen (this report section 4.5).*

Summary of national contributions on biological recovery

Biological recovery after reduced emissions of acidic components has been documented in most contributing countries (***Table 10***). Several regions have shown chemical recovery and the trends in biological recovery generally agree well with the chemical improvements. However, there are also examples on lack of responses or setbacks in the biological recovery process which may be related to other factors, such as climate change. Also, in some countries times series are still rather short.

The Czech contribution (Chapter 4.2) presents data on zooplankton, benthic animals and macrophytes in Bohemian forest lakes, of which all groups show a positive response in the recent years. The response was particularly evident in the zoobenthos communities resulting in increased species diversity. These results are encouraging, as the lakes of this region were among the most severely acidified worldwide.

In Finland (Chapter 4.3), chemical recovery of acid-sensitive lakes has taken place since the early 1990s. This resulted in biological recovery. Fish populations showed a significant improvement for perch as a response to improved water quality conditions. For more acid-sensitive species, evidently more time and further reductions in acidifying emissions are needed for a distinct recovery. The monitoring revealed that catchment characteristics and climatic and hydrological variability also play an important role in affecting the recovery process and trends. Changes in temperature, precipitation, and runoff pattern can all affect surface water chemistry and consequently the biological responses.

German data (Chapter 4.4) show that although reduction of sulphur and nitrogen emissions improved the acidification situation in formerly heavily affected regions such as the Ore Mountains and Harz, detrimental acidification effects are still present. Other areas less exposed to acidic depositions such as the Black Forest and parts of the eastern Bavaria have the potential to reach stable, near natural conditions in the long run.

Biological recovery in Norwegian streamwaters (Chapter 4.5) has been evident for more than a decade. The trends found in many localities are highly significant. The recovery in Norway has resulted in both increased densities of acid-sensitive benthic animal species and a marked increase in biodiversity. The response in Norway is clearer than results from other regions in Europe, as presented in this report. This may be explained by several factors: First, the Norwegian monitoring data cover a long period, which also includes the 1980´s when the freshwater biota was heavily affected by the acidity of the surface waters. Second, the data are based on running water fauna, which generally gives a quicker response than the fauna from better buffered lakes. Third, the Norwegian catchments are characterised by a surface water of very low concentrations of electrolytes and DOC and a thin layer of sediments making the localities both more susceptible to acidification and quicker to respond to decreasing acid deposition.

In Sweden (Chapter 4.6), water chemistry, phytoplankton, and littoral benthic invertebrate assemblage data from minimally disturbed (reference) lakes and acidified lakes studied during the last two decades show that assemblage composition of acidified lakes has become more similar to those of reference lakes. Changes in phytoplankton and littoral invertebrate assemblage composition were significant when measured with presence-absence data, but differences were not significant when biovolume or abundance data were used. The strongest differences were noted for phytoplankton assemblages. Interannual variations in phytoplankton and invertebrate species composition were related to climatic and to water chemical variables. The Swedish study shows that biological recovery is complex and the influence of climatic variability is poorly understood.

In Switzerland (Chapter 4.7), benthic recovery could not be detected by time trend analysis. There are, however, a few indications of a small number of newly appearing species in some of the localities. These appearances are still not significant. The Swiss timeseries start in or after 2000. It is therefore possible that some recovery already occurred before invertebrate sampling started and that a future prolongation of the timeseries will give significant responses.

Conclusions

The national contributions to the documentation of time trends in biological recovery differ considerably in time span of records, targeted groups of biota, and type of variable considered. However, some general patterns emerge. Almost all contributions report evidence of recovery for fish, zoobenthos and other biota (algae and water plants) which is attributed to improved water quality, although other factors – climate – also contribute to explaining temporal variations. Increasing organic acidity related to higher DOC in surface waters could also slow down biological recovery. Additionally, in almost all contributions it is underlined that these developments are not the endpoint and that more biological recovery can be expected. Recovery from acidification is unlikely to result in the pre-acidification biological community.

The endpoints that might be reached are of course not known, but comparison with ‘reference sites’ suggests that species diversity in fully restored aquatic ecosystems could be much higher than is presently observed in aquatic systems that are undergoing recovery from acidification.

***Table 10**. Summary of findings from national reporting on biological recovery. Colour coding for trends:* 🟩*, only positive trends;* ▨ *mixture of positive and no trend;.* ░ *no trends.*

Region	Country	Water body	Biota	Biological parameter	Period	Trends	recovery potential reached?	Cause of recovery	other relevant factors
North Nordic	Finland	8 lakes, 12 streams	Fish	Abundance, population structure	1993-2005	▨	no	improved water quality	climate
South Nordic	Norway	5 rivers	Zoobenthos	Species number, abundance, acidification index	1982-2010	🟩	no	improved water quality	seasalt episodes reduce recovery
	Sweden	8 lakes	Phytoplankton	Species number, abundance, richness	1988-2008	▨	no	improved water quality	temperature and NAO index
			Zoobenthos	Species number, abundance	1988-2008	🟩	no	improved water quality	temperature and NAO index
	Finland	30 lakes	Fish	Abundance, population structure	1985-2007	▨	no	improved water quality	climate, sensitivity of fish species
		29 lakes	Zoobenthos	Communities	1985-2001	▨	no	improved water quality	
		30 lakes	Periphyton, phytoplankton	Communities	1985-2001	▨	no	improved water quality	
East Central Europe	Czech republic	8 lakes	Phytoplankton	Chlorophyll	1990-2009	🟩	no	improved water quality	
			Zooplankton	Species number, abundance	1990-2009	🟩	no	improved water quality	
			Zoobenthos	Species number, abundance	1990-2009	🟩	no	improved water quality	
			Macrophytes	Abundance	2004-2010	▨	no	improved water quality	
	Germany	lakes, streams	Zoobenthos	Species number, abundance acidification index	1982-2010	🟩	no	improved water quality	time lags, climate
West Central Europe	Germany	lakes, streams	Zoobenthos	Species number, abundance acidification index	1982-2010	▨	no	improved water quality	time lags, climate
Alps	Switzerland	4 lakes	Zoobenthos	Species number, abundance	2000-2009	░	no information	Short time series	climate (drought)
		3 rivers	Zoobenthos	Species number, abundance	2000-2009	░	no information	water chemistry of sufficient quality at start of monitoring	climate (drought), short time series

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4.2 Czech republic: Trends in biological recovery from acid stress in the Bohemian Forest lakes

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Eight small glacial lakes, situated on forested slopes of the Bohemian Forest (Šumava in Czech, Böhmerwald in German) along the border between Bohemia (Czech Republic), Bavaria (Germany) and Austria at altitudes between 930–1180 m a.s.l., have represented one of the most severely acidified lake areas worldwide (Kopáček *et al.* 2002, Majer *et al.* 2003, Vrba *et al.* 2003). With a background of 140-year of hydrobiological research and palaeolimnological studies, we were able to document significant changes in the lake water chemistry and consequent changes in the plankton and benthos composition (in particular the conspicuous reduction in crustacean and/or insect species), as well as the extinction of fish (e.g., Vrba *et al.* 2003). A clear reversal in water chemistry trends, following the decline in atmospheric deposition with certain hysteresis, has been documented in each Bohemian Forest lake after >80% and 35% reduction in S and N deposition, respectively (e.g., Kopáček *et al.* 2002). In theory, such a remarkable drop in atmospheric deposition could release the ecosystems from the acid stress and result in biological recovery. Biotic response of the lake ecosystems, however, has lagged for a decade or even longer. The first signs of any plankton recovery appeared in the late 1990s (Vrba *et al.* 2003), while lake biota have been recovering more remarkably during the last decade (Nedbalová *et al.* 2006, Čtvrtlíková *et al.* 2009).

Plankton

A significant increase in phytoplankton biomass was observed in some lakes, where absence because of Cladocera in the open water (pelagial) there was no efficient top-down effect of filter-feeding herbivores (Rachelsee and Plešné Lake, ***Figure 16***). A remarkable 10-100 fold increase in abundance of acid-tolerant rotifers followed the increasing food sources in such lakes (e.g. Fig. 4 in Nedbalová *et al.* 2006) and newcomer rotifer species appeared in recovering lakes (***Figure 16***). *Ceriodaphnia quadrangula* used to be the first and only species of Cladocera returning to some recovering lakes (Černé Lake 1997, littoral of Prášilské Lake 2002, Čertovo Lake 2007, and Rachelsee 2009). Both the species composition of zooplankton (***Figure 16***) and the entire food web structure have been greatly affected by populations of invertebrate predators in the fishless lakes.

Of particular importance was *Cyclops abyssorum*, a copepod species formerly inhabiting most of the Bohemian Forest lakes. The species did not survive the period of acidification in the lakes Černé, Čertovo, Rachelsee, and Plešné, whereas it has occurred continuously in the lakes Prášilské and Grosser Arbersee. In 2004, a whole-lake experiment was carried out by implanting *Daphnia longispina* and *Cyclops abyssorum* from Prášilské Lake to Plešné Lake. The objective was to learn whether the chemical reversal in Plešné Lake has proceeded so far to make possible survival of this extirpated species (Kohout and Fott 2006). 2005-2006 numbers of *Cyclops* increased exponentially and in 2008–2010 summer values of large copepodites and adults fluctuated between 700–1500 ind.m^{-3}. On the other hand, the introduced *Daphnia* have not appeared until now. The paper on this experiment is under preparation.

Benthos

There was documented a remarkable increase in species diversity of stoneflies, mayflies and caddyflies, including a return of acid-sensitive species to the littoral of most Bohemian Forest lakes during last two decades (***Figure 16***).

Macrophytes

Critical endangered glacial relicts, quillworts *Isoëtes lacustris* and *I. echinospora*, survived the 30-year period of severe acidification in Černé and Plešné lakes, respectively. Both populations failed to reproduce and decreased until 2004 (***Figure 17***). Laboratory experiments and field studies have recently proven that ionic aluminium (Al) interfered with sporeling development and might have caused impairment of *Isoëtes* reproduction over the long acidification period (Čtvrtlíková *et al.* 2009). Similar Al concentrations have been critical for sporeling survival of both species. Despite improving chemistry in both lakes, the recovery of the *Isoëtes* populations has differed. While the plant stand of *I. lacustris* has not yet started any renewal due to a failure of sporeling survival, a reproduction boom of the *I. echinospora* population has been observed since 2005 (***Figure 17***). Due to the distinct germination length (Čtvrtlíková, unpubl. results), the sensitive reproduction stages of either species are exposed to different Al doses during the seasons. Thus, besides the key role of Al in phosphorus availability, food web structure, and plankton dynamics in the Bohemian Forest lakes (Vrba *et al.* 2006), harmful Al also forms a bottleneck for quillwort reproduction and recovery from acid stress.

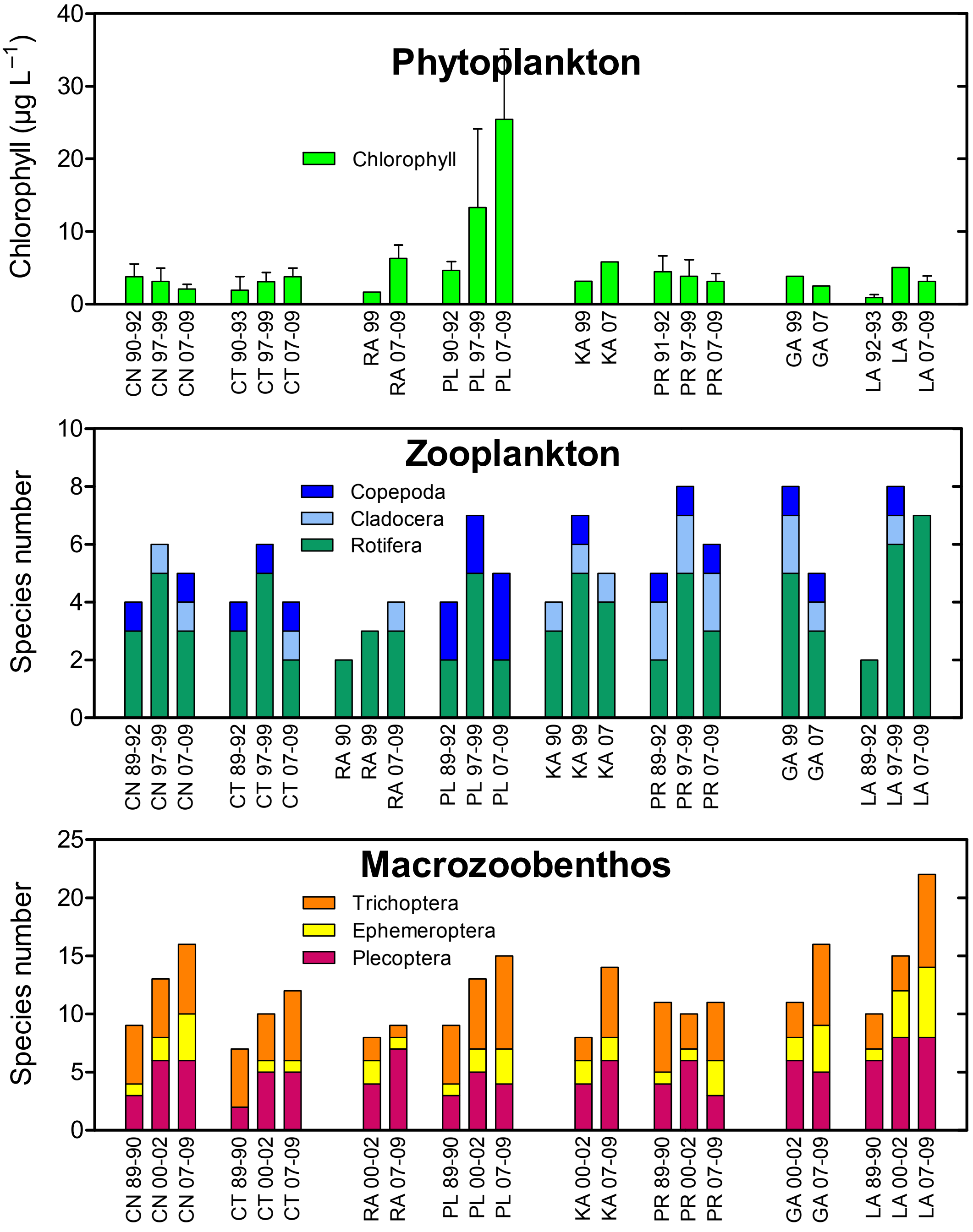

Figure 16. *Trends in biological recovery of eight acidified lakes in the Bohemian Forest (ICP Waters codes in parentheses) ordered according to their decreasing (from left to right) acidity (Nedbalová et al. 2006): CN – Černé Lake (CZ01), CT – Čertovo Lake (CZ02), RA – Rachelsee (DE17), PL – Plešné Lake (CZ03), KA – Kleiner Arbersee, PR – Prášilské Lake (CZ04), GA – Grosse Arbersee, LA – Laka Lake (CZ05). Sampling periods are not the same for all lakes and biota; data are averages of summer values for the periods (single values are available for most sampling of German lakes: RA, KA, and GA). Phytoplankton biomass is expressed as chlorophyll* a *concentrations in the epilimnion (error bars indicate SD of multiple summer data where available); zooplankton diversity includes only species occurring in the open water; and benthos diversity includes only littoral species.*

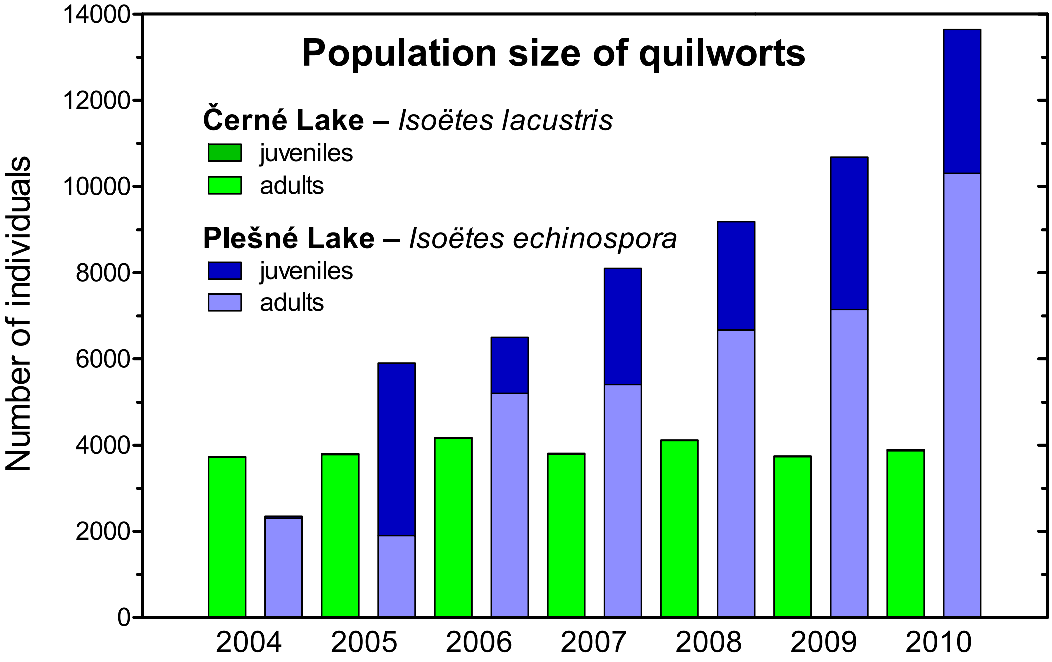

Figure 17. *Trends in reproduction recovery of two quillwort specie (macrophytes) in two Bohemian Forest lakes.*

Acknowledgements

The data from the German lakes have been collected owing to a long-term fruitful cooperation with the following institutions: Nationalpark Bayerischer Wald, Bayerisches Landesamt für Umwelt, WWA Deggensdorf and WWA Regensburg.

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4.3 Finland: Recovery trends in surface waters chemistry and biota

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Awareness of lake acidification in Finland

The multidisciplinary Finnish Acidification Research Progamme (HAPRO) in 1985-1990 documented that a considerable number of small headwater lakes in Finland were acidified (Forsius *et al.* 1990a,b), and acidification of waters had affected species composition and biodiversity of biological communities such as periphytic diatoms (Eloranta 1990), surface diatom assemblages (Huttunen and Turkia 1990), macrophytes (Heitto 1990), phytoplankton (Kippo-Edlund and Heitto 1990), benthic invertebrates (Meriläinen and Hynynen 1990), zooplankton (Sarvala and Halsinaho 1990) and fish (Rask and Tuunainen 1990). The national lake status survey in 1987 estimated the number of acidic (Gran alkalinity ≤ 0 µeq l^{-1}) lakes to be 4900 (Forsius *et al.* 1990b), and the fish status survey (lake area ≥ 0.01 km^2) in 1985-1987 reported declines and even extirpation of sensitive fish species in small lakes due to anthropogenic acidification in south and central Finland (Rask and Tuunainen 1990). The number of lakes in south and central Finland in which fish populations were affected due to acidification was estimated to be 2200-4400, and of those 1000-2000 fish populations was estimated to be lost (Rask *et al.* 1995a). The most damaged populations were roach (*Rutilus rutilus* L.), and to a smaller extent European perch (*Perca fluviatilis* L.), the most common species in small lakes in south and central Finland. The fish status survey of Finnish lakes (lake area ≥ 0.04 km^2) in connection with the Northern Europe Lake Survey in 1995 suggested that ca. 700 fish populations were lost and 1200 affected, mainly roach populations (Tammi *et al.* 2003a). Both fish surveys suggested that perch was not extinct in any lakes. However, some strongly acidified lakes were found outside the coverage of the surveys that had lost their perch populations (Rask and Tuunainen 1990, Nyberg *et al*. 2010). In a fish status and water chemistry survey carried out at 103 sites in three areas in north-eastern Lapland in 1991-1993, Lappalainen *et al*. (1995) found evidence of acid-induced damage in local minnow (*Phoxinus phoxinus*) populations in acid-sensitive low alkalinity lakes (< 50 µeq l^{-1}) in the Vätsäri area, which has been exposed to acidic deposition from industrial emissions on the Kola Peninsula.

Monitoring of acidification

Water chemistry

The chemical monitoring of lake acidification in Finland started in 1979 with 28 small forest lakes (Roila 1992). The monitoring network was extended in 1987 in connection with the national survey of lake acidification by subjectively chosen lakes from the survey lake population. The monitoring network was refined and extended in 1990, and comprised ca. 160 lakes located throughout the country, and the concept of the Regional Monitoring of Lake Acidification (RMLA) was also adopted as the monitoring strategy for acid-sensitive lakes in Finland (Mannio 2001a,b, Vuorenmaa 2007). In 2006, the monitoring network was optimized by reducing the lake group to 31 intensively, seasonally monitored lakes, in which eight lakes belong to UNECE ICP Waters network and three lakes belong to UNECE ICP Integrated Monitoring network. The results of chemical monitoring given in this report refer to samples taken at 1 meter depth.

Biological monitoring

Regular biological monitoring of acidified lakes in Finland has primarily been that of fish abundance and population structure. The fish monitoring in acidified lakes, mainly with perch and roach populations, started in the year 1985 with three-year interval monitoring, in connection with HAPRO. The integrated monitoring of water chemistry and fish populations in acidified lakes was started in 1990 with 21 lakes, which were selected from common lake acidification monitoring sites, representing the four levels of fish population response to acidification (Rask *et al*. 1995a):

(1) Perch extinct (lake n=4; pH<5; Al_{lab} 80-280 µg l^{-1})
(2) Perch affected (lake n=5; pH 4.8-5.5; Al_{lab} 50-160 µg l^{-1}
(3) Roach extinct (lake n=6; pH 5.2-6.0; Al_{lab} 25-135 µg l^{-1})
(4) Roach affected (lake n=6; pH 5.3-6.4; Al_{lab} 5-40 µg l^{-1})

In order to examine the biological recovery at different trophic levels, a project: 'Recovery processes in acidified Finnish headwater lakes (REPRO)' was conducted in 2001-2003. A subset of 30 lakes in south and central Finland from the 140 headwater lakes of the HAPRO project (1984-1988) were re-sampled for chemical parameters and resurveyed for fish (Tammi *et al*. 2004), macrozoobenthos (Hynynen and Meriläinen 2005) and littoral periphytic diatoms (Kwandrans and Eloranta 2004, Eloranta and Kwandrans 2005, Kwandrans 2007). For benthic invertebrates, palaeolimnological samples were also taken in order to assess whether subfossil chironomids have value in the monitoring of long-term changes in the degree of acidity in small forest lakes (Hynynen and Meriläinen 2005).

Trends in recovery from acidification

Chemical recovery

Sulphur deposition has been the major driving force in the anthropogenic acidification of surface waters in Finland as well as in other Nordic Countries (Skjelkvåle *et al*. 2001) and elsewhere in Europe (e.g. Prechtel *et al*. 2001, Wright *et al*. 2005). Following the general decreasing trend in sulphur deposition in Finland since the late 1980s (e.g.Vuorenmaa 2004), the regional recovery of acid-sensitive lakes, indicated by decreasing concentrations of sulphate and increase in alkalinity throughout Finland, was first observed in the early 1990s (Mannio and Vuorenmaa 1995). A significant decrease in sulphate concentrations and increase both in alkalinity (measured) and ANC (calculated) over the period 1990-2005 was detected in 94% and 63% of the study lakes, respectively (Figure 1). The significant increase in pH in lakes is less common (30% of the lakes) than the increase in alkalinity, but in the most affected areas in south Finland pH has had the highest increase, with significant increase in about 60% of the lakes. Accompanying the increases in pH, concentrations of labile aluminium are decreasing in the formerly most acidic but now recovering lakes (Tammi *et al*. 2004; Vuorenmaa and Forsius 2008). There are no indications of increasing nitrate concentrations in acid-sensitive Finnish forest lakes, and NO_3-N concentrations are decreasing in most of the monitoring lakes (Vuorenmaa and Forsius 2008). The recovery of lakes from acidification has been the strongest and most consistent in south Finland (***Figure 18***), where lakes have been exposed to highest S deposition load, and it is this region that has also showed strongest emission reduction responses in deposition (Vuorenmaa 2004, Vuorenmaa and Forsius 2008). Sulphur deposition in south Finland has declined by about 70% since the late 1980s (Vuorenmaa 2007). Chemical recovery is progressing even in the most acidified lakes, but the buffering capacity of many lakes is still low (Gran alkalinity and/or charge-balance ANC commonly between -20 and 20 µeq l^{-1}) and still sensitive to acidic episodes and any future increase in acid deposition (Vuorenmaa 2007).

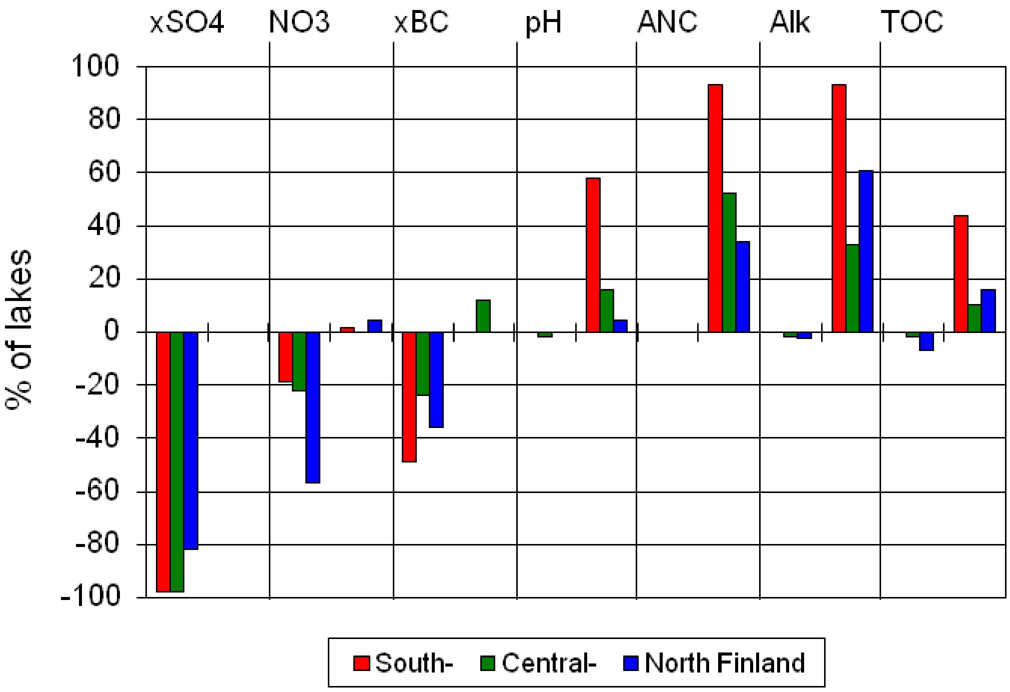

Figure 18. *Percent of RMLA lakes in south (n=59), central (n=58) and north (n=44) Finland showing significant increasing or decreasing trends (Kendall-τ, $p < 0.05$) of key water quality variables for the period 1990-2005. xSO4 and xBC refer to non-marine sulphate and non-marine base cations (Ca+Mg) respectively.*

Contrary to minerogenic acidification, organic acidity has generally increased in acid-sensitive Finnish lakes throughout the 1990s and 2000s, indicated by increased TOC concentrations (Vuorenmaa *et al.* 2006; Monteith *et al.* 2007). Elevated TOC concentrations in lakes were observed particularly in the late 2000s, due to rainy summers and autumns in 2004 and 2006. Leaching of humic material from the catchment, inducing higher organic acidity in the lakes, can be important factor suppressing recovery of pH and alkalinity in acid-sensitive Finnish lakes (Wright *et al.* 2006; Vuorenmaa and Forsius 2008; Nyberg *et al.* 2010). In spite of progressing recovery in acidified lakes (***Table 11***), there are signs of backlash in recovery of buffering capacity in the study lakes in south Finland since the year 2004, indicated by decreasing alkalinity (measured) and ANC (calculated) (***Figure 19***). Organic acid surges, together with diluted and low-buffered runoff water, may have delayed the increase of buffering capacity of lakes.

Biological recovery

Fish

At the same time in the early 1990s or in some cases even earlier than the measured chemical changes, the first signs of recovery in affected perch populations were recorded in south Finland, which had experienced poor reproduction periods and populations close to extinction in many acidified lakes in the 1970s and 1980s (Nyberg *et al.* 1995, Rask *et al.* 1995b). New strong year-classes of perch appeared in affected populations in the mid 1990s, perch NPUE (mean number of fish in one NORDIC survey net in one night) increased correspondingly, showing recovery of perch reproduction (***Figure 20***, ***Table 12***). Since then, the structure of most of the affected perch populations has turned normal. Increases in pH and alkalinity, and particularly decreases in labile aluminium concentrations towards levels tolerable for acid-sensitive species are mainly responsible for this positive development (e.g. Tammi *et al*. 2004) (***Table 11***). Successful re-establishment of perch populations into previously heavily acidified and fishless lakes in south Finland also emphasizes the significant impact of the chemical recovery of the lakes (Nyberg *et al*. 2010). Positive responses have been observed also from Minnow populations in lakes in north-eastern Finland in the Vätsäri area, which have experienced recovery during the 2000s along with decreased SO_2 emissions from the smelters on the Kola Peninsula (Tammi *et al*. 2003b, Lappalainen *et al*. 2007).The recovery process of roach, which is a more acid-sensitive species, has not been clear (***Table 12***), and in some acidic lakes, roach populations have probably not survived the turning point of the acidification-recovery process (Tammi *et al*. 2004). This suggests that water quality seems to be still critical for success of roach populations, and recovery in the future will depend on further reductions in acidifying emissions and consequent improvement of water conditions. Confounding factors, such as organic acid surges together with diluted and low-buffered runoff water, may also have restricted the recovery of acid-sensitive fish species such as roach. However, in a few acidic lakes that were inhabited by sparse roach populations during the 1985-1995, some reproduction was observed in the late 1990s (Nyberg *et al.* 2001) (***Figure 21***). Three consecutive fish status surveys were carried out at 20 sites (8 lakes and 12 streams) in north-eastern Finland in the Vätsäri area in 1993, 2000 and 2005 (Lappalainen et al. 1995; Tammi et al. 2003b; Lappalainen et al. 2007). It was shown that local minnow populations experienced recovery during the 2000s along with decreased SO2 emissions from the smelters on the Kola Peninsula (Tammi et al. 2003b, Lappalainen et al. 2007).

Table 11. *Trends of key chemical acidification variables during the period 1987-2009 and mean concentration of alkalinity (Gran method), pH and labile aluminium (Al_{lab}) for the two comparative periods (1987 and 2005-2009 for alkalinity and pH, 1987 and 2005 for Al_{lab}) in 12 integrated water chemistry and fish monitoring lakes in south Finland. For the annual change ($\mu eq\ l^{-1}\ yr^{-1}$) a statistically significant trend (seasonal Kendall-test, $p < 0.05$) is denoted with an asterisk (*). The monitoring lakes are presented by four lake groups, representing four levels of fish population response to acidification. Group 1: perch extinct, group 2: perch affected, group 3: roach extinct, group 4: roach affected.*

Lake groups	Trend data	Alka-linity	ANC	H^+	xSO_4	Alkalinity ($\mu eq\ l^{-1}$)		pH (pH-unit)		Al_{lab} ($\mu g\ l^{-1}$)	
		$\mu eq\ l^{-1}\ yr^{-1}$				1987	2005-2009	1987	2005-2009	1987	2005
Group 1											
Iso Lehmälampi	1987-2009	1.38*	0.81	-0.39*	-3.61*	-36	-4	4.8	5.4	83	25
Iso Majaslampi	1987-2007	1.76*	2.16*	-0.44*	-3.71*	-33	-3	4.8	5.2	141	34
Hauklampi	1987-2007	1.44*	2.67*	-0.68*	-5.70*	-19	-4	4.9	5.4	276	64
Group 2											
Munajärvi	1987-2006	1.20*	1.50	-0.73*	-2.75*	-15	-1	4.9	5.3	111	40
Orajärvi	1987-2006	1.00*	1.52	-0.59*	-4.77*	-37	-3	4.9	5.5	161	< 10
Saaren Musta	1987-2005	1.13*	1.68*	-0.23	-4.84*	-11	-3	5.2	5.1	66	54
Group 3											
Simijärvi	1987-2009	1.30*	0.18	-0.06*	-2.56*	28	21	5.9	6.3	35	< 10
Isojärvi	1987-2006	2.00*	2.83*	-0.06*	-4.90*	53	45	5.6	6.0	28	< 10
Pitkäjärvi	1987-2006	2.00*	1.25*	-0.15*	-3.33*	1	28	5.2	6.1	40	11
Group 4											
Saarijärvi	n.d.										
Vitsjön	1987-2009	3.00*	3.09*	-0.02*	-5.12*	7	67	6.1	6.5	< 10	< 10
Kattilajärvi	1987-2009	1.21*	0.38	-0.03*	-4.55*	10	38	6.0	6.3	19	< 10

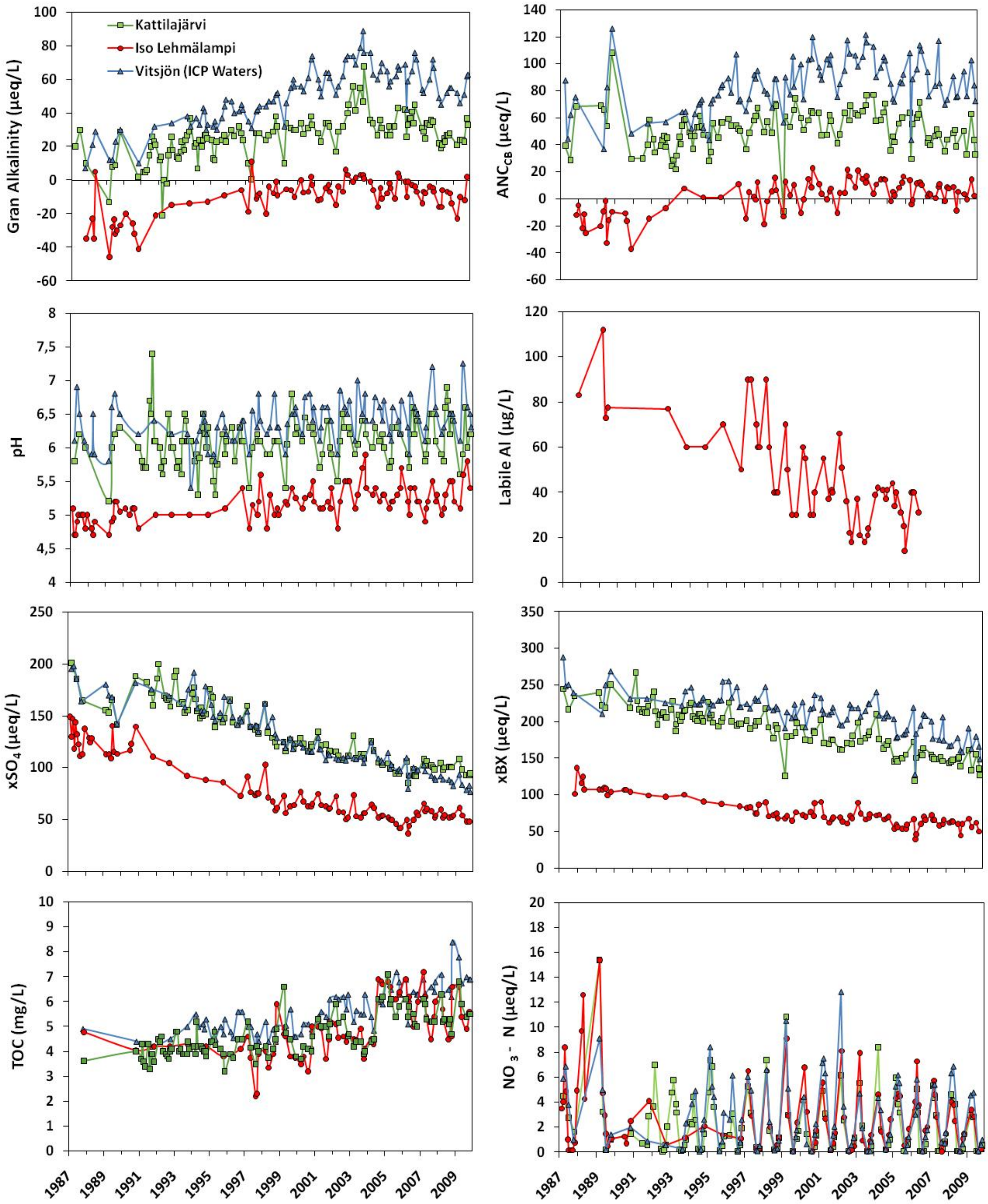

Figure 19. *Long-term trends of key chemical acidification parameters in three fish monitoring lakes (L. Kattilajärvi, L. Iso Lehmälampi and L. Vitsjön) in south Finland (1987-2009). Lake Vitsjön belongs to UNECE ICP Waters network. xSO4 and xBC refer to non-marine sulphate and non-marine base cations (Ca+Mg) respectively.*

Table 12. *Changes of NPUE and mean weight in perch and roach populations between the periods 1985-1988 and 2001-2007 in 12 integrated water chemistry and fish monitoring lakes. The monitoring lakes are presented by four lake groups, representing four levels of fish population response to acidification. Group 1: perch extinct, group 2: perch affected, group 3: roach extinct, group 4: roach affected. Biological trends are presented as: population recovering (+), no change (0), population declining(-), no data (n.d). NPUE= mean number of fish in one NORDIC survey net in one night. *= positive trend after reintroduction of perch to lakes that had lost their populations due to acidification.*

Lake groups	Trend data	NPUE (n of fish/Nordic net/night)				Mean weight (g)				Trend	
		Perch		Roach		Perch		Roach		Perch	Roach
		1985-1988	2001-2007	1985-1988	2001-2007	1985-1988	2001-2007	1985-1988	2001-2007		
Group 1											
Iso Lehmälampi	1985-2007	1.2	37			98	50			+	
Iso Majaslampi	1985-2007	0.1	6.1			583	152			+*	
Hauklampi	1985-2007	2.3	14.5			192	62			+*	
Group 2											
Munajärvi	1985-2001	0.5	22.8			462	64			+	
Orajärvi	1985-2007	9.5	41.5			293	44			+	
Saaren Musta	1985-2001	15.3	32.9			80	29			+	
Group 3											
Simijärvi	1985-1995	3.1	n.d.	0.1	n.d.	37	n.d.	120		0	_
Isojärvi	1985-2001	24.0	35.0	4.3	0	25	37	94		0	_
Pitkäjärvi	1985-2001	8.5	36.6	0.4	0	35	44	97		0	_
Group 4											
Saarijärvi	1985-2007	22.0	19.2	9.6	4.0	33	26	105	78	0	0/+
Vitsjön	1985-2007	14.5	20.0	15.5	11.7	30	54	80	65	0	0/+
Kattilajärvi	1985-2007	53.5	13.0	6.5	9.7	22	25	62	32	0	0/+

Clear recovery of perch populations in Finnish lakes was detected in the early 1990s, at the same time or even earlier than the measured chemical changes. Thus, factors other than decreased acid deposition apparently affected the occurrence of suitable conditions for the reproduction of perch at that time. Favourable thermal conditions, such as warm springs and growing seasons and the mild winters of 1989, 1990 and 1992, may have had a beneficial impact on the reproduction of perch (Rask *et al.* 1995b). Higher temperatures during the growing season in boreal waters may produce strong year classes and promote growth (Hokanson 1977, Koli *et al.* 1985). In addition, the autumn of 1989 was dry, resulting in low water tables in spring 1990. Permafrost was weak during that winter, allowing melting waters to percolate through soils, resulting in less acid input to the lakes in spring. The mild winters and thin snow cover in 1991 and 1992 may also have contributed to the weaker acidic pulse during the spring melt runoff. This suggests an important role of climatic factors in determining biological responses.

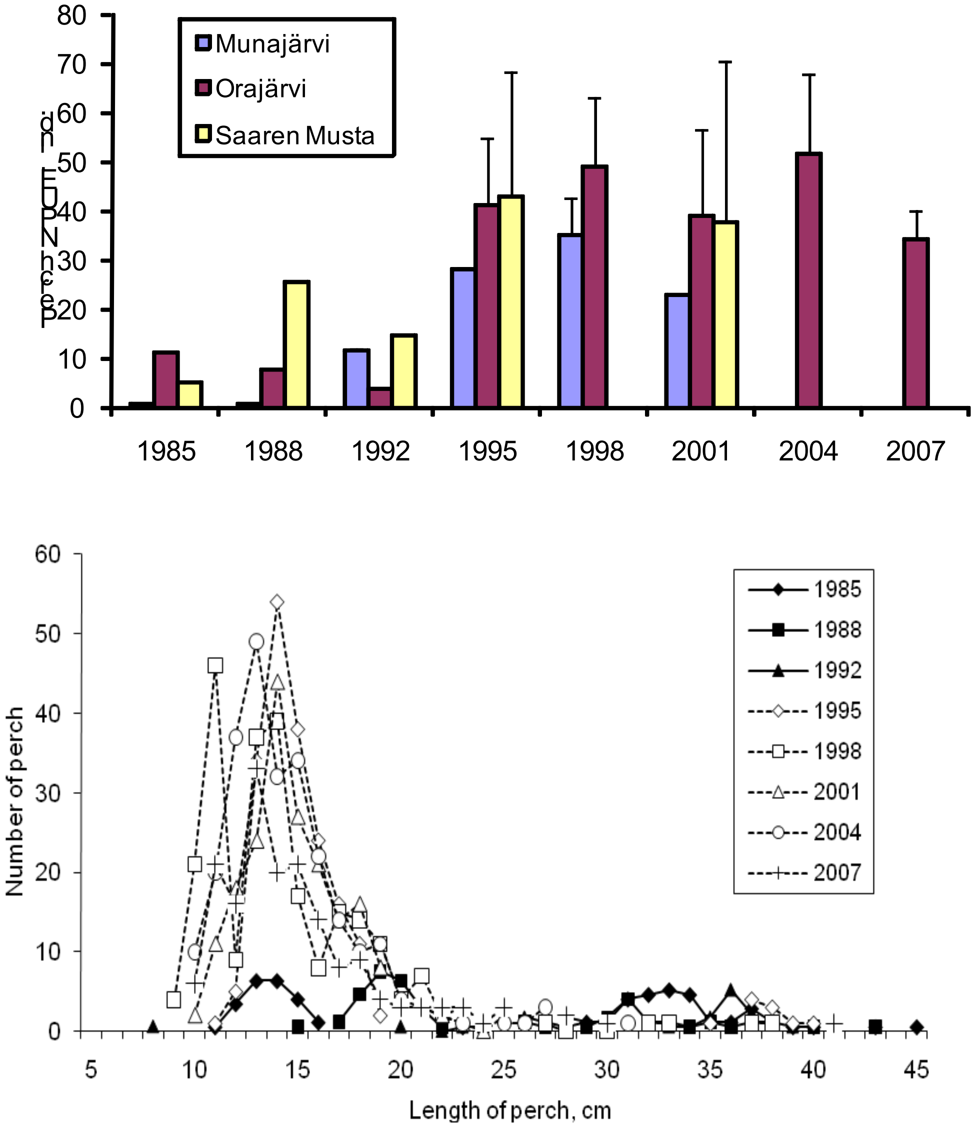

Figure 20. *Trends in unit catches of perch (number of perch per NORDIC gillnet per night, mean ± SD) in three lakes (L. Munajärvi, L. Orajärvi and L. Saaren Musta) recovering from acidification (top) and in length frequency distribution of perch in one of the lakes, L. Orajärvi (bottom).*

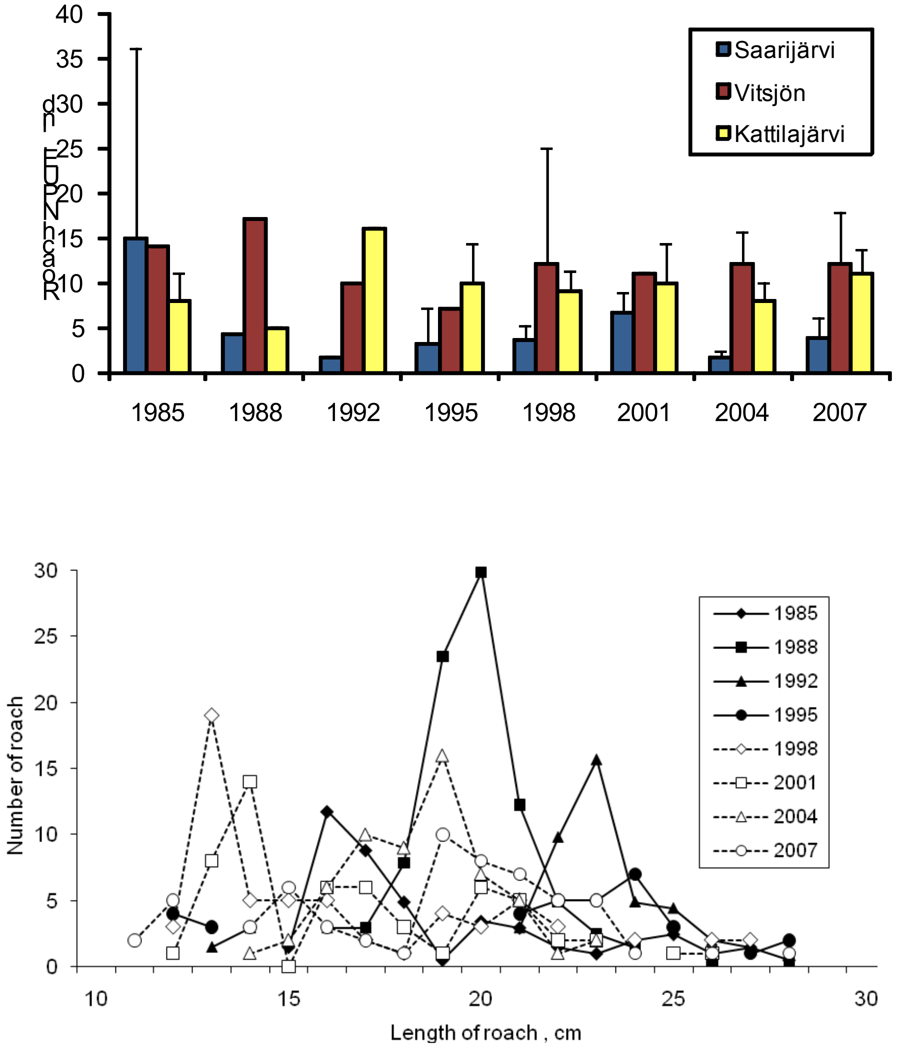

Figure 21. *Trends in unit catches of roach (number of roach per NORDIC gillnet per night, mean ± SD) in three lakes (L. Saarijärvi, L. Vitsjön and L. Kattilajärvi) recovering from acidification (top) and in length frequency distribution of roach in one of the lakes, L. Vitsjön (bottom).*

Other biota

In addition to fish populations, biological recovery from acidification has been observed at other trophic levels. Acid-sensitive and moderately acid-sensitive benthic invertebrate species showed a slight recovery between 1985 and 2001 in the formerly most acidic (pH ≤5.5) but now recovering lakes in south and central Finland (Hynynen and Meriläinen 2005). The most significant factors correlating to the response of benthic communities were increased pH and decreased labile aluminium concentration of the lakes. The results revealed that a recovery is progressing in acid-impacted lakes, but evidently more time is needed for a distinct recovery of acid-sensitive benthic species to take place. Palaeolimnological chironomid analysis revealed structural similarity between the present and pristine chironomid assemblages. This implies that no major changes in chironomid communities of these acidic lakes have occurred during the past centuries (Hynynen and Meriläinen 2005). Comparison of diatom-inferred lake water pH (DI-pH) between the HAPRO-period (1985) and the resurvey (2001) results clearly indicated an increase in pH and alkalinity (Kwandrans 2007) in 30 lakes in south and central Finland. Littoral periphytic diatoms indicated the most clear positive pH-change (median increase 0.5-0.75 pH units) in the lakes in south Finland, where empirical evidence of chemical recovery has been the strongest. In central Finland both negative and positive DI-pH changes were found. Many lakes in central Finland and some lakes in south Finland did not show clear changes in diatom communities. These lakes tended to be naturally acid due to high humus concentrations and organic acidity and some had even lower pH than in the 1980s indicating the sensitivity to pH changes due to low alkalinity (low ionic strength water with high humus content and acid mineral soils

Conclusions

The chemical recovery of Finnish acid-sensitive lakes has taken place since the early 1990s, and has resulted in biological recovery, the ultimate intention of emission abatement policy. Perch, which has good tolerance and adaptation ability, is responding most to improved water quality conditions. For more acid-sensitive species, evidently more time and further reductions in acidifying emissions are needed for a distinct recovery.

Catchment characteristics and climatic and hydrological variability also play an important role in affecting the recovery process and trends. Changes in temperature, precipitation and runoff pattern, can all affect surface water chemistry and – consequently - biological responses. The potential impact of mobilization and export of DOC and organic acidity may become particularly important in Finnish conditions because of the large stores of organic matter in boreal forest soils.

The importance and value of an integrated monitoring approach including physical, chemical and biological variables is clearly indicated, and continual environmental monitoring is needed as a scientific basis for further actions in air pollution policy.

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4.4 Germany: Long-term trends in biota at ICP Waters sites

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Germany has contributed data to the international UNECE monitoring program ICP Waters for more than 20 years. At selected surface water sites in acid-sensitive regions chemical and biological parameters are investigated to document the impact of atmospheric pollution, in particular sulphur deposition.

During this time period emissions from power plants were significantly reduced leading to a reduction of acidifying deposition, especially sulphur. At many sites water sulphate concentrations are at levels that are less likely to induce any further adverse effects in the aquatic system.
However, despite the reduction in sulphur and nitrogen deposition, surface water acidification is still a problem in some German regions. On the one hand, other factors (natural disasters like storms or insect invasions) may influence biological recovery; on the other hand, there is a time lag between pollutant reduction and improvement of aquatic systems.

The monitoring programme for assessing long-term trends on biota includes regular sampling of macrozoobenthos (MZB) in lakes and rivers in a number of acid-sensitive areas in Germany (***Figure 22***). The following metrics were calculated to assess the impact of acidification (implemented in ASTERICS/ PERLODES, the German official software tool for the assessment of the ecological status of streams and rivers with MZB according the EU-WFD) on macrozoobenthos:

- Number of species
- Number of Ephemeroptera and Trichoptera species
- Percentage of indicative taxa
- Acid Class according Braukmann & Biss (2004)
- Acidity Index according Hendrikson &Medin (1986)

In all cases with a positive trend the number of MZB-species increased in most cases the Acid Class according to Braukmann & Biss (2004) (an acidity index) also increased the acidity index according Hendrikson & Medin (1986) (***Table 13***).

Results from the German ICP Waters monitoring programme can be summarized as follows:
Reduction of sulphur and nitrogen emissions improved the acidification situation in formerly heavily affected regions such as the Ore Mountains (Erzgebirge) and Harz. However, detrimental acidification effects are still present.

Other areas less exposed to S- and N-deposition such as the Black Forest (Schwarzwald) and parts of eastern Bavaria (Bavarian Forest and Upper Palatinate Forest) have the potential to reach stable, near natural conditions in the long run.

***Table 13**. Trends of selected parameters in acid-sensitive regions in Germany (Schaumburg et al. 2010). For each region the number of investigated sites in lakes and streams are indicated*

Region (lakes/ rivers)	pH	SO4	Biology*	Comment
Lauenburgische Seenplatte (1/0)	o	+	not analyzed	changes in pH influenced by hydrology and only to a lesser extent by atmospheric pollutants; acidification is masked by eutrophication
Harz (0/1)	o	+	+	despite massive reduction in sulfur deposition SO4-concentration still high, no significant improvement of acidification detectable
Rothaargebirge (0/2)	+	+	+	overall positive trends in chemistry and biology, but at one site still acidification problems with tendency of deterioration during the last years
Hunsrück (0/2)	+	+	o	improving sulfate and aluminum concentrations, but despite improving pH-situation surface waters are periodically or permanently acid
Schwarzwald (0/3)	+	+	+	positive trend, 2 out of 3 investigated sites had recovered from acidification
Bayerischer Wald (1/4)	+	+	+	overall positive trend, but deterioration due to storm damages and insect calamities resulting in elevated nitrate concentrations
Oberpfälzer Wald (0/2)	+/o	+	+	positive trend at one site with increasing species number and pH improvement
Fichtelgebirge (0/3)	+	+	+	positive trend; however, region with the highest acidification problems in Bavaria
Erzgebirge mit Vogtland (2/4)	+	+	+	positive trend at all investigated sites due to significant sulfate and nitrate reduction

+ = improvement, o = unchanged, - deterioration *only qualitative assessment of trend

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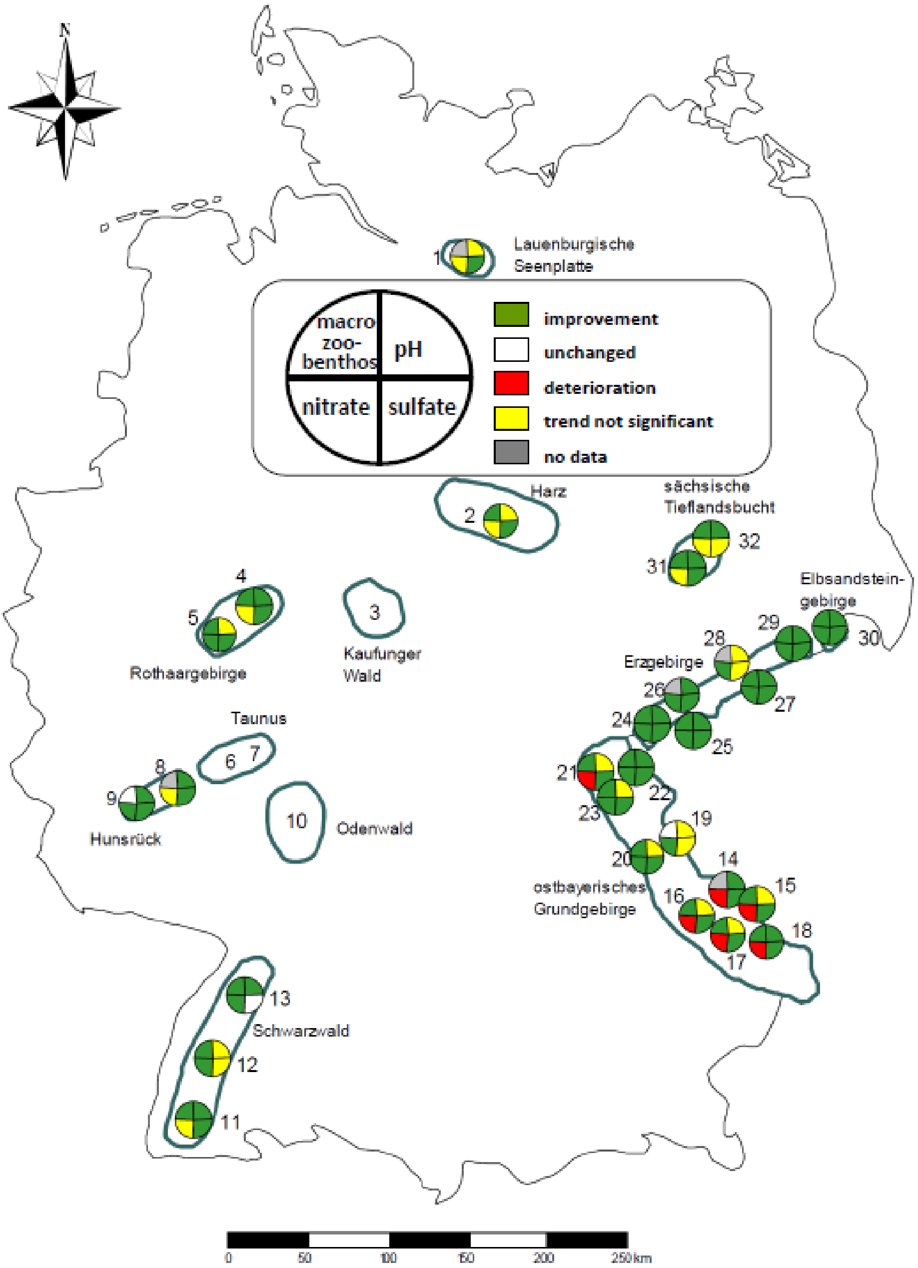

Layout and cartography: B. Kifinger, Geo-Ökologie, Weilheim i.Obb.
Sites 3,6,7,10 have been eliminated from the monitoring programme in 2004

***Figure 22**. Trends of selected parameters in acid-sensitive regions in Germany (Schaumburg et al., 2008)*

4.5 Norway: Trends in recovery of benthic invertebrate communities

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Introduction

The monitoring of benthic invertebrates in connection with acid precipitation is performed under the national monitoring programme "Monitoring of long-range transboundary air pollution". Data from this project date back to the early 1980s. During the period of study, the concentrations of sulphate in precipitation at the various sites decreased by 72 to 90 % in south Norway (Anonymous 2011). Similarly, the nitrate and ammonium concentration show significant decreases in precipitation at most sites, between 26 % and 46 % reduction for nitrate and 47 % to 63 % for ammonium since 1980. Invertebrate data from the last three decades thus cover a very important period regarding the situation of acidification in Norway. Invertebrates are useful for monitoring of acidification due to high biodiversity, a wide spectre of tolerance levels, and shorter life cycles than, for example, fish.

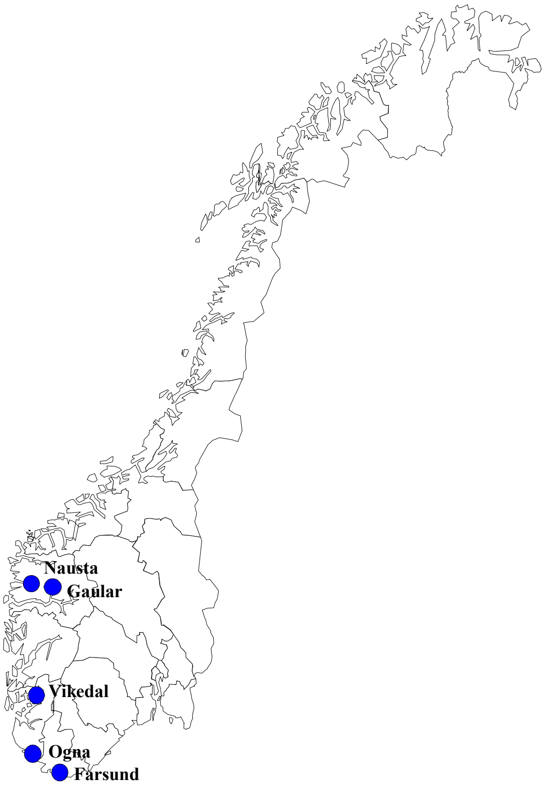

***Figure 23**. Location of monitoring rivers for benthic surveys in Norway.*

The regional benthic surveys in running waters include the monitoring of five rivers (***Figure 23***). This monitoring was started in 1981 at two adjacent catchments in the Farsund area. During the next three years, four other rivers were added to the monitoring network. All sites were sampled annually each spring and autumn using qualitative sampling methods (Raddum and Fjellheim, 1984, Fjellheim and Raddum, 1990, Raddum *et al.* 1988, Larsen *et al.* 1996, Lien *et al.* 1996).

The Norwegian monitoring data has produced some of the longest time series of benthic invertebrate data in the world. This gives an excellent platform for studies of long-term trends in the development of benthic assemblages following the reduction of acidifying emissions in Europe.

An important tool to assess the biological state of a sampling location is the use of the acidification score. The acidification score is based on presence/absence of sensitive benthic animal species (Fjellheim and Raddum, 1990). The score, which gives the mean index of the different sites within the watershed, ranges between 0 (highly acidified) to 1.0 (unacidified).

Results and discussion

The development of the acidification score in the different watersheds (***Figure 24***) shows that the acidification damage in all watersheds has significantly declined. In the recent years the differences in damage of invertebrate communities, based on the acidification score, have been smaller between the investigated rivers.

The biological recovery has stagnated during the last ten years. This must be considered natural, as sulphur emissions in Europe shows the same trends.

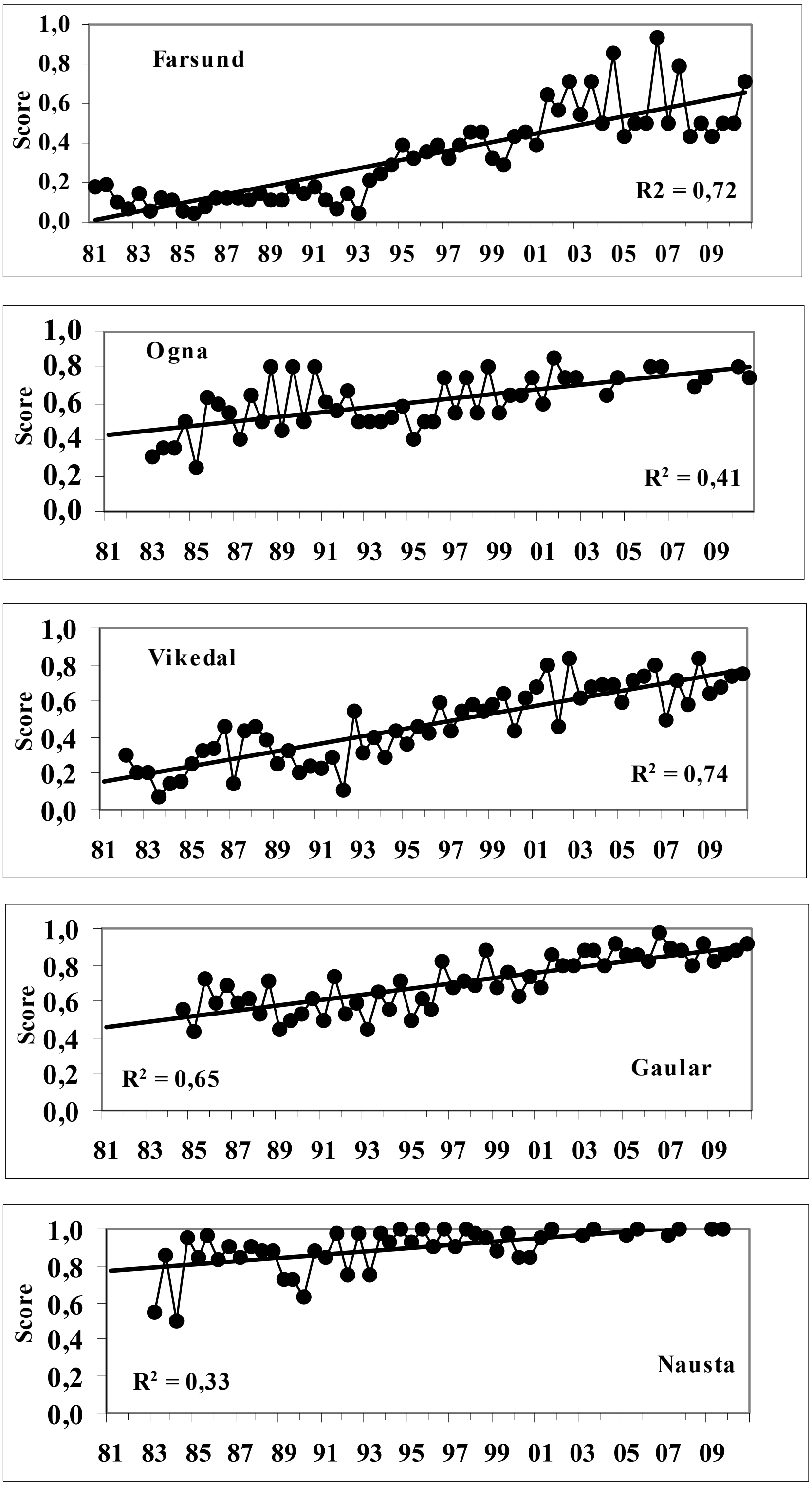

***Figure 24.**Acidification score (see text for explanation) for invertebrates in the monitored rivers ranged from south (top) to north (bottom). The score is described by Fjellheim and Raddum (1990).*

The fauna in the localities that are subject to this study have responded to the improved water quality with increased species diversity. The succession is mainly due to colonisation by acid-sensitive species. In the following we will give some examples.
The development of sensitive taxa in the Farsund watershed has been very positive since 1990. In 2010 we found nine sensitive taxa in the watershed compared to three in 1990. The increased biological diversity is shown by an increased EPT diversity (***Figure 25***). This EPT index is calculated by the sum of species within the insect groups Ephemeroptera (mayflies), Plecoptera (stoneflies) and Trichoptera (caddisflies).

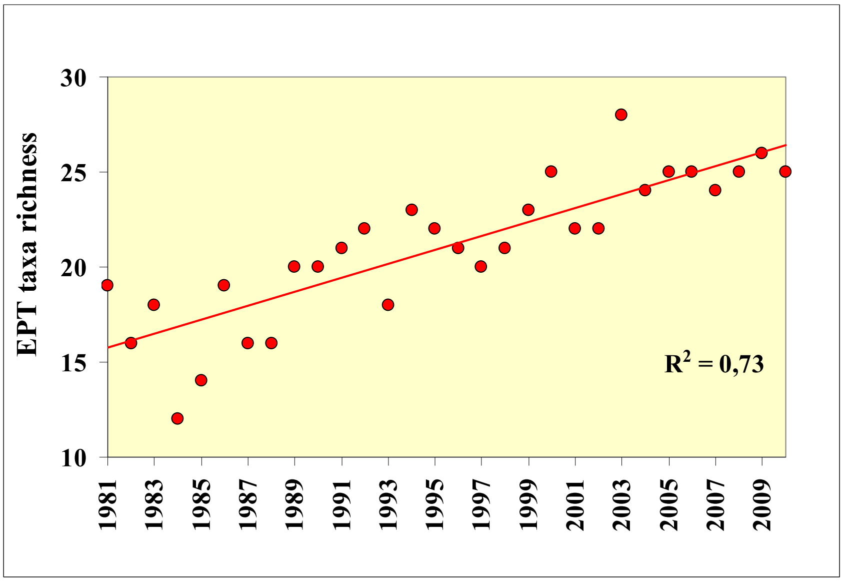

Figure 25. *Development of the EPT taxa richness in the Farsund area.*

The caddisflies *Hydropsyche siltalai* and *Wormaldia sp.* are examples of sensitive species that colonised running water localities at Farsund (***Figure 26***). Their response seems to be closely related to the substantial improvement in water quality after 1996, with increased pH and reduced concentrations of toxic inorganic aluminium (Hesthagen *et al.* 2011). Regional assessment shows that the critical pH limit for *H. siltalai* in oligotrophic waters in southern Norway lies between 5.0 and 5.5 (Fjellheim and Raddum, 1990; Larsen *et al.*, 1996). Typically, the species was absent from large areas in southernmost Norway in the 1980s (Andersen *et al.*, 1990). After 1990, the species has reappeared in this region, which is both due to a general reduction in sulphur deposition as well as the liming of most of the large salmon rivers in the region (cf. Hesthagen *et al.*, 2011).

The more acid-sensitive mayfly *Baetis rhodani* shows, by contrast, an unstable recovery process. After reappearing in 1995, the species was periodically absent in 1998, 1999, 2000, 2005 and 2008. Periodical absence is probably caused by acidic episodes. These may be caused by seasalt episodes (Raddum *et al.* 2007). The water quality in Farsund is still too unstable to permit a permanent establishment of this species. Moderately sensitive species, however, show stable populations.

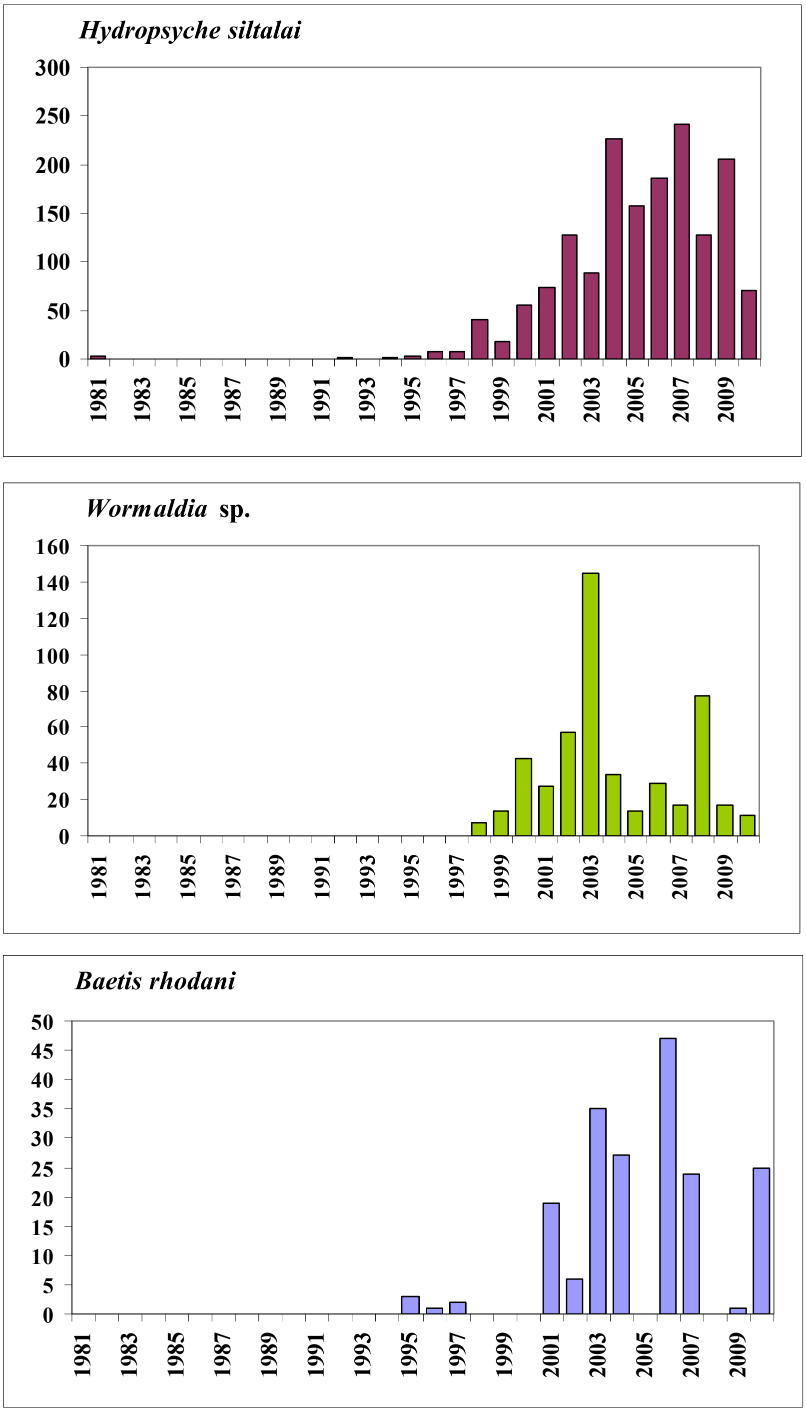

Figure 26*. Total number of the caddisflies Hydropsyche siltalai and Wormaldia sp. and the mayfly Baetis rhodani in the Farsund area during 1981-2010.*

The upper, unlimed part, of River Vikedal has to some extent recovered. The watershed must still be considered affected by acidification and several normally abundant benthic animal species are still missing in many localities. The biological diversity of the localities will increase if water quality improves. The situation in recent years shows a positive development with a reduced acidification damage and increased biological diversity (***Figure 27***).

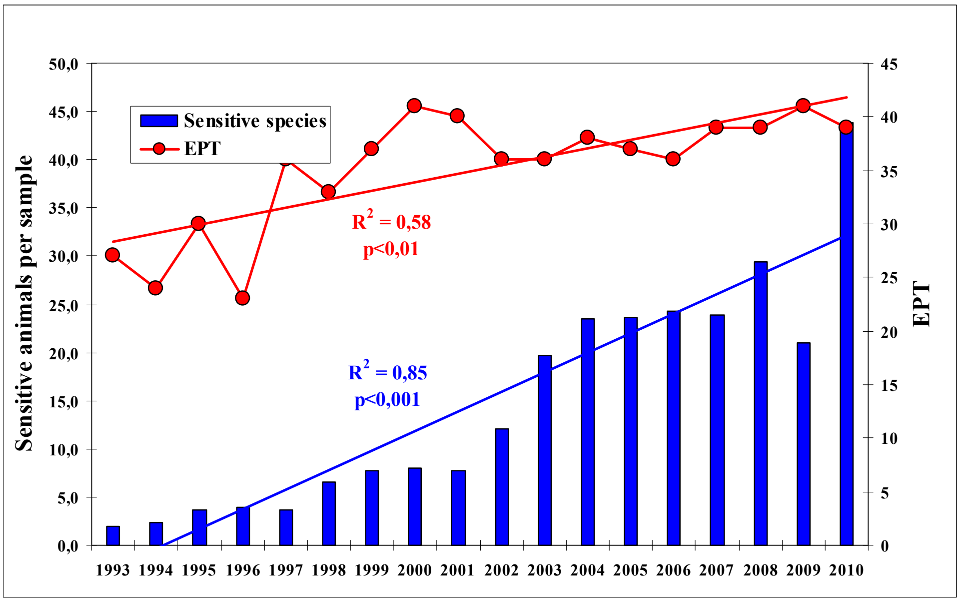

Figure 27. *Number of sensitive animals per sample and EPT taxa richness in the upper unlimed part of River Vikedal in the period 1993 - 2010.*

The lower part of the unlimed River Vikedal has shown a marked recovery during the monitoring period. The development of the strongly sensitive mayfly *B. rhodani* is an example of this (***Figure 28***). Here the species was recorded only sporadically in the period 1982 to 1994. After 1995, the population of the species grew significantly ($p<0.001$). Today the population of this species is stable.

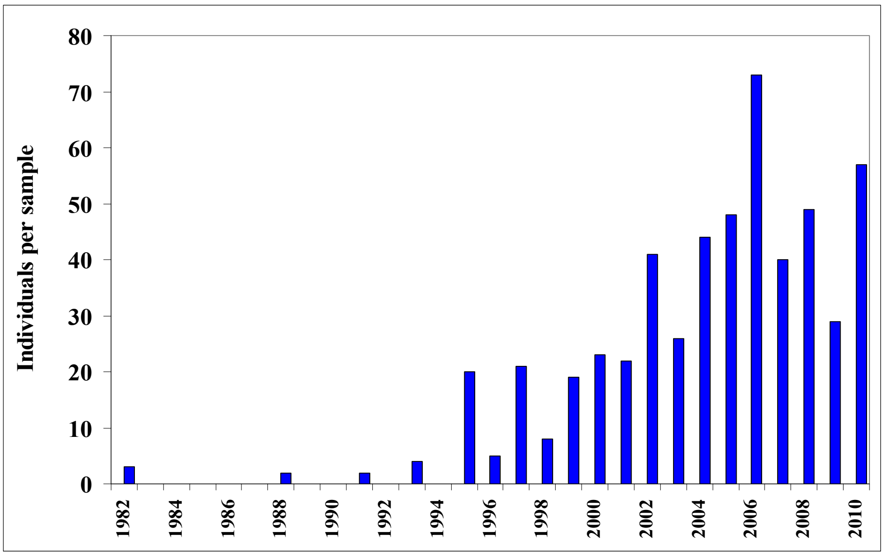

Figure 28*. Numbers of the acid-sensitive mayfly Baetis rhodani in an unlimed locality in the main River Vikedalselva during 1982-2010.*

The faunal succession in formerly acidified rivers in South Norway is marked, but has not yet reached an endpoint. We expect future faunal assemblages to be more diverse, as found in nearby limed rivers. The immigration and succession of animals is dependent of many factors, both biological and abiotic (Raddum and Fjellheim, 2002, 2003). Especially slow colonisers, like many species of snails are dependent on a stable water quality over time.

The positive development of the communities of acid-sensitive animals is related to reduced air pollution resulting from the international agreements from 1979 and onwards. Thus the changed benthic invertebrate communities in formerly strongly acidified regions of South Norway represent one of many positive signals to society that reduced emissions of pollutants are favourable to nature.

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4.6 Sweden: Tracing recovery under changing climate; response of phytoplankton and invertebrate assemblages to decreased acidification

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From:
Johnson R.K. and D.G. Angeler. 2010. Tracing recovery under changing climate: response of phytoplankton and invertebrate assemblages to decreased acidification. Journal of North American Benthological Society, 29: 1472-1490.

Introduction

Acidification of inland waters has been recognized as a major environmental problem for more than three decades, and increasing awareness that emissions from burning of fossil fuels were causing biodiversity loss in lakes and streams led to international action plans to protect and restore natural resources (Stoddard *et al.* 1999). Subsequent reductions in the emissions of sulphur dioxide and NO_x compounds resulted in improved air quality, and increases in surface water pH and alkalinity have been attributed to the decreased deposition of these acidifying compounds (e.g., Stoddard *et al.* 1999, Skjelkvåle *et al.* 2005). However, whilst a growing body of literature supports chemical recovery of surface waters, empirical evidence of biological recovery is scarce and findings are equivocal (Skjelkvåle *et al.* 2003, Stendera and Johnson 2008, Ormerod and Durance 2009). Furthermore, because biological recovery is often the ultimate goal of legislative action, failure to achieve biological objectives means that acidification is still considered a foremost problem affecting the biodiversity of inland surface waters in northern Europe (Johnson *et al.* 2003) and elsewhere (e.g., Monteith *et al.* 2005, Kowalik *et al.* 2007, Burns *et al.* 2008).

Water chemistry, phytoplankton, and littoral benthic invertebrate assemblage data from four minimally disturbed (reference) and four acidified lakes studied during the last two decades were used to track recovery pathways from acidification. We were interested in studying 1) chemical and biological recovery of acidified lakes as indicated by increased pH and diversity (e.g., biological assemblages of acidified lakes were expected to be more dissimilar to those of reference sites at the start than at the end of the study period, and dissimilarity decreases with time), 2) trajectories of biological recovery between assemblages, 3) long-term assemblage trends, indicated by between-year shifts in assemblage composition (measured as Euclidean distance), and their relation to decreased acidity, and 4) the footprints of climatic variability in recovery from acidification.

Methods

In the late 1980s, Sweden initiated long-term monitoring of multiple habitats and trophic levels of lakes to follow the effects of acidification and recovery of regionally representative lake ecosystems (Johnson 1999). Two lake categories were monitored: 1) minimally disturbed reference lakes (sensu Stoddard *et al.* 2006) to determine natural, interannual changes and 2) acidified lakes (defined as exceedence of critical load of $S > 0$) to assess the degree of degradation and subsequent recovery. Samples for analysis of water chemistry and phytoplankton and littoral invertebrate assemblages have been collected during the last 21 years (1988–2008) from eight lakes in the boreonemoral or mixed forest ecoregion of southern Sweden. Four lakes with a mean annual minimum $pH > 6.2$ were considered minimally disturbed reference lakes (Allgjuttern, Fräcksjön, Stora Envättern, Stora Skärsjön), and four lakes with mean annual maximum $pH < 5.8$ (Brunnsjön, Härsvatten, Övre Skärsjön, Rotehogstjärnen) were judged as being acidified based on estimates of exchange of preindustrial alkalinity for anthropogenic SO_4^{2-} (Persson 1996). All eight lakes are relatively small (mean lake surface area = 0.47 ± 0.52 km^2), shallow (mean depth = 5.8 ± 2.5 m), and nutrient poor (mean annual PO_4-P concentration < 6.5 µg/L). Catchment land use/cover also is similar between reference and acidified lakes (% forest: mean = 66%, range = 54–85%; % mire mean = 11%, range = 4–20%). More information on these lakes is available from: http://www.ma.slu.se.

Surface water samples (0.5 m) were collected six to eight times a year (monthly during the open-water phase and once in February) at a mid-lake station in each lake, and analysed for variables indicative of acidity (pH and SO_4^{2-} concentration), nutrients (PO_4-P), and water color (absorbance 420 nm of filtered water). All physicochemical analyses were done at the Department of Aquatic Sciences and Assessment following international (ISO) or European (EN) standards when available (Wilander *et al.* 2003). Annual mean values were used in statistical analyses to downweigh seasonal differences. *Phytoplankton* was sampled in August of each year by taking a water sample from the epilimnion (0–4 m) with a Plexiglas® tube sampler (diameter = 3 cm), and a subsample was taken and preserved with Lugol's Iodine solution supplemented with acetic acid. Phytoplankton counts were made using an inverted light microscope and the modified Utermöhl technique, and taxa were identified to the lowest taxonomic unit possible (usually species), and species-specific biovolume measures were calculated from geometric formulae. In 1992, enumeration of phytoplankton changed from counts of predominating taxa to counts of all species in a sample; therefore, phytoplankton counts made before 1992 are not included here. *Benthic invertebrates* were collected from wind-exposed, vegetation-free littoral habitats in late autumn (October–November) each year. Five replicate samples were taken by standardized kick sampling with a hand net (0.5-mm mesh size). Samples were preserved in 70% ethanol in the field and processed in the laboratory by sorting against a white background with 10× magnification. Invertebrates were identified to the lowest taxonomic unit possible (generally to species level) and counted with dissecting and light microscopes.

Results and Discussion

Trends in water chemistry - Acidified lakes had lower pH than reference lakes (Wilcoxon signed-rank test, $p< 0.001$). Mean annual pH was 5.3 ± 0.41 for the four acidified compared to 6.6 ± 0.17 for the four reference lakes. Mean surface water pH increased significantly with time in five of the eight lakes during the 21-year period. However, acidic episodes occurred during the recovery period. Annual minimum pH was < 5.0 (mean: 4.4 ± 0.41) in the acidified lakes compared to > 6.0 (mean: 6.2 ± 0.17) for the reference lakes. Significant decreases in SO_4 concentration were noted for all eight of the lakes. Likewise, increasing trends in lake water color were noted for all eight of the lakes, with acidified lakes having higher water color. No temporal trends or differences were noted between acidified and reference lakes for PO_4-P concentration.

Trends in phytoplankton and littoral benthic invertebrate assemblages - Assemblage composition (species presence–absence data) but not dominance patterns (invertebrate abundance and phytoplankton biovolume) of acidified lakes became more similar to those of reference lakes (distance decreased with time), indicating that detection of recovery varies as a function of chosen metrics. Moreover, acidified lakes had more pronounced shifts in assemblage composition than did reference lakes. Mean between-year shifts (Euclidean distance) of assemblage composition supported the idea that changes were stronger in acidified than in reference lakes. Changes in phytoplankton and littoral invertebrate assemblage composition were significant when measured with Sørensen similarity (presence–absence data) (Wilcoxon signed-rank test, $p< 0.05$), but differences were not significant when biovolume or abundance data were used (Bray–Curtis dissimilarity). The strongest differences were noted for phytoplankton assemblages; acidified lakes had a mean Euclidean distance of almost twice that of reference lakes (acidified: 0.037 ± 0.039, reference: 0.019 ± 0.016; Sørensen similarity). Between-year shifts in littoral invertebrate assemblages were 0.035 ± 0.024 for acidified and 0.027 ± 0.025 for reference lakes (Sørensen similarity).

Patterns of biological recovery - Recovery patterns of phytoplankton and invertebrates were highly variable among acidified lakes. Regarding phytoplankton, species evenness and biovolume-based assemblage composition (NMDS 1, Bray–Curtis) showed the most consistent recovery for Brunnsjön and Övre Skärsjön. These metrics were either within or close to the 95% confidence interval reflecting target conditions in neutral lakes. In contrast, phytoplankton in lake Rotehogstjärnen was most resistant to recovery. Species richness, biovolume, Simpson diversity, and species composition (NMDS, Sørensen) showed different degrees of variability approaching reference conditions. As a result of this strong variability between and idiosyncratic responses within lakes, most comparisons in an ANOVA analysis were not significant, except the significant temporal variation detected for biovolume-based assemblage composition (NMDS 1, Bray Curtis). None of the metrics had a significant treatment x year interaction, i.e., no consistent recovery occurred across all acidified lakes.

Regarding the invertebrates, better recovery responses were observed compared with phytoplankton. Most structural metrics indicated that acidified lakes were within or close to the 95% confidence interval representing reference conditions. Brunnsjön and Övre Skärsjön seemed to have a better recovery response, whereas Härsvatten had a poorer response. Both composition- and abundance-based assemblage similarity showed variable degrees of approaching assemblage structure in reference lakes.

Environmental drivers - High between-year shifts in phytoplankton assemblage composition were negatively correlated with temperature, water color, and pH. Likewise, interannual shifts in invertebrate assemblage composition were negatively correlated with temperature and pH. In addition, $NO_2 + NO_3$ concentration and the NAO_{winter} index were significantly correlated with the temporal turnover of benthic invertebrate assemblages. Hence, while trends in water chemistry showed unequivocal recovery, responses of phytoplankton and invertebrate assemblages, measured as between-year shifts in assemblage composition, were correlated with interannual variability in climate (e.g., North Atlantic Oscillation, [NAO], water temperature) in addition to decreased acidity (***Table 14***) The NAO is an important regional-level driver of climate in southern Sweden, with positive NAO values signifying higher precipitation and, thus, more variable hydraulic and chemical conditions. Correlation of between-year Euclidean distance of invertebrate assemblages and NAO_{winter} index values were significant for acidified lakes, and greater between-year distance of invertebrate assemblages was associated with warmer winters (i.e., positive NAO values). This finding implies that recovery might be offset by effects of interannual, climate-driven changes on local lake variables, lending support to our 4th hypothesis (climatic effects subsume the signal of biological recovery). In addition to climate-mediated imprints on recovery, a further plausible explanation for the marked interannual variability in assemblage structure is that acidic episodes can inflict recurrent stress on lake assemblages. This conjecture is supported by data showing that annual mean pH was < 5.5 during several of the 21 years of study in the acidified lakes.

Table 14. *Spearman correlation (ρ) of between-year Euclidean distance of phytoplankton and littoral invertebrate assemblages in 4 acidified and 4 reference lakes and selected water chemistry variables (annual means between 1988 and 2008) and the North Atlantic Oscillation winter index (NAO_{winter}). Sørensen similarity and Bray–Curtis dissimilarity were used in a nonmetric multidimensional scaling ordination. * = $p < 0.05$, ** = $p < 0.01$, *** = $p < 0.001$*

Variable	Phytoplankton		Invertebrates	
	Sørensen similarity	Bray–Curtis dissimilarity	Sørensen similarity	Bray–Curtis dissimilarity
Temperature	–0.336***	0.094	–0.249**	–0.027
pH	–0.318***	–0.076	–0.281***	–0.112
NO_2+NO_3 (µg/L)	0.044	0.167	0.109	–0.167*
PO4-P (µg/L)	0.035	0.015	0.061	–0.015
Water color	0.057	–0.307***	–0.069	0.065
$NAO_{winter\ index}$	0.115	0.073	0.249**	–0.055

Clear shifts in lake assemblages of phytoplankton (Findlay 2003), zooplankton (Frost *et al.* 2006), and epilithic algae (Battarbee *et al.* 1988, but see Vinebrooke *et al.* 2003) have been related to decreased acidity, whereas results for other groups, such as fish and benthic invertebrate assemblages, have been more equivocal (Burns *et al.* 2008, Stendera and Johnson 2008). Our study further highlights the complexity of biological recovery responses to acidification. The finding that recovery pathways and trajectories of individual acidified lakes and the environmental drivers explaining these changes differed among assemblages shows that biological recovery is complex and the influence of climatic variability is poorly understood. This complexity is reflected in strongly idiosyncratic changes over time as a function of the response variable chosen to track change for both assemblages.

Although our results are of general interest concerning recovery of ecosystems from disturbance, many of our findings are particularly relevant to lake management. Our finding that phytoplankton and littoral invertebrate assemblages responded differently to improved water quality among acidified lakes adds to the growing body of literature underpinning the use of multiple groups of organisms when a priori knowledge of response (recovery) signatures is poor (Stendera and Johnson 2008; Johnson and Hering 2009). Use of phytoplankton or invertebrate assemblages alone would have led us to a biased conclusion regarding biological recovery. Not only choice of taxonomic group, but also the choice of the response variable influenced our ability to detect recovery. Assemblage composition (NMDS ordination) was powerful for detecting changes, but the dissimilarity measure used (whether based on presence–absence or biovolume/abundance) influenced the results. Finally, one of our most interesting findings was the congruence of trends in water chemistry and biology in acidified and reference lakes. Our results show the influence of factors other than decreased acidity, such as the underlying importance of climate, as drivers of recovery trajectories and pathways.

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4.7 Switzerland: Recovery of macroinvertebrates in acid-sensitive freshwaters in southern Switzerland

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Introduction

Precipitation in southern Switzerland is mainly determined by warm, humid air masses originating from the Mediterranean Sea, passing over the Po Plain, getting enriched with pollutants, and colliding with the Alps. Because of its particular lithology (base-poor rocks especially gneiss) and high altitudes (thin soil layer) the buffer capacity of lakes southern Switzerland is low and is therefore very sensitive to acidification.

In order to study the impact of acid deposition on acid-sensitive freshwaters, the water quality of wet deposition and of 20 alpine mountain lakes have been monitored since the 1980's. After the year 2000, 3 rivers were also sampled regularly. Chemical recovery as a consequence of reduced sulphur emissions has been discussed in detail by Steingruber and Colombo (2006) and Steingruber and Colombo (2010). Since biological recovery is the ultimate goal of emission reduction, sampling of macroinvertebrates has been added to the monitoring programme in the year 2000. The analyses of its trends are here presented for the first time.

Study Site

The study area is located in the Central Alps in the northern part of canton Ticino, southern Switzerland. In order to study the influence of transboundary air pollution, the freshwater systems were selected in remote areas far from local pollution sources. Biological monitoring occurred in 4 alpine mountain lakes and 3 rivers. The lake watersheds are mainly constituted by bare rocks with vegetation often confined to small areas of Alpine meadow. The lakes are situated between an altitude of 1690 m and 2130 m and are characterized by intensive irradiation, a short vegetation period, a long period of ice coverage (usually from November to June) and by low nutrient concentrations. Laghetto Superiore has an outflowing stream that connects with Laghetto Inferiore. Laghetto Starlarescio, being very shallow, has wetland characteristics. The sampling points of the rivers are located at lower altitudes (610-918 m), implying larger catchment areas and therefore less sensitivity towards acidification than lakes. The main geographic, morphometric and geologic parameters of the monitored lakes and rivers are shown in ***Table 15***.

***Table 15**. Lake and river parameters*

Type	Lake name	Longitude	Latitude	Altitude m a.s.l.	Catchment area km^2	Lake area ha	Max depth m
Lake	Lago del Starlaresc da Sgiof (Starlarescio)	8°46’25’’	46°16’26’’	1875	0.23	1.1	6
Lake	Lago di Tomè (Tomeo)	8°41’23’’	46°21’47’’	1692	2.94	5.8	38
Lake	Laghetto Inferiore (Inferiore)	8°35’34’’	46°28’34’’	2074	1.82	5.6	33
Lake	Laghetto Superiore (Superiore)	8°35’05’’	46°28’34’’	2128	1.25	8.3	29
River	Maggia	8°38’ 8’’	46°21’16’’	610	189		
River	Vedeggio	8°59’24’’	46°07’45’’	740	20		
River	Verzasca	8°47’33’’	46°21’24’	918	27		

Methods

Lake water was sampled for chemical analysis 3 times a year (beginning of summer, end of summer, autumn). River water was sampled monthly since the year 2000. For detecting trends in water chemistry a seasonal Mann-Kendall test was performed for the period 2000-2010 (Dietz and Kileen, 1981). The two sided test for the null hypothesis that no trend is present was rejected for p-values below 0.05. For the lakes two seasons were chosen: season 1 (June-July) and season 2 (September-November). Estimates for temporal variations in lake and river water chemistry were quantified with the seasonal Kendall slope estimator (Gilbert, 1987). Since the detection limit of aluminium decreased during the monitoring period from 7 µg/l (2000) to 3 µg/l (2004) and finally to 0.2 µg/l (2005), in order to avoid that this improvement leads to an erroneous decreasing aluminium concentration trend, a minimum aluminium concentration of 7 µg/l was used for the data analysis.

Macroinvertebrates have been collected by “kick sampling” according to the ICP Waters Manual (NIVA, 1996). Sampling in rivers Maggia, Vedeggio and Verzasca occurred 4-8 times a year from 2000 to 2009, while in lakes samples were collected from the littoral and the out let 2-3 times per year from 2002 to 2009. Macroinvertebrates were conserved in 70% ethanol and then identified to the lowest possible taxon: Ephemeroptera, Plecoptera, Trichoptera, Heteroptera, Odonata mostly to genus or species and Oligochaeta and Diptera to family. The number of collected individuals increased over the monitored period. In order to avoid that species appearing for the first time during the monitoring period, simply because of the higher probability to find them, are misinterpreted as a result of changing species composition over time, we “downweighted” each sample to the sample size of the first year. In this way individuals that “could” have been detected from the beginning if the sample size were larger, disappear in the data. This procedure is used for all time trend analyses. The number of individuals was then transformed in relative abundances and for rivers averaged for 2 season every year (season 1: February-June, season 2: July-November). Temporal trends in biology were studied by different metrics: the total taxa number, the number of taxa belonging to the orders Ephemeroptera, Plecoptera, Trichoptera (EPT), the number of taxa belonging to the 5 indicator values of acid sensitivity according to Braukmann and Biss (2004) and their relative abundance. Finally a multivariate statistic using Canoco for Windows 4.5 (ter Braak and Smilauer, 2002) was performed. All the taxa numbers in the trend analysis refer to the most inclusive taxa, in order to avoid that the same species appears in more than 1 taxon, confounding the statistics (e.g. individuals of Baetis rhodani, that could not be determined down to species, may appear as Baetis sp.). Abundance data refer to the entire dataset.

To investigate if changes over time in water chemistry are reflected in the macroinvertebrate community, a multivariate analysis was performed as described in Halvorsen *et al.* 2003, First a detrended correspondence analysis (DCA) identified the length of the first axis to be less than 2 standard deviations. Consequently a redundancy analysis (RDA) was selected as most appropriate. The environmental variables used in the analysis were pH, concentrations of calcium (meq/m^3), total alkalinity=alkalinity-acidity (meq/m^3) and soluble aluminium (µg/l). All water chemistry variables except pH and alkalinity in lakes (because of the presence of negative values) were log-transformed. A linear variable called Time ranging from 0 to 19 and 23 in rivers and lakes, respectively, was added. Furthermore a dummy variable called Season was considered. For rivers average values of the period February-June were coded as Spring=1 and average values of the period July-November were coded as Spring=2. For lakes 3 seasons were considered: beginning of summer (season 1), summer (season 2) and autumn (season 3). During the analysis the relative abundances of species of list 1 and 2 were log-transformed to reduce the skewness and heteroscedasticity in the data. Downweighting of rare species was not used. Species were centered and samples centered and standardized. Furthermore the RDA analysis was focused on interspecies correlations and species scores were divided by standard deviation. To evaluate the significance of the RDA analysis a Monte-Carlo permutation test under a reduced model (999 permutations) was performed on both the first axis and the canonical axis combined. First, chemical variables were included as the only environmental variables to test whether changes in water chemistry were reflected in the macroinvertebrate community. Second, only the variable Time was included to test the correlation of the benthic community with time. If both chemistry and time were significant in determining the composition of macroinvertebrates, a RDA analysis was run with both chemical and time variables to calculate the proportion of variation in species composition attributable to the long-term recovery in freshwater chemistry with the formula described by Halvorsen *et al.* (2003). In order to test the influence of seasonality on benthic community the same procedure as described above was used substituting the variable.

Results and Discussion

Chemical parameters used in the multivariate analyses measured in rivers and lakes from year 2000 to 2010 are presented in ***Figure 29***. Note the different scales for rivers and lakes. Having a larger catchment, rivers are better buffered than lakes. pH, calcium and alkalinity were the higher in river Maggia, followed by Vedeggio and Verzasca. Average pH's were 7.4, 7.1, 6.8, average concentrations of calcium 398, 237, 104 (meq/m3) and of alkalinity 284, 152, 63 (meq/m3). Minimum values of pH, concentrations of calcium and alkalinity are characteristics of high discharge occurring mostly in spring (April-June) and sometimes in autumn (September-November). Concentrations of aluminium are mostly close to the detection limit, and high values are limited to single high discharge events. As discussed in Steingruber and Colombo (2006), differences in catchment areas and geology are the main cause for differences in concentrations among rivers. The catchment area of river Maggia is 7 and 10 times larger than the watersheds of river Verzasca and Vedeggio, respectively, implying a longer average water residence time and higher average weathering rate related to increased buffering capacity in the watershed of river Maggia. Differences in water chemistry of rivers Vedeggio and Verzasca are more related to their different catchment geology.

The lakes can be divided in 2 groups by water chemistry: lakes Inferiore and Superiore with pH's mostly above 6 and total alkalinities around 30 meq/m^3 and lakes Starlarescio and Tomeo with pH's always below 6 and total alkalinities around 0 meq/m^3. As a result of low pH values, concentrations of soluble aluminium are high in lakes Starlarescio and Tomeo and mostly close to the detection limit in lakes Inferiore and Superiore. River chemistry depends very much on the hydrology (Steingruber and Colombo, 2006). From year 2003 to 2007 a very dry period caused an increase of average alkalinity, calcium and pH (Steingruber and Colombo, 2010). In Figure 1 an increase of minimum values of pH, calcium and alkalinity can be observed during that period. In lakes the phenomena was less pronounced.

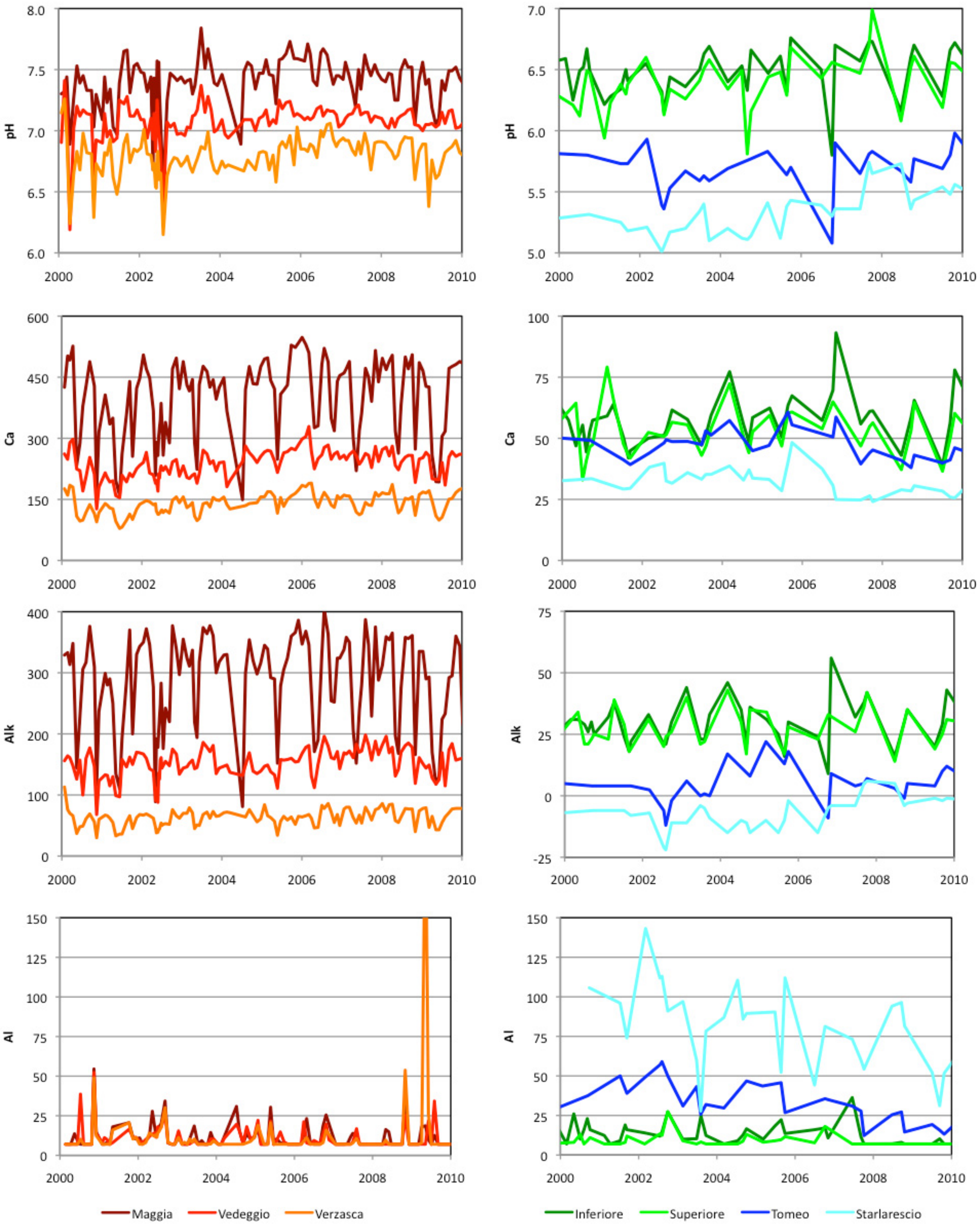

***Figure 29**. pH, calcium (meq/m^3), total alkalinity (meq/m^3) and soluble aluminium (µg/l) in rivers Maggia, Vedeggio, Verzasca and lakes Inferiore, Superiore, Starlarescio, Tomeo.*

A trend analysis for the period 2000 to 2010 revealed significant increasing concentrations of alkalinity in river Verzasca and lakes Superiore and Starlarescio (***Table 16***). pH increased significantly only in lake Starlarescio. Concentrations of aluminium decreased significantly only in lake Tomeo, where concentrations decreased from around 50 µg/l to below 25 µg/l approaching concentrations in lakes Inferiore and Superiore. It is possible that especially for rivers, whose chemistry depends very much on hydrology, the results of the trend may have been "disturbed" by the succession of a dry period (2003-2007) after a wet period (1998-2003).

Table 16. *Results from trend analyses (significant trends in red) during the period 2000-2010. p corresponds to the probability level obtained with the seasonal Mann-Kendall test and the rate (meq* m^{-3} yr^{-1} *for anions and cations, mg* l^{-1} yr^{-1} *for* Al_{sol}*) with the seasonal Kendall slope estimator.*

Type	**Name**	**pH**		**Ca^{2+}**		**Alkalinity**		**Al_{sol}**	
		p	rate	p	rate	p	rate	p	rate
River	Maggia	0.661	0.00	0.234	4.18	0.799	0.02	0.159	0.00
River	Vedeggio	0.307	0.00	0.059	3.58	0.061	2.21	0.057	0.00
River	Verzasca	0.371	0.00	0.011	3.00	0.017	1.21	0.424	0.00
Lake	Inferiore	0.323	0.01	0.429	0.41	0.173	0.33	0.068	-0.28
Lake	Superiore	0.299	0.02	0.560	0.40	0.036	0.56	0.070	-0.25
Lake	Starlarescio	0.024	0.04	0.081	-0.56	0.044	1.14	0.078	-3.93
Lake	Tomeo	0.546	0.01	0.255	-0.46	0.290	0.49	0.012	-3.06

According to Braukmann and Biss (2004) in rivers with pH's mostly above 6.5, often around 7 and not decreasing below 6, acid-sensitive species should be present. If acidic episodes occur but pH does not fall below 5.5, acid-sensitive species start to disappear and moderate acid-sensitive species become more important. Chemical analysis of river waters suggests that in river Maggia and Vedeggio after year 2000, acidification should not have been a problem for benthic community, and therefore it is expected to find acid-sensitive species. pH in river Verzasca never decrease below 6, but being more acidified than the other two rivers, during acidic episodes pH may have dropped further, so that acid-sensitive species may be found less frequently. Rivers Maggia, Vedeggio and Verzasca were already quite well buffered from the beginning of the monitoring period, and therefore it is not very likely to find a biological recovery.

In lakes Inferiore and Superiore it is expected that acid-sensitive species are missing (pH around 6.5-7.0, rarely below 5.5) favoring moderate acid-sensitive species. Lake Tomeo falls in the next category (pH 6.5-5.5, sometimes below) with the presence of acid-tolerant species. In lake Starlarescio, at the beginning of the monitoring, even acid-tolerant species may have disappeared, and only acid-resistant and very acid-resistant organisms are likely to have been present. Acid-resistant species may survive at pH around 5.5, sometimes below 5.0-4.3. From the temporal variations of pH and aluminium no great change of benthic community in lakes Inferiore and Superiore is expected. Although the variations of pH in lake Tomeo do not suggest the reappearance of moderately sensitive species, the significant decrease in concentrations of aluminium may have improved conditions for biology. Also the improvement of chemistry in lake Starlarescio is insufficient for a shift to the next higher acidification class of Braukmann and Biss (2004), but some signs of improvement may also be observed in the composition of benthic community.

Invertebrate communities differ between rivers and lakes and among them, as seen by the average abundances of the main macroinvertebrate groups and the number of identified taxa during the whole monitoring period (only most inclusive species considered) (***Table 17***). Rivers are mainly inhabited by Coleoptera, Diptera, Ephemeroptera and Plecoptera. The out lets of lakes Inferiore and Superiore are dominated Oligochaeta, Diptera and Plecoptera. In the out lets of lakes Tomeo and Starlarescio, Diptera and Plecoptera and Diptera dominate, respectively. . At all sites Diptera are mainly represented by Chironomidae (data not shown). There is a progressive disappearance of first Coleoptera, Ephemeroptera and then Plecoptera with increasing pH. The taxa number also decreased along the pH gradient. However, differences in biology between rivers and lake out lets are not only due to their different chemistry. In fact, because of the high altitudes and therefore extreme physical-chemical conditions the population of macroinvertebrates in Alpine lakes is expected to be generally poor (Fjellheim *et al.*, 2000; Hieber, 2002; Marchetto *et al.*, 2004). Littoral sites are dominated by Diptera followed by Oligochaeta. Because Diptera and Oligochaeta were determined only to the family level, most individuals in littoral samples where therefore not identified to genus and/or species making a trend analysis of the littoral dataset less interesting. Data from littoral sites are therefore omitted in further analysis.

Table 17. *Average relative abundances of the main macroinvertebrate groups and the taxa number (only most inclusive taxa) during the period 2000-2010. Values were derived from the original dataset (not downweighted).*

	Rivers			Lakes							
	Maggia	Vedeggio	Verzasca	Inferiore		Superiore		Starlarescio		Tomeo	
Site				Out let	Littoral	Out let	Littoral	Out let	Littoral	Out let	Littoral
Oligochaeta	1%	1%	1%	27%	18%	31%	20%	2%	11%	1%	25%
Coleoptera	9%	10%	19%	0%	1%	0%	1%	0%	0%	1%	1%
Diptera	29%	21%	17%	46%	71%	44%	60%	83%	79%	61%	60%
Ephemeroptera	32%	27%	41%	1%	0%	3%	0%	0%	0%	0%	0%
Plecoptera	21%	30%	18%	16%	2%	18%	4%	5%	0%	32%	1%
Trichoptera	3%	9%	2%	2%	1%	2%	2%	2%	1%	2%	7%
Other	4%	2%	2%	9%	7%	3%	13%	8%	10%	3%	6%
Identified taxa	94	111	82	39	37	37	32	35	20	33	29

The number of taxa and the number of EPT taxa through time is shown in ***Figure 30***. During the last 10 years the number of taxa in rivers was fairly constant. The number of EPT taxa decreased after year 2004 in rivers Vedeggio and Verzasca and after year 2007 in river Maggia. In lake out lets the trends is less clear, but the taxa number seemed to have decreased in lakes Inferiore and Tomeo after year 2004 and the number of EPT taxa in all lakes. The number of taxa and the number of EPT taxa do not suggest a recovery in rivers nor in lakes.

A more detailed analysis of indicator organisms of the acid status of river and lake out lets is presented in ***Figure 31*** and ***Figure 32***. For each river and lake out let the temporal evolution of the number of acid-sensitive taxa (Indicator 1), moderate acid-sensitive taxa (Indicator 2), acid-tolerant taxa (Indicator 3), acid-resistant taxa (Indicator 4), very acid-resistant taxa (Indicator 5) according to the classification list of Braukmann and Biss (2004) and their relative abundances are presented. In all rivers, although acid-sensitive species are present, moderately sensitive species prevail in number. Their relative abundance dominates compared to the other indicator organisms. From the chemical data it is therefore expected a slightly better situation for biology (presence of more acid-sensitive species). During the last 10 years the number of moderately sensitive taxa decreased in river Maggia, increased during the dry years (2003-2007) and then decreased in river Vedeggio and remained constant in river Verzasca. The number of the other indicator organisms remained also constant over time. The abundance of the moderately sensitive taxa decreased in river Maggia. The reason was the decrease in abundance of Ecdyonurus helveticus-Gr. In river Vedeggio and Verzasca the abundance of moderately sensitive and acid-tolerant taxa decreased. This result is caused by a decrease in the abundance of identified Baetis alpinus and an increase of Baetis sp. indicating a problem in identifying this species, that has to be resolved. In lakes Inferiore and Superiore, as expected from pH, moderate acid-sensitive taxa are occasionally present. In the more acidic lakes Starlarescio and Tomeo only acid-resistant and very acid-resistant species were identified. The expected acid-resistant taxa were not found in lake Tomeo. In all lakes very acid resistant taxa have the highest abundance and it seems to decrease over time. This decrease was caused by Nemoura mortoni in lake Inferiore, by Nemoura mortoni and Leuctra sp. in lake Superiore and by Leuctra sp in lake Starlarescio. With exception of the decrease in abundance of acid very resistant species in all lakes, which may be a sign of recovery, the study of indicator organisms did not indicate improvement of biology in the studied rivers and lakes.

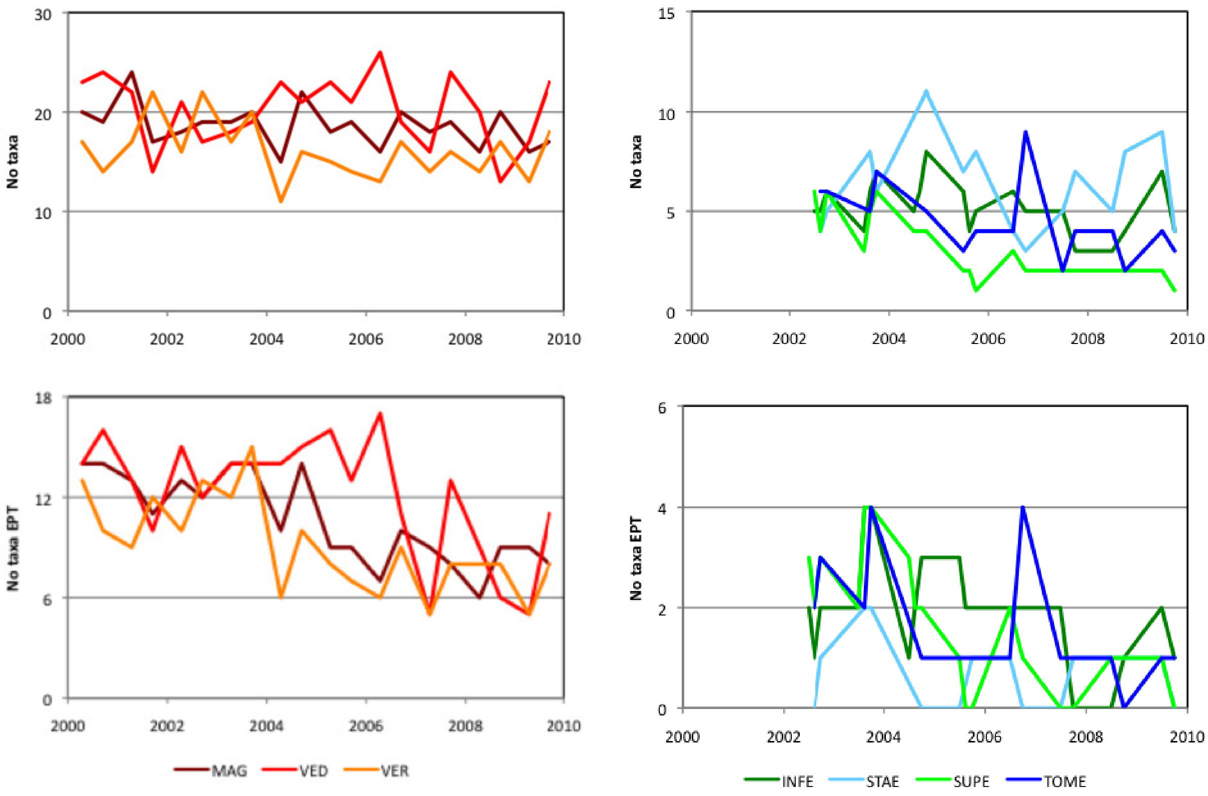

Figure 30. *Number of taxa and number of taxa belonging to the orders Ephemeroptera, Plecoptera, Trichoptera in rivers and outlets of lakes.*

Interestingly, the RDA analysis showed significant time trends for the biology of all lakes, but only in river Vedeggio and lakes Inferiore and Tomeo has the biological community changed also significantly with chemistry. From the $\sum\lambda$ values reported in ***Table 18*** and the formula proposed by Halvorsen *et al.* (2003), the variation of species that can be explained by a change of water chemistry with time can be derived: 11% in river Vedeggio, 5% in lake Inferiore and 39% in lake Tomeo, respectively. However, for river Vedeggio and lake Inferiore a seasonal variation of chemistry explain 11% and 9% of the variation respectively, in the macroinvertebrate structure indicating that seasonal variation may “disturb” temporal variations in benthic community.

A correlation analysis between environmental variables indicate that in river Vedeggio Time is significantly correlated with calcium (0.508) and alkalinity (0.479) and Season with aluminium (0.590) and alkalinity (0.448). The increase in aluminium during summer/autumn may be caused by washout/erosion during storm events, while the reason for increased calcium during time may be increased weathering, or the result of the increased number of alkaline rain events during the wet years (1998-2002) or simply because of less dilution during the dry years 2003-2007. The RDA analysis showed that the abundance of Enchitraeidae and Empididae increased with time while Limoniidae, Ecdyonurus helveticus-Gr, Rhithrogena sp., Perla sp. decreased with time. From these taxa only Empididae seem to correlate significantly with calcium. The absence of an increase in the abundance of acid-sensitive species in river Vedeggio with time suggests that the observed temporal change in benthic community explained by chemistry may not be caused by chemical recovery in the sense of increased pH and alkalinity.

In lake Inferiore only aluminium correlates with time (-0.494). pH, calcium and alkalinity correlates with season (0.413, 0.719, 0.592, respectively) being lower at the beginning of summer immediately after snow melt compared to autumn values. The RDA analysis showed that Enchitraeidae and Chironomidae increased with time, while Nemoura mortoni decreased. From these species only the latter was correlated with aluminium. Their abundance decreased significantly with decreasing aluminium concentrations. Nemoura mortoni is an acid-resistant species, its decrease may indicate a beginning recovery; however concentrations of aluminium in lake Inferiore were already low at the beginning of the monitoring period so that the decrease in the abundance of Nemoura mortoni may be related to something else.

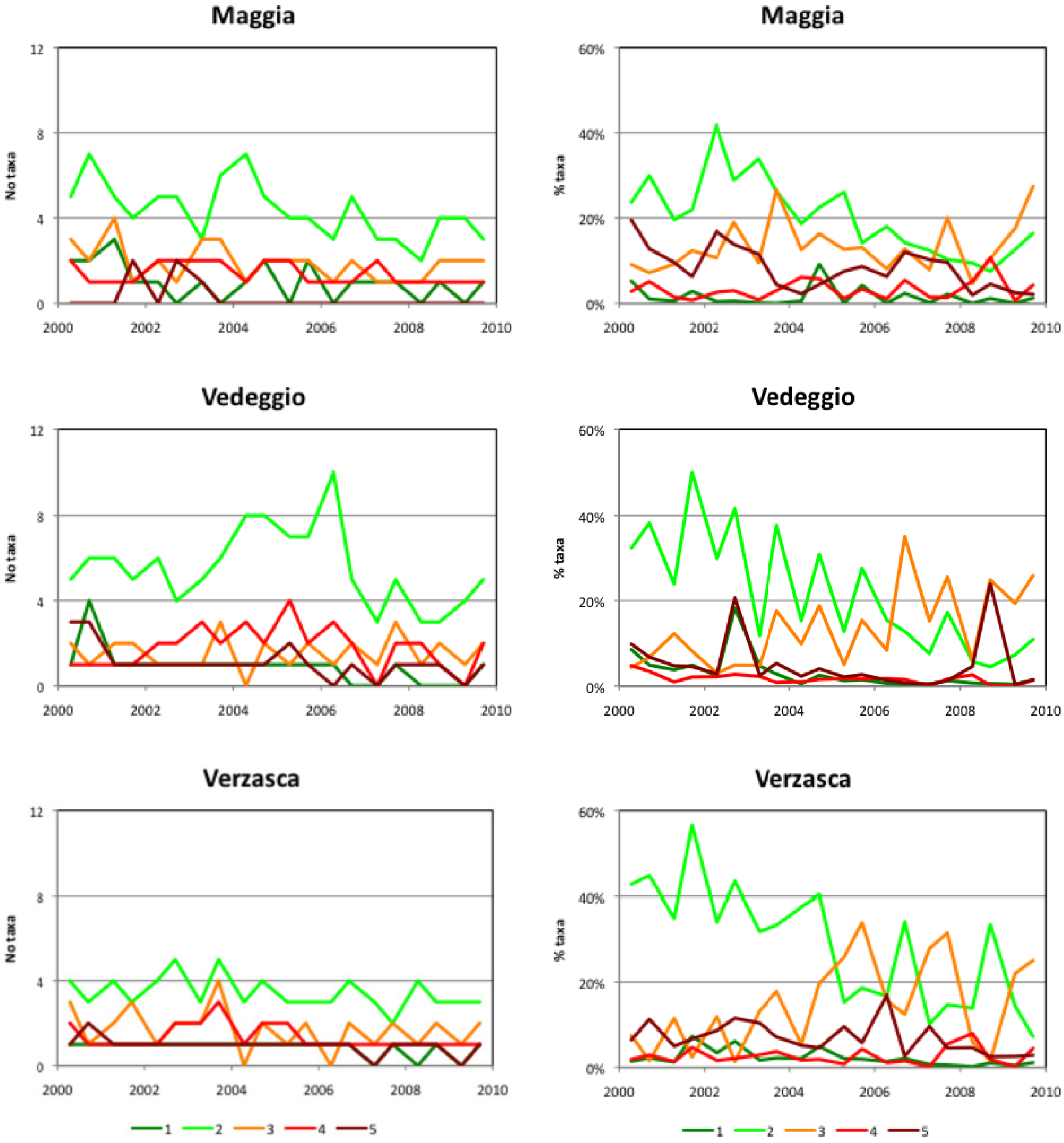

Figure 31. *Number of indicator organisms by class according to Braukmann and Biss (2004) and their relative abundances in rivers.*

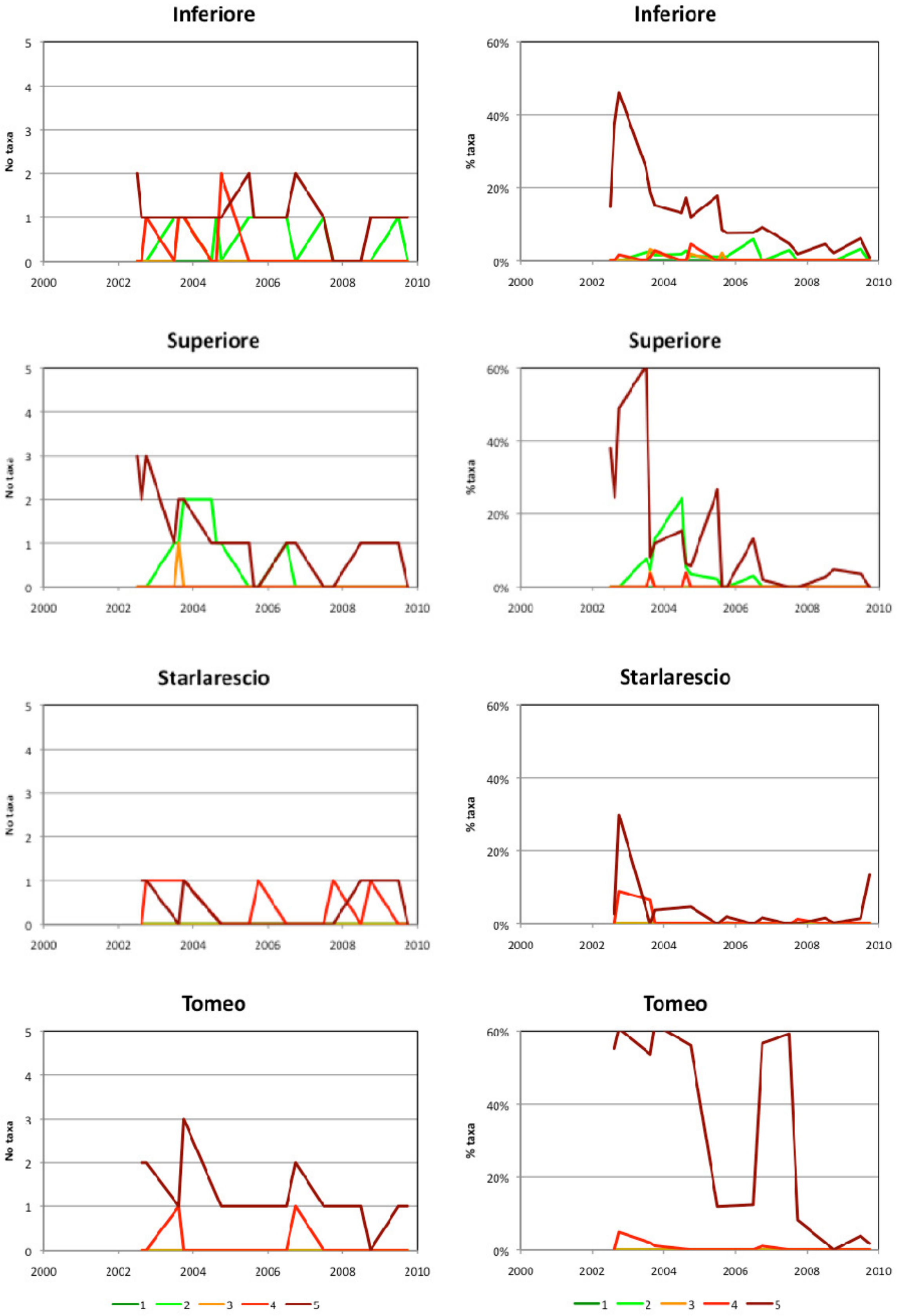

Figure 32. Number of indicator organisms by class according to Braukmann and Biss (2004) and their relative abundances in lake outlets.

In lake Tomeo pH and alkalinity were positively (0.653, 0.498, respectively) and calcium and aluminium negatively correlated with time (-0.598, -0.780, respectively). The RDA analysis of species variation indicated significant increase of Chironomidae and decrease of Leuctra sp. with time. The first being mainly negatively correlated with alkalinity and the second mainly positively correlated with calcium. No species correlated significantly with pH or aluminium. Similarly to what observed for lake Inferiore the decrease of the very acid resistant taxon Leuctra sp. can indicate an improvement of biology, but since at the same time no less tolerant taxa seem to appear or increase their abundance, it is difficult to ascribe this to recovery.

The RDA analysis showed significant temporal changes in benthic community at all sampling sites, in river Vedeggio and lakes Inferiore and Starlarescio also connected with chemical variations, but a clear sign for recovery was not found. It is possible that the time trends observed in lakes are the result of five years of poor rain, with decreased discharge in the outlet favoring more littoral lake species such as Oligochaeta and Diptera. Also in the other lakes Oligochaeta and Diptera tended to increase and Plecoptera to decrease. The same could have happened in rivers as well.

Table 18. *Results from the RDA analysis for the different environmental variabels. $\sum\lambda$ is the sum of all canonical eigenvalues, p the significance valu,e and n.s.stands for not significant.*

Type	Name	Chemistry		Time		Season		Chemistry+Time		Chemistry+Season	
		p	∑λ	p	∑λ	p	∑λ	P	∑λ	p	∑λ
River	Maggia	n.s.		0.001	0.233	n.s.					
River	Vedeggio	0.001	0.358	0.001	0.236	0.002	0.162	0.001	0.484	0.001	0.407
River	Verzasca	n.s.		0.001	0.306	n.s.					
Lake	Inferiore	0.018	0.342	0.034	0.130	0.003	0.195	0.006	0.424	0.004	0.446
Lake	Superiore	n.s.		0.035	0.129	0.006	0.218				
Lake	Starlarescio	n.s.		0.015	0.147	n.s.					
Lake	Tomeo	0.004	0.643	0.004	0.395	n.s.		0.008	0.649		

Conclusion

None of the time trend analyses presented here indicates that a benthic recovery is occurring at the sampling sites. Recovery may have begun at some sites but has not been identified, because of the very small number of the newly appearing species. These were excluded from the analysis because of the "downweighting" to the initial sample size. Especially in lakes Tomeo and Starlarescio, where concentrations of aluminium decreased to levels less toxic for benthic organisms, a recovery might have started or will occur soon. Comparing their benthic community with those of the less acidified lakes Inferiore and Superiore, especially in lake Tomeo but also in lake Starlarescio, the appearance of a few Ephemeroptera species or of moderately sensitive taxa might be expected as a sign of recovery. Looking at the original dataset none of these signs can be observed. However, after autumn 2006 each out let sample of lake Tomeo contains at least 1 individual of Crenobia alpina, that can be defined as "moderately acid-sensitive" according to Fjellheim and Raddum (1990). Since in out lets of lakes Inferiore and Superiore Crenobia alpina was sampled from the beginning of the sampling period, the appearance may be an initial sign of recovery. The appearance in every spring sample of the moderate acid-sensitive Protonemoura nimborum after 2005 in lake Inferiore, and after 2004 in lake Superiore, or the occasionally appearance of Perlodes sp. after 2003 in lake Superiore and after 2004 in lake Inferiore may also be first signs of improved water conditions. However, in order to see if these appearances are significant we still have to wait some years.

Although Oligochaeta and Chironomidae are in general considered as acid-tolerant, their identification only to family level might have hidden important changes in the benthic community and early signs of recovery especially in lakes, where they are abundant.

In addition, it is possible that some recovery has already occurred before invertebrate sampling started. Finally, other existing temporal variations than chemistry like the effects of climate change (increased temperature, increased or decreased precipitation), may also influence the composition of benthic communities and overlap and confound recovery from acidification.

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Appendix A. Trends in precipitation chemistry at the individual sites

***Mann Kendall Test**:*

Z:	The presence of a statistically significant trend is evaluated using the *Z v*alue. A positive (negative) value of *Z* indicates an upward (downward) trend. The statistic *Z* has a normal distribution.
Signific:	Significance level corresponding to *** if trend at 〈 = 0.001 level of significance ** if trend at 〈 = 0.01 level of significance * if trend at 〈 = 0.05 level of significance + if trend at 〈 = 0.1 level of significance

The Sen method is used to estimate the true slope of a linear trend, i.e. change per unit time (*t*)period (in this case a year); $f(t) = Qt + B$. Relative change at individual sites is calculated as the ratio Qt/B.

Table A1. *Trends in non sea-salt SO_4 concentration in precipitation at the individual sites.*

EMEP															
Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R	-4.89798	***	-0.0478	1.3016	-70 %	-2.68328	**	-0.069	1.393	-50 %	-2.86217	**	-0.0478	1.2942	-37 %
CH0002R	-5.10789	***	-0.01617	0.485167	-63 %	-3.04105	**	-0.03767	0.622667	-60 %	-2.68328	**	-0.01071	0.412286	-26 %
CZ0001R	-4.6181	***	-0.0485	1.295	-71 %	-3.57771	***	-0.10289	1.489	-69 %					
CZ0003R	-4.82801	***	-0.0464	1.1754	-75 %	-2.86217	**	-0.069	1.2795	-54 %	-2.5044	*	-0.02233	0.788833	-28 %
DE0001R	-4.68807	***	-0.03633	0.936	-74 %	-1.78885	+	-0.023	0.9025	-25 %	-2.86217	**	-0.02438	0.762938	-32 %
DE0003R	-4.68807	***	-0.02062	0.576846	-68 %	-1.96774	*	-0.01767	0.577333	-31 %	-2.68328	**	-0.01143	0.4485	-25 %
DE0005R	-5.10789	***	-0.03161	0.8135	-74 %	-3.39882	***	-0.037	0.8355	-44 %	-2.5044	*	-0.02367	0.705	-34 %
DK0005R	-4.89798	***	-0.0395	0.9995	-75 %	-3.57771	***	-0.0804	1.1355	-71 %	-2.14663	*	-0.01767	0.6935	-25 %
FI0004R	-4.6181	***	-0.01367	0.433	-60 %	-3.39882	***	-0.0245	0.47	-52 %	-2.5044	*	-0.00838	0.354813	-24 %
FI0017R	-3.56853	***	-0.0321	0.9215	-66 %	-2.14663	*	-0.06175	1.064875	-58 %					
FI0022R	-3.71302	***	-0.00775	0.293	-50 %	-3.04105	**	-0.02525	0.3825	-66 %	-1.70625	+	-0.0035	0.242	-14 %
FR0008R	-4.26824	***	-0.01573	0.518182	-58 %	-3.21994	**	-0.02614	0.549714	-48 %					
FR0009R	-3.39568	***	-0.01544	0.556778	-53 %	-2.32551	*	-0.0305	0.6165	-49 %					
FR0010R	-3.28864	**	-0.01	0.418	-45 %	-2.32551	*	-0.01763	0.451625	-39 %					
GB0002R	-4.54812	***	-0.01858	0.493167	-72 %	-2.32551	*	-0.01663	0.492	-34 %	-2.14663	*	-0.01467	0.422	-35 %
HU0002R	-1.67931	+	-0.03914	1.355571	-55 %						-1.96774	*	-0.05783	1.584667	-36 %
IT0004R	-4.37588	***	-0.03758	1.082333	-66 %						-3.04105	**	-0.03833	1.074667	-36 %
LT0015R	-4.33821	***	-0.07975	1.5915	-95 %	-2.5044	*	-0.1965	2.2155	-89 %	-2.14663	*	-0.03767	0.977833	-39 %
LV0010R	-3.91838	***	-0.033	0.871	-72 %	-2.5044	*	-0.07925	1.05075	-75 %	-2.14663	*	-0.0262	0.7848	-33 %
NO0001R	-4.72595	***	-0.02667	0.738333	-69 %	-2.42467	*	-0.03425	0.77925	-44 %	-2.32551	*	-0.02063	0.650813	-32 %
NO0015R	-3.78307	***	-0.00467	0.141667	-63 %	-3.04105	**	-0.01071	0.163071	-66 %					
NO0039R	-2.90558	**	-0.00233	0.106	-42 %	-1.70625	+	-0.003	0.1105	-27 %					
PL0002R	-4.82801	***	-0.04653	1.329133	-67 %	-3.39882	***	-0.0755	1.43525	-53 %					
SE02-14	-5.03792	***	-0.035	0.852	-78 %	-2.5044	*	-0.04625	0.903	-51 %	-3.04105	**	-0.01783	0.626	-28 %
SE0005R	-3.28864	**	-0.00929	0.296571	-59 %	-2.68328	**	-0.0295	0.3945	-75 %					
SE0011R	-5.31781	***	-0.04009	0.969182	-79 %	-3.04105	**	-0.06663	1.07375	-62 %	-3.21994	**	-0.01833	0.6335	-29 %
RS0005R	-3.49856	***	-0.08988	2.524	-68 %						-3.49856	***	-0.08988	2.524	-36 %

CAPMoN															
Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB	-3.35861	***	-0.01985	0.931385	-40 %						-2.5044	*	-0.0272	1.0411	-26 %
NL2BAB	-3.28864	**	-0.01492	0.599167	-47 %										
NS1JAC	-2.76555	**	-0.0165	0.8555	-37 %						-2.68328	**	-0.025	0.977	-26 %
NS1KEJ	-2.6589	**	-0.013	0.841	-29 %										
ON1ALG	-3.70847	***	-0.03213	1.766125	-35 %						-3.04105	**	-0.05167	2.0805	-25 %
ON1BON	-3.70847	***	-0.02283	1.103	-39 %						-2.14663	*	-0.0188	1.0324	-18 %
ON1CHA	-3.49856	***	-0.03655	1.896636	-37 %	-1.96774	*	-0.041	1.9	-22 %	-2.5044	*	-0.075	2.439	-31 %
ON1EGB	-3.71302	***	-0.04725	2.3365	-38 %										
ON1ELA															
ON1LON	-4.82801	***	-0.05925	2.8095	-40 %	-2.5044	*	-0.05925	2.806625	-21 %	-3.04105	**	-0.094	3.3915	-28 %
ON1WAR	-3.6385	***	-0.04871	2.339	-40 %						-2.14663	*	-0.0655	2.68825	-24 %
PQ1CPS	-3.28864	**	-0.028	1.179	-45 %						-2.68328	**	-0.041	1.375	-30 %
PA1PEN	-2.09913	*	-0.04063	2.54875	-30 %										

NADP															
Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR	-4.23585	***	-0.05875	2.54625	-44 %						-2.78388	**	-0.07333	2.72	-27 %
MA1QBB	-2.62553	**	-0.032	1.602	-38 %	-1.70625	+	-0.062	1.754	-35 %					
MD1WYE	-2.69554	**	-0.04182	2.073636	-38 %	-2.14663	*	-0.09	2.35	-38 %					
ME1BRM	-3.64744	***	-0.03143	1.287143	-46 %	-1.80334	+	-0.05556	1.448333	-38 %					
ME1HAN	-3.39568	***	-0.03	1.2	-48 %	-1.78885	+	-0.04	1.125	-36 %					
ME1SQU	-2.59636	**	-0.024	1.082	-42 %										
MI1CHS	-2.76895	**	-0.02	1.1	-35 %	-2.68328	**	-0.06667	1.293333	-52 %					
MI1DGL	-4.45135	***	-0.04	1.82	-42 %	-1.88586	+	-0.045	1.815	-25 %	-3.14309	**	-0.0675	2.1825	-31 %
MI1WEL	-3.81576	***	-0.043	1.978	-41 %										
NH1HBR	-4.54812	***	-0.04833	1.695	-54 %	-2.5044	*	-0.055	1.7525	-31 %	-1.78885	+	-0.045	1.645	-27 %
NY1ARF	-4.1283	***	-0.074	2.954	-48 %	-1.96774	*	-0.138	3.106	-44 %	-3.04105	**	-0.09	3.19	-28 %
NY1BSC	-3.5729	***	-0.05	1.99	-48 %	-2.14663	*	-0.1	2.185	-46 %	-2.06546	*	-0.044	1.948	-23 %
NY1HUN	-3.74575	***	-0.036	1.692	-40 %										
NY1WFM	-3.15256	**	-0.0425	1.5875	-51 %	-1.88586	+	-0.07667	1.768333	-43 %					
NY1WPT	-3.70847	***	-0.06	2.26	-50 %	-2.5044	*	-0.1	2.45	-41 %					
OH1DEL	-4.47815	***	-0.07278	2.903333	-48 %	-2.14663	*	-0.09	2.865	-31 %	-3.04105	**	-0.10222	3.367222	-30 %
PA1LDR	-2.27825	*	-0.04625	2.6875	-33 %	-2.06546	*	-0.10571	2.836429	-37 %					
PA1MLF	-3.91838	***	-0.05667	2.23	-48 %	-2.86217	**	-0.09333	2.288333	-41 %					
VA1VPI	-2.38194	*	-0.02222	1.692222	-25 %										
VT1BEN	-3.5729	***	-0.05667	2.08	-52 %	-2.5044	*	-0.105	2.285	-46 %					
VT1UND	-3.18564	**	-0.05429	1.911429	-54 %										

***Table A2**. Trends in hydrogen ion (H^+) concentration in precipitation at the individual sites.*

EMEP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R															
CH0002R	-3.84841	***	-3.8E-07	9.93E-06	-73 %										
CZ0001R	-4.75804	***	-1.7E-06	3.81E-05	-86 %	-3.57771	***	-3E-06	4.46E-05	-67 %					
CZ0003R	-4.47815	***	-1.8E-06	4.11E-05	-82 %	-2.14663	*	-1.8E-06	4.25E-05	-42 %	-1.96774	*	-8.5E-07	2.69E-05	-32 %
DE0001R	-2.93879	**	-8.7E-07	2.79E-05	-59 %	-2.5044	*	-2E-06	3.1E-05	-63 %					
DE0003R	-4.02581	***	-5.8E-07	1.91E-05	-58 %	-2.5044	*	-9E-07	1.94E-05	-46 %					
DE0005R	-3.70847	***	-9.1E-07	2.29E-05	-76 %	-2.14663	*	-9.8E-07	2.38E-05	-41 %					
DK0005R	-4.1283	***	-1.2E-06	2.65E-05	-87 %						-2.32551	*	-5.7E-07	1.8E-05	-32 %
FI0004R	-3.88578	***	-5.8E-07	2.57E-05	-43 %	-2.5044	*	-9.7E-07	2.69E-05	-36 %					
FI0017R															
FI0022R	-3.42859	***	-3.5E-07	2.2E-05	-30 %	-2.68328	**	-9.4E-07	2.43E-05	-38 %					
FR0008R															
FR0009R	-1.67931	+	-1.7E-07	1.26E-05	-26 %										
FR0010R															
GB0002R	-3.42859	***	-6.7E-07	2.03E-05	-63 %						-2.86217	**	-1.4E-06	3.16E-05	-44 %
HU0002R						-1.78885	+	-2.2E-06	2.03E-05	-108 %					
IT0004R	-4.89798	***	-2.1E-06	4.05E-05	-97 %	-2.86217	**	-2E-06	4.04E-05	-51 %	-2.32551	*	-2.1E-06	4.32E-05	-49 %
LT0015R						-2.14663	*	-5.8E-06	5.81E-05	-99 %					
LV0010R	-2.72887	**	-7E-07	2.76E-05	-48 %						-2.86217	**	-1.5E-06	3.9E-05	-37 %
NO0001R	-5.03792	***	-1.6E-06	4.38E-05	-68 %	-2.86217	**	-1.9E-06	4.55E-05	-41 %	-2.68328	**	-9.7E-07	3.46E-05	-28 %
NO0015R	-2.79885	**	-2.1E-07	7.06E-06	-56 %	-3.04105	**	-6.2E-07	9.06E-06	-69 %					
NO0039R	-4.30586	***	-2.1E-07	7.83E-06	-50 %	-2.14663	*	-2.1E-07	7.85E-06	-27 %	-1.88586	+	-2E-07	7.75E-06	-25 %
PL0002R	-4.47815	***	-1.5E-06	4.05E-05	-69 %	-3.39882	***	-2.7E-06	4.55E-05	-59 %					
SE02-14	-5.07602	***	-2E-06	4.82E-05	-79 %	-2.96349	**	-2.4E-06	4.95E-05	-49 %	-3.04105	**	-1.8E-06	4.51E-05	-39 %
SE0005R	-4.0633	***	-6.1E-07	1.85E-05	-63 %	-2.42467	*	-1E-06	2.04E-05	-50 %	-2.42467	*	-6.5E-07	1.91E-05	-34 %
SE0011R	-4.75804	***	-1.6E-06	3.93E-05	-78 %	-2.32551	*	-1.8E-06	4.05E-05	-44 %	-2.86217	**	-1.5E-06	3.7E-05	-40 %
RS0005R															

CAPMoN

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB	-4.47815	***	-0.00073	0.026849	-52 %	-2.14663	*	-0.00087	0.027177	-32 %	-2.32551	*	-0.00082	0.028118	-29 %
NL2BAB	-4.05833	***	-0.00044	0.016503	-51 %	-2.14663	*	-0.00054	0.016921	-32 %	-1.78885	+	-0.00042	0.016163	-26 %
NS1JAC	-3.6385	***	-0.00041	0.021038	-37 %						-2.14663	*	-0.00048	0.022134	-22 %
NS1KEJ	-3.70847	***	-0.00061	0.025575	-45 %						-2.14663	*	-0.00075	0.027773	-27 %
ON1ALG	-4.89798	***	-0.00124	0.03586	-66 %	-1.96774	*	-0.00096	0.034421	-28 %	-3.21994	**	-0.00111	0.034214	-32 %
ON1BON	-4.26824	***	-0.00076	0.024943	-58 %						-2.14663	*	-0.00057	0.021989	-26 %
ON1CHA	-5.10789	***	-0.00152	0.048245	-60 %	-2.5044	*	-0.00135	0.046653	-29 %	-3.39882	***	-0.00207	0.05616	-37 %
ON1EGB	-4.75804	***	-0.00161	0.046736	-65 %	-1.78885	+	-0.00142	0.045774	-31 %	-3.04105	**	-0.00177	0.048967	-36 %
ON1ELA	-4.19827	***	-0.00032	0.011573	-52 %						-2.5044	*	-0.00021	0.00976	-22 %
ON1LON	-4.89798	***	-0.0021	0.057365	-70 %	-2.32551	*	-0.00191	0.056275	-34 %	-3.21994	**	-0.00217	0.058105	-37 %
ON1WAR	-4.68807	***	-0.00178	0.055135	-61 %						-3.39882	***	-0.00203	0.058923	-34 %
PQ1CPS	-4.47815	***	-0.00081	0.02818	-55 %						-3.21994	**	-0.00117	0.033833	-35 %
PA1PEN	-4.33821	***	-0.00207	0.067728	-58 %	-2.5044	*	-0.00274	0.071346	-38 %	-2.32551	*	-0.0023	0.073683	-31 %

NADP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR	-4.26824	***	-0.00163	0.045108	-69 %						-2.32551	*	-0.00126	0.038821	-32 %
MA1QBB	-3.07873	**	-0.0009	0.039061	-44 %										
MD1WYE	-3.35861	***	-0.00127	0.049854	-49 %	-1.96774	*	-0.00175	0.050374	-35 %					
ME1BRM	-3.42859	***	-0.0009	0.032903	-52 %										
ME1HAN	-3.42859	***	-0.00079	0.028493	-53 %						-1.78885	+	-0.00095	0.032111	-30 %
ME1SQU	-2.51896	*	-0.00052	0.024768	-40 %										
MI1CHS	-4.33821	***	-0.00055	0.017849	-58 %	-2.86217	**	-0.00064	0.018472	-35 %					
MI1DGL	-4.40818	***	-0.00089	0.031911	-53 %						-3.04105	**	-0.00139	0.039581	-35 %
MI1WEL	-4.19827	***	-0.00122	0.035483	-66 %						-2.5044	*	-0.00117	0.034681	-34 %
NH1HBR	-4.6181	***	-0.00128	0.043673	-56 %	-1.78885	+	-0.00101	0.042489	-24 %	-2.68328	**	-0.00124	0.041719	-30 %
NY1ARF	-4.89798	***	-0.00195	0.060637	-61 %	-2.32551	*	-0.00281	0.062933	-45 %	-3.75659	***	-0.00229	0.066883	-34 %
NY1BSC	-4.40818	***	-0.00145	0.050092	-55 %	-2.32551	*	-0.00209	0.054353	-38 %	-2.68328	**	-0.00118	0.045197	-26 %
NY1HUN	-3.98836	***	-0.00112	0.040766	-52 %						-1.96774	*	-0.00108	0.040185	-27 %
NY1WFM	-4.33821	***	-0.00114	0.03839	-56 %	-1.96774	*	-0.00132	0.037616	-35 %	-2.5044	*	-0.00091	0.035119	-26 %
NY1WPT	-3.91838	***	-0.00157	0.053653	-55 %	-2.5044	*	-0.00215	0.054373	-40 %					
OH1DEL	-4.33821	***	-0.00163	0.052408	-59 %						-2.5044	*	-0.00213	0.061477	-35 %
PA1LDR	-3.56853	***	-0.00136	0.060645	-43 %										
PA1MLF	-4.26824	***	-0.00146	0.052383	-53 %	-2.86217	**	-0.00199	0.054101	-37 %					
VA1VPI	-3.35861	***	-0.00067	0.032937	-39 %										
VT1BEN	-4.1283	***	-0.00139	0.04796	-55 %	-2.32551	*	-0.00228	0.051889	-44 %					
VT1UND	-3.84841	***	-0.00145	0.043234	-64 %						-2.14663	*	-0.00104	0.03705	-28 %

***Table A3**. Trends in nitrate concentration in precipitation at the individual sites.*

EMEP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R	-1.85537	+	-0.00792	0.57425	-26 %	-1.70625	+	-0.012	0.576	-21 %					
CH0002R	-2.7367	**	-0.00531	0.339923	-30 %	-1.82321	+	-0.01063	0.36	-30 %					
CZ0001R	-2.09913	*	-0.00871	0.572	-29 %	-2.32551	*	-0.026	0.6385	-41 %					
CZ0003R	-3.77844	***	-0.01341	0.639824	-40 %	-2.86217	**	-0.02433	0.673667	-36 %					
DE0001R	-2.90558	**	-0.0124	0.6724	-35 %										
DE0003R	-2.79885	**	-0.0055	0.372	-28 %										
DE0005R	-2.76555	**	-0.00969	0.568438	-32 %						-2.24507	*	-0.026	0.801	-32 %
DK0005R	-1.81925	+	-0.00763	0.647625	-22 %										
FI0004R	-2.55551	*	-0.0028	0.2612	-20 %	-2.32551	*	-0.0075	0.27825	-27 %					
FI0017R															
FI0022R															
FR0008R															
FR0009R						-2.14663	*	-0.01175	0.39975	-29 %					
FR0010R															
GB0002R	-2.06795	*	-0.00289	0.233222	-24 %										
HU0002R	-1.88922	+	-0.00912	0.561118	-31 %	-2.14663	*	-0.02956	0.642333	-46 %					
IT0004R	-1.81925	+	-0.01044	0.749222	-26 %										
LT0015R	-3.18564	**	-0.01867	0.762	-47 %	-2.32551	*	-0.05975	0.98925	-60 %					
LV0010R	-2.51896	*	-0.01108	0.593	-36 %	-2.5044	*	-0.032	0.6365	-50 %					
NO0001R	-3.60572	***	-0.01158	0.5565	-40 %	-1.70625	+	-0.01183	0.56225	-21 %					
NO0015R						-2.60428	**	-0.003	0.089	-34 %	1.788854	+	0.002375	0.042063	56 %
NO0039R															
PL0002R	-2.13543	*	-0.00667	0.539	-24 %	-1.96774	*	-0.02175	0.6145	-35 %					
SE02-14	-4.27347	***	-0.01425	0.6395	-42 %						-2.5044	*	-0.019	0.73	-26 %
SE0005R	-1.85765	+	-0.00183	0.1615	-22 %	-2.32551	*	-0.00771	0.184	-42 %					
SE0011R	-3.42859	***	-0.01333	0.673333	-38 %										
RS0005R						2.325511	*	0.024667	0.467833	53 %					

CAPMoN

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB	-2.16911	*	-0.01133	0.748333	-29 %						-2.32551	*	-0.03	1.0405	-29 %
NL2BAB	-2.45199	*	-0.00658	0.416583	-30 %										
NS1JAC	-1.81925	+	-0.0086	0.6302	-26 %						-2.68328	**	-0.027	0.9025	-30 %
NS1KEJ	-2.30905	*	-0.0104	0.7504	-26 %						-2.32551	*	-0.03567	1.141833	-31 %
ON1ALG	-2.16911	*	-0.01933	1.815333	-20 %						-2.86217	**	-0.0566	2.3353	-24 %
ON1BON	-2.17177	*	-0.013	1.048	-24 %						-2.42467	*	-0.03757	1.388214	-27 %
ON1CHA	-2.6589	**	-0.02929	1.883118	-30 %						-3.21994	**	-0.10925	3.007	-36 %
ON1EGB	-2.51896	*	-0.02929	2.151286	-26 %						-2.14663	*	-0.04583	2.313333	-20 %
ON1ELA															
ON1LON	-2.93879	**	-0.04878	2.605667	-36 %						-3.04105	**	-0.1025	3.32875	-31 %
ON1WAR	-3.28864	**	-0.039	2.386	-31 %						-2.14663	*	-0.06267	2.785167	-23 %
PQ1CPS	-2.4855	*	-0.01538	0.972875	-30 %						-3.21994	**	-0.0475	1.452	-33 %
PA1PEN	-2.6589	**	-0.03873	2.083636	-35 %						-1.96774	*	-0.08117	2.748333	-30 %

NADP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR	-2.90915	**	-0.02875	1.82	-30 %						-2.78388	**	-0.07	2.365	-30 %
MA1QBB	-2.72887	**	-0.03	1.42	-40 %										
MD1WYE	-4.09582	***	-0.03778	1.536667	-47 %	-2.42467	*	-0.04714	1.576429	-30 %					
ME1BRM	-3.11562	**	-0.02667	1.093333	-46 %						-2.14663	*	-0.03857	1.306429	-30 %
ME1HAN	-2.80228	**	-0.01846	0.842308	-42 %						-2.32551	*	-0.0325	1.05125	-31 %
ME1SQU	-2.97925	**	-0.02154	0.914615	-45 %						-2.14663	*	-0.03667	1.136667	-32 %
MI1CHS											-1.78885	+	-0.02875	1.3775	-21 %
MI1DGL	-1.9954	*	-0.02125	1.67375	-24 %						-2.68328	**	-0.0625	2.2825	-27 %
MI1WEL	-2.14156	*	-0.02417	1.920833	-24 %						-2.32551	*	-0.075	2.62	-29 %
NH1HBR	-3.39568	***	-0.04	1.49	-51 %						-2.32551	*	-0.05875	1.7975	-33 %
NY1ARF	-3.77844	***	-0.05222	2.167778	-46 %						-3.04105	**	-0.11125	2.95	-38 %
NY1BSC	-3.88578	***	-0.04333	1.626667	-51 %	-1.78885	+	-0.05	1.645	-30 %	-2.5044	*	-0.0675	1.9975	-34 %
NY1HUN	-3.33111	***	-0.03	1.44	-40 %						-2.14663	*	-0.06	1.865	-32 %
NY1WFM	-3.32567	***	-0.02857	1.307143	-42 %						-2.5044	*	-0.05429	1.652143	-33 %
NY1WPT	-2.83557	**	-0.035	1.6	-42 %	-2.14663	*	-0.054	1.663	-32 %					
OH1DEL	-2.79885	**	-0.045	2.06	-42 %						-3.39882	***	-0.104	2.913	-36 %
PA1LDR	-3.28864	**	-0.03625	1.92	-36 %						-2.5044	*	-0.06167	2.328333	-26 %
PA1MLF	-4.34887	***	-0.03889	1.71	-43 %	-3.06568	**	-0.035	1.7075	-20 %	-2.34434	*	-0.0525	1.9025	-28 %
VA1VPI	-2.56285	*	-0.01643	1.095714	-28 %						-1.96774	*	-0.03167	1.253333	-25 %
VT1BEN	-3.53571	***	-0.04533	1.808667	-48 %						-1.78885	+	-0.05571	1.907857	-29 %
VT1UND	-3.46569	***	-0.0425	1.5525	-52 %						-2.32551	*	-0.05429	1.74	-31 %

__Table A4__. Trends in ammonium concentration in precipitation at the individual sites.

EMEP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R	-1.74928	+	-0.0148	0.781	-36 %	-1.78885	+	-0.03143	0.877571	-36 %					
CH0002R	-2.41548	*	-0.00825	0.56	-28 %	-1.96774	*	-0.01088	0.567313	-19 %					
CZ0001R	-1.85537	+	-0.0115	0.7865	-28 %	-1.78885	+	-0.03729	0.872714	-43 %					
CZ0003R	-2.51896	*	-0.0094	0.7138	-25 %	-2.32551	*	-0.025	0.769	-33 %					
DE0001R															
DE0003R	-3.42859	***	-0.00941	0.444706	-40 %										
DE0005R	-3.07873	**	-0.01608	0.7215	-42 %						-1.96774	*	-0.024	0.827	-29 %
DK0005R						-2.32551	*	-0.059	0.887	-67 %					
FI0004R	-2.38194	*	-0.00322	0.198333	-31 %	-1.96774	*	-0.011	0.2315	-48 %	-1.70625	+	-0.00325	0.202375	-16 %
FI0017R	-3.49856	***	-0.01238	0.4565	-52 %	-1.78885	+	-0.01463	0.490125	-30 %					
FI0022R						-1.88586	+	-0.006	0.12	-50 %					
FR0008R	-2.37902	*	-0.01885	0.644	-56 %	-2.32551	*	-0.044	0.696	-63 %					
FR0009R	-1.95919	+	-0.0228	0.7924	-55 %	-3.04105	**	-0.08343	1.087286	-77 %	2.146625	*	0.0176	0.2271	77 %
FR0010R	-1.95919	+	-0.02283	0.701833	-62 %	-2.14663	*	-0.062	0.842	-74 %					
GB0002R															
HU0002R	-2.62553	**	-0.02025	0.78225	-49 %										
IT0004R															
LT0015R	-2.76555	**	-0.05535	1.263412	-83 %	-2.14663	*	-0.11638	1.511938	-77 %					
LV0010R															
NO0001R	-3.56853	***	-0.01143	0.494429	-44 %	-1.78885	+	-0.01122	0.5	-22 %					
NO0015R															
NO0039R															
PL0002R															
SE02-14	-1.74928	+	-0.00763	0.57125	-25 %										
SE0005R															
SE0011R	-3.70847	***	-0.02075	0.77775	-51 %	-2.14663	*	-0.035	0.8545	-41 %					
RS0005R						-1.96774	*	-0.049	1.089	-45 %					

CAPMoN

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB															
NL2BAB															
NS1JAC															
NS1KEJ	1.925385	+	0.001273	0.090909	27 %										
ON1ALG											-1.96774	*	-0.006	0.5425	-11 %
ON1BON															
ON1CHA															
ON1EGB															
ON1ELA	2.065413	*	0.007154	0.297385	46 %										
ON1LON											-1.96774	*	-0.009	0.6675	-13 %
ON1WAR															
PQ1CPS															
PA1PEN															

NADP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR															
MA1QBB															
MD1WYE							*	-0.00857	0.282857	-30 %					
ME1BRM															
ME1HAN															
ME1SQU															
MI1CHS	2.085096	*	0.004615	0.280769	31 %										
MI1DGL															
MI1WEL															
NH1HBR															
NY1ARF															
NY1BSC															
NY1HUN															
NY1WFM															
NY1WPT															
OH1DEL											-2.02813	*	-0.01	0.505	-20 %
PA1LDR															
PA1MLF															
VA1VPI															
VT1BEN															
VT1UND															

Table A5. *Trends in calcium concentration in precipitation at the individual sites.*

EMEP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R	-4.05833	***	-0.10333	1.968333	-100 %	-2.86217	**	-0.17622	2.357	-75 %					
CH0002R															
CZ0001R	-2.23908	*	-0.0105	0.377	-53 %	-2.32551	*	-0.01825	0.411125	-44 %					
CZ0003R	-2.58893	**	-0.00945	0.351	-51 %										
DE0001R	-4.05833	***	-0.04047	0.960533	-80 %	-1.96774	*	-0.02625	0.960625	-27 %					
DE0003R	-1.9954	*	-0.005	0.286	-33 %										
DE0005R															
DK0005R						-1.78885	+	-0.04133	0.615	-67 %					
FI0004R															
FI0017R	-4.6181	***	-0.02433	0.527333	-88 %	-2.86217	**	-0.04014	0.603643	-67 %	-1.78885	+	-0.0084	0.3323	-25 %
FI0022R															
FR0008R											2.325511	*	0.006778	0.079944	85 %
FR0009R															
FR0010R															
GB0002R															
HU0002R	-3.1487	**	-0.05057	1.566	-61 %	-2.32551	*	-0.247	2.686	-92 %					
IT0004R											-2.68328	**	-0.04367	1.118	-39 %
LT0015R	-2.23908	*	-0.0275	0.9475	-55 %										
LV0010R	-3.67574	***	-0.02978	0.676667	-84 %	-2.14663	*	-0.0655	0.83025	-79 %					
NO0001R											1.788854	+	0.0036	0.0646	56 %
NO0015R											1.885856	+	0.008	0.015	533 %
NO0039R											2.146625	*	0.00575	0.021375	269 %
PL0002R	-3.28864	**	-0.00862	0.361692	-45 %	-2.68328	**	-0.016	0.392	-41 %					
SE02-14											1.788854	+	0.012	0.0645	186 %
SE0005R															
SE0011R															
RS0005R						-1.96774	*	-0.14183	2.46025	-58 %					

CAPMoN

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB	2.290475	*	0.0005	0.0355	27 %						-1.82321	+	-0.00067	0.052667	-13 %
NL2BAB															
NS1JAC	2.288581	*	0.000714	0.035571	38 %	1.922376	+	0.001	0.034	29 %	-1.70625	+	-0.00063	0.057438	-11 %
NS1KEJ	2.174435	*	0.000917	0.04275	41 %										
ON1ALG															
ON1BON															
ON1CHA															
ON1EGB															
ON1ELA	1.81925	+	0.002692	0.124615	41 %										
ON1LON															
ON1WAR															
PQ1CPS	1.927749	+	0.001	0.052	37 %										
PA1PEN															

NADP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR															
MA1QBB	1.677819	+	0.001	0.047	40 %										
MD1WYE						-2.04479	*	-0.0025	0.08125	-31 %					
ME1BRM															
ME1HAN															
ME1SQU						-1.95837	+	0	0.04	0 %					
MI1CHS															
MI1DGL															
MI1WEL															
NH1HBR						2.553605	*	0.002	0.04	50 %	-2.20368	*	-0.00143	0.073571	-19 %
NY1ARF											-2.08232	*	-0.0075	0.225	-33 %
NY1BSC															
NY1HUN															
NY1WFM															
NY1WPT															
OH1DEL											-2.10546	*	-0.005	0.2525	-20 %
PA1LDR															
PA1MLF															
VA1VPI															
VT1BEN															
VT1UND															

Table A6. *Trends in precipitation amount at the individual sites.*

EMEP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
AT0002R						1.96774	*	21.96667	345.2667	64 %					
CH0002R	1.81925	+	16.1	675.1	45 %	1.788854	+	44.25	600.875	74 %					
CZ0001R						2.146625	*	33.53475	632.7494	53 %					
CZ0003R															
DE0001R															
DE0003R															
DE0005R	2.309048	*	22.266	853.177	50 %										
DK0005R															
FI0004R															
FI0017R						-2.14663	*	-23.8708	688.45	-35 %					
FI0022R															
FR0008R															
FR0009R	2.518961	*	29.12483	827.2637	67 %	2.325511	*	83.219	610.887	136 %					
FR0010R															
GB0002R															
HU0002R															
IT0004R															
LT0015R															
LV0010R															
NO0001R															
NO0015R															
NO0039R															
PL0002R						1.788854	+	25.3875	444.4813	57 %					
SE02-14															
SE0005R	-2.09913	*	-16.508	563.107	-56 %						-2.32551	*	-49.4116	1048.389	-47 %
SE0011R															
RS0005R															

CAPMoN

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
NB1HAB															
NL2BAB															
NS1JAC															
NS1KEJ															
ON1ALG	-1.88922	+	-1.13846	132.2	-16 %										
ON1BON															
ON1CHA						2.065461	*	1.15	82.575	14 %					
ON1EGB															
ON1ELA															
ON1LON															
ON1WAR															
PQ1CPS															
PA1PEN															

NADP

Site	1990-2008					1990-1999					1999-2008				
	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change	Test Z	Signific.	Q	B	% change
IN1HTR															
MA1QBB															
MD1WYE															
ME1BRM	1.891537	+	1.1	98.4	21 %										
ME1HAN															
ME1SQU															
MI1CHS															
MI1DGL															
MI1WEL						-2.32551	*	-3.23333	110.35	-29 %					
NH1HBR	1.749279	+	1.14	110.94	20 %						1.788854	+	3.911111	79.05556	49 %
NY1ARF											2.325511	*	3	46.9	64 %
NY1BSC	2.275455	*	2.890909	120.2273	46 %						2.065461	*	4	102.3	39 %
NY1HUN											1.96774	*	2.85	77.75	37 %
NY1WFM	1.891537	+	1.2125	105.4125	22 %						2.065461	*	3	80.5	37 %
NY1WPT															
OH1DEL															
PA1LDR															
PA1MLF															
VA1VPI															
VT1BEN															
VT1UND	2.239077	*	1.777778	106.6889	32 %						2.683282	**	2.95	88.225	33 %

***Table A7.** Average trends in annual volume weighted mean and number of sites with significant positive or negative trend in the three different networks*

Comp	Network	Nr of sites	Period	decrease	increase	Average*	STD
SO4	EMEP	27	1990-2008	100 %	0 %	-60 %	11 %
			1990-1999	89 %	0 %	-51 %	17 %
			1999-2008	67 %	0 %	-19 %	12 %
	CAPMoN	13	1990-2008	92 %	0 %	-37 %	8 %
			1990-1999	15 %	0 %	-18 %	11 %
			1999-2008	62 %	0 %	-26 %	8 %
	NADP	21	1990-2008	100 %	0 %	-42 %	9 %
			1990-1999	71 %	0 %	-36 %	10 %
			1999-2008	29 %	0 %	-22 %	12 %
H	EMEP	24	1990-2008	74 %	0 %	-54 %	24 %
			1990-1999	63 %	0 %	-43 %	32 %
			1999-2008	37 %	0 %	-21 %	21 %
	CAPMoN	13	1990-2008	100 %	0 %	-57 %	10 %
			1990-1999	54 %	0 %	-28 %	6 %
			1999-2008	100 %	0 %	-32 %	8 %
	NADP	21	1990-2008	100 %	0 %	-53 %	11 %
			1990-1999	43 %	0 %	-29 %	10 %
			1999-2008	52 %	0 %	-27 %	12 %
NO3	EMEP	27	1990-2008	70 %	0 %	-23 %	12 %
			1990-1999	48 %	4 %	-23 %	22 %
			1999-2008	7 %	4 %	-9 %	21 %
	CAPMoN	13	1990-2008	92 %	0 %	-28 %	11 %
			1990-1999	0 %	0 %	2 %	12 %
			1999-2008	85 %	0 %	-41 %	11 %
	NADP	21	1990-2008	95 %	0 %	-41 %	12 %
			1990-1999	19 %	0 %	-11 %	16 %
			1999-2008	86 %	0 %	-38 %	11 %
NH4	EMEP	27	1990-2008	59 %	0 %	-27 %	20 %
			1990-1999	56 %	0 %	-32 %	25 %
			1999-2008	7 %	4 %	-7 %	18 %
	CAPMoN	13	1990-2008	0 %	15 %	9 %	15 %
			1990-1999	0 %	0 %	4 %	15 %
			1999-2008	15 %	0 %	-13 %	13 %
	NADP	21	1990-2008	0 %	5 %	-1 %	13 %
			1990-1999	5 %	0 %	-8 %	13 %
			1999-2008	5 %	0 %	-4 %	18 %
Ca	EMEP	27	1990-2008	37 %	0 %	-28 %	33 %
			1990-1999	33 %	0 %	-32 %	33 %
			1999-2008	7 %	19 %	-3 %	46 %
	CAPMoN	13	1990-2008	0 %	38 %	24 %	17 %
			1990-1999	0 %	8 %	14 %	17 %
			1999-2008	15 %	0 %	-3 %	23 %
	NADP	21	1990-2008	0 %	5 %	4 %	11 %
			1990-1999	5 %	5 %	11 %	26 %
			1999-2008	14 %	0 %	-7 %	23 %
Total precip amount	EMEP	27	1990-2008	4 %	11 %	8 %	32 %
			1990-1999	4 %	19 %	16 %	40 %
			1999-2008	4 %	0 %	0 %	33 %
	CAPMoN	13	1990-2008	8 %	0 %	2 %	7 %
			1990-1999	0 %	8 %	-2 %	12 %
			1999-2008	0 %	0 %	10 %	13 %
	NADP	21	1990-2008	0 %	24 %	15 %	13 %
			1990-1999	2 %	0 %	-1 %	13 %
			1999-2008	0 %	29 %	21 %	18 %

Appendix B. Chemistry data from ICP Waters sites

Table B1. *Sites in the ICP Waters programme anno 2008 with mean results for samples collected between 2005 and 2008 (n indicates the number of observations). Results from extra Ontario stations are presented in a table below.*

Country	ID	Station name	n	K25 mS/m	pH	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	SO4 mg/l	ANC µEq/l	Alk µEq/l	NH4-N µg/l N	NO3-N µg/l N	TOTN µg/l N	TOTP µg/l P	TOC/DOC mg/l C	H+ µEq/l
Belarus	BY01	Berezinsky Biosphere Reserve	28	36.14	7.76	50.41	14.07	3.95	1.71	7.36	23.58	3110	3157	663	1228	1903	59		0.02
Canada	CA01	Ontario, Algoma Region, Batchawana Lake	272	2.07	5.99	2.33	0.38	0.57	0.20	0.25	4.01	70	51	55	233	580	6	5.1	1.37
Canada	CA02	Ontario, Algoma Region, Wishart Lake	291	2.83	6.57	3.78	0.43	0.66	0.20	0.24	4.50	124	100	34	469	584	5	3.7	0.30
Canada	CA03	Ontario, Algoma Region, Little Turkey Lake	280	3.29	6.73	4.67	0.47	0.67	0.21	0.25	4.82	169	144	46	417	577	5	4.0	0.23
Canada	CA04	Ontario, Algoma Region, Turkey Lake	275	3.75	6.77	5.58	0.50	0.71	0.23	0.27	5.00	219	199	32	357	491	5	4.0	0.20
Canada	CA05	Quebec, Lac Veilleux	8	6.29	6.40	1.15	0.21	0.46	0.20	0.34	1.99	46	22	13	35			3.4	0.42
Canada	CA06	Quebec, Lac Josselin	8	5.91	6.11	1.00	0.17	0.43	0.13	0.31	2.33	26	12	13	38			3.5	0.82
Canada	CA07	Quebec, Lac Bonneville	8	5.60	5.35	0.73	0.19	0.37	0.05	0.41	2.65	-1	4	10	39			5.6	4.75
Canada	CA08	Quebec, Lac Laflamme	49		6.41	1.78	0.40	0.86	0.15	0.18	2.66	100	39	22	36			4.6	0.47
Canada	CA09	Quebec, Lac Macleod	7	4.69	5.73	0.77	0.19	0.43	0.05	0.39	2.34	12	8	10	36			4.8	2.25
Canada	CA10	Nova Scotia, Mount Tom Lake	8	2.52	4.70	0.28	0.29	2.25	0.23	3.41	1.21	19			20	228	10	9.1	20.19
Canada	CA11	Nova Scotia, Mountain Lake	8	2.09	5.22	0.29	0.28	2.39	0.21	3.53	1.47	15			24	150	6	4.4	6.33
Canada	CA12	Nova Scotia, Little Red Lake	8	3.51	4.38	0.27	0.31	2.65	0.26	3.68	1.42	25			20	285	12	15.6	41.43
Canada	CA13	Nova Scotia, Kejimkujik Lake	8	2.88	4.99	0.53	0.37	3.03	0.26	4.64	1.76	27			20	213	13	8.3	11.55
Canada	CA14	Nova Scotia, Beaverskin Lake	8	2.05	5.55	0.30	0.30	2.42	0.21	3.70	1.62	11			21	194	6	2.8	3.07
Canada	CA16	Ontario,Lake224	53	1.76	7.01	1.70	0.44	0.63	0.31	0.17	2.12	106	95	15	4	256	7	3.6	0.12
Canada	CA17	Ontario,Lake239	60	2.70	7.08	2.80	0.84	1.08	0.44	0.25	2.52	206	164	16	26	316	7	7.2	0.09
Canada	CA20	Ontario,Lake373	59	2.62	7.17	3.00	0.70	0.75	0.37	0.20	2.01	202	179	22	6	255	9	4.4	0.08
Switzerland	CH03	Lago di Tomè	11	1.05	5.68	0.95	0.08	0.36	0.17	0.33	1.36	12	8		341	561	2	0.3	2.46
Switzerland	CH05	Laghetto Inferiore	13	1.14	6.51	1.25	0.11	0.32	0.40	0.47	1.23	39	30		235	405	3	0.3	0.39
Switzerland	CH06	Laghetto Superiore	12	1.01	6.52	1.10	0.09	0.28	0.32	0.30	1.12	34	28		243	412	4	0.4	0.34
Switzerland	CH09	Lago Nero	11	1.72	6.82	2.06	0.16	0.38	0.47	0.84	2.11	65	62		168	403	4	0.4	0.17
Switzerland	CH19	Lago d'Alzasca	11	1.74	7.14	2.06	0.21	0.52	0.50	0.70	1.58	88	80		207	465	4	1.0	0.08
Switzerland	CH20	Lago del Starlaresc da Sgiof	13	1.08	5.44	0.63	0.10	0.33	0.19	0.47	1.20	0	1		284	531	3	0.8	3.94
Switzerland	CH26	Maggia	48	6.78	7.48	8.65	0.72	1.84	1.56	1.48	10.02	316	303		626	641		0.5	0.03
Switzerland	CH27	Vedeggio	48	4.88	7.12	5.19	1.02	1.70	0.63	0.98	6.62	187	163		1131	1200		0.6	0.08
Switzerland	CH28	Verzasca	48	2.62	6.87	3.04	0.24	0.76	0.60	0.20	3.90	78	67		777	823		0.3	0.14

Country	ID	Station name	n	K25 mS/m	pH	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	SO4 mg/l	ANC µEq/l	Alk µEq/l	NH4-N µg/l N	NO3-N µg/l N	TOTN µg/l N	TOTP µg/l P	TOC/DOC mg/l C	H+ µEq/l
Czech Rep.	CZ01	Bohemian Forest, Cerné	8	2.38	5.02	0.73	0.40	0.74	0.46	0.67	3.19	-28	-10		786		5	2.1	10.99
Czech Rep.	CZ02	Bohemian Forest, Certovo	8	2.33	4.72	0.35	0.27	0.58	0.26	0.51	3.24	-42	-25		439		13	3.5	20.94
Czech Rep.	CZ03	Bohemian Forest, Ple¿né	8	1.92	5.17	0.93	0.19	0.91	0.57	0.59	2.89	-4	-3		610		17	5.1	7.36
Czech Rep.	CZ04	Bohemian Forest, Prá¿ilské	8	1.58	5.19	0.50	0.29	0.63	0.25	0.50	2.46	1	-2		230		9	5.9	7.98
Czech Rep.	CZ05	Bohemian Forest, Laka	8	1.89	5.92	0.90	0.43	1.07	0.45	0.59	1.66	47	23		560		9	4.8	1.41
Czech Rep.	CZ06	Bohemian Forest, Zd´árské	7	2.73	6.23	1.99	0.50	2.44	0.46	0.63	5.71	117	65		62		27	9.4	1.07
Czech Rep.	CZ07	Lysina	48		4.27	1.91	0.43	2.32	0.52	1.25	10.02	-1	-59	12	26	456		18.7	60.88
Czech Rep.	CZ08	Uhlirska	45	56.89	5.77	4.64	0.93	3.19	0.52	1.00	13.16	146	62	14	124	595		7.9	7.19
Germany	DE01	Schwarzwald, Dürreychbach	47		6.13	2.40	0.64	1.04	2.64	2.94	3.55	50	69	16	1096			3.4	0.99
Germany	DE02	Fichtelgebirge, Eger	47		6.23	2.78	1.30	6.07	1.05	11.37	4.92	71	81		729				1.15
Germany	DE03	Rothaargebirge, Elberndorfer Bach	44	5.90	6.51	3.32	2.76	2.21	0.38	2.81	10.96	79		25	1283			1.9	0.49
Germany	DE04	Sächsische Tieflandsbucht, Ettelsbach	13	38.62	5.08	42.38	10.67	7.62	2.62	10.77	147.69	-119	54	51	1838	2	20	10.8	9.31
Germany	DE05	Schwarzwald, Goldersbach	52		6.73	2.34	0.51	1.26	1.57	2.04	3.05	104	119	17	400		8	5.0	0.25
Germany	DE06	Hunsrück, Gräfenbach	37	9.12	4.77	4.83	2.76	4.27	0.53	7.13	19.43	-158	22	12	3129		9	6.2	19.80
Germany	DE07	Erzgebirge, Grosse Pyra	46	5.31	4.73	3.47	0.79	2.15	1.02	1.36	13.91	-23	44	17	743	1	21	4.2	25.28
Germany	DE08	Bayerischer Wald, Grosse Ohe	82		6.27	1.93	0.65	1.51	0.58	1.21	2.78	68	129	20	1071		14	4.6	2.05
Germany	DE09	Sächsische Tieflandsbucht, Heidelbach	21	46.66	4.67	50.86	8.95	12.29	2.26	22.24	175.95	-474	38	197	675	2	23	7.6	56.53
Germany	DE10	Bayerischer Wald, Hinterer Schachtenbach	42		6.21	2.08	0.56	1.53	0.52	0.70	2.63	84	112	9	1085			3.3	1.90
Germany	DE12	Harz, Lange Bramke	109		6.44	3.45	1.83	1.81	0.71	2.95	3.28	228	77	121	629	1	8	1.0	0.53
Germany	DE13	Erzgebirge, Talsperre Neunzehnhain	21		6.95	13.21	6.21	7.95	1.51	14.42	35.70								0.13
Germany	DE17	Bayerischer Wald, Rachelsee	4		4.65	1.25	0.60	0.77	0.55	0.60	2.65	20	73	45	940				23.61
Germany	DE18	Fichtelgebirge, Röslau	36		5.59	2.19	0.58	3.05	0.53	1.65	8.82	31	63		613				6.61
Germany	DE21	Erzgebirge, Rote Pockau	44	9.69	5.96	7.46	2.70	4.27	1.05	3.79	28.59	61	98	20	608	1	25	7.4	1.88
Germany	DE23	Bayerischer Wald, Seebach	42		6.19	1.84	0.63	1.36	0.52	0.71	2.29	96	106	11	1024			3.7	2.12
Germany	DE24	Erzgebirge, Talsperre Sosa	24	56.23	5.71	4.08	1.85	2.55	0.93	2.34	15.58			52					3.16
Germany	DE25	Elbsandsteingebirge, Taubenbach	22	18.91	7.33	29.04	1.92	2.14	2.13	3.73	50.35	507	654	21	1326	2	21	3.4	0.06
Germany	DE26	Hunsrück, Traunbach 1	46	6.96	5.08	2.29	1.76	5.30	0.50	10.49	6.26	25	28	12	729		7	8.1	14.65
Germany	DE27	Bayerischer Wald, Vorderer Schachtenbach	42		6.36	2.27	0.61	1.76	0.57	0.74	2.78	127	151	10	1056			4.0	1.66
Germany	DE28	Oberpfälzer Wald. Waldnaab 2	11		6.54	3.20	1.48	2.66	0.98	1.72	10.34	143	128	20	957		22	3.1	0.40
Germany	DE29	Oberpfälzer Wald, Waldnaab 8	11		5.65	5.18	1.16	3.83	0.96	1.92	14.66	233	73	23	609		59	7.8	10.16
Germany	DE30	Erzgebirge, Wilde Weisseritz	45	8.31	6.74	7.97	1.77	3.21	1.46	4.21	19.10	149	261	32	782	1	22	5.0	0.35
Germany	DE31	Erzgebirge, Wolfsbach	45	18.07	7.09	14.18	4.81	10.55	1.98	18.13	26.09	404	452	22	2151	3	31	5.4	0.09
Germany	DE32	Rothaargebirge, Zinse	43	5.07	6.34	2.99	2.16	2.06	0.32	2.53	9.75	64		25	861			1.7	1.38

Country	ID	Station name	n	K25	pH	Ca	Mg	Na	K	Cl	SO4	ANC	Alk	NH4-N	NO3-N	TOTN	TOTP	TOC/DOC	H+
				mS/m		mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	µEq/l	µEq/l	µg/l N	µg/l N	µg/l N	µg/l P	mg/l C	µEq/l
Germany	DE33	Fichtelgebirge, Zinnbach	34		4.30	2.96	0.68	2.49	0.99	1.58	14.77	-59	33		701				52.61
Germany	DE35	Taunus, Rombach 4	51		6.94	3.40	1.02	2.52	2.10	5.55	3.80	136	170	14	625			1.3	0.17
Estonia	EE01	River Ahja, Kiidjärve	38	35.23	7.47	63.77	12.21	4.18	1.64	6.42	12.40	3846	3890	57	1042	1371	45		19.99
Finland	FI01	Hirvilampi	24	2.45	5.70	1.44	0.37	1.30	0.62	1.27	4.92	33	16	38	36	358	7	4.7	2.15
Finland	FI02	Vuorilampi	24	2.87	5.88	1.87	0.44	1.54	0.67	0.96	4.96	82	45	126	25	585	9	7.9	1.72
Finland	FI03	Mäkilampi	23	2.90	5.70	1.49	0.30	1.02	0.41	1.11	4.30	31	13	31	32	358	6	4.7	2.13
Finland	FI05	Lapland, Suopalampi	24	1.20	6.15	0.82	0.28	1.01	0.18	0.43	0.97	79	45	10	18	302	11	6.8	0.82
Finland	FI06	Lapland, Vasikkajärvi	25	0.73	5.92	0.31	0.10	0.50	0.12	0.42	1.29	10	12	6	5	125	4	1.6	1.29
Finland	FI07	Vusimaa, Vitsjön	23	3.70	6.42	1.76	0.80	2.80	0.55	3.43	5.03	86	59	24	43	340	5	6.5	0.45
Finland	FI08	N-Karelia,Kakkisenlampi	35	0.51	5.36	0.28	0.08	0.32	0.17	0.24	1.30	4	-1	7	15	172	5	3.5	5.32
Finland	FI09	Sonnanen	25	3.31	6.74	2.96	0.47	1.57	0.57	2.35	3.56	127	85	15	23	236	4	2.7	0.20
Italy	IT01	Piemonte, Lake Paione Inferiore	7	1.36	6.65	1.52	0.13	0.35	0.37	0.11	1.77	47	45	5	338	401	2	0.3	0.23
Italy	IT02	Piemonte, Lake di Mergozzo	5	6.00	7.05	6.13	1.47	2.34	1.16	1.61	7.43	309	277	11	689	873	4	1.7	0.10
Italy	IT03	Piemonte, Lake Paione Superiore	7	0.93	6.23	0.90	0.08	0.24	0.28	0.12	1.40	19	17	10	262	367	4	0.7	0.59
Italy	IT04	Piemonte, River Cannobino	48	5.27	7.37	4.54	1.58	2.23	0.79	1.88	5.77	250	243	12	709	800	7	1.0	0.05
Italy	IT05	Piemonte, River Pellino	48	5.90	7.27	4.99	1.24	3.71	0.59	2.97	4.34	242	233	7	1563	1658	14	0.8	0.05
Italy	IT06	Piemonte, River Pellesino	48	5.97	7.20	4.41	1.00	4.59	0.68	4.60	3.14	207	193	18	1638	1760	37	1.2	0.07
Latvia	LV03	Liela Jugla, Zaki	36	42.89	7.90	64.71	16.54	4.30	2.41	5.78	66.89	3232	3779	61	703	1436	51	14.5	0.01
Latvia	LV04	Tulija, Zoseni	34	33.87	7.90	55.20	14.01	3.38	2.51	3.67	14.49	3682	3788	126	426	1510	71	14.7	0.02
Latvia	LV05	Zvirbuli stream, hydrosite	22	7.38	4.07	3.32	1.06	1.74	0.54	2.35	3.07	207	56	244	70	1356	34	53.4	200.13
Latvia	LV07	Amula mouth	36	49.87	7.88	64.89	24.47	5.64	3.37	6.52	23.47	4863	5080	67	648	1300	58	14.2	0.02
Latvia	LV08	Tervete	36	67.17	7.94	100.50	29.49	7.51	3.69	20.29	75.81	5366	6167	45	4305	4945	46	7.0	0.01
Norway	NO01	Birkenes, Aust Agder	208	3.23	4.83	0.76	0.26	2.91	0.09	4.75	2.76	-10	1	16	87	283	4	5.9	16.58
Norway	NO03	Langtjern, Buskerud	208	1.30	5.04	0.91	0.13	0.57	0.07	0.44	1.01	49	2	10	11	260	5	11.4	10.06
Norway	NO04	Dalelv, Finnmark	208	3.79	6.34	1.61	0.86	3.82	0.28	5.99	3.93	73	46	20	13	171	4	3.6	0.53
Norway	NO10	Storgama, Telemark	201	1.28	5.00	0.48	0.08	0.75	0.04	1.01	1.03	12	1	12	31	261	4	5.8	11.21
Norway	NO11	Kårvatn, Møre og Romsdal	206	1.25	6.35	0.65	0.19	1.28	0.13	1.98	0.60	37	28	4	24	68	2	0.9	0.48
Poland	PL01	Tatra Mountains, Dlugi Staw Gasienicowy	44	9.33	6.06	1.83	0.13	0.31	0.09	0.28	2.04	37	45	32	425				1.24
Poland	PL02	Tatra Mountains, Zielony Staw Gasienicowy	44	11.45	6.27	2.48	0.19	0.36	0.14	0.27	1.88	97	106	37	206				0.65
Poland	PL03	Karkonosze, Maly Stav	53	5.36	6.01	1.18	0.26	1.18	0.22	0.50	3.48	32	37	25	262				1.38
Poland	PL04	Wielki Staw	52	4.85	5.32	0.88	0.15	0.91	0.20	0.53	3.18	-3	7	24	324				6.03
Sweden	SE01	Delångersån Iggersund	35	4.44	7.03	3.70	1.15	2.80	0.82	2.70	3.74	263	199	9	74	291	8	5.9	0.10
Sweden	SE02	Alsterån Getebro	36	6.46	6.76	5.13	1.33	4.97	1.28	6.40	6.55	288	166	19	133	539	24	13.9	0.19

Country	ID	Station name	n	K25	pH	Ca	Mg	Na	K	Cl	SO4	ANC	Alk	NH4-N	NO3-N	TOTN	TOTP	TOC/DOC	H+
				mS/m		mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	µEq/l	µEq/l	µg/l N	µg/l N	µg/l N	µg/l P	mg/l C	µEq/l
Sweden	SE03	Alsterån Strömsborg	12	6.51	6.83	5.17	1.23	5.25	1.25	6.12	6.88	296	158	12	109	586	14	14.2	0.16
Sweden	SE05	Tväringen	12	2.59	6.77	2.77	0.66	1.40	0.58	0.78	1.82	208	131	11	11	248	6	7.9	0.18
Sweden	SE06	Stensjön	24	1.68	6.39	1.29	0.37	1.26	0.27	0.75	1.59	101	49	16	19	229	5	6.7	0.45
Sweden	SE08	Brunnsjön	23	5.53	5.52	3.60	1.25	4.30	0.84	6.41	7.12	155	3	34	95	683	12	22.0	3.24
Sweden	SE09	Fiolen	24	4.82	6.66	2.82	0.99	3.69	1.66	5.93	5.28	137	74	21	144	556	11	8.2	0.23
Sweden	SE10	Storasjö	12	2.91	5.48	1.30	0.53	3.01	0.42	3.63	2.72	89	-1	22	28	388	13	11.1	4.17
Sweden	SE11	Fräcksjön	23	6.08	6.48	3.12	1.08	6.17	0.82	10.77	4.28	137	66	17	63	358	8	9.6	0.37
Sweden	SE12	Härsvatten	23	5.22	4.80	0.60	0.71	5.88	0.43	10.60	3.66	-26	-25	32	84	290	3	3.8	16.03
UK	UK01	Scotland, Loch Coire nan Arr	8	4.55	6.12	0.74	0.76	5.63	0.30	11.49	1.50	-10	24		33	129		3.6	0.94
UK	UK04	Scotland, Lochnagar	13	1.83	5.55	0.45	0.32	1.81	0.18	2.71	2.03	-1	2		190	273		2.8	3.03
UK	UK07	Scotland, Round Loch of Glenhead	13	3.38	5.13	0.59	0.50	3.59	0.31	6.75	2.08	-9	-6		143	333		5.1	8.06
UK	UK10	England, Scoat Tarn	13	3.19	5.17	0.51	0.50	3.28	0.22	6.15	2.28	-19	-5		188	273		2.0	7.14
UK	UK15	Wales, Llyn Llagi	12	2.73	5.70	0.87	0.50	3.46	0.14	6.25	1.91	17	4		81	281		3.4	2.40
UK	UK21	N.Ireland, Blue Lough	13	4.76	4.91	0.55	0.56	5.23	0.51	9.34	2.81	-28	-12		273	554		6.0	12.86
USA	US05	Maine, Little Long Pond	4	1.85	5.75	0.72	0.27	1.97	0.25	2.46	2.57	27	10	14	0			2.4	1.80
USA	US06	Maine, Tilden Pond	4	1.96	6.27	1.13	0.33	2.14	0.27	2.20	2.10	78	46	23	6			3.7	0.54
USA	US100	Howe, Vermont	12	1.60	6.11	1.43	0.31	0.60	0.38	0.48	2.64	57	46		111			4.8	1.01
USA	US102	Forester, Vermont	12	10.38	5.42	1.54	0.29	15.04	0.47	26.05	3.47	-51	7		63			3.8	5.06
USA	US103	Paine Run, Virginia	103	2.05	5.78	0.60	0.62	0.51	1.86	0.80	5.23	17	6	7	25				2.02
USA	US104	Piney River, Virginia	112	3.55	7.06	2.97	1.45	1.79	0.28	1.00	3.16	256	229	22	34				0.10
USA	US105	Staunton River, Virginia	102	1.83	6.61	1.35	0.34	1.42	0.43	0.77	2.23	100	85	13	2				0.31
USA	US11	New York, Adirondack Mnt., Arbutus	47	2.34	6.37	2.55	0.45	0.71	0.25	0.34	4.53	91	67	11	96			5.1	0.51
USA	US12	New York, Adirondack Mnt., Constable	47	2.00	5.27	1.40	0.25	0.58	0.32	0.29	3.99	18	15	18	205			6.0	8.29
USA	US123	Haystack, Vermont	13	1.27	5.06	0.60	0.18	0.34	0.10	0.43	2.25	-15	-7		252			1.1	9.48
USA	US125	Lily, Vermont	1	1.52	6.28	1.01	0.37	0.82	0.43	0.70	3.00	45	36		3				0.52
USA	US126	Little, Winhall, Vermont	1	1.30	5.40	0.97	0.21	0.42	0.26	0.30	2.00	11	6		410				3.98
USA	US13	New York, Adirondack Mnt., Dart Lake	47	1.82	5.70	1.52	0.26	0.60	0.29	0.38	3.74	27	20	20	204			4.6	2.58
USA	US14	New York, Adirondack Mnt., Heart Lake	47	1.63	6.36	1.75	0.24	0.52	0.08	0.27	3.17	51	44	19	103			2.3	0.53
USA	US15	New York, Adirondack Mnt., Lake Rondaxe	47	2.09	6.23	1.91	0.37	0.77	0.34	0.39	3.82	64	52	22	187			4.1	0.81
USA	US16	New York, Adirondack Mnt., Moss Lake	47	2.43	6.42	2.33	0.46	0.87	0.37	0.42	4.17	86	74	19	234			3.9	0.60
USA	US17	New York, Adirondack Mnt., Otter Lake	47	1.87	5.65	1.31	0.34	0.67	0.22	0.36	4.31	13	20	8	214			2.5	3.43
USA	US23	New York, Catskill Mnt., E. Branch Neversink, Head	153	1.89	5.05	0.98	0.43	0.29	0.23	0.43	3.86	-11	-4	25	307			2.9	10.65
USA	US24	New York, Catskill Mnt., Rondout Creek	108	1.79	5.43	1.31	0.43	0.34	0.62	0.49	4.14	13	5	27	275			2.2	5.39

Country	ID	Station name	n	K25	pH	Ca	Mg	Na	K	Cl	SO4	ANC	Alk	NH4-N	NO3-N	TOTN	TOTP	TOC/DOC	H+
				mS/m		mg/l	mg/l	mg/l	mg/l	mg/l	mg/l	µEq/l	µEq/l	µg/l N	µg/l N	µg/l N	µg/l P	mg/l C	µEq/l
USA	US25	W Br Neversink R At Winnisook, Catskills	148	2.05	4.85	0.76	0.34	0.25	0.18	0.40	3.98	-35	-10	25	315			2.7	16.30
USA	US26	Biscuit Brook, Catskills	184	1.87	5.85	1.98	0.43	0.30	0.22	0.46	3.84	34	21	25	363			2.4	2.19
USA	US27	Little Hope Pond, Adirondacks	47	1.88	5.78	1.85	0.48	0.60	0.28	0.29	2.80	96	44	30	32			11.7	2.03
USA	US28	Big Hope Pond, Adirondacks	47	2.01	6.22	1.95	0.52	0.73	0.29	0.66	3.00	96	54	21	33			8.5	0.70
USA	US29	East Copperas Pond, Adirondacks	47	1.80	4.59	0.59	0.12	0.14	0.38	0.24	1.42	17	-16	141	31			12.0	27.11
USA	US30	Sunday Pond, Adirondacks	47	1.13	5.03	0.52	0.18	0.07	0.27	0.19	2.49	-8	-3	15	18			1.5	9.65
USA	US31	Sochia Pond, Adirondacks	47	1.13	4.82	0.33	0.09	0.09	0.14	0.22	1.48	-8	-6	99	34			4.1	15.57
USA	US32	Marcy Dam Pond, Adirondacks	47	1.75	5.81	1.53	0.25	0.61	0.06	0.25	3.23	22	25	11	387			3.1	2.25
USA	US33	Grass Pond, Adirondacks	47	1.64	4.56	0.47	0.14	0.11	0.28	0.22	1.31	11	-19	53	33			9.4	27.95
USA	US34	Little Clear Pond, Adirondacks	47	1.75	6.46	2.02	0.53	0.10	0.41	0.19	1.26	126	103	141	23			7.0	0.39
USA	US35	Loon Hollow Pond, Adirondacks	45	2.04	4.65	0.51	0.12	0.36	0.21	0.27	3.58	-38	-15	30	171			5.0	23.42
USA	US36	Willys Lake, Adirondacks	45	1.93	4.87	0.90	0.15	0.41	0.27	0.28	4.00	-27	-5	14	244			3.2	14.54
USA	US37	Woods Lake, Adirondacks	46	1.86	5.93	2.03	0.22	0.41	0.23	0.28	3.70	41	28	22	250			4.4	1.85
USA	US38	Middle Settlement Lake, Adirondacks	47	1.44	5.59	0.90	0.19	0.60	0.23	0.25	3.15	14	16	54	78			3.3	3.78
USA	US39	Grass Pond, Adirondacks	47	2.03	5.79	1.46	0.31	0.86	0.30	0.29	4.03	30	30	18	308			4.1	3.59
USA	US40	Middle Branch Lake, Adirondacks	45	1.93	6.32	1.65	0.37	0.86	0.33	0.27	3.61	71	56	18	73			4.2	0.78
USA	US41	Limekiln Lake, Adirondacks	47	1.85	6.13	1.71	0.30	0.67	0.23	0.57	3.55	44	34	11	163			3.4	1.03
USA	US42	Squaw Lake, Adirondacks	46	1.53	5.89	1.33	0.33	0.38	0.20	0.27	3.32	30	21	21	117			3.5	1.62
USA	US43	Indian Lake, Adirondacks	46	1.71	5.02	1.00	0.24	0.39	0.17	0.24	3.20	4	3	18	182			5.8	11.02
USA	US44	Brook Trout Lake, Adirondacks	45	1.54	5.43	0.95	0.23	0.43	0.18	0.27	3.49	-2	6	31	159			2.6	5.36
USA	US45	Lost Pond, Adirondacks	45	1.91	5.09	1.07	0.27	0.58	0.17	0.24	3.39	9	7	35	264			6.2	11.38
USA	US46	South Lake, Adirondacks	47	1.66	5.50	1.13	0.24	0.48	0.20	0.26	3.31	6	9	18	289			3.0	5.39
USA	US47	North Lake, Adirondacks	47	1.87	5.24	1.21	0.28	0.51	0.23	0.25	3.54	8	8	12	315			5.0	8.12
USA	US48	Willis Lake, Adirondacks	29	2.31	6.04	2.48	0.44	0.90	0.11	0.77	3.47	106	77	21	33			6.4	1.02
USA	US49	Long Pond, Adirondacks	47	2.41	4.58	0.99	0.35	0.47	0.31	0.31	3.22	29	-16	31	24			15.4	27.86
USA	US50	Carry Pond, Adirondacks	45	1.01	5.50	0.64	0.15	0.20	0.33	0.24	1.77	16	14	107	32			3.5	3.69
USA	US51	Lake Colden, Adirondacks	47	1.79	5.18	1.28	0.20	0.46	0.07	0.22	3.28	-5	8	13	447			4.3	7.58
USA	US52	Avalanche Lake, Adirondacks	47	1.93	5.03	1.28	0.19	0.42	0.10	0.23	3.03	-10	6	27	562			5.7	10.78
USA	US53	Little Simon Pond, Adirondacks	47	2.67	6.57	3.30	0.30	0.63	0.21	0.34	4.51	97	92	11	288			3.6	0.85
USA	US54	Raquette Lake Reservoir, Adirondacks	47	2.26	5.48	1.80	0.41	0.77	0.34	0.36	4.25	54	33	17	189			8.5	5.40
USA	US55	G Lake, Adirondacks	47	1.61	5.61	1.07	0.27	0.49	0.16	0.25	3.34	7	14	12	240			2.8	4.88
USA	US57	Sagamore Lake, Adirondacks	47	2.29	5.87	2.00	0.50	0.79	0.31	0.32	4.41	64	40	10	255			7.8	2.05
USA	US58	Black Pond Outlet, Adirondacks	47	3.43	6.97	3.48	1.06	1.04	0.36	0.32	4.05	217	209	18	70			3.9	0.12

Country	ID	Station name	n	K25 mS/m	pH	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	SO4 mg/l	ANC µEq/l	Alk µEq/l	NH4-N µg/l N	NO3-N µg/l N	TOTN µg/l N	TOTP µg/l P	TOC/DOC mg/l C	H+ µEq/l
USA	US59	Windfall Pond Outle, Adirondacks	47	2.30	6.24	2.44	0.45	0.53	0.27	0.29	4.34	71	58	18	264			4.7	0.79
USA	US60	Queer Lake, Adirondacks	47	1.79	5.76	1.56	0.28	0.44	0.29	0.30	3.96	22	18	28	200			3.4	2.63
USA	US61	Big Moose Lake, Adirondacks	47	1.81	5.61	1.45	0.25	0.59	0.29	0.38	3.77	23	17	20	204			4.7	3.26
USA	US62	Cascade Lake Outlet, Adirondacks	47	2.66	6.46	2.60	0.51	0.99	0.37	0.35	4.36	103	93	10	285			4.2	0.45
USA	US63	Little Echo Pond, Adirondacks	47	2.54	4.34	0.55	0.32	0.23	0.18	0.41	1.91	16	-39	84	19			15.4	45.56
USA	US64	Squash Pond Outlet, Adirondacks	42	2.37	4.55	0.70	0.14	0.42	0.24	0.26	3.59	-30	-21	28	268			7.4	28.78
USA	US65	West Pond Outlet, Adirondacks	47	1.82	5.00	1.13	0.23	0.53	0.23	0.24	3.24	23	4	36	102			8.4	11.42
USA	US66	Bubb Lake Outlet, Adirondacks	47	1.92	6.26	1.71	0.37	0.74	0.33	0.27	3.82	57	44	11	167			3.3	0.59
USA	US67	Owen Pond, Adirondacks	47	3.08	6.65	3.47	0.62	0.82	0.18	0.37	4.71	125	113	17	440			5.1	0.25
USA	US68	Jockeybush Lake, Adirondacks	47	1.72	5.42	1.18	0.29	0.45	0.17	0.29	3.84	-2	8	18	282			2.4	4.96
USA	US69	Clear Pond, Adirondacks	47	2.30	6.83	2.69	0.32	0.88	0.08	0.27	3.68	111	101	10	73			3.6	0.17
USA	US70	Nate Pond, Adirondacks	45	2.25	6.37	2.19	0.54	0.74	0.31	0.29	4.27	89	64	15	118			5.6	0.57
USA	US71	Bean Pond, Maine	9	1.56	6.19	1.48	0.42	0.82	0.34	0.46	2.06	97	51	6	2			7.6	0.74
USA	US72	Bracey Pond, Maine	9	1.90	6.70	2.19	0.35	1.28	0.31	1.25	1.13	142	102	15	4			5.6	0.23
USA	US73	Anderson Pond, Maine	3	1.49	5.79	0.59	0.23	1.67	0.20	1.84	2.12	30	13	27	5			2.3	1.70
USA	US74	Mud Pond, Maine	8	2.46	4.74	0.42	0.22	1.85	0.23	2.42	3.09	-8	-17	9	0			5.3	18.91
USA	US75	Salmon Pond, Maine	9	1.93	6.46	1.26	0.33	1.85	0.24	2.00	2.07	77	55	10	6			3.0	0.35
USA	US76	Wiley Pond, Maine	9	2.21	6.66	3.55	0.59	0.74	0.16	0.35	1.75	215	147	5	3			10.1	0.24
USA	US77	Second Pond, Maine	9	1.99	6.40	1.59	0.39	1.60	0.28	1.21	2.35	105	63	9	3			4.8	0.43
USA	US78	Abol Pond, Maine	9	2.76	6.92	3.17	0.39	1.59	0.85	0.53	2.65	211	179	4	6			2.4	0.12
USA	US79	Duck Pond, Maine	9	1.88	4.55	0.20	0.13	0.69	0.20	1.10	1.03	3	-29	3	4			7.1	28.58
USA	US80	Jellison Hl Pd, Maine	9	2.04	6.16	1.29	0.36	1.91	0.25	1.80	2.66	77	39	0	1			5.2	0.78
USA	US81	Crystal Pond, Maine	9	0.91	5.50	0.30	0.13	0.65	0.16	0.85	1.22	8	-1	7	8			2.4	3.19
USA	US82	Newbert Pond, Maine	8	2.59	4.74	1.36	0.40	1.37	0.34	1.34	1.80	92	-7	19	17			18.0	19.50
USA	US83	Partridge Pond, Maine	9	1.44	6.12	0.93	0.23	1.42	0.25	1.10	2.07	59	30	5	6			3.8	0.85
USA	US84	Benner Run, Mid-Apps	53	2.69	5.91	1.60	0.61	1.50	0.80	3.37	4.49	18	18	11	98			1.5	1.99
USA	US85	Linn Run, Mid-Apps	53	3.40	5.99	3.56	0.67	0.67	0.43	0.98	9.14	43	48	16	210			1.1	1.72
USA	US86	Roberts Run, Mid-Apps	53	2.74	5.46	1.81	0.81	0.60	0.60	0.83	7.80	7	8	16	77			1.7	6.43
USA	US87	Stone Run, Mid-Apps	53	2.58	5.35	1.70	0.67	0.57	0.49	0.78	7.75	-10	-3	14	42			1.2	7.06
USA	US88	Baldwin Creek, Mid-Apps	53	3.56	6.23	3.30	0.99	0.57	0.69	1.11	9.38	44	49	11	288			0.9	0.75
USA	US89	Bourn, Vermont	12	1.12	5.64	0.64	0.28	0.50	0.35	0.31	1.98	29	13		81			5.2	2.66
USA	US90	Grout, Vermont	12	1.30	6.14	1.00	0.29	0.49	0.43	0.43	2.32	40	29		70			3.9	0.91
USA	US91	Hardwood, Vermont	12	1.54	6.41	1.57	0.43	0.44	0.11	0.24	2.66	69	50		61			5.1	0.41

Country	ID	Station name	n	K25 mS/m	pH	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	SO4 mg/l	ANC µEq/l	Alk µEq/l	NH4-N µg/l N	NO3-N µg/l N	TOTN µg/l N	TOTP µg/l P	TOC/DOC mg/l C	H+ µEq/l
USA	US92	Little ¿ Woodford, Vermont	12	1.50	5.30	0.82	0.21	0.58	0.37	0.34	3.30	-2	1		230			2.1	6.06
USA	US93	Stamford, Vermont	12	1.59	5.69	1.13	0.26	0.75	0.30	0.43	3.15	23	15		235			3.3	2.64
USA	US94	Stratton, Vermont	1	1.16	5.96	0.88	0.23	0.44	0.33	0.30	2.10	29	20		130				1.10
USA	US95	Sunset, Vermont	12	1.32	6.17	0.96	0.26	0.68	0.19	0.62	2.83	23	23		60			1.9	0.78
USA	US96	Big Mud, Vermont	12	1.41	5.37	1.13	0.27	0.38	0.29	0.33	2.39	36	13		91			7.7	5.14
USA	US97	Branch, Vermont	12	1.36	4.95	0.53	0.20	0.46	0.32	0.31	2.36	6	-5		107			5.7	11.75
USA	US98	Beaver Pond, Vermont	12	1.79	6.46	2.21	0.27	0.56	0.19	0.37	2.60	92	66		75			6.1	0.38

Table B2. *Mean results for samples collected between 2005 and 2008 (n indicates the number of observations) from extra Ontario stations. Andrew Paterson (Ontario Ministry of Environment) provided volume weighted data from 8 lakes from the Dorset area (first 8 rows). Bill Keller (Laurentian University and Ontario Ministry of Environment) provided data from 5 lakes from the Sudbury area (last 5 rows).*

Country	ID	Station name	n	K25 mS/m	pH	Ca mg/l	Mg mg/l	Na mg/l	K mg/l	Cl mg/l	SO4 mg/l	ANC µEq/l	Alk µEq/l	NH4-N µg/l N	NO3-N µg/l N	TOTN µg/l N	TOTP µg/l P	TOC/DOC mg/l C	H+ µEq/l
Canada	29230	Blue Chalk Lake	19		6.612	2.31	0.66	0.78	0.36	0.38	4.82	100.36	96.29	22.81	10.75			2.20	0.33
Canada	29231	Chub Lake	20		5.672	1.84	0.54	0.72	0.27	0.40	5.46	46.49	20.69	17.45	48.04			5.94	2.47
Canada	29232	Crosson Lake	20		5.693	1.63	0.47	0.62	0.35	0.37	4.91	39.48	19.23	24.84	59.31			5.12	2.28
Canada	29233	Dickie Lake	38		6.012	3.05	0.68	2.57	0.39	5.54	5.13	66.36	37.83	27.51	22.71			5.74	1.17
Canada	29234	Harp Lake	40		6.268	2.75	0.85	2.09	0.48	2.63	6.25	98.87	74.79	12.75	100.62			4.05	0.67
Canada	29235	Heney Lake	21		5.852	1.51	0.44	0.97	0.27	1.30	4.69	25.12	11.19	19.00	13.90			3.97	1.73
Canada	29236	Plastic Lake	37		5.603	1.34	0.36	0.51	0.18	0.35	5.00	9.59	7.01	14.57	8.51			2.32	2.92
Canada	29237	Red Chalk Lake	19		6.308	2.07	0.69	0.74	0.37	0.35	5.04	81.12	70.75	12.15	85.00			3.18	0.73
Canada	29377	Clearwater	12	6.53	6.28	4.27	1.08	11.07	0.56	9.55	10.09	316.98		20.17	9.00	185.00	3.68	2.67	0.53
Canada	29400	Sans Chambre	12	2.06	6.257	1.73	0.51	0.75	0.33	0.26	5.41	50.35		34.18	18.91	209.75	7.56	2.43	0.57
Canada	29411	Whitepine (Aurora)	11	1.81	5.231	1.12	0.41	0.59	0.29	0.20	5.78	-3.42		16.55	7.82	126.55	4.62	2.81	5.94
Canada	35405	Whitepine (Mcleod)	12	2.04	6.234	1.73	0.57	0.70	0.27	0.18	5.83	43.69		18.33	9.33	164.83	5.96	3.30	0.59
Canada	35406	Swan Lake	24	4.41		3.21	0.64	2.54	0.30	6.47	6.95	2.71		45.29	11.63	292.99	7.61	2.78	

Appendix C. Sites without trend analysis

Site	Trend 1990-1999	Insensitive to acidification	Record too short	Possible other sources of sulphate	Other disturbances in the catchment	Trend 1999-2008	Insensitive to acidification	Record too short	Possible other sources of sulphate	Other disturbances in the catchment
BY01	No	*	*			Only sulphate	*			
EE01	Only sulphate	*				Only sulphate	*			
LV01	Only sulphate	*				No	*	*		
LV02	Only sulphate	*				No	*	*		
LV03	Only sulphate	*				Only sulphate	*			
LV04	Only sulphate	*				Only sulphate	*			
LV05	Yes					No				*
LV06	No	*	*			No	*	*		
LV07	No	*	*			No	*			*
LV08	No	*	*			No	*	*		
PL03	No		*			No		*		
PL04	No		*			No		*		
CH03	No		*			Yes				
CH05	No		*			Yes				
CH06	No		*			Yes				
CH09	No		*			Yes				
CH19	No		*			Yes				
CH20	No		*			Yes				
CH26	No		*			Yes				
CH27	No		*			Yes				
CH28	No		*			Yes				
DE04	No			*		No			*	
DE09	No			*		No			*	
DE11	Yes					No		*		
DE13	No		*			No		*		
DE14	No				*	No				*
DE15	Yes					No		*		
DE16	No		*			No		*		
DE17	Yes					No				*
DE19	Yes					No		*		

Site	Trend 1990-1999	Insensitive to acidification	Record too short	Possible other sources of sulphate	Other disturbances in the catchment	Trend 1999-2008	Insensitive to acidification	Record too short	Possible other sources of sulphate	Other disturbances in the catchment
DE20	Yes					No		*		
DE22	Yes					No		*		
DE25	No			*		No			*	
DE34	Yes					No		*		
DE35	No		*			Yes				
CA19	No		*			Yes		*		
US82	No		*			Yes				
US83	No		*			Yes				
US25	No		*			Yes				
US94	No		*			Yes				
US101	No		*			Yes				
US122	No		*			Yes				
US125	No		*			Yes				
US126	No		*			Yes				
US124	No		*			Yes				

Appendix D. Results of Mann-Kendall trend analysis for individual sites

Table D1. *Results of Mann-Kendall trend analysis for individual sites 1990-1999. Slope is annual change in the unit given for each parameter, p is the probability that there is no trend and n is the number of years with observations. Significant results (p<0.05) are written in bold. Results from extra Ontario stations are given in a table below.*

1990-1999		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Adirondacks	US11	**10**	**0.002**	**-2.68**	10	0.089	-0.63	10	0.325	0.00	**10**	**0.003**	**-4.47**	10	0.788	-0.51	**10**	**0.003**	**-2.24**	10	0.929	0.01	10	0.325	0.01
Adirondacks	US12	**10**	**0.025**	**-2.45**	10	0.245	-1.27	10	0.531	0.00	**10**	**0.025**	**-3.20**	**10**	**0.025**	**1.76**	10	0.531	1.64	10	0.072	0.10	**10**	**0.040**	**-0.76**
Adirondacks	US13	**10**	**0.003**	**-2.97**	10	0.128	-1.04	10	0.655	0.00	**10**	**0.003**	**-2.20**	**10**	**0.003**	**1.38**	10	0.421	0.76	10	0.089	0.10	10	0.089	-0.52
Adirondacks	US14	**10**	**0.001**	**-1.87**	10	0.788	-0.29	10	0.788	0.00	**10**	**0.025**	**-2.12**	10	0.929	0.08	10	0.325	-1.44	10	0.655	-0.01	10	0.531	0.01
Adirondacks	US15	**10**	**0.000**	**-2.39**	**10**	**0.009**	**-1.71**	10	0.180	-0.01	**10**	**0.040**	**-1.99**	**10**	**0.016**	**1.50**	10	0.788	0.73	10	0.089	0.06	10	0.531	-0.13
Adirondacks	US16	**10**	**0.009**	**-2.32**	**10**	**0.040**	**-1.55**	10	0.128	-0.01	10	0.060	-3.47	10	0.060	0.72	10	0.421	-1.07	10	0.531	0.02	10	0.929	-0.02
Adirondacks	US17	**10**	**0.003**	**-2.02**	10	0.060	-1.33	10	0.245	-0.01	**10**	**0.002**	**-2.36**	**10**	**0.025**	**1.45**	10	0.788	0.22	10	0.209	-0.03	**10**	**0.001**	**-0.39**
Adirondacks	US27	8	0.458	-1.36	**8**	**0.006**	**0.84**	**8**	**0.006**	**0.01**	8	0.458	-1.58	8	0.322	0.71	8	0.621	-0.60	8	0.805	0.02	8	0.216	0.82
Adirondacks	US28	8	0.621	-0.47	8	0.083	0.31	8	0.138	0.00	8	1.000	-0.27	8	0.083	1.24	8	1.000	-0.17	8	0.322	0.18	8	0.805	0.09
Adirondacks	US29	8	0.083	1.31	**8**	**0.048**	**0.18**	8	0.138	0.00	8	0.805	-0.17	8	0.621	-0.29	**8**	**0.026**	**-1.56**	8	0.805	-0.08	8	0.621	0.26
Adirondacks	US30	8	0.216	-1.15	8	0.805	-0.15	8	0.805	0.00	**8**	**0.026**	**-2.07**	8	0.621	0.31	8	0.138	-1.17	8	0.138	-0.11	8	0.458	0.33
Adirondacks	US31	**8**	**0.026**	**-2.52**	8	0.216	0.46	8	0.138	0.01	8	0.216	-0.53	**8**	**0.048**	**2.17**	**8**	**0.048**	**0.85**	8	1.000	0.00	8	0.083	-1.80
Adirondacks	US32	**8**	**0.003**	**-2.92**	8	0.458	0.26	8	0.322	0.01	8	0.216	-2.84	8	0.458	1.05	8	0.621	-0.38	8	1.000	0.00	8	0.138	-0.17
Adirondacks	US33	**8**	**0.003**	**-1.69**	8	0.458	0.23	8	0.138	0.01	**8**	**0.026**	**-0.77**	8	0.083	1.01	8	0.621	0.47	8	0.322	0.10	8	0.458	-0.28
Adirondacks	US34	**8**	**0.048**	**-1.28**	8	0.458	0.29	8	0.216	0.01	**8**	**0.003**	**22.55**	**8**	**0.001**	**25.56**	**8**	**0.013**	**23.98**	8	0.901	-0.01	**8**	**0.003**	**-2.15**
Adirondacks	US36	**8**	**0.003**	**-4.44**	8	0.805	-0.20	8	0.322	0.00	**8**	**0.003**	**-2.65**	8	0.138	1.43	8	0.138	1.95	8	0.322	-0.09	8	0.216	-0.75
Adirondacks	US37	**8**	**0.013**	**-4.18**	8	0.138	1.36	**8**	**0.026**	**0.01**	**8**	**0.006**	**-7.01**	8	0.083	-2.38	8	0.083	-5.41	8	0.621	-0.06	8	0.138	0.24
Adirondacks	US38	**8**	**0.048**	**-2.08**	8	0.805	0.30	8	0.458	0.00	8	0.138	-0.46	8	0.083	1.61	8	0.805	0.10	8	0.805	-0.01	8	0.138	-0.35
Adirondacks	US39	**8**	**0.013**	**-1.32**	8	1.000	-0.14	8	1.000	0.00	8	0.138	-1.28	8	0.805	0.86	8	0.805	-1.09	8	1.000	0.00	8	0.322	-0.16
Adirondacks	US40	8	0.322	-0.66	8	0.621	0.33	8	0.458	0.00	8	0.322	-0.80	8	0.621	2.01	8	0.621	-0.70	8	0.805	-0.01	8	0.621	0.13
Adirondacks	US41	**8**	**0.003**	**-3.11**	8	1.000	-0.06	8	0.621	0.00	8	0.138	-2.68	**8**	**0.026**	**2.00**	8	0.805	0.63	8	1.000	0.00	8	1.000	-0.02
Adirondacks	US42	**8**	**0.003**	**-5.32**	8	0.138	-1.19	8	0.458	-0.01	**8**	**0.006**	**-4.76**	**8**	**0.013**	**2.08**	8	0.322	0.70	8	0.805	0.04	8	0.621	-0.02
Adirondacks	US43	**8**	**0.013**	**-4.92**	8	0.322	-1.39	8	0.458	-0.01	**8**	**0.006**	**-3.52**	8	0.083	1.16	8	0.458	0.59	8	0.805	0.05	8	0.138	-0.65
Adirondacks	US44	**8**	**0.001**	**-4.34**	**8**	**0.048**	**-1.41**	8	0.083	-0.01	**8**	**0.013**	**-2.75**	8	0.083	1.85	8	0.083	2.98	8	0.216	0.16	8	0.322	-0.63
Adirondacks	US45	8	0.083	-1.36	8	0.138	-2.69	8	0.216	-0.02	**8**	**0.013**	**-3.86**	8	0.138	1.71	8	1.000	-0.01	8	0.805	-0.04	8	0.083	-1.02
Adirondacks	US46	**8**	**0.006**	**-2.30**	8	1.000	0.00	8	0.322	0.01	**8**	**0.048**	**-2.03**	8	0.138	1.51	8	1.000	-0.16	8	1.000	0.00	8	0.458	-0.71

1990-1999		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Adirondacks	US47	8	0.083	-1.37	8	0.621	-0.81	8	1.000	0.00	**8**	**0.048**	**-3.25**	8	0.216	0.97	8	1.000	-0.19	8	1.000	-0.01	8	0.322	-0.34
Adirondacks	US48	8	0.621	0.04	8	0.083	0.23	8	0.083	0.00	8	0.322	-1.87	8	0.458	1.87	8	0.458	-2.15	8	0.621	0.17	8	0.805	-0.04
Adirondacks	US49	**8**	**0.003**	**-4.76**	8	0.805	0.13	8	0.138	0.00	**8**	**0.006**	**-4.44**	8	0.458	0.63	8	0.621	0.48	8	0.621	0.13	8	0.458	-0.38
Adirondacks	US50	8	0.216	-1.23	8	0.805	-0.13	8	0.621	0.00	**8**	**0.013**	**-2.07**	8	0.322	1.12	8	0.458	-1.01	8	0.083	-0.03	8	0.458	-0.46
Adirondacks	US51	**8**	**0.048**	**-3.18**	8	1.000	0.19	8	0.458	0.00	8	0.083	-2.91	**8**	**0.013**	**1.03**	8	0.805	-0.05	8	0.805	-0.06	**8**	**0.006**	**-0.38**
Adirondacks	US52	**8**	**0.048**	**-4.16**	8	0.216	1.09	8	0.083	0.01	8	0.083	-3.85	8	0.216	0.29	8	0.805	-0.49	8	0.805	0.02	8	0.805	0.15
Adirondacks	US53	**8**	**0.006**	**-3.80**	8	0.083	1.26	**8**	**0.013**	**0.01**	**8**	**0.048**	**-10.32**	8	0.138	-4.63	**8**	**0.048**	**-8.48**	8	0.805	-0.04	8	0.138	0.16
Adirondacks	US54	**8**	**0.026**	**-3.38**	8	1.000	-0.02	8	0.621	0.00	8	0.216	-3.68	8	0.458	1.03	8	0.805	-1.07	8	0.322	-0.09	8	1.000	0.00
Adirondacks	US55	**8**	**0.013**	**-2.07**	8	0.621	0.96	8	0.458	0.01	8	0.138	-2.41	8	0.216	1.80	8	0.805	-1.16	8	0.458	-0.01	8	0.805	-0.26
Adirondacks	US57	**8**	**0.048**	**-2.84**	8	0.322	-0.97	8	0.458	0.00	8	0.083	-4.68	8	1.000	-0.02	8	0.621	-1.47	8	0.621	0.07	8	0.805	0.03
Adirondacks	US58	**10**	**0.025**	**-0.97**	10	0.929	0.06	10	0.788	0.00	10	0.089	-3.00	10	0.325	-0.40	10	0.180	-2.30	10	0.060	-0.12	10	0.421	0.00
Adirondacks	US59	10	0.060	-2.49	10	0.245	-1.31	10	0.655	0.00	10	0.089	-1.93	**10**	**0.016**	**2.77**	10	0.929	0.48	10	0.531	0.03	10	0.325	-0.05
Adirondacks	US60	**8**	**0.013**	**-3.45**	8	0.805	0.10	8	0.458	0.00	**8**	**0.006**	**-4.70**	8	0.138	0.56	8	0.621	-0.43	8	0.621	-0.03	8	0.621	-0.10
Adirondacks	US61	**10**	**0.016**	**-2.41**	**10**	**0.025**	**-1.95**	10	0.245	-0.01	**10**	**0.016**	**-2.83**	**10**	**0.003**	**1.41**	10	0.788	0.17	10	0.421	0.05	**10**	**0.009**	**-0.53**
Adirondacks	US62	**10**	**0.009**	**-2.31**	**10**	**0.006**	**-1.70**	**10**	**0.040**	**-0.01**	**10**	**0.016**	**-5.07**	10	0.655	-0.52	10	0.421	-3.75	**10**	**0.031**	**0.10**	10	0.655	-0.01
Adirondacks	US63	**10**	**0.016**	**-1.42**	10	0.929	0.00	10	0.421	0.00	10	0.089	0.97	**10**	**0.002**	**2.02**	**10**	**0.040**	**1.02**	**10**	**0.025**	**0.28**	**10**	**0.040**	**-1.49**
Adirondacks	US64	**10**	**0.016**	**-2.37**	10	0.531	-1.15	10	0.929	0.00	**10**	**0.002**	**-1.78**	**10**	**0.002**	**1.08**	10	0.531	0.57	10	0.128	0.08	**10**	**0.025**	**-0.57**
Adirondacks	US65	**10**	**0.016**	**-3.05**	10	0.128	-0.46	10	0.788	0.00	**10**	**0.003**	**-5.71**	**10**	**0.040**	**-1.63**	**10**	**0.016**	**-3.79**	10	0.128	0.19	10	0.089	0.69
Adirondacks	US66	**10**	**0.006**	**-1.88**	10	0.531	-0.36	10	0.929	0.00	10	0.060	-2.22	10	0.089	1.20	10	0.531	-0.62	10	0.325	0.04	10	0.655	-0.01
Adirondacks	US67	8	0.083	-3.58	8	0.458	1.17	8	0.216	0.01	8	0.138	-6.53	8	0.805	0.34	8	0.322	-2.89	8	0.322	-0.05	8	1.000	0.00
Adirondacks	US68	**8**	**0.001**	**-3.10**	8	0.322	1.59	8	0.083	0.02	**8**	**0.026**	**-1.56**	**8**	**0.003**	**1.28**	8	1.000	-0.08	8	0.083	-0.08	**8**	**0.048**	**-0.67**
Adirondacks	US69	10	0.060	-1.38	10	0.929	0.05	10	0.788	0.00	10	0.060	-3.56	10	0.180	-1.55	**10**	**0.025**	**-3.46**	10	0.421	-0.04	10	0.245	0.00
Adirondacks	US70	8	0.138	-0.97	8	1.000	0.15	8	0.458	0.00	**8**	**0.048**	**-2.75**	8	0.458	-0.55	**8**	**0.048**	**-4.56**	8	0.621	-0.03	8	0.805	0.03
Alps	CH03																								
Alps	CH05																								
Alps	CH06																								
Alps	CH09																								
Alps	CH19																								
Alps	IT01	**10**	**0.000**	**-1.96**	10	0.929	0.00	10	0.060	0.01	**10**	**0.006**	**-2.22**	10	0.060	0.81	10	0.655	-0.59				10	0.531	-0.02
Alps	IT02	**10**	**0.000**	**-4.16**	**10**	**0.048**	**0.40**	**10**	**0.002**	**0.00**	9	0.211	-1.86	**10**	**0.004**	**2.44**	9	1.000	0.33				10	0.649	0.00
Alps	IT03	**10**	**0.009**	**-1.26**	10	0.245	0.58	**10**	**0.002**	**0.01**	10	0.245	-1.04	9	0.144	1.24	10	0.531	-0.34				10	0.089	-0.11
Alps	IT04	**10**	**0.006**	**-2.48**	10	0.531	-0.33	**10**	**0.025**	**0.00**	10	0.788	-1.22	10	0.325	0.83	10	0.531	1.90				10	0.128	0.00
Alps	IT05	10	0.180	-0.70	10	0.128	2.27	**10**	**0.040**	**0.01**	10	0.128	4.67	10	0.180	2.90	10	0.531	1.68				**10**	**0.040**	**0.00**
Alps	IT06	**10**	**0.000**	**-2.21**	**10**	**0.016**	**0.95**	**10**	**0.002**	**0.01**	10	0.325	-1.73	10	0.531	0.43	10	0.421	-0.69				10	0.128	0.00

1990-1999		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Appalachians	US23	**10**	**0.009**	**-2.96**	**10**	**0.040**	**-1.31**	10	0.245	-0.01				10	0.089	-0.50				**10**	**0.025**	**0.20**	10	0.655	0.18
Appalachians	US24	**10**	**0.006**	**-2.75**	10	0.060	-1.17	10	0.325	-0.01				10	0.325	-0.26				**10**	**0.040**	**0.19**	10	0.421	0.10
Appalachians	US25																								
Appalachians	US26	**10**	**0.001**	**-3.83**	10	0.060	-2.09	10	0.180	-0.01				10	0.929	-0.08				10	0.089	0.08	10	0.128	-0.06
Appalachians	US35	**8**	**0.013**	**-4.48**	8	0.621	0.22	8	0.138	0.01	8	0.083	-1.41	**8**	**0.026**	**2.48**	8	0.216	1.85	8	0.216	-0.15	**8**	**0.013**	**-2.11**
Appalachians	US84	10	0.325	1.18	10	0.929	-0.03	10	0.929	0.00				**10**	**0.003**	**1.58**				10	0.929	-0.01	10	0.590	-0.06
Appalachians	US85	**10**	**0.040**	**-1.74**	10	0.180	-0.92	10	0.245	0.00				10	0.128	2.98				10	0.788	0.01	10	0.531	-0.11
Appalachians	US86	**10**	**0.009**	**-3.65**	10	0.655	-0.13	10	0.245	0.00				10	0.060	2.27				10	0.788	-0.04	10	0.788	0.02
Appalachians	US87	**10**	**0.025**	**-3.46**	10	0.788	0.09	10	0.325	0.00				10	0.245	1.09				10	0.655	-0.02	10	0.655	-0.18
Appalachians	US88	10	0.180	2.81	10	0.245	-3.23	10	0.245	-0.01				**10**	**0.009**	**3.19**				10	0.531	0.03	10	0.788	0.00
Blue Ridge Mountains	US103	10	0.531	0.40	10	0.128	-1.40	10	0.128	-0.01				10	0.531	-0.16							10	0.245	-0.03
Blue Ridge Mountains	US104	8	0.216	-1.26	**8**	**0.013**	**-3.07**	**8**	**0.013**	**-0.02**				8	0.216	3.37							8	0.083	0.00
Blue Ridge Mountains	US105	**10**	**0.025**	**0.51**	**10**	**0.002**	**-0.59**	**10**	**0.009**	**-0.01**				10	0.531	-1.17							10	0.788	0.00
ECEurope	CZ01	**10**	**0.000**	**-4.21**							**10**	**0.016**	**-1.68**	9	0.835	0.57							**10**	**0.040**	**-0.49**
ECEurope	CZ02	**10**	**0.003**	**-6.89**	10	0.128	-1.09	**10**	**0.040**	**0.01**	**10**	**0.009**	**-1.98**	10	0.128	1.08	**10**	**0.000**	**6.46**				10	0.531	-0.33
ECEurope	CZ03	**10**	**0.006**	**-9.15**	10	0.421	-1.09	10	0.788	0.00	**10**	**0.006**	**-2.98**	9	0.917	-0.18	**10**	**0.006**	**7.28**				**10**	**0.016**	**-1.00**
ECEurope	CZ04	**10**	**0.002**	**-4.07**	10	0.531	-0.77	10	0.180	0.01	10	0.421	-0.70	10	0.719	-0.07	**10**	**0.002**	**4.45**				10	0.060	-0.69
ECEurope	CZ05	10	0.180	-1.01	10	0.421	0.52	10	0.325	0.01	10	0.325	-0.46	10	0.325	-1.50	10	0.929	-0.14				10	0.325	0.13
ECEurope	CZ06	**10**	**0.040**	**-1.38**	10	0.929	0.04	10	0.531	0.00	10	0.060	-1.35	10	0.421	-2.98	10	0.531	-1.19				10	0.655	0.02
ECEurope	CZ07	**10**	**0.001**	**-26.99**	**10**	**0.009**	**-0.01**	10	0.128	0.00	**10**	**0.002**	**-13.63**	**10**	**0.040**	**3.71**	**10**	**0.001**	**13.90**	7	0.881	-0.12	**10**	**0.040**	**-3.60**
ECEurope	CZ08	7	0.293	-4.70	7	0.051	-0.03	7	0.099	0.00	7	0.051	-7.23	7	0.652	4.75	7	0.652	-2.68				7	0.453	0.62
ECEurope	DE02	10	0.325	-0.94	10	1.000	0.00	10	0.325	0.00	10	0.531	0.46	10	0.421	2.08				10	0.929	0.00	10	0.531	0.13
ECEurope	DE04	8	0.458	-69.29	**8**	**0.013**	**9.74**	**8**	**0.006**	**0.00**	**8**	**0.003**	**-136.95**	**7**	**0.033**	**-12.30**	8	0.216	-99.18				8	0.138	-7.95
ECEurope	DE07	7	0.051	-39.27	**8**	**0.006**	**-7.42**	7	0.881	0.00	7	0.652	-3.92	8	0.105	-4.20	**7**	**0.011**	**59.51**				8	0.216	-2.48
ECEurope	DE08	**9**	**0.012**	**-1.66**	10	0.180	1.33	**9**	**0.012**	**0.01**	9	0.677	-0.21				7	0.176	2.91	8	0.901	0.00	10	0.060	-0.20
ECEurope	DE09	8	1.000	1.37	8	0.322	1.64	8	0.458	0.00	**7**	**0.011**	**-138.11**				7	0.293	-289.73				**8**	**0.013**	**-40.46**
ECEurope	DE10	**10**	**0.025**	**-1.27**	**10**	**0.016**	**3.59**	10	0.060	0.02	10	0.180	1.13										10	0.325	-0.09
ECEurope	DE13				**7**	**0.024**	**-13.7**																7	0.881	0.02
ECEurope	DE17	**10**	**0.001**	**-6.22**	10	0.421	0.91	**10**	**0.016**	**0.02**	**10**	**0.025**	**-2.08**				10	0.060	2.31	10	0.856	0.00	10	0.128	-0.83
ECEurope	DE18	**10**	**0.006**	**-8.02**	10	0.369	-0.30	**10**	**0.025**	**0.00**	**10**	**0.040**	**-3.02**	10	0.857	-0.20				**10**	**0.004**	**0.16**	10	0.788	-0.76
ECEurope	DE21	8	0.216	-32.8	8	0.216	-4.59	8	0.805	0.00	8	0.322	-9.16	8	0.621	0.51	8	0.805	20.15				**8**	**0.048**	**-2.24**
ECEurope	DE23	**10**	**0.009**	**-1.70**	**10**	**0.025**	**-1.58**	10	0.655	0.00	**10**	**0.025**	**-1.01**										10	0.089	-0.33
ECEurope	DE24	9	0.144	-13.0	**9**	**0.004**	**-2.87**	9	0.532	0.00	9	0.061	-23.56										10	0.089	-0.77
ECEurope	DE25																								

1990-1999		SO4* (μeqv/L)			NO3 (μeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (μeqv/L)			Alkalinity (μeqv/L)			ANC (μeqv/L)			TOC_DOC (mg/L)			H+ (μeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
ECEurope	DE27	**10**	**0.002**	**-2.12**	10	0.421	0.79	10	0.089	0.01	10	0.788	0.61										10	0.929	0.00
ECEurope	DE28	9	1.000	-0.15	9	0.297	-1.15	9	0.835	-0.01	9	0.532	18.38	9	0.211	9.77	9	0.677	15.35	8	0.383	0.15	9	0.532	-0.06
ECEurope	DE29	9	0.144	-6.38	9	0.297	-0.78	9	0.835	0.00	9	0.404	10.60	8	0.083	57.00	9	0.144	23.04	8	0.458	0.47	9	1.000	0.10
ECEurope	DE30	**9**	**0.007**	**-63.2**	9	0.095	-3.75	**9**	**0.022**	**0.01**				7	0.293	-25.15							9	0.677	-0.60
ECEurope	DE31	**8**	**0.006**	**-55.3**	**8**	**0.006**	**-22.4**	8	0.322	0.00	**8**	**0.003**	**-53.14**	8	1.000	-1.34	**8**	**0.026**	**27.43**				8	0.216	-0.01
ECEurope	DE33	9	0.095	-10.0	**9**	**0.004**	**-5.73**	**9**	**0.022**	**-0.01**	**9**	**0.004**	**-5.45**	8	0.621	0.99				9	0.095	0.11	9	0.144	-4.52
ECEurope	PL01	**8**	**0.048**	**-2.83**	8	0.083	-1.33	8	1.000	0.00	8	0.083	-5.16	7	0.293	1.62	8	0.805	-0.26						
ECEurope	PL02	**8**	**0.003**	**-3.15**	8	1.000	-0.06	**8**	**0.006**	**0.01**	**8**	**0.003**	**-6.57**	8	0.216	-1.20	**8**	**0.003**	**-4.01**				7	0.453	0.01
Maine/Atlantic	CA10	9	0.095	-1.03	9	0.297	0.00	**9**	**0.037**	**0.00**	9	0.404	0.53	9	0.835	-0.14	9	0.297	2.71	8	0.138	0.70	9	0.144	2.03
Maine/Atlantic	CA11	**10**	**0.025**	**-2.19**	10	1.000	0.00	10	0.089	0.00	10	0.929	-0.15	10	0.421	0.23	10	0.929	-0.04	8	0.805	0.03	10	0.421	0.17
Maine/Atlantic	CA12	7	0.652	-1.57	7	0.734	0.00	7	0.051	0.00	7	0.881	0.01	7	0.761	-0.10	7	0.176	4.57	7	0.176	1.46	7	0.099	2.96
Maine/Atlantic	CA13	10	0.245	-1.40	**10**	**0.004**	**0.06**	10	0.060	0.00	10	0.655	-0.15	10	0.929	-0.01	10	0.089	1.33	9	0.211	0.24	**10**	**0.031**	**0.43**
Maine/Atlantic	CA14	**10**	**0.003**	**-1.15**	10	0.125	0.00	**10**	**0.006**	**0.00**	**10**	**0.040**	**-0.54**	10	0.856	0.00	10	0.089	0.57	9	0.116	0.16	10	0.281	-0.06
Maine/Atlantic	US05	**10**	**0.002**	**-1.32**	10	0.501	0.00	10	0.631	0.00	**10**	**0.009**	**-1.12**	10	0.325	-0.21	10	0.929	0.04	10	0.316	-0.03	10	0.245	-0.07
Maine/Atlantic	US06	10	0.128	-0.74	10	0.241	0.03	10	0.089	0.00	**10**	**0.025**	**-0.91**	10	0.472	-0.20	10	0.655	-0.89	10	0.528	-0.02	10	0.788	0.00
Maine/Atlantic	US71	**7**	**0.024**	**-3.12**	7	0.129	0.10	7	0.099	0.00	7	0.099	-6.47	7	0.051	-5.43	7	0.099	-5.54	7	0.176	0.41	7	0.176	0.09
Maine/Atlantic	US72	7	0.881	0.26	7	0.453	-0.09	7	0.453	0.00	7	0.293	-3.03	7	0.881	-0.14	7	0.453	-1.77	7	0.293	0.07	7	0.881	0.01
Maine/Atlantic	US73	**10**	**0.025**	**-1.00**	10	0.925	0.00	10	0.926	0.00	10	0.060	-1.15	10	0.180	-0.29	10	0.531	0.25	10	0.787	-0.02	10	0.857	0.00
Maine/Atlantic	US74	**10**	**0.001**	**-1.72**	10	0.787	-0.02	10	0.857	0.00	**10**	**0.006**	**-1.20**	10	0.325	0.51	10	0.531	0.23	10	0.472	0.04	10	0.655	-0.26
Maine/Atlantic	US75	10	0.060	-0.36	**10**	**0.031**	**0.06**	**10**	**0.020**	**0.00**	10	0.421	-0.32	10	0.531	-0.22	10	0.325	-1.43	10	0.472	0.03	10	0.325	-0.01
Maine/Atlantic	US76	7	0.176	-3.75	7	0.167	0.07	7	0.099	0.00	7	0.881	-2.69	7	0.652	2.87	7	0.652	-0.32	7	0.881	-0.08	7	0.652	0.01
Maine/Atlantic	US77	7	0.099	-2.10	7	0.224	0.12	7	0.176	0.00	7	0.051	-3.35	7	0.293	-0.35	7	0.453	-1.83	7	0.293	0.06	7	0.881	0.00
Maine/Atlantic	US78	7	0.652	-0.68	**7**	**0.033**	**0.08**	**7**	**0.033**	**0.00**	7	0.051	-3.41	7	0.652	-2.00	7	0.176	-4.26	**7**	**0.033**	**-0.09**	7	0.881	0.01
Maine/Atlantic	US79	**7**	**0.024**	**-1.30**	7	0.453	0.04	7	0.453	0.00	**7**	**0.024**	**-1.61**	7	0.176	0.54	7	0.652	-0.35	7	0.068	0.11	7	0.176	-0.98
Maine/Atlantic	US80	7	0.453	-0.95	**7**	**0.046**	**0.08**	**7**	**0.033**	**0.00**	**7**	**0.011**	**-3.44**	7	0.652	-0.49	7	0.051	-3.37	7	0.176	0.03	7	0.881	0.00
Maine/Atlantic	US81	7	0.051	-1.00	7	0.543	0.02	7	0.543	0.00	7	0.051	-1.81	7	0.176	-0.62	7	0.099	-1.77	7	0.176	-0.06	7	0.652	-0.03
Maine/Atlantic	US82																								
Maine/Atlantic	US83																								
NoNordic	FI05	10	0.089	0.68	10	0.531	0.06	10	0.655	0.00	10	0.929	0.02	**10**	**0.040**	**2.16**	10	0.655	-0.69	10	0.531	0.18	10	0.128	-0.05
NoNordic	FI06	**10**	**0.025**	**-0.66**	10	0.788	-0.03	10	0.788	0.00	10	0.060	-1.15	**10**	**0.016**	**1.97**	10	0.245	-1.04	10	0.180	0.07	10	0.325	-0.08
NoNordic	FI08	**10**	**0.000**	**-2.76**	10	0.788	-0.02	10	0.531	0.00	**10**	**0.009**	**-0.61**	**10**	**0.003**	**1.30**	**10**	**0.001**	**3.11**	**10**	**0.009**	**0.19**	**10**	**0.000**	**-0.67**
NoNordic	NO04	**10**	**0.000**	**-2.75**	**10**	**0.025**	**0.06**	**10**	**0.009**	**0.00**	**10**	**0.016**	**-1.67**	10	0.180	1.34	10	0.325	1.45	10	0.325	0.05	**10**	**0.003**	**-0.06**
NoNordic	SE01	**10**	**0.006**	**-2.59**	10	0.655	-0.07	10	0.788	0.00	10	0.245	0.85	10	0.089	1.03	**10**	**0.025**	**2.16**	10	0.060	0.05	**10**	**0.025**	**-0.01**
NoNordic	SE05	**10**	**0.025**	**-2.22**	10	0.531	0.03	10	0.180	0.00	**10**	**0.040**	**-1.63**	10	0.089	-1.71	10	0.788	0.57	10	0.929	0.04	10	0.180	0.01

1990-1999		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
NoNordic	SE06	10	0.180	-0.89	10	0.421	0.03	10	0.325	0.00	10	0.655	-0.32	10	0.788	-0.14	10	0.929	-0.01	10	0.788	-0.02	10	0.421	-0.01
SoNordic	FI01	**10**	**0.000**	**-5.57**	10	0.128	0.33	10	0.089	0.00	**10**	**0.002**	**-3.43**	10	0.060	1.42	**10**	**0.009**	**3.74**	10	0.089	0.09	10	0.788	-0.22
SoNordic	FI02	**10**	**0.002**	**-6.18**	10	0.245	0.34	10	0.128	0.00	**10**	**0.040**	**-2.87**	10	0.180	2.75	10	0.089	3.10	10	0.421	0.05	10	0.089	-0.32
SoNordic	FI03	**10**	**0.000**	**-5.16**	10	0.929	0.05	10	0.531	0.00	**10**	**0.040**	**-8.73**	10	0.128	-0.63	10	0.929	0.75	10	0.128	0.17	10	0.655	0.15
SoNordic	FI07	**10**	**0.000**	**-5.59**	10	0.089	0.27	**10**	**0.040**	**0.00**	**10**	**0.002**	**-2.06**	**10**	**0.000**	**2.33**	**10**	**0.001**	**3.98**	10	0.176	0.05	10	0.369	-0.01
SoNordic	FI09	**7**	**0.024**	**-5.12**	7	0.652	-0.12				7	0.176	-1.66	**7**	**0.024**	**2.45**				7	0.881	0.00	7	0.293	0.02
SoNordic	NO01	**10**	**0.025**	**-2.60**	10	0.180	-0.41	10	0.655	0.00	**10**	**0.009**	**-1.86**	9	0.107	0.02	10	0.089	2.91	10	0.531	0.05	**10**	**0.006**	**-1.04**
SoNordic	NO03	**10**	**0.006**	**-3.18**	10	0.089	-0.04	10	0.060	0.00	10	0.128	-1.59	10	0.128	0.23	**10**	**0.009**	**1.84**	**10**	**0.009**	**0.28**	**10**	**0.016**	**-0.50**
SoNordic	NO10	**10**	**0.001**	**-3.32**	10	0.421	-0.21	10	0.180	0.00	10	0.128	-0.63	7	0.362	0.03	**10**	**0.000**	**3.43**	**10**	**0.016**	**0.14**	**10**	**0.000**	**-1.41**
SoNordic	NO11	10	0.089	-0.26	**10**	**0.006**	**0.10**	**10**	**0.000**	**0.01**	10	0.655	0.21	10	0.089	1.05	10	0.245	0.75	10	0.655	0.00	10	0.180	-0.01
SoNordic	SE02	**10**	**0.000**	**-18.4**	10	0.421	0.11	**10**	**0.003**	**0.00**	**10**	**0.009**	**-7.16**	**10**	**0.016**	**6.56**	**10**	**0.003**	**10.97**	**10**	**0.016**	**0.41**	10	0.060	-0.02
SoNordic	SE03	**10**	**0.009**	**-16.6**	10	0.060	-0.41	10	0.531	0.00	10	0.929	0.02	10	0.089	10.75	**10**	**0.016**	**17.03**				**10**	**0.040**	**-0.04**
SoNordic	SE08	**10**	**0.002**	**-26.7**	10	0.655	0.08	**10**	**0.040**	**0.00**	**10**	**0.003**	**-22.49**	10	0.180	-1.70	**10**	**0.040**	**2.98**	**10**	**0.040**	**1.00**	10	0.531	0.09
SoNordic	SE09	**10**	**0.009**	**-4.10**	10	0.245	-0.38	10	0.531	0.00	**10**	**0.003**	**-3.07**	10	0.072	1.22	**10**	**0.006**	**3.06**	10	0.531	-0.02	**10**	**0.009**	**-0.03**
SoNordic	SE10	**10**	**0.001**	**-8.46**	**10**	**0.025**	**0.25**	**10**	**0.003**	**0.00**	**10**	**0.006**	**-3.06**	10	0.929	-0.17	**10**	**0.025**	**4.75**	**10**	**0.016**	**0.50**	10	0.060	-0.24
SoNordic	SE11	**10**	**0.000**	**-8.79**	10	0.788	0.06	10	0.128	0.00	**10**	**0.009**	**-7.27**	**10**	**0.003**	**1.95**	**10**	**0.009**	**4.54**	**10**	**0.025**	**0.34**	**10**	**0.016**	**-0.03**
SoNordic	SE12	**10**	**0.000**	**-6.40**	**10**	**0.003**	**-0.69**	10	0.655	0.00	**10**	**0.001**	**-4.84**	10	0.204	-5.03	10	0.325	1.18	10	0.655	0.09	**10**	**0.002**	**-2.33**
UK	UK01	10	0.245	-0.64	10	0.531	-0.08	10	0.245	0.01	10	0.128	1.00	10	0.531	0.63	10	0.245	0.87	**10**	**0.009**	**0.15**	10	0.180	0.02
UK	UK04	**10**	**0.016**	**-0.82**	10	0.245	0.93	10	0.060	0.02	10	0.325	-0.26	10	0.072	-0.25	10	0.421	-0.64	10	0.089	0.07	10	0.788	0.06
UK	UK07	10	0.128	-2.37	10	0.089	0.46	**10**	**0.006**	**0.02**	10	0.325	-0.44	10	0.531	0.25	10	0.325	0.71	**10**	**0.003**	**0.13**	10	0.089	-0.25
UK	UK10	10	0.531	-0.28	10	0.531	-0.12	10	0.788	0.00	10	0.060	-1.18	10	0.176	0.31	10	0.788	0.13	**10**	**0.002**	**0.13**	**10**	**0.025**	**-0.23**
UK	UK15	10	0.245	-1.85	10	0.788	0.09	10	0.180	0.01	10	0.089	-1.71	10	0.655	0.12	10	0.655	0.30	**10**	**0.004**	**0.10**	**10**	**0.009**	**-0.54**
UK	UK21	10	0.089	-3.69	10	0.788	0.30	10	0.089	0.02	10	0.089	-1.34	**10**	**0.025**	**1.00**	10	0.929	0.16	**10**	**0.012**	**0.18**	10	0.128	-0.58
Ontario	CA01	10	0.089	-1.44	10	0.929	-0.02	10	0.929	0.00	**10**	**0.009**	**-2.88**	10	0.325	0.79	**10**	**0.025**	**-1.84**	10	0.325	0.04	10	0.788	-0.01
Ontario	CA02	10	0.180	-1.47	10	0.929	0.03	10	0.531	0.00	10	0.060	-3.20	10	0.929	-0.20	10	0.060	-2.54	10	0.929	0.00	10	0.655	0.00
Ontario	CA03	10	0.060	-2.21	10	0.929	-0.05	10	0.655	0.00	**10**	**0.003**	**-3.71**	10	0.929	0.20	**10**	**0.016**	**-1.76**	10	0.655	0.01	10	0.325	0.00
Ontario	CA04	**10**	**0.009**	**-2.10**	10	0.929	0.06	10	0.531	0.00	**10**	**0.006**	**-3.99**	10	0.788	-1.85	**10**	**0.009**	**-1.85**	10	0.245	0.05	10	0.531	0.00
Ontario	CA16	**10**	**0.000**	**-2.37**	10	0.929	0.00	10	0.655	0.00	**10**	**0.003**	**-2.14**	10	0.325	0.86	10	0.788	0.10	**10**	**0.016**	**0.05**	10	0.421	0.00
Ontario	CA17	**10**	**0.000**	**-5.62**	10	0.655	0.03	10	0.325	0.00	**10**	**0.003**	**-3.49**	10	0.245	-0.54	10	0.788	0.24	10	0.060	0.11	10	0.788	0.00
Ontario	CA19																								
Ontario	CA20	**8**	**0.001**	**-2.03**	**8**	**0.016**	**0.01**	**8**	**0.006**	**0.00**	**8**	**0.003**	**-2.53**	9	1.000	-0.06	8	0.322	-1.30	9	0.532	0.01	8	0.216	0.00
Vermont/Quebec	CA05	**10**	**0.000**	**-3.41**	**10**	**0.015**	**-0.27**	10	0.421	0.00	10	0.325	-0.49	10	0.128	1.57	**10**	**0.003**	**3.76**	10	0.417	0.03	10	0.421	-0.02
Vermont/Quebec	CA06	**10**	**0.000**	**-3.53**	10	0.241	-0.09	10	0.788	0.00	**10**	**0.040**	**-0.85**	**10**	**0.009**	**1.26**	**10**	**0.002**	**3.92**	10	0.151	0.04	10	0.929	0.00
Vermont/Quebec	CA07	**10**	**0.000**	**-3.70**	10	0.421	-0.11	10	0.531	0.00	**10**	**0.006**	**-1.60**	10	0.325	0.65	**10**	**0.025**	**2.36**	10	0.128	0.17	10	0.531	-0.23

1990-1999		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Vermont/Quebec	CA08	**10**	**0.003**	**-2.85**	**10**	**0.016**	**-0.39**	10	0.421	0.00	10	0.060	-2.97	10	0.655	0.22	10	0.929	0.26	**8**	**0.026**	**0.15**	10	0.325	0.01
Vermont/Quebec	CA09	**10**	**0.001**	**-4.01**	10	0.784	0.00	10	0.245	0.00	**10**	**0.040**	**-1.23**	10	0.929	0.03	**10**	**0.040**	**2.29**	10	0.421	0.11	10	0.655	-0.05
Vermont/Quebec	US100	**10**	**0.000**	**-3.28**	10	0.209	-0.47	10	0.325	0.00	10	0.089	-2.76	10	0.929	0.29	10	0.655	0.76				10	0.128	0.05
Vermont/Quebec	US102	**7**	**0.002**	**-10.1**	7	0.129	-0.20	7	1.000	0.00	7	0.293	-9.46	7	0.099	1.70	7	0.881	1.14				7	0.293	-1.21
Vermont/Quebec	US122																								
Vermont/Quebec	US123	**10**	**0.000**	**-2.29**	**10**	**0.025**	**-0.53**	10	0.655	0.00	**10**	**0.003**	**-1.36**	10	0.245	0.45	**10**	**0.003**	**1.12**				10	0.325	-0.34
Vermont/Quebec	US125																								
Vermont/Quebec	US126																								
Vermont/Quebec	US89	**10**	**0.006**	**-2.67**	10	0.060	-0.29	10	0.128	0.00	**10**	**0.009**	**-1.88**	10	0.655	0.27	10	0.421	0.82				10	0.180	-0.21
Vermont/Quebec	US90	**10**	**0.001**	**-2.40**	10	0.180	-0.10	10	0.788	0.00	**10**	**0.025**	**-1.92**	10	0.531	0.26	10	0.421	0.55				10	0.421	0.02
Vermont/Quebec	US91	10	0.089	-0.90	10	0.180	-0.14	10	0.325	0.00	**10**	**0.006**	**-2.08**	10	0.180	-1.16	10	0.180	-1.05				10	0.788	0.00
Vermont/Quebec	US92	**10**	**0.002**	**-3.24**	**10**	**0.016**	**-1.78**	**10**	**0.016**	**-0.01**	**10**	**0.006**	**-2.64**	10	0.655	-0.21	**10**	**0.025**	**1.68**				10	0.531	-0.12
Vermont/Quebec	US93	**10**	**0.040**	**-2.31**	**10**	**0.001**	**-2.53**	**10**	**0.000**	**-0.02**	**10**	**0.009**	**-1.24**	**10**	**0.025**	**1.62**	**10**	**0.025**	**2.32**				10	0.089	-0.20
Vermont/Quebec	US94																								
Vermont/Quebec	US95	**10**	**0.006**	**-2.47**	10	0.590	-0.02	10	0.531	0.00	**10**	**0.006**	**-2.11**	**10**	**0.040**	**-1.25**	10	0.788	-0.21				10	0.421	0.02
Vermont/Quebec	US96	9	0.061	-4.34	9	0.532	-0.17	9	0.835	0.00	9	0.144	-3.66	9	0.404	0.61	9	0.211	1.37				9	0.061	-0.70
Vermont/Quebec	US97	**10**	**0.003**	**-3.32**	**10**	**0.040**	**-0.45**	10	0.060	0.00	**10**	**0.003**	**-1.38**	10	0.128	0.45	**10**	**0.003**	**1.92**				10	0.089	-0.55
Vermont/Quebec	US98	**7**	**0.024**	**-2.47**	7	0.881	0.12	7	0.176	0.00	7	0.099	-3.44	7	0.652	-0.67	7	0.881	0.35				7	0.453	0.01
WCEurope	DE01	10	0.089	-0.38	10	0.421	-0.43	10	0.788	0.00	**10**	**0.003**	**9.55**	9	0.144	9.28	**10**	**0.016**	**11.35**	10	0.180	0.34	**10**	**0.006**	**-0.79**
WCEurope	DE03	**10**	**0.009**	**-5.03**	10	0.929	0.07	10	0.325	0.00	10	0.245	-5.08				8	0.805	0.64	10	0.106	0.06	9	0.677	0.05
WCEurope	DE05	**10**	**0.000**	**-3.76**	10	0.245	-1.14	10	0.128	0.01	10	0.128	4.18	**9**	**0.007**	**18.69**	10	0.089	8.27	10	0.590	-0.04	10	0.089	-0.03
WCEurope	DE06	**10**	**0.001**	**-15.7**	**10**	**0.003**	**-12.1**	**10**	**0.003**	**-0.01**	**10**	**0.025**	**-9.72**	10	0.309	0.00	**10**	**0.003**	**18.52**	**10**	**0.016**	**0.21**	10	0.788	-1.47
WCEurope	DE11	10	0.655	-0.71	**10**	**0.025**	**-1.25**	10	0.325	-0.01	10	0.325	1.79	9	0.144	7.92	10	0.089	4.04	10	0.106	-0.06	10	0.655	0.01
WCEurope	DE12	10	0.325	-0.51	**10**	**0.025**	**-1.38**	**10**	**0.016**	**0.00**	**10**	**0.003**	**-7.84**				**10**	**0.040**	**-5.93**	9	0.061	-0.10	10	0.089	0.10
WCEurope	DE14	**9**	**0.007**	**-32.6**	**9**	**0.007**	**-5.44**	9	0.404	0.00	**9**	**0.012**	**-81.22**	**9**	**0.008**	**-16.67**	9	0.211	-40.29				9	0.095	2.53
WCEurope	DE15	**9**	**0.037**	**-19.7**	**9**	**0.037**	**-1.66**	9	0.677	0.00	9	0.297	-6.35	9	0.885	0.00	9	0.144	14.65				9	0.211	2.37
WCEurope	DE16				**8**	**0.045**	**0.05**													**8**	**0.048**	**0.45**	8	0.083	-2.02
WCEurope	DE19	10	0.655	-2.97	10	0.325	3.37	10	0.089	0.00	**10**	**0.003**	**12.13**	10	0.373	0.00	**10**	**0.016**	**21.87**				10	0.655	0.55
WCEurope	DE20	10	0.180	-3.10	10	0.089	1.04	**10**	**0.025**	**0.01**	10	0.060	7.16	**10**	**0.040**	**-6.01**	10	0.060	11.87				10	0.788	-0.05
WCEurope	DE22	**9**	**0.037**	**-20.2**	9	0.095	-9.14	9	0.532	0.00	9	0.061	-6.99	8	0.127	0.00	**9**	**0.022**	**23.90**				9	0.532	-4.57
WCEurope	DE26	**10**	**0.016**	**-6.00**	10	0.180	-2.60	10	0.128	0.00	10	0.788	1.60	10	0.784	0.00	**10**	**0.009**	**10.86**	10	0.060	-0.07	10	0.655	1.78
WCEurope	DE32	**10**	**0.009**	**-5.42**	10	0.180	-1.28	10	0.531	0.00	**10**	**0.040**	**-6.84**				8	1.000	-0.61	10	0.421	0.03	9	0.404	0.12
WCEurope	DE34	**10**	**0.025**	**-10.5**	**10**	**0.016**	**1.20**	**10**	**0.003**	**0.00**	10	0.128	-4.86	**10**	**0.043**	**5.56**	10	0.788	-1.34				10	0.929	-0.58

Table D2. *Results of trend analysis of data from 13 extra Ontario stations for the time span 1990-1999. Andrew Paterson (Ontario Ministry of Environment) provided data from the 8 Dorset lakes. Bill Keller (Laurentian University and Ontario Ministry of Environment) provided data from the 5 Sudbury lakes.*

		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	Station Name	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Ontario (Dorset)	Blue Chalk Lake	**10**	**0.003**	**-2.53**	10	0.325	0.08	10	0.089	0.00	10	0.325	-1.54	10	0.788	-0.24	10	0.325	0.57	10	0.180	0.01	10	0.531	0.00
Ontario (Dorset)	Chub Lake	**10**	**0.003**	**-5.31**	10	0.929	-0.01	10	0.245	0.00	**10**	**0.003**	**-5.74**	10	0.788	-0.05	10	0.929	-0.05	10	0.421	-0.02	10	0.089	-0.07
Ontario (Dorset)	Crosson Lake	**10**	**0.003**	**-6.02**	10	0.929	0.01	10	0.060	0.00	**10**	**0.009**	**-4.78**	10	0.325	-0.22	10	0.929	-0.02	10	0.180	-0.05	10	0.180	-0.07
Ontario (Dorset)	Dickie Lake	**10**	**0.040**	**-6.48**	10	0.788	-0.07	10	0.788	0.00	10	0.060	-3.49	10	0.089	1.19	**10**	**0.003**	**2.19**	10	0.325	-0.07	**10**	**0.025**	**-0.08**
Ontario (Dorset)	Harp Lake	**10**	**0.009**	**-4.92**	10	0.089	0.17	**10**	**0.016**	**0.00**	**10**	**0.009**	**-3.57**	10	0.325	-0.55	10	0.421	0.53	10	0.421	0.03	10	0.060	-0.01
Ontario (Dorset)	Heney Lake	**10**	**0.002**	**-5.79**	10	0.421	-0.06	10	0.531	0.00	10	0.060	-3.16	**10**	**0.003**	**2.30**	10	0.128	2.59	10	0.245	0.05	**10**	**0.003**	**-0.14**
Ontario (Dorset)	Plastic Lake	**10**	**0.001**	**-3.40**	10	0.531	-0.05	10	0.929	0.00	**10**	**0.003**	**-3.38**	10	0.929	0.03	10	0.325	-0.19	10	0.245	-0.02	10	0.325	-0.02
Ontario (Dorset)	Red Chalk Lake	**10**	**0.000**	**-3.93**	10	0.421	-0.14	10	0.421	0.00	**10**	**0.025**	**-3.31**	10	0.655	0.18	10	0.325	0.44	10	0.180	-0.02	**10**	**0.025**	**-0.02**
Ontario (Sudbury)	Clearwater	**10**	**0.000**	**-13.0**	10	0.531	-0.10	10	0.788	0.00	**10**	**0.001**	**-11.8**	**10**	**0.000**	**3.72**	**10**	**0.016**	**5.36**	**10**	**0.002**	**0.16**	**10**	**0.000**	**-1.81**
Ontario (Sudbury)	Sans Chambre	**10**	**0.000**	**-8.35**	**10**	**0.009**	**0.11**	**10**	**0.006**	**0.00**	**10**	**0.000**	**-8.41**	**10**	**0.009**	**-1.34**	10	0.128	-1.35	10	0.180	0.07	10	0.325	-0.05
Ontario (Sudbury)	Whitepine (Aurora)	**10**	**0.000**	**-5.24**	10	0.531	0.04	10	0.128	0.00	**10**	**0.003**	**-2.20**	**10**	**0.006**	**2.20**	**10**	**0.006**	**2.26**	10	0.128	0.02	**10**	**0.009**	**-0.64**
Ontario (Sudbury)	Whitepine (Mcleod)	**10**	**0.002**	**-4.83**	10	0.325	-0.05	10	0.245	0.00	**10**	**0.000**	**-2.83**				**10**	**0.025**	**1.95**	10	0.928	0.00	**10**	**0.001**	**-0.19**
Ontario (Sudbury)	Swan Lake	**10**	**0.000**	**-9.92**	10	0.788	0.01	10	0.655	0.00	**10**	**0.016**	**-14.4**				**10**	**0.016**	**2.84**	**10**	**0.040**	**0.16**			

Table D3. *Results of Mann-Kendall trend analysis for individual sites, 1999-2008. Slope is annual change in the unit given for each parameter, p is the probability that there is no trend and n is the number of years with observations. Significant results (p<0.05) are written in bold.*

1999-2008		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Adirondacks	US11	**10**	**0.003**	**-3.53**	10	0.655	-0.04	10	0.421	0.00	10	0.060	-3.38	10	0.060	-1.17	10	0.421	-0.71	10	0.180	0.08	**10**	**0.006**	**0.04**
Adirondacks	US12	**10**	**0.000**	**-3.45**	10	0.245	-0.37	10	0.929	0.00	**10**	**0.040**	**-3.06**	10	0.788	0.08	10	0.929	0.20	10	0.060	0.13	10	0.929	0.00
Adirondacks	US13	**10**	**0.002**	**-2.85**	10	0.180	-0.60	10	0.325	0.00	**10**	**0.003**	**-1.50**	**10**	**0.009**	**0.53**	**10**	**0.016**	**1.03**	10	0.245	0.07	10	0.655	-0.03
Adirondacks	US14	**10**	**0.003**	**-1.69**	10	0.788	-0.05	10	0.531	0.00	**10**	**0.009**	**-1.92**	10	0.089	-0.36	10	0.788	-0.22	**10**	**0.016**	**-0.03**	10	0.531	0.02
Adirondacks	US15	**10**	**0.003**	**-2.42**	10	0.245	-0.30	10	0.325	0.00	10	0.128	-1.34	10	0.788	0.67	10	0.421	0.88	10	0.245	0.05	10	0.531	0.00
Adirondacks	US16	**10**	**0.002**	**-2.61**	10	0.655	0.16	**10**	**0.006**	**0.01**	**10**	**0.016**	**-2.47**	10	0.655	-0.07	10	0.788	-0.03	10	0.531	0.02	10	0.089	0.02
Adirondacks	US17	**10**	**0.002**	**-2.54**	**10**	**0.016**	**0.34**	**10**	**0.002**	**0.00**	10	0.060	-0.91	10	0.325	0.65	10	0.245	0.69	10	0.180	0.03	10	0.788	0.02
Adirondacks	US27	**10**	**0.016**	**-3.13**	**10**	**0.009**	**-0.55**	**10**	**0.009**	**0.00**	10	0.089	-1.91	10	0.531	-0.17	10	0.421	0.82	10	0.788	0.02	10	0.929	0.01
Adirondacks	US28	**10**	**0.009**	**-2.62**	**10**	**0.040**	**-0.12**	10	0.421	0.00	10	0.929	0.06	10	0.245	0.81	10	0.128	1.55	10	0.531	0.03	10	0.929	0.00
Adirondacks	US29	**10**	**0.009**	**-3.29**	10	0.089	-0.15	10	0.929	0.00	**10**	**0.001**	**-1.74**	10	0.531	-0.07	**10**	**0.025**	**2.15**	10	0.089	0.29	10	0.655	0.16
Adirondacks	US30	10	0.325	0.33	10	0.788	0.00	10	0.421	0.00	**10**	**0.001**	**-1.01**	**10**	**0.040**	**-0.29**	**10**	**0.001**	**-1.20**	**10**	**0.003**	**-0.14**	**10**	**0.016**	**0.41**
Adirondacks	US31	**10**	**0.001**	**-1.63**	10	0.421	-0.12	10	0.929	0.00	**10**	**0.009**	**-0.32**	10	0.180	1.05	**10**	**0.003**	**1.37**	10	0.788	-0.03	10	0.531	-0.40
Adirondacks	US32	**10**	**0.002**	**-2.36**	10	0.060	1.16	**10**	**0.016**	**0.01**	10	0.180	-0.87	10	0.421	-0.21	10	0.929	-0.19	10	0.369	-0.02	10	0.325	-0.03
Adirondacks	US33	**10**	**0.006**	**-0.77**	10	0.655	-0.05	10	0.929	0.00	10	0.421	-0.13	10	0.325	-0.40	**10**	**0.040**	**0.52**	10	0.369	0.05	10	0.421	0.27
Adirondacks	US34	**10**	**0.001**	**-1.12**	10	0.128	0.09	10	0.128	0.00	**10**	**0.000**	**-10.8**	**10**	**0.000**	**-9.18**	**10**	**0.000**	**-10.0**	**10**	**0.025**	**-0.06**	**10**	**0.000**	**0.03**
Adirondacks	US36	**10**	**0.001**	**-2.80**	10	0.128	-0.27	10	0.325	0.00	**10**	**0.000**	**-1.63**	10	0.531	0.28	**10**	**0.009**	**1.46**	**10**	**0.040**	**0.17**	10	0.788	0.01
Adirondacks	US37	**10**	**0.016**	**-2.76**	10	0.245	-0.18	10	0.325	0.00	**10**	**0.025**	**-2.03**	10	0.128	0.53	10	0.060	1.16	10	0.245	0.07	10	0.180	0.06
Adirondacks	US38	**10**	**0.000**	**-2.63**	10	0.089	-0.11	10	0.128	0.00	**10**	**0.040**	**-0.65**	**10**	**0.016**	**1.06**	**10**	**0.003**	**1.62**	**10**	**0.002**	**0.05**	10	0.421	-0.02
Adirondacks	US39	**10**	**0.002**	**-2.28**	10	0.421	0.30	10	0.128	0.01	10	0.089	-1.71	10	0.325	-0.96	10	0.788	-1.06	10	0.421	-0.02	10	0.788	0.05
Adirondacks	US40	**10**	**0.009**	**-1.72**	10	0.655	-0.08	10	0.531	0.00	10	0.089	-1.59	10	0.655	-0.53	10	0.788	-0.61	10	0.655	-0.02	10	0.655	0.01
Adirondacks	US41	**10**	**0.000**	**-3.01**	10	0.089	-0.49	10	0.929	0.00	**10**	**0.016**	**-2.24**	10	0.128	0.55	10	0.245	1.23	10	0.245	0.03	10	0.929	0.02
Adirondacks	US42	**10**	**0.000**	**-2.94**	10	0.128	0.31	**10**	**0.025**	**0.01**	**10**	**0.001**	**-1.92**	10	0.929	-0.02	10	0.245	0.17	10	0.421	-0.02	10	0.245	0.07
Adirondacks	US43	**10**	**0.002**	**-3.61**	10	0.655	0.04	**10**	**0.003**	**0.01**	**10**	**0.009**	**-1.94**	**10**	**0.016**	**0.61**	**10**	**0.006**	**1.31**	10	0.245	0.06	10	0.929	0.00
Adirondacks	US44	**10**	**0.002**	**-1.73**	**10**	**0.016**	**0.82**	**10**	**0.009**	**0.01**	**10**	**0.006**	**-1.53**	10	0.089	-0.69	10	0.325	-0.60	10	0.180	-0.11	10	0.421	0.10
Adirondacks	US45	**10**	**0.016**	**-2.45**	10	0.325	0.50	**10**	**0.040**	**0.01**	10	0.180	-1.15	10	0.421	0.68	10	0.325	0.79	10	0.788	0.01	10	0.788	-0.02
Adirondacks	US46	**10**	**0.001**	**-2.41**	10	0.180	-0.50	10	0.421	0.00	**10**	**0.016**	**-2.51**	10	0.655	-0.27	10	0.325	-0.80	10	0.241	0.02	10	0.531	0.17
Adirondacks	US47	**10**	**0.001**	**-2.98**	10	0.655	0.02	10	0.128	0.01	**10**	**0.003**	**-2.02**	10	0.089	0.42	10	0.089	0.94	10	0.788	-0.01	10	0.788	-0.07
Adirondacks	US48	**9**	**0.022**	**-2.69**	9	1.000	-0.01	9	0.144	0.00	9	0.061	-4.08	9	0.211	-1.11	9	0.211	-1.85	**9**	**0.004**	**-0.60**	9	0.532	0.01
Adirondacks	US49	**10**	**0.009**	**-2.02**	**10**	**0.016**	**-0.12**	**10**	**0.025**	**0.00**	10	0.180	-1.13	10	0.531	-0.37	10	0.180	1.16	10	0.128	0.18	10	0.325	0.64
Adirondacks	US50	**10**	**0.000**	**-4.57**	10	0.531	-0.05	10	0.060	0.00	**10**	**0.009**	**-1.05**	**10**	**0.016**	**2.22**	**10**	**0.003**	**3.71**	**10**	**0.007**	**0.21**	**10**	**0.025**	**-0.90**
Adirondacks	US51	**10**	**0.006**	**-2.71**	10	0.929	0.06	10	0.060	0.01	**10**	**0.025**	**-1.27**	10	0.089	0.47	10	0.180	0.99	10	0.421	0.02	10	0.421	-0.14

1999-2008		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Adirondacks	US52	**10**	**0.006**	**-2.37**	10	0.655	-0.51	10	0.180	0.01	10	0.089	-1.21	**10**	**0.040**	**0.64**	10	0.325	0.88	10	0.929	-0.01	10	0.655	-0.12
Adirondacks	US53	**10**	**0.000**	**-2.39**	10	0.180	-0.71	10	0.421	0.00	10	0.929	0.27	10	0.245	2.54	10	0.089	3.32	10	0.421	0.02	10	0.655	-0.02
Adirondacks	US54	**10**	**0.000**	**-4.41**	10	0.325	-0.45	10	0.929	0.00	**10**	**0.016**	**-2.72**	10	0.531	0.35	10	0.245	2.13	10	0.531	0.10	10	0.325	-0.16
Adirondacks	US55	**10**	**0.001**	**-2.41**	10	0.128	0.72	**10**	**0.040**	**0.01**	**10**	**0.003**	**-2.19**	10	0.531	-0.57	10	0.325	-0.59	10	0.421	-0.03	10	0.180	0.23
Adirondacks	US57	**10**	**0.006**	**-3.40**	10	0.531	0.17	10	0.180	0.01	**10**	**0.040**	**-2.31**	10	0.929	0.02	10	0.531	1.14	10	0.655	0.04	10	0.245	0.06
Adirondacks	US58	**10**	**0.000**	**-2.66**	10	0.655	0.15	10	0.245	0.00	**10**	**0.009**	**-3.82**	10	0.060	-2.03	10	0.325	-1.57	10	0.245	0.02	**10**	**0.009**	**0.01**
Adirondacks	US59	**10**	**0.006**	**-3.05**	10	0.531	-0.21	10	0.128	0.00	10	0.060	-2.59	10	0.655	-0.19	10	0.245	1.22	10	0.655	0.04	10	0.788	0.00
Adirondacks	US60	**10**	**0.000**	**-2.58**	**10**	**0.025**	**-0.22**	10	0.325	0.00	**10**	**0.003**	**-1.76**	10	0.128	0.44	**10**	**0.040**	**0.94**	**10**	**0.016**	**0.08**	10	0.325	0.14
Adirondacks	US61	**10**	**0.001**	**-3.02**	10	0.180	-0.51	10	0.421	0.00	**10**	**0.003**	**-1.67**	10	0.089	0.55	**10**	**0.006**	**1.04**	10	0.325	0.04	10	0.929	0.00
Adirondacks	US62	**10**	**0.025**	**-1.97**	**10**	**0.040**	**0.35**	**10**	**0.006**	**0.01**	10	0.325	-1.88	10	0.421	1.51	10	0.929	0.16	10	0.421	-0.05	10	0.788	0.01
Adirondacks	US63	10	0.788	0.06	10	0.655	0.02	10	0.929	0.00	10	0.245	-0.53	10	0.531	-0.63	10	0.531	-0.37	10	0.060	-0.23	10	0.421	0.10
Adirondacks	US64	**10**	**0.016**	**-1.63**	10	0.929	0.19	10	0.245	0.01	**10**	**0.016**	**-0.61**	10	0.060	0.74	10	0.128	0.92	10	0.655	0.04	10	0.245	-0.40
Adirondacks	US65	10	0.089	-1.55	10	0.788	-0.10	10	0.655	0.00	10	0.089	-1.06	10	0.421	0.15	10	0.421	0.43	10	0.531	-0.03	10	0.655	-0.04
Adirondacks	US66	**10**	**0.025**	**-1.42**	10	0.788	-0.06	10	0.531	0.00	**10**	**0.001**	**-2.50**	**10**	**0.016**	**-1.41**	10	0.128	-1.41	10	0.128	-0.04	10	0.060	0.02
Adirondacks	US67	**10**	**0.003**	**-3.76**	10	0.531	-0.35	10	0.180	0.00	**10**	**0.025**	**-5.00**	10	0.531	-0.91	10	0.929	-0.15	10	0.421	0.05	10	0.929	0.00
Adirondacks	US68	**10**	**0.000**	**-2.50**	10	0.325	0.40	10	0.089	0.01	**10**	**0.009**	**-1.53**	10	0.089	0.46	10	0.531	0.29	10	0.531	0.02	10	0.655	0.07
Adirondacks	US69	**10**	**0.000**	**-3.17**	10	0.180	0.13	**10**	**0.009**	**0.00**	**10**	**0.016**	**-3.06**	10	0.325	-0.50	10	0.929	-0.10	10	0.531	0.02	10	0.325	0.00
Adirondacks	US70	**10**	**0.002**	**-3.65**	10	0.421	-0.18	10	0.421	0.00	**10**	**0.040**	**-3.59**	10	0.325	-1.95	10	0.788	-0.52	10	0.325	0.06	10	0.245	0.02
Alps	CH03	**9**	**0.012**	**-0.65**	9	0.211	-1.41	9	0.404	-0.01	9	0.677	-0.41	9	0.463	0.19	9	0.144	1.47	7	0.881	0.00	9	0.404	0.05
Alps	CH05	**10**	**0.009**	**-1.33**	10	0.421	-0.34	10	0.245	0.00	10	0.655	0.30	10	0.929	-0.21	10	0.421	1.38	9	0.835	0.00	10	0.788	0.01
Alps	CH06	**10**	**0.003**	**-1.46**	10	0.180	-0.71	10	0.245	0.00	10	0.655	0.25	**10**	**0.020**	**0.90**	**10**	**0.006**	**2.43**	9	0.835	0.00	10	0.128	-0.02
Alps	CH09	9	0.404	0.40	9	0.835	-0.02	9	0.532	0.00	9	0.211	0.67	9	0.917	0.25	9	0.532	0.70	8	0.458	0.02	9	0.835	0.00
Alps	CH19	9	0.144	-0.99	9	0.404	-0.36	9	0.677	0.00	9	0.404	1.55	9	0.095	1.83	9	0.061	2.66	**8**	**0.048**	**0.09**	9	0.144	-0.01
Alps	CH20	**9**	**0.007**	**-1.08**	9	0.404	-0.44	9	0.835	0.00	9	0.532	-0.74	9	0.060	0.00	9	0.144	1.26	9	0.835	0.01	**9**	**0.037**	**-0.40**
Alps	CH26	9	0.144	3.79	9	0.095	1.39	9	0.532	0.00	9	0.061	10.39	9	0.211	3.34	9	0.211	6.35	9	0.835	0.01	9	0.095	0.00
Alps	CH27	9	0.404	1.23	9	0.404	0.32	9	0.677	0.00	**9**	**0.037**	**9.00**	**9**	**0.004**	**3.78**	**9**	**0.037**	**6.02**	9	0.532	0.01	9	0.061	0.00
Alps	CH28	9	0.095	0.92	9	0.532	0.53	9	1.000	0.00	**9**	**0.012**	**3.87**	**9**	**0.012**	**1.68**	**9**	**0.012**	**4.29**	9	1.000	0.00	**9**	**0.037**	**-0.01**
Alps	IT01	10	0.209	-0.45	**10**	**0.025**	**-1.17**	10	0.245	-0.01	10	0.180	1.83	9	0.075	1.30	**10**	**0.009**	**4.54**				10	0.060	-0.02
Alps	IT02	**10**	**0.001**	**-3.79**	10	0.060	0.87	**10**	**0.016**	**0.01**	**10**	**0.025**	**6.09**	**9**	**0.001**	**4.00**	**10**	**0.002**	**9.88**				10	0.321	0.00
Alps	IT03	**10**	**0.040**	**-0.61**	**10**	**0.000**	**-1.43**	**10**	**0.006**	**-0.01**	**10**	**0.016**	**1.22**	**9**	**0.007**	**1.32**	**10**	**0.000**	**3.15**				**10**	**0.040**	**-0.10**
Alps	IT04	10	0.325	1.38	10	0.421	-0.87	**10**	**0.040**	**-0.01**	10	0.325	5.90	9	0.211	4.90	10	0.060	5.35				10	0.245	0.00
Alps	IT05	10	0.325	-0.54	**10**	**0.040**	**-2.22**	10	0.325	0.00	**10**	**0.016**	**4.41**	**9**	**0.022**	**4.75**	**10**	**0.009**	**5.74**				**10**	**0.040**	**0.00**
Alps	IT06	**10**	**0.025**	**-0.98**	10	0.655	-0.45	10	0.180	0.00	**10**	**0.016**	**6.28**	9	0.095	4.36	10	0.060	5.18				10	0.245	0.00
Appalachians	US23	**10**	**0.002**	**-1.99**	10	0.128	1.41	10	0.060	0.01				**10**	**0.001**	**1.32**				10	0.180	-0.04	**10**	**0.025**	**-0.73**

1999-2008		SO4* (μeqv/L)			NO3 (μeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (μeqv/L)			Alkalinity (μeqv/L)			ANC (μeqv/L)			TOC_DOC (mg/L)			H+ (μeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Appalachians	US24	**10**	**0.001**	**-1.90**	10	0.655	0.35	10	0.325	0.01				10	0.421	0.38				10	0.531	0.03	10	0.655	-0.17
Appalachians	US25	**10**	**0.000**	**-2.02**	10	0.089	1.21	**10**	**0.016**	**0.01**				**10**	**0.003**	**1.82**				10	0.788	0.00	10	0.089	-0.81
Appalachians	US26	**10**	**0.001**	**-2.49**	10	0.089	1.30	**10**	**0.025**	**0.02**				10	0.180	-0.86				10	0.180	0.05	10	0.060	0.14
Appalachians	US35	10	0.060	-0.82	10	0.245	-0.52	10	0.180	0.00	**10**	**0.002**	**-0.87**	10	0.655	0.13	10	0.180	0.88	10	0.089	0.13	10	0.180	0.27
Appalachians	US84	**10**	**0.016**	**-2.06**	10	0.128	-0.20	10	0.421	0.00				10	0.788	0.23				10	0.421	-0.02	10	0.655	0.00
Appalachians	US85	**10**	**0.006**	**-2.42**	**10**	**0.040**	**-1.61**	10	0.060	-0.01				10	0.325	1.95				**10**	**0.016**	**-0.02**	10	0.531	-0.07
Appalachians	US86	10	0.245	-1.05	10	0.128	0.19	10	0.245	0.00				10	0.325	1.19				**10**	**0.009**	**-0.10**	10	0.788	-0.02
Appalachians	US87	**10**	**0.040**	**-2.22**	10	0.325	0.14	10	0.245	0.00				10	0.929	-0.04				10	0.151	-0.03	10	0.929	0.01
Appalachians	US88	10	0.245	1.01	**10**	**0.006**	**-1.94**	**10**	**0.009**	**-0.01**				10	0.245	-0.92				10	0.180	-0.02	10	0.788	0.01
Blue Ridge Mountains	US103	8	0.138	-0.57	8	0.805	0.02	8	1.000	0.00				8	0.621	-0.20							8	0.621	0.03
Blue Ridge Mountains	US104	8	0.458	-0.52	8	0.083	-0.77	8	0.216	-0.01				8	0.805	1.35							8	0.322	0.00
Blue Ridge Mountains	US105	8	0.805	0.05	8	0.083	-0.07	**8**	**0.048**	**0.00**				8	0.805	0.39							8	0.805	0.00
ECEurope	CZ01	**10**	**0.006**	**-3.11**	10	0.060	-0.95	10	0.180	0.00	10	0.180	-0.78	9	0.249	0.43	**10**	**0.025**	**2.74**	10	0.325	0.07	**10**	**0.040**	**-1.17**
ECEurope	CZ02	**10**	**0.006**	**-3.50**	**10**	**0.016**	**-0.92**	10	0.325	0.00	**10**	**0.016**	**-0.98**	9	0.677	0.56	**10**	**0.025**	**3.50**	10	0.369	0.04	**10**	**0.016**	**-2.46**
ECEurope	CZ03	**10**	**0.002**	**-3.74**	**10**	**0.006**	**4.61**	**10**	**0.001**	**0.05**	10	0.128	1.31	9	0.297	0.99	10	0.929	0.19	**10**	**0.048**	**0.13**	10	0.089	-0.41
ECEurope	CZ04	10	0.128	-1.40	10	0.245	-0.69	10	0.325	-0.01	10	0.180	-0.68	9	0.144	1.48	10	0.655	1.61	10	0.089	0.17	**10**	**0.040**	**-1.14**
ECEurope	CZ05	10	0.788	-0.50	10	0.180	1.15	**10**	**0.040**	**0.01**	10	0.929	-0.17	9	0.677	0.24	10	0.655	-0.74	10	0.325	0.20	10	0.128	-0.14
ECEurope	CZ06	10	0.325	-1.74	10	0.824	0.00	10	0.788	0.00	10	0.531	-2.13	9	0.532	1.14	10	0.788	1.04	10	0.531	0.35	10	0.655	-0.02
ECEurope	CZ07	**10**	**0.009**	**-12.6**	10	0.325	0.00	10	0.325	0.00	**10**	**0.040**	**-5.09**	10	0.325	1.51	**10**	**0.002**	**5.96**	**10**	**0.040**	**0.74**	10	0.245	-0.86
ECEurope	CZ08	10	0.788	-0.68	10	0.325	0.43	10	0.531	0.00	10	0.421	1.72	10	0.325	2.26	10	0.531	2.47				10	0.245	-0.43
ECEurope	DE02	8	0.322	0.43	8	0.458	-0.21	7	0.293	0.00	10	0.929	-0.20	7	0.881	-0.21	7	0.099	7.82				**10**	**0.040**	**-0.43**
ECEurope	DE04	7	0.881	-20.4				7	0.176	0.00	7	0.652	6.82				**7**	**0.024**	**58.14**				7	0.453	-0.80
ECEurope	DE07	**10**	**0.000**	**-14.0**	**10**	**0.025**	**-1.79**	10	0.325	0.00	**10**	**0.003**	**-8.87**	8	0.615	-0.30	**10**	**0.006**	**15.82**	9	0.835	0.01	10	0.531	-0.66
ECEurope	DE08	8	0.621	0.38	10	0.245	-1.42	8	0.621	0.00	9	0.677	0.61				8	0.458	4.75				10	0.325	-0.19
ECEurope	DE09	8	0.138	177				8	0.083	0.00	8	0.216	102.53				8	0.458	-37.81	7	0.652	-0.31	8	0.216	7.12
ECEurope	DE10	8	0.805	0.42	**10**	**0.003**	**-6.62**	**8**	**0.013**	**-0.02**	9	0.211	1.81				**8**	**0.006**	**19.36**				10	0.089	-0.37
ECEurope	DE13																								
ECEurope	DE17	8	0.138	-1.76				8	0.458	-0.01	8	0.083	9.12				**8**	**0.026**	**22.24**				8	0.458	-2.10
ECEurope	DE18	**8**	**0.001**	**-2.69**	8	0.138	-1.43	7	0.881	0.00	10	0.531	-0.38	7	0.530	-0.92	7	0.051	8.21				10	0.180	-1.22
ECEurope	DE21	10	0.060	-12.0	**10**	**0.000**	**-5.60**	**10**	**0.000**	**-0.01**	**10**	**0.003**	**-21.6**	8	0.262	1.25	10	0.788	1.98				10	0.245	-0.21
ECEurope	DE23	7	0.453	-0.31	10	0.531	0.39	7	0.176	0.01	7	0.176	3.37							9	0.677	0.07	10	0.531	-0.08
ECEurope	DE24	**8**	**0.001**	**-18.1**							**8**	**0.003**	**-21.5**										**8**	**0.013**	**-0.83**
ECEurope	DE25	8	0.322	-10.5				8	0.621	0.00	8	0.458	12.61				**8**	**0.026**	**41.90**				8	0.805	0.00
ECEurope	DE27	7	0.051	-1.15	10	0.245	-1.56	7	0.453	0.01	8	0.621	1.17				7	0.293	6.56	7	0.293	-0.10	10	0.325	-0.06

1999-2008		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
ECEurope	DE28	9	0.144	5.93	9	0.144	-2.21	8	0.216	-0.02	9	0.835	9.77	8	0.083	-15.31	8	1.000	2.43				10	0.929	-0.01
ECEurope	DE29	9	0.211	-4.80	9	0.677	0.60	8	0.621	0.00	9	0.677	6.32	**8**	**0.026**	**-12.95**	8	0.621	7.78	9	0.404	0.18	10	0.929	0.05
ECEurope	DE30	**9**	**0.007**	**-13.0**	**9**	**0.004**	**-3.29**	**9**	**0.012**	**0.00**	9	0.211	-6.31	7	0.881	5.15	**9**	**0.012**	**20.50**	9	0.144	0.27	9	0.211	0.03
ECEurope	DE31	10	0.128	-22.4	**10**	**0.040**	**-5.82**	10	0.531	0.00	**10**	**0.016**	**-22.8**	8	1.000	-0.27	10	0.128	23.31	9	0.835	0.01	10	0.788	0.00
ECEurope	DE33	**8**	**0.003**	**-12.2**	**9**	**0.004**	**-2.13**	7	0.881	0.00	**10**	**0.040**	**-5.72**	**7**	**0.021**	**-3.89**	**7**	**0.024**	**15.27**	9	0.211	0.15	**10**	**0.006**	**-5.04**
ECEurope	PL01	**7**	**0.011**	**-2.17**	7	0.099	-2.31	7	0.453	0.00	7	0.881	1.58				7	0.099	6.08				7	0.881	0.08
ECEurope	PL02	**7**	**0.011**	**-2.30**	7	0.176	-0.88	7	0.652	-0.01	7	0.453	-0.74				7	0.051	3.21				7	0.176	0.09
ECEurope	PL03																								
ECEurope	PL04																								
Maine/Atlantic	CA10	10	0.060	-1.27				10	0.060	0.01	10	0.180	-0.32				10	0.655	0.30	10	0.325	0.08	10	0.089	-0.49
Maine/Atlantic	CA11	**10**	**0.000**	**-1.47**	10	0.681	0.00	10	0.060	0.00	**10**	**0.040**	**-1.16**				10	0.655	-0.63	**10**	**0.031**	**0.16**	10	0.929	-0.02
Maine/Atlantic	CA12	**10**	**0.006**	**-1.63**				**10**	**0.006**	**0.00**	10	0.421	-0.37				10	0.655	-0.55	10	0.421	0.09	10	0.089	-0.77
Maine/Atlantic	CA13	**10**	**0.040**	**-1.33**	10	0.862	0.00	**10**	**0.040**	**0.00**	**10**	**0.040**	**-1.22**				10	0.421	-0.82	10	0.128	0.16	10	0.788	-0.09
Maine/Atlantic	CA14	**10**	**0.000**	**-1.35**	10	0.569	0.00	**10**	**0.009**	**0.00**	10	0.089	-0.63				10	0.325	-1.04	10	0.281	0.07	10	0.089	-0.12
Maine/Atlantic	US05	**10**	**0.003**	**-1.46**	10	0.357	0.00	10	0.416	0.00	7	0.652	-0.29	10	0.929	-0.02	7	0.453	2.24	**9**	**0.037**	**0.10**	10	0.857	0.00
Maine/Atlantic	US06	**10**	**0.006**	**-1.20**	10	0.652	0.01	10	0.531	0.00	7	0.881	0.18	10	0.176	-1.00	7	0.293	1.98	9	0.345	0.08	10	0.655	0.02
Maine/Atlantic	US71	**9**	**0.037**	**-3.58**	9	0.273	-0.02	9	0.273	0.00				9	0.677	-0.44				9	1.000	0.00	9	0.835	0.00
Maine/Atlantic	US72	**10**	**0.025**	**-1.96**	10	1.000	0.00	10	0.719	0.00	7	0.176	-5.66	10	0.089	-1.73	7	0.652	-0.89	10	0.325	0.25	10	0.180	-0.01
Maine/Atlantic	US73	**9**	**0.037**	**-0.96**	9	0.345	-0.02	9	0.532	0.00				9	0.833	0.05				8	0.527	0.02	9	0.600	0.02
Maine/Atlantic	US74	**10**	**0.009**	**-2.37**	**10**	**0.001**	**-0.06**	**10**	**0.001**	**0.00**	7	0.099	-0.69	10	0.180	-0.70	7	0.652	0.65	10	0.089	0.12	10	0.929	0.37
Maine/Atlantic	US75	**10**	**0.009**	**-0.51**	10	0.209	-0.04	10	0.245	0.00	7	0.453	-0.31	**10**	**0.040**	**-1.27**	7	0.881	-0.23	**10**	**0.009**	**-0.11**	10	0.531	0.00
Maine/Atlantic	US76	**10**	**0.040**	**-1.72**	10	0.857	0.00	10	0.929	0.00	7	0.652	2.91	10	0.655	0.88	7	0.652	2.24	10	0.369	-0.07	10	0.531	0.00
Maine/Atlantic	US77	**10**	**0.040**	**-0.98**	10	0.180	-0.04	10	0.325	0.00	7	0.881	-0.40	10	0.655	-0.28	7	0.453	0.74	10	0.929	-0.02	10	0.245	-0.01
Maine/Atlantic	US78	10	0.325	-0.22	10	0.717	0.01	10	0.717	0.00	7	0.881	1.18	10	0.128	-3.56	7	0.881	-0.56	10	0.717	0.01	10	0.929	0.00
Maine/Atlantic	US79	**9**	**0.001**	**-3.05**	9	0.061	-0.07	9	0.211	0.00	7	0.881	-0.74	**9**	**0.022**	**1.49**	**7**	**0.024**	**2.85**	**9**	**0.046**	**0.21**	**9**	**0.037**	**-1.31**
Maine/Atlantic	US80	**10**	**0.016**	**-1.80**	10	0.212	-0.03	10	0.212	0.00	7	0.453	-0.23	10	0.788	0.22	7	0.051	2.70	**10**	**0.040**	**0.16**	10	0.531	-0.01
Maine/Atlantic	US81	**10**	**0.009**	**-1.19**	10	0.204	0.04	10	0.146	0.00	7	0.453	-0.35	10	0.929	-0.03	7	0.293	0.34	10	0.058	-0.06	**10**	**0.006**	**-0.09**
Maine/Atlantic	US82	9	0.144	-4.51	9	0.532	-0.06	9	0.297	0.00				9	0.116	1.50				9	0.835	0.19	9	0.144	-0.48
Maine/Atlantic	US83	**10**	**0.009**	**-1.23**	10	0.128	-0.02	10	0.245	0.00	7	0.176	0.63	10	0.325	-0.40	7	0.051	2.72	10	0.128	0.11	10	0.531	-0.01
NoNordic	FI05	9	0.404	-0.40	9	0.404	-0.11	9	0.532	0.00	9	0.404	0.52	9	0.677	-0.97	9	0.532	0.74	9	0.677	0.10	9	0.532	0.01
NoNordic	FI06	**9**	**0.022**	**-0.67**	**9**	**0.046**	**-0.18**	9	0.095	0.00	**9**	**0.005**	**-0.89**	**9**	**0.002**	**0.98**	9	0.532	-0.23	9	0.673	-0.01	9	0.677	-0.04
NoNordic	FI08	**9**	**0.000**	**-1.56**	9	0.095	-0.13	9	0.835	0.00	9	0.404	-0.30	9	0.061	0.84	9	0.061	1.15	9	0.095	0.21	9	0.677	0.22
NoNordic	NO04	**10**	**0.009**	**-2.19**	10	0.128	-0.06	10	0.128	0.00	10	0.788	-0.11	**10**	**0.025**	**1.16**	10	0.089	1.89	10	0.089	0.04	**10**	**0.001**	**-0.03**
NoNordic	SE01	**9**	**0.022**	**-1.91**	**9**	**0.037**	**-0.53**	9	0.144	0.00	9	0.297	-0.40	**9**	**0.007**	**3.58**	**9**	**0.012**	**2.95**	9	0.532	-0.07	**9**	**0.022**	**0.00**

1999-2008		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
NoNordic	SE05	9	0.677	-0.16	**9**	**0.037**	**-0.07**	9	0.095	0.00	9	0.144	4.13	9	0.345	2.87	9	0.144	4.43	9	0.835	0.02	**9**	**0.022**	**-0.02**
NoNordic	SE06	**9**	**0.004**	**-1.54**	9	0.144	-0.07	9	0.835	0.00	9	0.677	-0.10	**9**	**0.037**	**1.56**	**9**	**0.007**	**2.41**	9	0.835	-0.01	9	0.297	-0.02
SoNordic	NO01	**10**	**0.016**	**-2.01**	10	0.421	-0.28	10	0.655	0.00	10	0.180	-0.59	10	0.421	0.08	10	0.245	0.94	10	0.325	0.08	10	0.929	0.06
SoNordic	FI01	**9**	**0.022**	**-4.35**	9	0.677	-0.28	9	0.677	0.00	**9**	**0.012**	**-1.12**	9	0.532	0.23	9	0.095	2.36	**9**	**0.037**	**0.13**	9	0.144	-0.15
SoNordic	FI02	9	0.211	-2.43	**9**	**0.000**	**-0.64**	**9**	**0.001**	**-0.01**	9	0.404	-1.08	9	1.000	0.01	9	0.404	1.36	9	0.144	0.26	9	1.000	0.01
SoNordic	FI03	**9**	**0.001**	**-2.71**	9	0.297	0.10	9	0.144	0.00	**9**	**0.007**	**-2.88**	9	0.463	-0.19	9	0.404	-1.32	9	0.173	-0.05	9	0.211	-0.14
SoNordic	FI07	**9**	**0.000**	**-4.39**	9	0.532	0.05	9	0.144	0.00	**9**	**0.012**	**-5.44**	9	1.000	-0.12	9	0.297	-1.37	**9**	**0.004**	**0.22**	9	0.677	0.00
SoNordic	FI09	**9**	**0.012**	**-2.47**	9	0.061	0.13	**9**	**0.037**	**0.00**	9	0.835	0.20	9	0.095	1.38	9	0.061	2.72	9	0.061	0.08	9	0.297	-0.01
SoNordic	NO03	**10**	**0.000**	**-1.42**	**10**	**0.000**	**-0.04**	10	0.788	0.00	10	0.245	-0.88	10	0.929	0.00	10	0.531	0.67	10	0.089	0.14	10	0.929	-0.01
SoNordic	NO10	**10**	**0.009**	**-1.26**	**10**	**0.009**	**-0.47**	10	0.060	-0.01	10	0.245	-0.27	**10**	**0.031**	**0.05**	**10**	**0.009**	**1.51**	10	0.060	0.07	**10**	**0.009**	**-0.47**
SoNordic	NO11	10	0.060	-0.44	10	0.060	-0.10	10	0.929	0.00	10	0.929	0.01	10	0.655	0.14	10	0.531	0.32	10	0.929	0.00	10	0.089	-0.01
SoNordic	SE02	**9**	**0.037**	**-10.6**	9	0.144	0.44	**9**	**0.004**	**0.01**	9	0.095	-12.8	9	0.061	4.64	9	0.835	0.74	9	0.095	0.17	9	0.211	-0.01
SoNordic	SE08	9	0.095	-8.98	9	0.297	-0.07	9	0.211	0.00	9	0.532	-3.42	9	0.532	0.34	**9**	**0.037**	**2.08**	9	0.095	0.62	9	0.061	-0.10
SoNordic	SE09	**9**	**0.000**	**-6.43**	**9**	**0.037**	**0.88**	**9**	**0.022**	**0.01**	**9**	**0.002**	**-3.58**	**9**	**0.007**	**3.15**	9	0.211	4.28	9	0.211	0.17	9	0.211	-0.01
SoNordic	SE10	**9**	**0.004**	**-4.51**	9	0.297	-0.09	9	0.677	0.00	**9**	**0.037**	**-1.77**	9	0.532	0.27	**9**	**0.037**	**3.84**	9	0.835	0.06	9	0.297	-0.15
SoNordic	SE11	**9**	**0.007**	**-5.15**	**9**	**0.007**	**-0.54**	9	0.297	0.00	9	0.061	-3.31	9	1.000	-0.14	9	0.835	-0.20	9	1.000	-0.02	9	0.297	-0.02
SoNordic	SE12	**9**	**0.002**	**-6.44**	9	0.144	-0.21	**9**	**0.007**	**0.01**	**9**	**0.022**	**-1.68**	**9**	**0.012**	**2.64**	9	0.211	3.02	9	1.000	-0.01	**9**	**0.012**	**-1.12**
UK	UK01	10	0.151	-0.90	**10**	**0.016**	**0.16**	10	0.369	0.05	10	0.245	-1.44	9	0.144	0.88	10	0.245	-2.22	9	0.835	0.07	9	0.835	-0.01
UK	UK04	**10**	**0.016**	**-0.67**	10	0.128	-0.59	10	0.788	0.00	**10**	**0.025**	**-1.03**	10	0.060	0.47	10	0.788	-0.11	**10**	**0.031**	**0.11**	10	0.128	-0.21
UK	UK07	10	0.788	-0.15	**10**	**0.009**	**0.54**	**10**	**0.016**	**0.01**	10	0.531	0.60	10	0.590	-0.25	10	0.655	-0.40	**10**	**0.040**	**0.25**	10	0.929	-0.10
UK	UK10	10	0.128	-0.76	**10**	**0.016**	**-0.68**	10	0.531	-0.01	10	0.531	-0.20	10	1.000	0.00	10	0.929	-0.01	10	0.281	0.07	10	0.421	-0.25
UK	UK15	**10**	**0.040**	**-0.88**	10	0.128	0.34	10	0.089	0.01	10	0.929	-0.13	10	0.929	-0.01	10	0.531	-0.56	9	0.211	0.10	10	0.788	0.02
UK	UK21	**10**	**0.016**	**-1.41**	**10**	**0.040**	**-0.91**	10	0.655	0.00	10	0.060	-0.81	10	0.655	-0.42	10	0.929	0.19	10	0.325	0.14	**10**	**0.016**	**-0.52**
Ontario	CA01	**10**	**0.003**	**-1.85**	10	0.421	-0.13	10	0.421	0.00	10	0.060	2.01	7	0.176	1.12	**10**	**0.006**	**4.51**	10	0.655	-0.03	10	0.421	0.02
Ontario	CA02	**10**	**0.016**	**-0.94**	10	0.325	0.29	10	0.245	0.00	**10**	**0.025**	**4.03**	7	0.176	1.74	**10**	**0.009**	**4.94**	10	0.128	-0.08	10	0.128	0.01
Ontario	CA03	**10**	**0.016**	**-0.81**	10	0.531	0.30	10	0.245	0.00	10	0.060	3.13	7	0.881	-0.40	**10**	**0.025**	**4.11**	10	0.421	-0.02	**10**	**0.040**	**0.01**
Ontario	CA04	10	0.325	-0.43	10	0.421	0.42	10	0.325	0.00	**10**	**0.003**	**4.50**	7	0.293	1.54	**10**	**0.006**	**5.31**	10	0.421	0.03	**10**	**0.006**	**0.01**
Ontario	CA16	**10**	**0.001**	**-1.02**	10	0.655	0.01	10	0.531	0.00	9	0.532	0.31	10	0.421	0.79	9	0.095	1.30	8	0.458	0.04	10	0.655	0.00
Ontario	CA17	**10**	**0.009**	**-1.41**	10	0.421	0.02	10	0.180	0.00	10	0.788	0.20	10	0.060	-0.87	**10**	**0.040**	**1.11**	**9**	**0.037**	**-16.67**	**10**	**0.016**	**-0.01**
Ontario	CA20	**10**	**0.003**	**-0.69**	10	0.089	0.03	10	0.060	0.00	**10**	**0.003**	**2.32**	10	0.180	0.60	**10**	**0.009**	**3.04**	9	0.404	-4.58	10	0.089	0.00
Vermont/Quebec	CA05	10	0.325	-1.41	10	0.407	-0.18	10	0.929	0.00	10	0.245	-0.49	10	0.281	0.50	10	0.929	0.08	**10**	**0.024**	**0.08**	10	0.788	0.00
Vermont/Quebec	CA06	10	0.325	-0.90	10	0.247	-0.04	10	0.655	0.00	**10**	**0.000**	**-1.30**	10	0.147	-0.33	10	0.245	-1.27	**10**	**0.005**	**0.09**	10	0.531	0.02
Vermont/Quebec	CA07	**10**	**0.025**	**-2.03**	10	0.083	-0.24	10	0.245	0.00	**10**	**0.016**	**-2.99**	10	0.652	-0.33	10	0.180	-1.21	10	0.241	0.07	10	0.128	0.28
Vermont/Quebec	CA08	**10**	**0.002**	**-0.93**	10	0.929	0.01	10	0.421	0.00	10	0.325	-0.74	10	0.128	-1.86	10	0.655	-0.44	10	0.325	0.06	10	0.929	-0.01

1999-2008		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	ID	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Vermont/Quebec	CA09	10	0.245	-0.75	10	0.351	0.00	10	0.929	0.00	10	0.060	-1.14	10	0.281	0.25	10	0.128	-0.74	10	0.321	0.03	10	0.655	-0.09
Vermont/Quebec	US100	**10**	**0.025**	**-1.02**	**10**	**0.002**	**0.86**	**10**	**0.006**	**0.01**	**10**	**0.016**	**-1.63**	10	0.788	0.30	10	0.089	-2.56	10	0.151	-0.10	10	0.929	0.00
Vermont/Quebec	US102	10	0.421	-0.82	10	0.151	0.35	10	0.106	0.06	10	0.655	-0.36	**10**	**0.025**	**1.82**	10	0.128	-5.96	10	0.151	0.19	**10**	**0.025**	**-1.26**
Vermont/Quebec	US123	**10**	**0.002**	**-1.40**	**10**	**0.040**	**1.31**	**10**	**0.016**	**0.02**	10	0.655	-0.09	10	0.788	-0.03	10	0.655	-0.15	10	0.719	0.02	10	0.531	-0.05
Vermont/Quebec	US89	**10**	**0.006**	**-1.80**	10	0.106	0.18	**10**	**0.016**	**0.00**	10	0.180	-0.46	10	0.060	1.13	10	0.325	0.76	10	0.369	-0.03	**10**	**0.040**	**-0.32**
Vermont/Quebec	US90	**10**	**0.025**	**-1.04**	10	0.236	0.11	10	0.060	0.00	**10**	**0.016**	**-1.32**	10	0.421	-0.20	10	0.128	-0.88	10	0.125	0.06	10	0.655	-0.01
Vermont/Quebec	US91	**10**	**0.003**	**-2.06**	10	0.710	0.00	10	0.655	0.00	10	0.060	-1.05	10	0.655	0.34	10	0.929	0.31	10	0.590	-0.03	10	0.128	-0.02
Vermont/Quebec	US92	**10**	**0.003**	**-2.18**	**10**	**0.012**	**1.58**	**10**	**0.003**	**0.02**	10	0.245	-0.62	10	0.421	0.22	10	0.929	0.05	10	0.586	0.03	10	0.655	-0.09
Vermont/Quebec	US93	**10**	**0.001**	**-1.78**	**10**	**0.016**	**1.03**	**10**	**0.003**	**0.01**	**10**	**0.040**	**-1.24**	10	0.421	-0.70	10	0.421	-1.02	10	0.788	0.01	10	0.531	0.11
Vermont/Quebec	US95	**10**	**0.002**	**-1.72**	10	0.928	0.00	10	0.421	0.00	**10**	**0.025**	**-1.68**	10	0.245	0.41	10	0.421	-0.51	10	0.531	0.02	10	0.325	-0.01
Vermont/Quebec	US96	**10**	**0.031**	**-1.86**	10	0.281	0.30	10	0.281	0.01	10	0.060	-0.70	**10**	**0.048**	**0.78**	10	0.655	0.49	10	0.180	-0.08	10	0.089	-0.22
Vermont/Quebec	US97	**10**	**0.002**	**-1.96**	10	0.060	0.70	10	0.060	0.01	**10**	**0.016**	**-0.91**	10	0.089	0.61	10	0.929	0.17	10	0.927	0.00	10	0.089	-0.28
Vermont/Quebec	US98	**10**	**0.000**	**-3.77**	10	0.928	0.00	10	0.531	0.00	**10**	**0.000**	**-5.14**	10	0.180	-0.78	**10**	**0.016**	**-1.58**	**10**	**0.048**	**0.07**	10	0.655	0.01
WCEurope	DE01	10	0.655	-0.15	10	0.128	-0.89	10	0.531	0.00	**10**	**0.002**	**-5.45**				**10**	**0.016**	**7.46**	10	0.929	0.02	**10**	**0.025**	**-0.08**
WCEurope	DE03	8	0.083	-3.24	**8**	**0.003**	**4.37**	**8**	**0.001**	**0.01**	8	0.322	-4.79										10	0.060	-0.11
WCEurope	DE05	10	0.788	0.33	**10**	**0.025**	**-1.06**	**10**	**0.016**	**-0.01**	**10**	**0.040**	**-5.02**				10	0.531	-1.72	**10**	**0.016**	**0.08**	10	0.655	-0.03
WCEurope	DE06	10	0.180	-3.51				**8**	**0.048**	**0.02**	10	0.089	7.17	**8**	**0.002**	**-4.62**	**8**	**0.048**	**11.95**	10	0.128	-0.14	10	0.531	-0.28
WCEurope	DE11																								
WCEurope	DE12				**9**	**0.022**	**1.93**				9	0.297	3.11							7	0.881	0.00	**9**	**0.012**	**-0.18**
WCEurope	DE14																								
WCEurope	DE16																								
WCEurope	DE19																								
WCEurope	DE26	**10**	**0.006**	**-5.88**	**8**	**0.048**	**1.92**	**8**	**0.048**	**0.02**	10	0.655	-1.33	8	0.322	-4.79	**8**	**0.026**	**7.89**	10	0.788	0.04	10	0.325	-1.26
WCEurope	DE32	**8**	**0.026**	**-3.73**				**8**	**0.003**	**0.01**	8	0.083	-7.65										10	0.655	-0.06
WCEurope	DE34																								
WCEurope	DE35	7	0.099	-21.5	**7**	**0.024**	**-12.4**	7	0.453	0.01	**7**	**0.011**	**-37.7**				7	0.293	5.41	7	0.051	-0.21	7	0.453	-0.16

Table D4. *Results of trend analysis of data from 13 extra Ontario stations for the time span 1999-2008. Andrew Paterson (Ontario Ministry of Environment) provided data from the 8 Dorset lakes. Bill Keller (Laurentian University and Ontario Ministry of Environment) provided data from the 5 Sudbury lakes.*

		SO4* (µeqv/L)			NO3 (µeqv/L)			NO3/(NO3+SO4*)			Ca*+Mg* (µeqv/L)			Alkalinity (µeqv/L)			ANC (µeqv/L)			TOC_DOC (mg/L)			H+ (µeqv/L)		
Region	Station Name	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope	n	p	slope
Ontario (Dorset)	Blue Chalk Lake	**9**	**0.001**	**-1.78**	9	0.211	-0.12	9	0.297	0.00	9	0.144	-2.34	9	0.061	2.28	9	0.835	-0.39	9	0.532	0.01	9	1.000	0.00
Ontario (Dorset)	Chub Lake	**9**	**0.007**	**-1.93**	**9**	**0.037**	**-0.21**	9	0.095	0.00	**9**	**0.037**	**-1.59**	**9**	**0.037**	**0.84**	9	0.532	0.60	**9**	**0.037**	**0.12**	9	0.677	0.01
Ontario (Dorset)	Crosson Lake	9	0.061	-1.97	9	0.677	-0.06	9	1.000	0.00	**9**	**0.022**	**-1.62**	**9**	**0.022**	**1.34**	9	0.095	0.59	9	0.404	0.05	9	0.677	-0.02
Ontario (Dorset)	Dickie Lake	9	1.000	0.03	9	0.297	-0.10	9	0.404	0.00	9	0.835	-0.16	9	0.144	1.00	9	0.532	-0.62	9	0.211	0.06	9	0.404	0.01
Ontario (Dorset)	Harp Lake	9	0.095	-0.70	9	1.000	0.00	9	1.000	0.00	9	1.000	0.10	**9**	**0.022**	**1.12**	9	0.297	1.02	**9**	**0.022**	**0.02**	9	0.532	0.02
Ontario (Dorset)	Heney Lake	9	0.835	-0.11	**9**	**0.012**	**-0.13**	**9**	**0.007**	**0.00**	**9**	**0.022**	**-1.83**	9	0.249	-0.72	**9**	**0.012**	**-1.78**	9	0.297	0.03	9	0.297	0.02
Ontario (Dorset)	Plastic Lake	**9**	**0.004**	**-1.62**	9	0.297	-0.08	9	0.532	0.00	**9**	**0.012**	**-1.84**	**9**	**0.037**	**0.68**	9	0.677	0.24	9	0.061	0.03	9	0.677	0.08
Ontario (Dorset)	Red Chalk Lake	**9**	**0.000**	**-1.96**	9	0.211	-0.17	9	0.677	0.00	**9**	**0.037**	**-1.69**	9	0.095	1.33	9	0.532	0.42	**9**	**0.022**	**0.07**	9	0.211	0.03
Ontario (Sudbury)	Clearwater	8	0.138	-0.71	8	0.322	-0.04	8	0.533	0.00	8	0.322	-1.80				**8**	**0.026**	**8.08**	**8**	**0.016**	**0.10**	**8**	**0.026**	**-0.04**
Ontario (Sudbury)	Sans Chambre	8	0.138	2.01	8	0.083	-0.23	8	0.083	0.00	8	0.216	1.87				8	0.458	1.38	**8**	**0.048**	**-0.07**	8	0.621	-0.01
Ontario (Sudbury)	Whitepine (Aurora)	8	0.083	-1.96	8	0.083	-0.18	8	0.216	0.00	**8**	**0.001**	**-3.14**				8	0.621	-1.38	8	1.000	0.00	8	0.216	0.24
Ontario (Sudbury)	Whitepine (Mcleod	**8**	**0.003**	**-3.08**	8	1.000	-0.01	8	0.621	0.00	8	0.458	-0.53				8	0.083	2.67	**8**	**0.026**	**0.08**	8	0.458	0.01
Ontario (Sudbury)	Swan Lake	10	0.128	-2.09	10	0.788	-0.05	10	0.929	0.00	**10**	**0.003**	**-4.30**				10	0.929	-0.31	10	0.929	0.00			

Appendix E. Results of regional trend analysis

Table E1. *Results of regional trend analysis 1990-1999. Median trend slopes in µeqv/L/y (mg/L/y for TOC_DOC); p is the probability that there is no consistent unidirectional trend (Regional Kendall Test) and n is the total number of observations (yearly means) for all sites included from each region.*

1990-1999		SO4*			NO3			NO3/(NO3+SO4*)	
Region	p	Slope	n	p	Slope	n	p	Slope	n
Adirondacks	0.000	-2.31	434	0.020	-0.18	434	0.086	0.001	434
Alps	0.000	-2.12	60	0.010	0.49	60	0.000	0.007	60
Appalachians	0.000	-2.77	48	0.190	-0.28	48	0.704	0.000	48
Blue Ridge Mountains	0.240	0.4	28	0.000	-0.96	28	0.000	-0.013	28
ECEurope	0.000	-3.39	198	0.130	-0.53	76	0.007	0.004	76
Maine_Atlantic	0.000	-1.15	145	0.000	0.02	145	0.000	0.001	145
NoNordic	0.000	-1.5	70	0.220	0.03	70	0.057	0.001	70
SoNordic	0.000	-5.29	147	0.450	0.03	147	0.000	0.002	147
UK	0.000	-1.09	60	0.400	0.18	60	0.000	0.012	60
Ontario	0.000	-3.98	180	0.440	0.01	180	0.000	0.001	180
Vermont_Quebec	0.000	-3.04	166	0.000	-0.24	166	0.017	-0.001	166
WCEurope	0.000	-4.64	60	0.000	-1.14	60	0.381	-0.001	60

		Ca*+Mg*			Alkalinity			ANC	
	p	Slope	n	p	Slope	n	p	Slope	n
Adirondacks	0.000	-2.39	424	0.000	0.98	434	0.070	-0.36	424
Alps	0.100	-1.08	60	0.000	1.1	60	1.000	0	60
Appalachians	0.110	-1.41	8	0.000	1.39	48	0.270	1.85	8
Blue Ridge Mountains				0.960	-0.03	28			
ECEurope	0.000	-1.33	189	0.930	-0.07	131	0.000	3.06	129
Maine_Atlantic	0.000	-1	128	0.100	-0.22	99	0.910	0.03	128
NoNordic	0.000	-0.7	70	0.000	1.13	70	0.060	1.12	70
SoNordic	0.000	-2.72	147	0.000	0.48	147	0.000	3.23	140
UK	0.020	-0.63	60	0.960	0	30	0.400	0.16	60
Ontario	0.000	-3.66	180	0.170	0.32	130	0.170	0.23	180
Vermont_Quebec	0.000	-1.77	166	0.020	0.4	166	0.000	1.42	166
WCEurope	0.250	-2.37	70	0.090	2.38	20	0.000	11.4	40

		TOC_DOC			H+	
	p	Slope	n	p	Slope	n
Adirondacks	0.120	0.02	434	0.000	-0.05	434
Alps				0.240	0	60
Appalachians	0.640	0.01	48	0.530	-0.03	48
Blue Ridge Mountains				0.870	0	28
ECEurope	0.820	0.13	28	0.000	-0.16	198
Maine_Atlantic	0.140	0.03	145	0.750	0	145
NoNordic	0.010	0.06	70	0.000	-0.02	70
SoNordic	0.000	0.11	147	0.000	-0.06	147
UK	0.000	0.13	60	0.010	-0.16	60
Ontario	0.010	0.02	180	0.000	-0.02	180
Vermont_Quebec	0.000	0.1	50	0.150	-0.02	166
WCEurope	0.680	0.02	50	1.000	0	70

Table E2. *Results of regional trend analysis 1999-2008. Median trend slopes in µeqv/L/y (mg/L/y for TOC_DOC); p is the probability that there is no consistent unidirectional trend (Regional Kendall Test) and n is the total number of observations (yearly means) for all sites included from each region.*

1999-2008	SO4*			NO3			NO3/(NO3+SO4*)		
Region	p	Slope	n	p	Slope	n	p	Slope	n
Adirondacks	0.000	-2.4	499	0.290	-0.03	499	0.000	0.002	499
Alps	0.000	-0.8	60	0.000	-1.13	60	0.188	-0.002	60
Appalachians	0.000	-1.2	50	0.690	-0.12	50	0.810	0.000	50
Blue Ridge Mountains	0.280	-0.3	24	0.060	-0.11	24	0.099	-0.002	24
ECEurope	0.000	-1.7	201	0.260	-0.16	74	0.022	0.004	74
Maine_Atlantic	0.000	-1.4	167	0.060	0	167	0.231	0.000	167
NoNordic	0.000	-1.1	64	0.000	-0.11	64	0.008	-0.001	64
SoNordic	0.000	-2.6	139	0.000	-0.07	139	0.089	0.001	139
UK	0.000	-0.9	60	0.860	0.05	60	0.133	0.006	60
Ontario	0.000	-1.3	164	0.000	-0.07	164	0.219	0.000	164
Vermont_Quebec	0.000	-1.6	172	0.000	0.09	172	0.000	0.004	172
WCEurope	0.078	-1.6	60	0.000	1.97	59	0.000	0.013	59

	Ca*+Mg*			Alkalinity			ANC		
	p	Slope	n	p	Slope	n	p	Slope	n
Adirondacks	0.000	-1.62	489	0.250	0.07	499	0.000	0.55	489
Alps	0.000	3.49	60	0.000	3.29	60	0.000	4.86	60
Appalachians	0.000	-0.87	10	0.750	0.02	50	0.280	0.77	10
Blue Ridge Mountains				1.000	-0.02	24			
ECEurope	0.040	-0.6	192	0.730	0.28	133	0.000	2.81	133
Maine_Atlantic	0.000	-0.97	130	0.060	-0.42	117	0.160	-0.67	130
NoNordic	0.260	-0.22	64	0.000	0.99	64	0.000	1.24	64
SoNordic	0.000	-1.43	139	0.000	0.21	139	0.000	1.09	130
UK	0.030	-0.39	60	0.470	0.25	30	0.320	-0.43	60
Ontario	0.070	-0.5	164	0.000	0.96	114	0.000	1.2	164
Vermont_Quebec	0.000	-1.07	172	0.150	0.15	172	0.000	-0.45	172
WCEurope	0.629	-0.95	69	0.020	-4.81	20	0.100	-3.65	40

	TOC_DOC			H+		
	p	Slope	n	p	Slope	n
Adirondacks	0.020	0.01	499	0.010	0.01	499
Alps				0.000	0	60
Appalachians	0.230	-0.02	50	0.750	0.02	50
Blue Ridge Mountains				0.720	0	24
ECEurope	0.030	0.23	30	0.000	-0.22	201
Maine_Atlantic	0.030	0.05	167	0.000	-0.01	167
NoNordic	0.260	0.02	64	0.000	-0.01	64
SoNordic	0.000	0.08	139	0.000	-0.02	139
UK	0.000	0.13	60	0.150	-0.1	60
Ontario	0.000	0.02	164	0.020	0.01	164
Vermont_Quebec	0.000	0.07	50	0.040	-0.02	172
WCEurope	0.470	0.02	49	0.000	-0.09	69

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