ICP Waters Report 119/2015

Chemical and biological recovery in acid-sensitive waters: trends and prognosis

Lago Leit. Photo: Nicola Bomio-Pacciorini

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
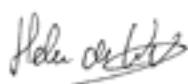
REPORT

Title	Report No..	Date
Chemical and biological recovery in acid-sensitive waters: trends and prognosis	6847-2015	09.04.2015
	ICP Waters report 119/2015	Pages 97
	Project No. 10300	

Author(s)	Topic group	Distribution
Garmo Ø.A., De Wit, H.A. and Fjellheim A.	Effects of air pollution	Open
		Printed
	Geographical area	NIVA
	Europe and North America	

Client(s)	Client ref.
The Norwegian Environment Agency United Nations Economic Commission for Europe (UNECE)	

Abstract

We present a trend analysis of surface water chemistry in in acid-sensitive lakes and rivers in Europe and North America, for the period 2000 to 2011, combined with a prognosis for water chemistry in Europe in 2020. Also, trends in freshwater biology in acidified surface waters in Europe are presented. Declining emissions and deposition of sulphur between 2000 and 2011 have lowered sulphate concentrations in surface waters. The clearest indication that water chemistry is getting less hostile to acid sensitive organisms is increased pH. Several examples of partial biological recovery are presented, but also examples where communities of aquatic invertebrates show few signs of recovery. We expect that climate variability will confound effects of reduced sulphur emissions on chemical and biological recovery in the next decades.

4 keywords, Norwegian	4 keywords, English
1. Luftforurensing	1. Air pollution
2. Vannkjemi	2. Surface water chemistry
3. Biologisk gjenhenting	3. Biological recovery
4. Trender	4. Trends

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ISBN 978-82-577-6582-8

CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

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

**Chemical and biological recovery in acid-sensitive
waters: trends and prognosis**

Prepared at the ICP Waters Programme Centre,
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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). The Programme Centre's work is supported financially by the Norwegian Environment Agency who leads the programme, and by 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. Analyses of trends in water chemistry and biology are a key approach in ICP Waters.

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 in Europe and North America. The network is thus well poised to document changes that result from implementation of the protocols.

In this report, trends in surface water chemistry and biology are presented, in addition to a prognosis of future water chemistry based on current legislation in the revised Gothenburg protocol from 2012.

We thank Jussi Vuorenmaa and Sirpa Kleemola from ICP Integrated Monitoring (at the Finnish Environment Institute), for kindly supplying data for method validation for the prognosis of future water chemistry, and Max Posch at the Coordination Centre for Effects (CCE) for supplying EMEP data. We would like to thank all those who contributed with comments to the draft report.

Oslo, April 2015

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Contents

Summary

The previous trend report from ICP Waters in 2011 concluded that the most distinct changes in water chemistry occurred before 2000 as a result of large reductions in deposition, paving the way for biological recovery. In the current report we consider changes in water chemistry after year 2000 and give a prognosis for development in the near future based on current legislation in the revised Gothenburg protocol from 2012. Norway, Switzerland and the Czech Republic provide reports on the state of recovery of the zoobenthos community, while Finland contributes results from monitoring of fish populations.

The analysis of trends in water chemistry was restricted to non-marine sulphate and base cations, nitrate, alkalinity, charge-balance ANC, dissolved organic carbon (DOC) and pH, all variables that play major roles in acidification and recovery. The prognosis for water chemistry in the near future was based on emission ceilings in the Gothenburg protocol, the EMEP chemical transport model and a modified version of the steady-state model that is used to calculate critical load of acid deposition. The validity of this approach was tested with data from the ICP Integrated Monitoring programme, using measured deposition instead of modelled deposition from EMEP, and found acceptable. The analysis of the biological state of recovery from acidification comprises presence/absence of sensitive species, abundance and biodiversity.

Sulphate concentrations in headwaters declined markedly between 2000 and 2011 as a result of decreasing sulphur emissions. Nitrate has decreased in the Alps over the whole period, but elsewhere the pattern is mixed. Clear changes in nitrate on a regional scale are not apparent in any region except the Alps. Consistently increasing trends in both alkalinity and ANC are not evident in most regions, despite the decrease in sulphate, but positive trends still dominate. The clearest indication that water chemistry is recovering from acidification is widespread pH increase. The clearest indications of biological recovery come from the sites with the longest time series or largest absolute improvements in chemical water quality. The report presents only one example of full recovery on species level, namely the relatively acid-tolerant perch (*Perca fluviatilis*) in Finland. Elsewhere and for other species the pattern is mixed with many zoobenthos communities showing partial recovery (Norway, Czech Republic) and no clear indications of recovery (Switzerland).

Sulphur deposition is predicted to decrease further until 2020 (EMEP scenario of Current Legislation), but changes are expected to be smaller than those observed between years 2000 and 2010. Increases in ANC are expected, but changes will be subtle and smaller than interannual variations between 2000 and 2011. The interannual variation in ANC is caused by variations in climate and deposition, i.e. precipitation, sulphur and seasalt deposition. Water chemistry in 2020 will not reach a status that will support full biological recovery in acid-sensitive surface waters throughout Europe. Within the next two decades, we expect that variations in climate will confound effects of continued decreased acid deposition on recovering lakes and rivers, leading to variations in water chemistry that may hamper 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 national 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

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1.2 The current report

The current report is a follow-up to the 2011-trend report from ICP Waters, documenting trends in precipitation chemistry, surface water chemistry and surface water biology between the years 1990 and 2008 (Skjelkvåle and De Wit, 2011). We concluded that the acidity of lakes and rivers had been reduced as a result of the decrease in sulphate emissions, paving way for the recovery of aquatic biological communities that have been damaged owing to acidification. The report documented ongoing biological recovery, but did not show evidence of full recovery anywhere. Here, we raise the question whether current legislation of emissions of acidifying components is enough to produce sufficient water quality, so that full biological recovery might be expected. Additionally, we ask whether climate change and climatic variability now play a more prominent role for water quality than previously, when changes in atmospheric chemistry were the most important control for acid-sensitive species in acid-sensitive surface waters.

In the current report, we forecast expected water quality in 2020, given current legislation (the revised Gothenburg protocol from 2012) to reduce emissions of acidifying components. We compare the expected water quality in 2020 with current water quality, i.e. the most recent water records in the ICP Waters database from 2000 to 2011. Also, we document the most recent trends in surface water chemistry in the given period, and present updated trends in biological recovery.

The report is divided into three parts:
1) a trend analysis of surface water chemistry in Europe and North America in acid-sensitive waters for the most recent data, i.e. from 2000 onwards.
2) a prognosis for water chemical status in 2020 using predicted deposition for 2020, based on the 2012 revision of the Gothenburg protocol, and
3) a trend analysis of freshwater biology in acid-sensitive lakes and rivers.

Surface water chemistry trends have been analysed at the ICP Waters programme centre based on data delivered to ICP Waters by national focal centres. The programme centre for the International Cooperative Programme on Integrated Monitoring of Air Pollution Effects on Ecosystems (ICP IM) provided measured deposition and water chemistry for acid sensitive ICP IM catchments. These data were used to test if water chemistry in 2020 could be predicted using estimated average runoff volume, EMEP modelled sulphur deposition and the assumption of a steady state between sulphur deposition and runoff of SO_4.

The ICP Waters focal centres in Finland, Czech Republic and Switzerland, and the programme subcentre in Norway present results from a trend analysis of biological indicators of acidification and recovery.

2. Trends in water chemistry

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2.1 Methods

2.1.1 Selection of ICP Waters sites for analysis

Sites in the ICP Waters database display 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. Only sites where data were available from at least 9 out of 12 years (2000-2011) were included in the statistical analysis and model exercise (see sections 2.1.3 and 2.1.4).

2.1.2 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 charge balance

2.1.3 Variables and statistical method

Data from ICP Waters are suitable for assessment of 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 (see e.g. Skjelkvåle and De Wit, 2011 and references therein).

Our analysis of surface water response to changing deposition was restricted to 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 + Mg) are mobilised by weathering reactions and cation exchange that neutralise acids in watersheds. Deposition from air may also be a (usually minor) source for Ca and Mg in catchments. Base cations will respond indirectly to changes in SO_4^{2-} and NO_3^{-}.

3)   **Acidity**, including pH, measured 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 fulvic acid), derived through degradation of natural organic matter in catchment soils and wetlands.

Both SO_4^{2-} and base cation concentrations were sea salt corrected by subtracting the marine contribution estimated from the ratio of the ion to Cl in seawater (Lyman and Fleming, 1940) (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.

Annual means were used in the statistical analyses. The pH was back calculated from arithmetic mean of H^+. The frequency of observations per station varied from a single annual observation in some lakes to weekly sampling in some streams, 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.

The Mann Kendall test (MKT) (Hirsch and Slack, 1984) was used to detect monotonic trends based on the value of the test statistic (Z-score). This method is robust against outliers, missing data and does not require normal distribution of data. Slopes were calculated using the Sen estimator (Sen 1968).

2.1.4 Estimate of water chemistry in Europe in year 2020

This estimate relied on deposition of oxidised sulphur (S_{dep}) in the years 2005 and 2020 as predicted by the EMEP chemical transport model (Simpson et al., 2012). The EMEP model results were provided by Maximilian Posch (Coordination Centre for Effects) and had a gridded spatial resolution of 0.25 degrees latitude and 0.5 degrees longitude, i.e. the grids were approximately 28*28 km. The scenario for 2020 is based on emission ceilings defined by the Gothenburg protocol which was revised in 2012.

Surface water concentrations of non-marine sulphate, base cations and ANC were estimated for each ICP Waters station as described below (derivation and information about equations can be found in Henriksen and Posch, 2001). First the background concentration of non-marine sulphate ($[SO_4^*]_0$) was considered to be a function of measured non-marine base cation concentration ($[BC^*]$) (Eq 1).

$$[SO_4^*]_0 = 8 + 0.17[BC^*] \qquad \text{(Eq 1)}.$$

The concentration of non-marine sulphate at time t ($[SO_4^*]_t$) was estimated with Eq 2.

$$[SO_4^*]_t = [SO_4^*]_0 + K\frac{S_{dep,t}}{\bar{Q}} \qquad\qquad \text{(Eq 2)},$$

where $\bar{Q}$ is average annual runoff (30-year mean) and $K = \frac{[SO_4^*]_{2005,measured}-[SO_4^*]_0}{\frac{S_{dep,2005}}{\bar{Q}}}$ is a catchment-specific

scaling factor. The assumption implicit here is that there is a dynamic equilibrium between oxidised sulphur deposited in the catchment and sulphate in run off, i.e. a steady state (a constant ratio between deposition and runoff of oxidised sulphur). The concentration of nitrate ($[NO_3]_t$) was assumed to be constant and equal to the average measured concentration (2000-2012). The background concentration of nitrate ($[NO_3]_0$) was set to zero. Next, the so-called F-factor, which indicates how much of the strong acid deposition that is neutralised in the catchment, was estimated with Eq 3[1] (Brakke et al., 1990).

$$F = \sin((\frac{\pi}{2})\overline{Q}[BC^*]/S) \qquad\qquad \text{(Eq 3)},$$

where S is the base cation flux when F is equal to 1, here set to 400 mEq/m²/yr. The average value of F for the time span 2000-2012 was used in subsequent calculations. Next $[BC^*]_0$ can be estimated from the definition of the F-factor (Eq 4), using measured $[BC^*]$ and $[SO_4^*]$ as $[BC^*]_t$ and $[SO_4^*]_t$, respectively

$$F = \frac{[BC^*]_t-[BC^*]_0}{[SO_4^*]_t-[SO_4^*]_0+[NO_3]_t-[NO_3]_0} \qquad\qquad \text{(Eq 4)}$$

Furthermore, by keeping F constant, $[BC^*]_t$ can be calculated from $[SO_4^*]_t$, thus allowing forecasts (and hindcasts) of ANC.

2.2 Results

2.2.1 Water chemistry trends 2000 - 2011

Several papers reporting trend analyses of acidification variables in the regions covered by the current report have been published recently. The reader is referred to these for a more detailed discussion of the developments in each region (Borg and Sundbom, 2014; Futter et al., 2014; Monteith et al., 2014; Oulehle et al., 2013; Rogora et al., 2013; Waller et al., 2012). References to papers from before 2011 covering these and other regions can be found in the previous ICP Waters trend report (Skjelkvåle and De Wit, 2011).

In the current report, data from a total of 197 stations were included in the analysis: 91 in Europe (Figure 1) and 106 in North America (Figure 2). The overall patterns for Europe and North America will be presented first and then the results from the various regions.

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[1] Another expression (Posch et al., 1993) where the F-factor is independent of the (variable) $[BC^*]$, was tested, but gave rather similar F-factors as Eq 3 (y=0.94. R²=0.82).

Figure 1. Map showing the location of the 91 European stations. The different colours of the markers indicate grouping of the stations into geographical regions.

Figure 2. Map showing the location of the 106 North American stations. The different colours of the markers indicate grouping of the stations into geographical regions.

The results of the individual trend analysis of European and North American stations are shown in
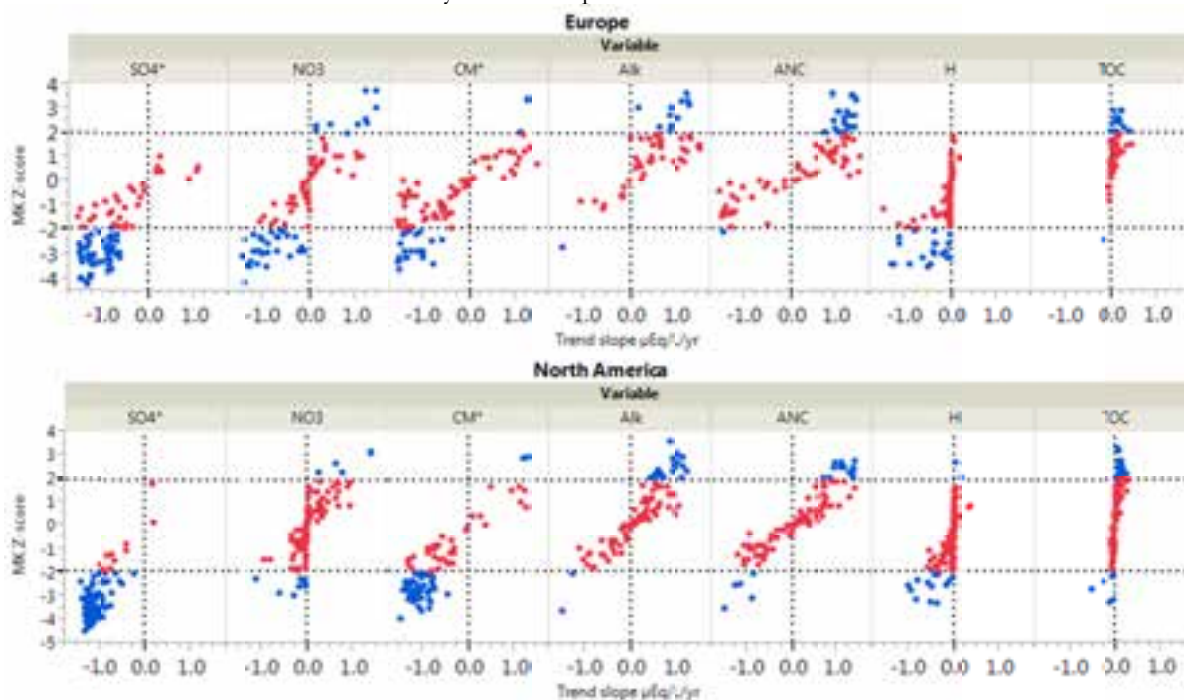
Figure 3. The patterns are similar on both continents for these stations and this time span, and are similar to those presented in Skjelkvåle and De Wit (2011) and Garmo et al. (2014). In Europe and North America 60 % and 90 % of sites, respectively, show significant decrease in non-marine sulphate, while none show an increasing trend. Most sites show no unidirectional trend for nitrate; for those who do there are slightly more negative trend slopes than positive in Europe and the other way around in North America. A minority (20 %) of European sites and a majority (60 %) of North American sites show decreasing concentration of non-marine base cations. Increasing trends in pH are much more prevalent than decreasing trends on both continents, but the majority of sites show no uni-directional trend. A total of 12-23 % of European sites shows significant increases in pH alkalinity, ANC and DOC, whereas declines are observed at some North American stations. When assessing the results one should be aware that lack of statistical significance does not imply that there is no real change, but that it cannot be confirmed with more than 95 % confidence using the chosen statistical test.
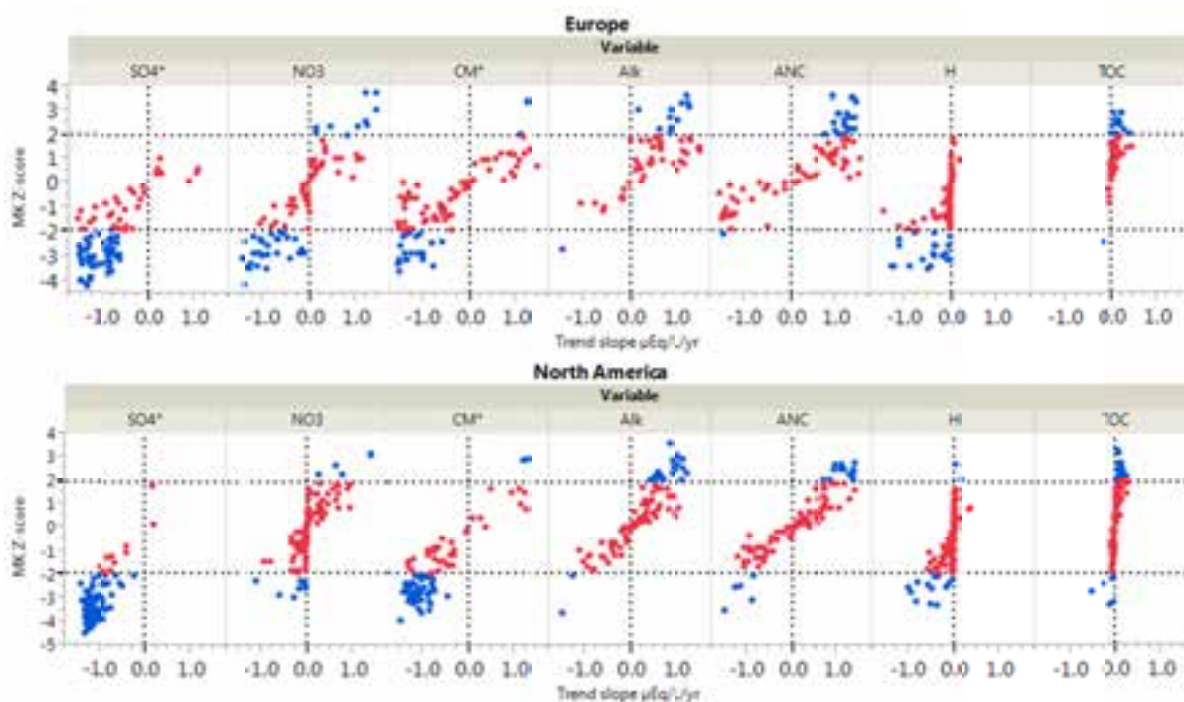
Figure 3. Mann Kendall (MK) Z-scores (Y-axis) and trend slope (X-axis) for SO₄*, NO₃, sum of non-marine calcium and magnesium CM*), alkalinity, Acid Neutralizing Capacity (ANC), H⁺, and total or dissolved organic carbon (TOC) for all the analysed sites between 2000 and 2011. A MK Z-score > 1.96 or < -1.96 implies that the slope is significantly different from 0 at the 0.05 confidence level (blue colour). The x-axes are arctan transformed.

Trends in sulphate by region
In most regions there has been a clear decrease in the average concentration of non-marine sulphate. The largest absolute changes occurred in the Central European regions with an average decline of 60-70 µEq/L between 2000 and 2011 (Figure 4). The non-marine sulphate concentration has decreased by 30-50 percent in all regions except the Alps, UK+Ireland, Blue Ridge Mountains and Ontario. The distribution of the time series slopes confirms that concentrations tend to decrease in the large majority of stations in all regions (Figure 5).

Figure 4. Annual mean concentration of non-marine sulphate in stations from regions in Europe and North America between 2000 and 2011. The blue lines are cubic splines with a lambda of 0.05. Sulphate concentrations in 2011 were extrapolated for 6 stations in East Central Europe.

Figure 5. Distribution of non-marine sulphate Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

Trends in nitrate by region

The deposition of nitrogen (nitrate and reduced forms) has also decreased, but is now higher than that of sulphur in most parts of Europe (Helliwell et al., 2014). Most of the nitrogen is, however, retained in the terrestrial catchments and is a minor contributor to acidification at the large majority of sites. The Alps and, possibly, the Blue Ridge Mountains display uni-directional decrease in nitrate for the whole time span (Figure 6). Decreasing concentrations in the Alps has been attributed to decreasing deposition but also to hydrological factors (Rogora et al., 2012). Clear increases on a regional scale are not apparent in any of the regions. There are regions where trend slopes tend to be positive (West Central Europe, Baltics) or negative (the Alps), but in most regions trend slopes are almost evenly distributed between positive and negative (Figure 7).

Figure 6. Annual mean concentration of nitrate in stations from regions in Europe and North America between 2000 and 2011. The blue lines are cubic splines with a lambda of 0.05. Nitrate concentrations in 2011 were extrapolated for 6 stations in East Central Europe.

Figure 7. Distribution of nitrate Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

Trends in base cations (Ca+Mg) by region

One of the expected responses of catchments to decreasing sulphur inputs is the reduced leaching of base cations because of increased pH and base cation scavenging by cation exchange sites (Galloway et al., 1983). All European regions except the Alps display decreasing trends in base cations (Figure 8). In North America, however, decline is only evident in the Adirondacks. This is also reflected in the distribution of individual trend slopes (Figure 9). Increasing trends in relatively well-buffered lakes in the Alps have been attributed to increased weathering rates due to climate change (Rogora et al., 2013).
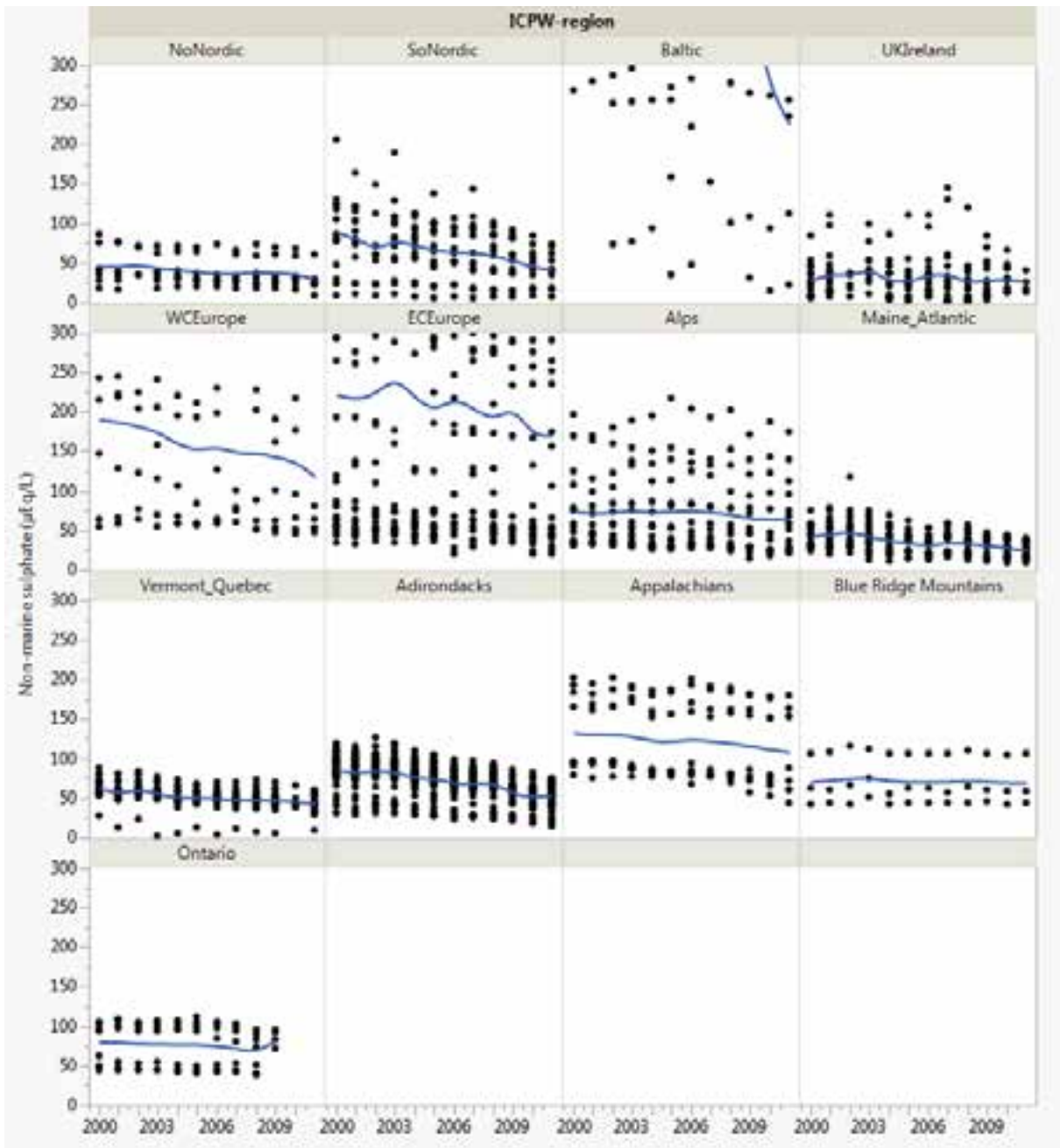
Figure 8. Annual mean concentration of non-marine base cations in stations from regions in Europe and North America between 2000 and 2011. The blue lines are cubic splines with a lambda of 0.05. Base cation concentrations in 2011 were extrapolated for 6 stations in East Central Europe.

Figure 9. Distribution of base cation (Ca+Mg) Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

Trends in alkalinity and ANC by region
Measured alkalinity and calculated charge balance ANC are widely used as indices of the extent of acidification and the susceptibility of natural waters to acidification. ANC is a surrogate for alkalinity, thus their temporal and regional patterns are similar. The ANC tends to be higher than alkalinity because organic anions are not included in calculation of charge-balance. With the exceptions of the East Central European region and the Alps, consistent increasing trends for both variables on the regional level are not evident (Figure 10) despite the reduced sulphate concentration. Even so, statistical analysis of individual time series show that there are more positive than negative slopes in Europe, while the distribution in North American regions is more even, especially for ANC (Figure 11 and Figure 12).
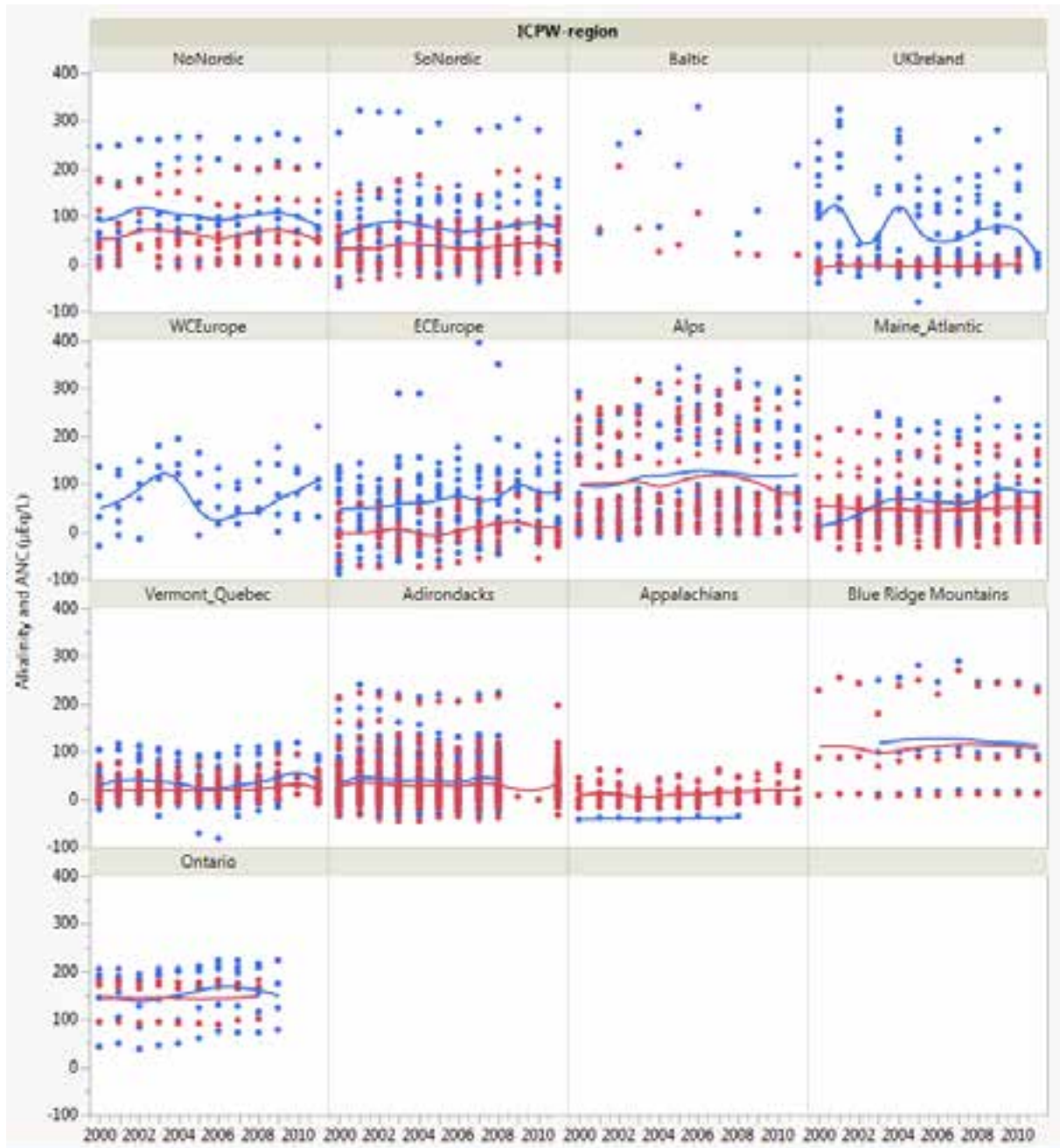
Figure 10. Annual mean ANC (blue) and alkalinity (red) in stations from regions in Europe and North America between 2000 and 2011. The lines are cubic splines with a lambda of 0.05. Alkalinity and ANC are extrapolated for 6 stations in East Central Europe for 2011.
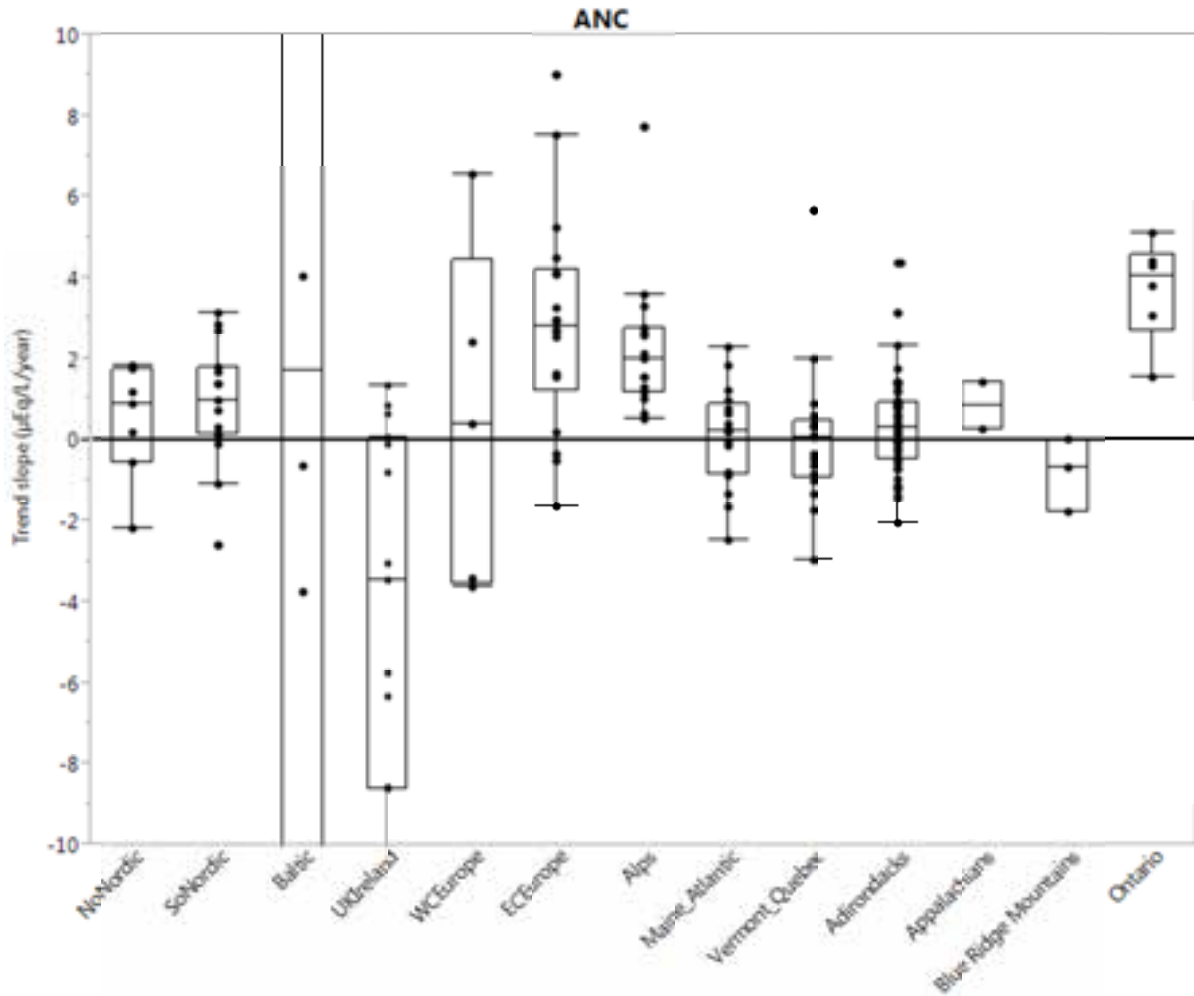
Figure 11. Distribution of acid neutralising capacity (ANC) trend slopes in regions of Europe and North America between year 2000 and 2011. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.
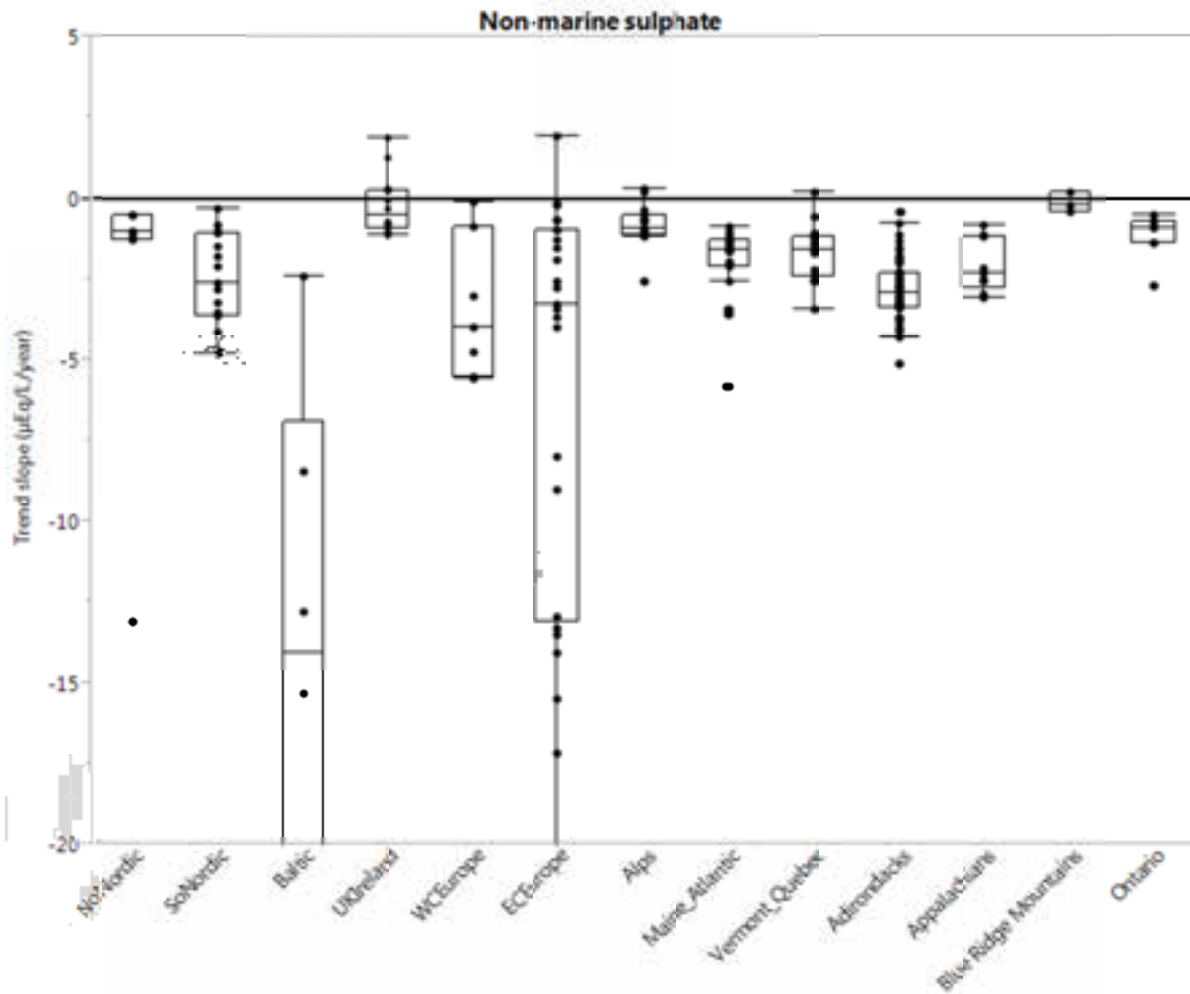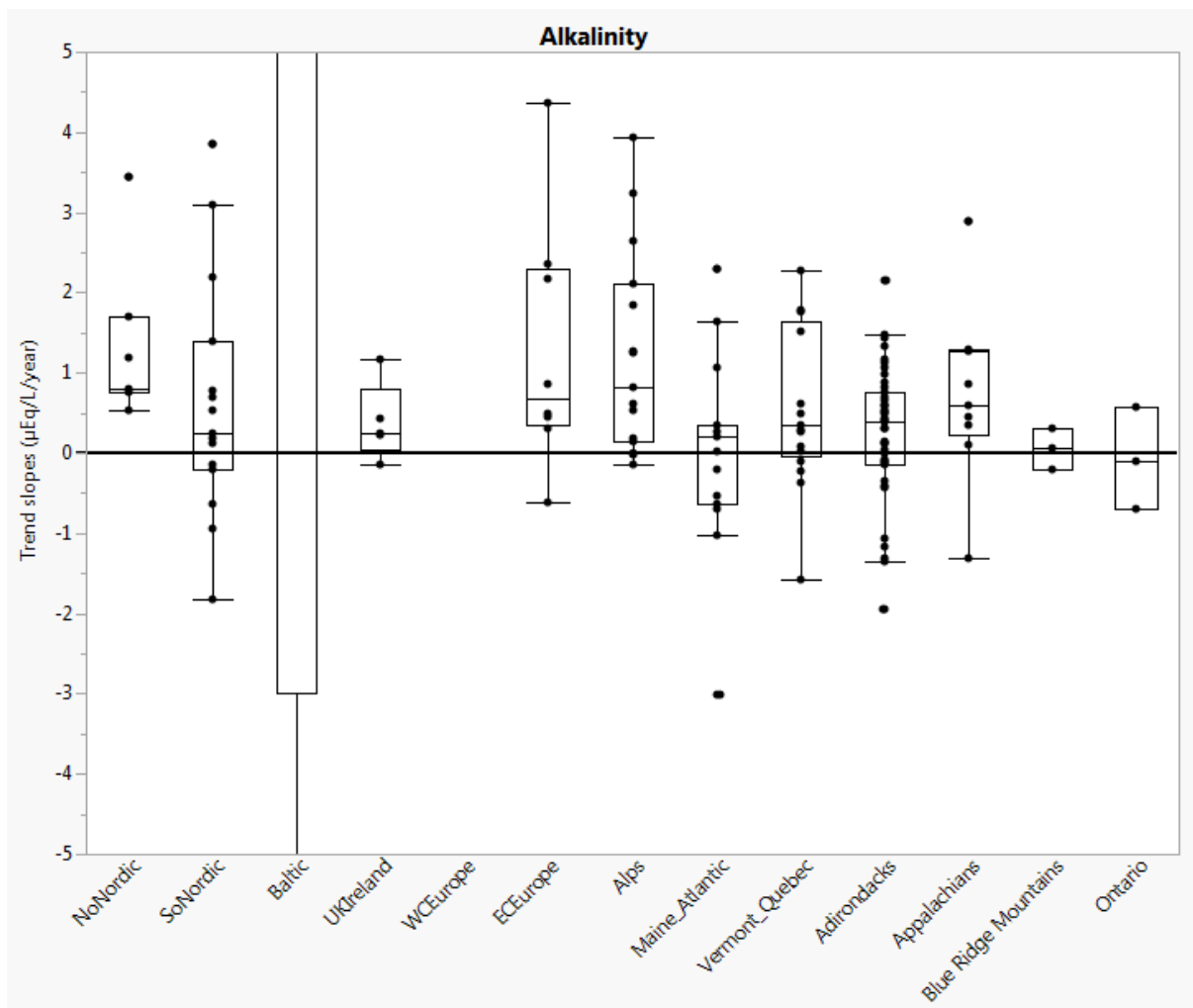
Figure 12. Distribution of alkalinity Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

Trends in DOC by region
DOC is a key component of aquatic chemistry, e.g., as an indicator of natural organic acidity and as a complexing agent that reduces the aquatic toxicity of aluminium. DOC has received considerable attention in recent years because of rising levels in many regions (see e.g. Monteith et al., 2007; Hruška et al., 2009), which raises questions regarding pre-industrial and future state of acidification (Erlandsson et al., 2010; Hruška et al., 2014). Increasing regional average DOC is evident in the European regions, but the same is only observed for the Maine-Atlantic region in North America (Figure 13). The distribution of individual slopes suggests that positive trends are more common than negative also in Vermont-Quebec and the Adirondacks (Figure 14). Other workers have found significant DOC increases in the Adirondacks lakes between 1993 and 2011(Lawrence et al., 2013).
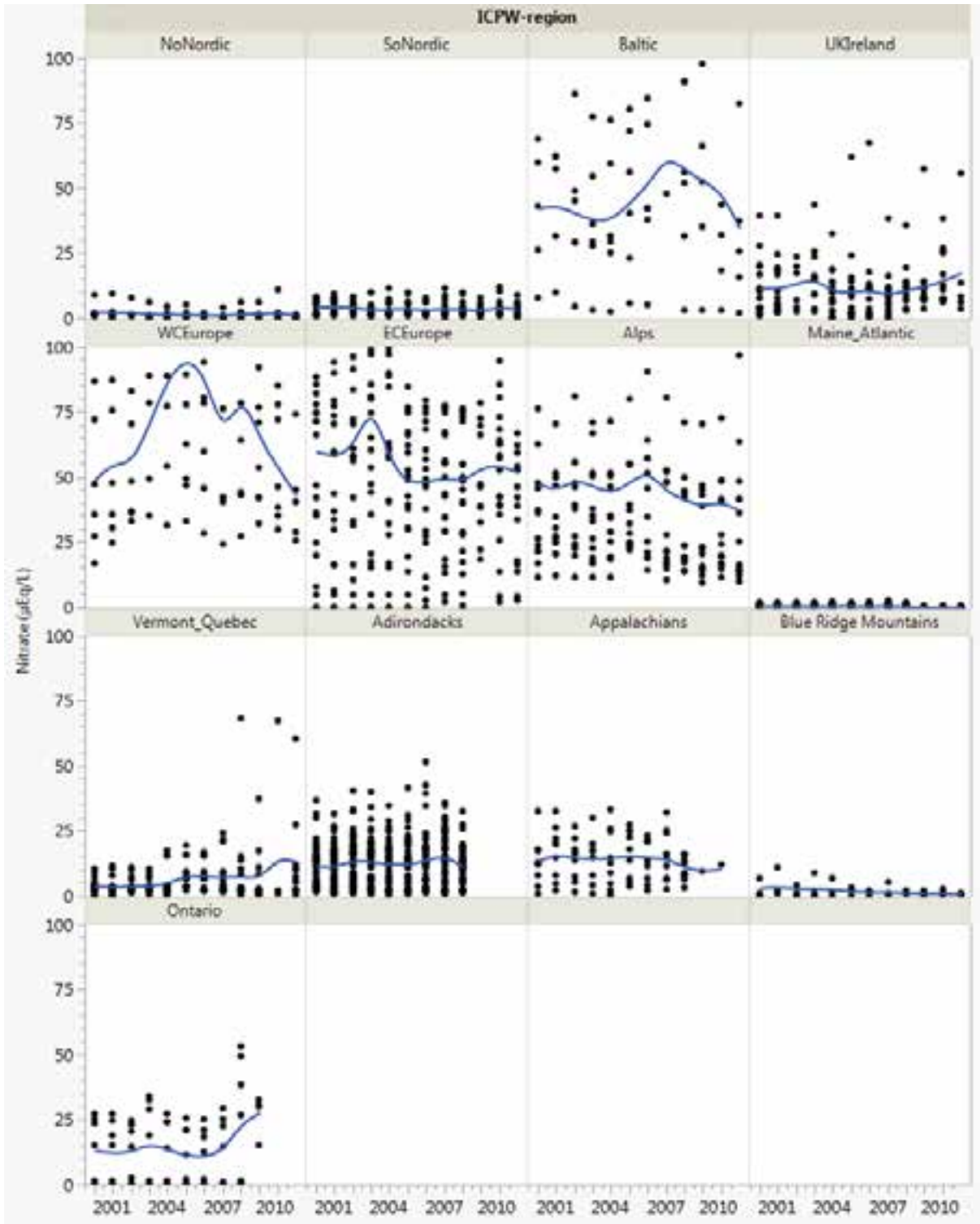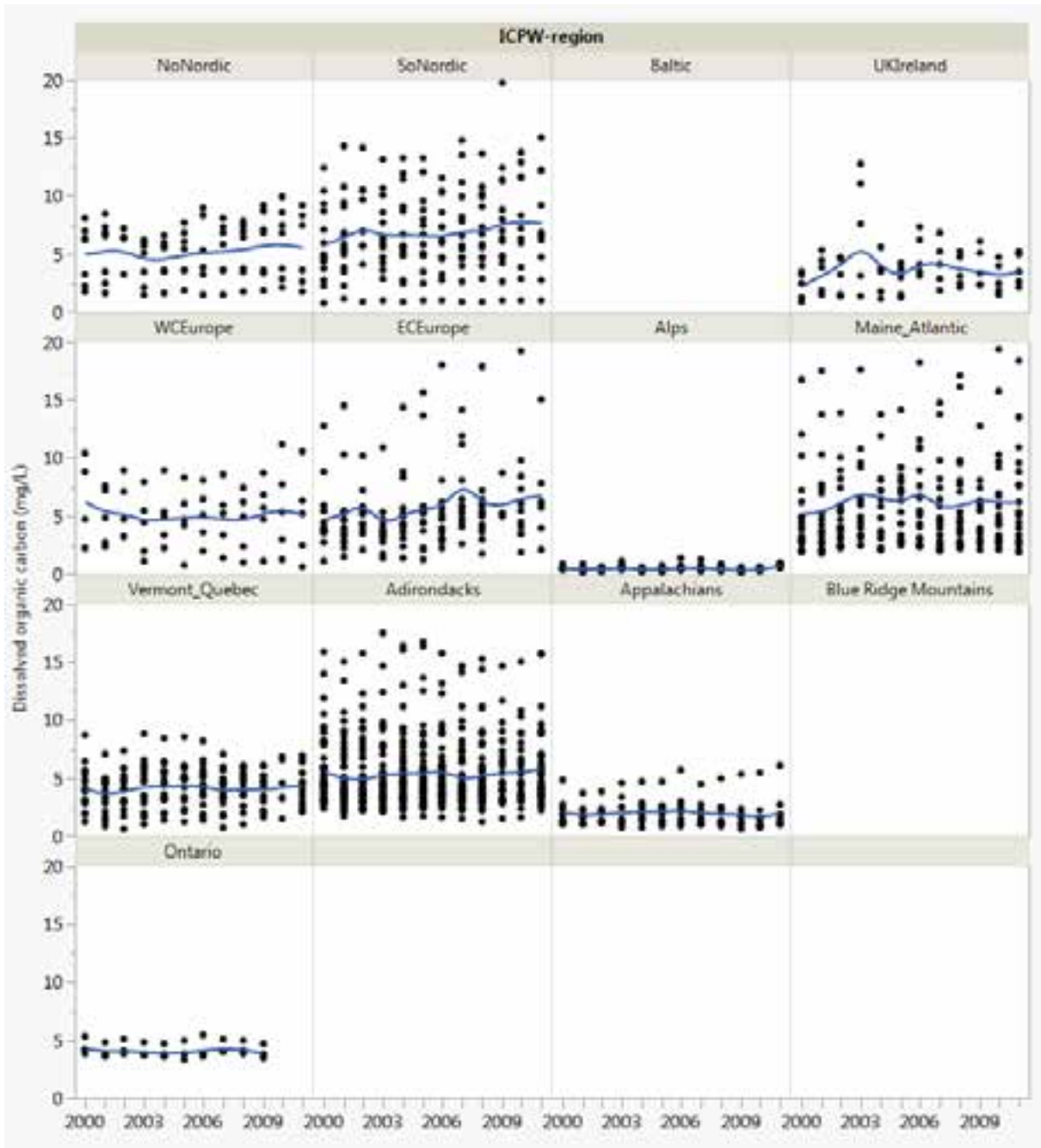
Figure 13. Annual mean concentration of dissolved organic carbon in stations from regions in Europe and North America between 2000 and 2011. The blue lines are cubic splines with a lambda of 0.05. Note that the panels present results from some stations that were not included in the statistical analysis because data series were shorter than 9 years.
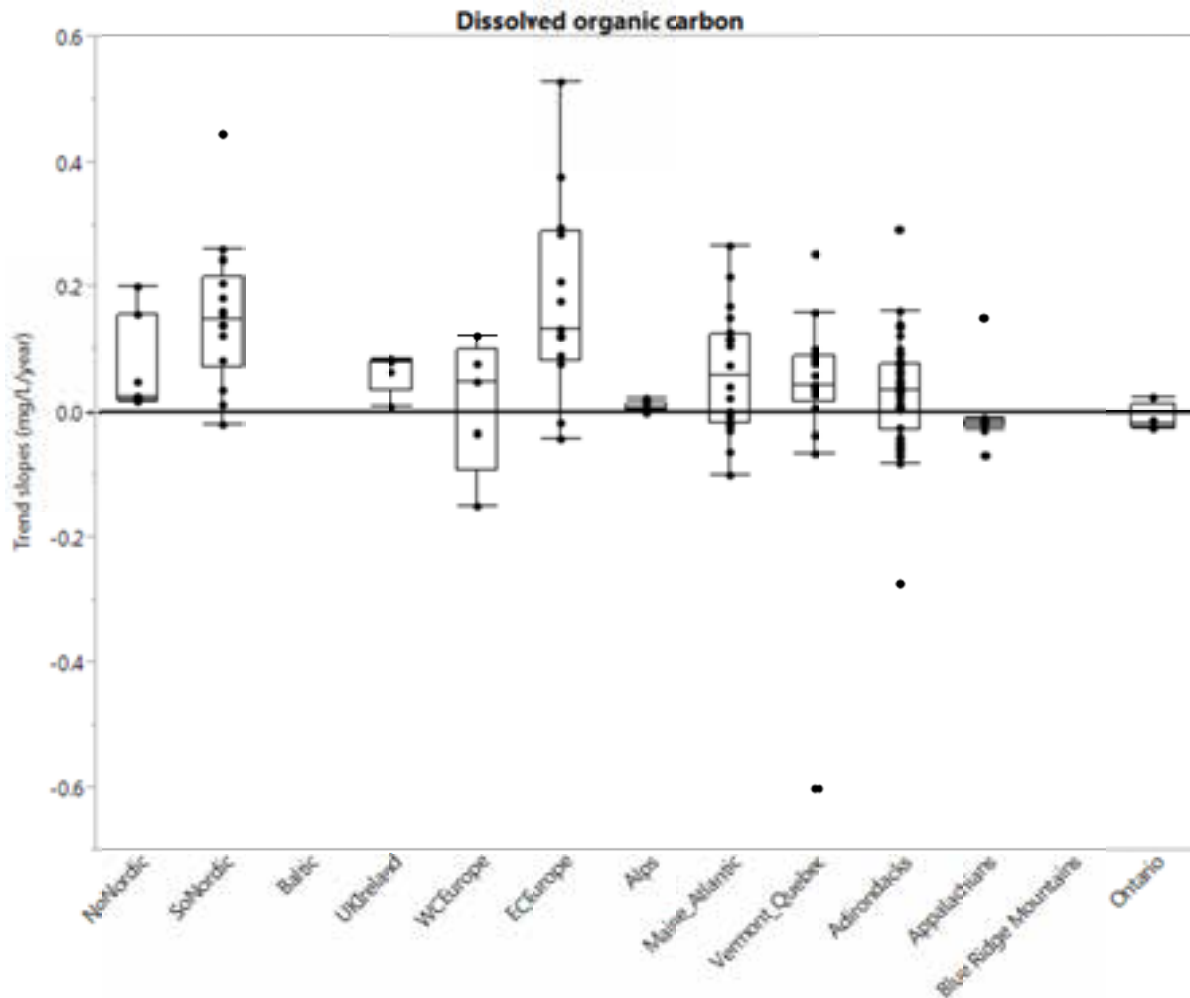
Figure 14. Distribution of dissolved organic carbon Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

Trends in pH (H$^+$) by region
All regions except the North Nordic region, the Blue Ridge Mountains and Ontario show decreasing average H$^+$ ion concentrations, i.e. increasing pH (Figure 15). The distribution of individual time series slopes confirms this pattern (Figure 16). Deposition of acidifying substances kills organisms by lowering the pH in surface waters. This happens either because the H$^+$ concentrations reach toxic levels or, more often, because low pH affect the solubility and speciation of toxic metals like aluminium (Gensemer and Playle, 1999). Increasing pH is a clear indicator of chemical recovery from acidification and shows that living conditions for vulnerable biota are improving.
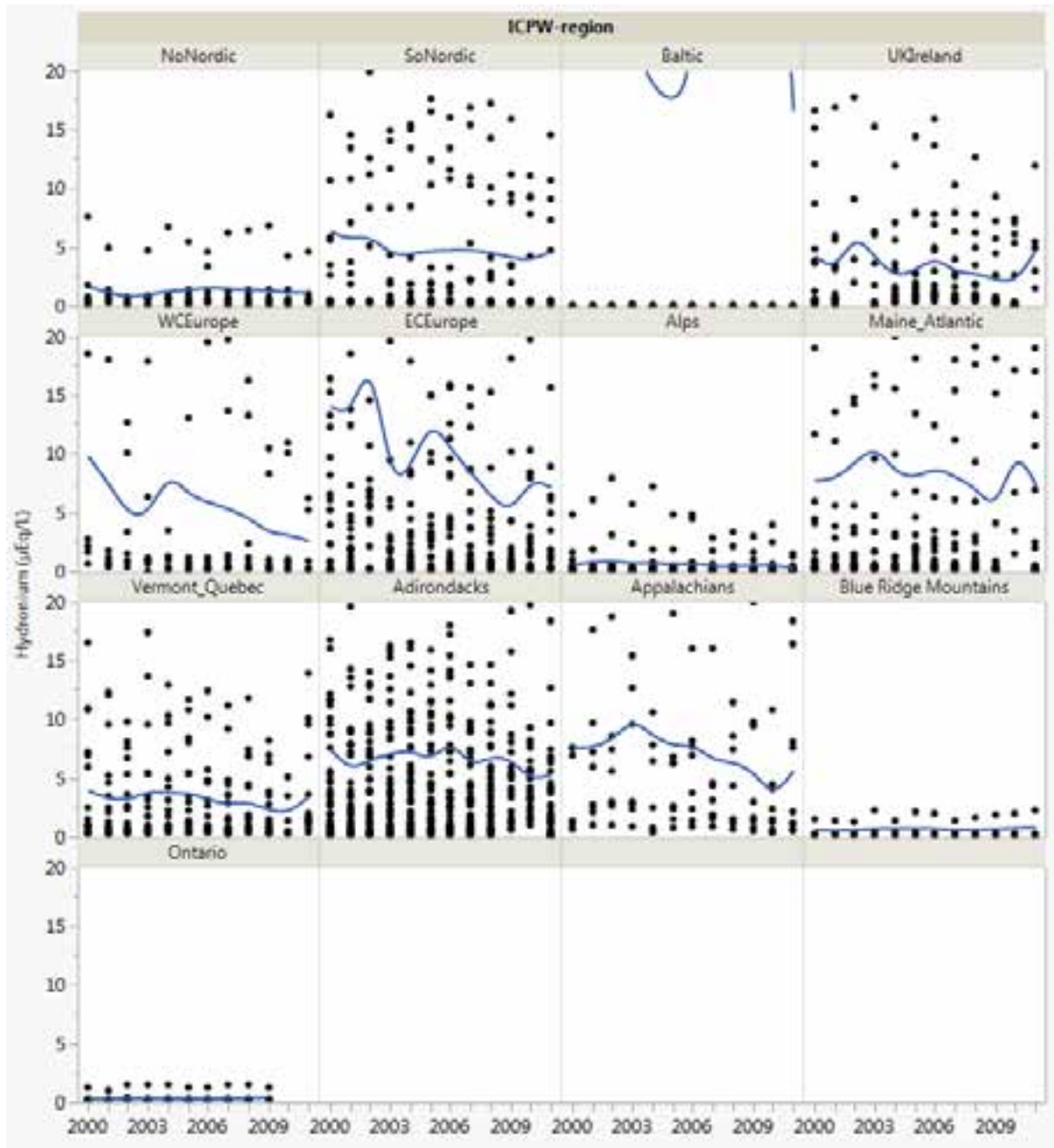
Figure 15. Annual mean concentration of H+ in stations from regions in Europe and North America between 2000 and 2011. The blue lines are cubic splines with a lambda of 0.05.
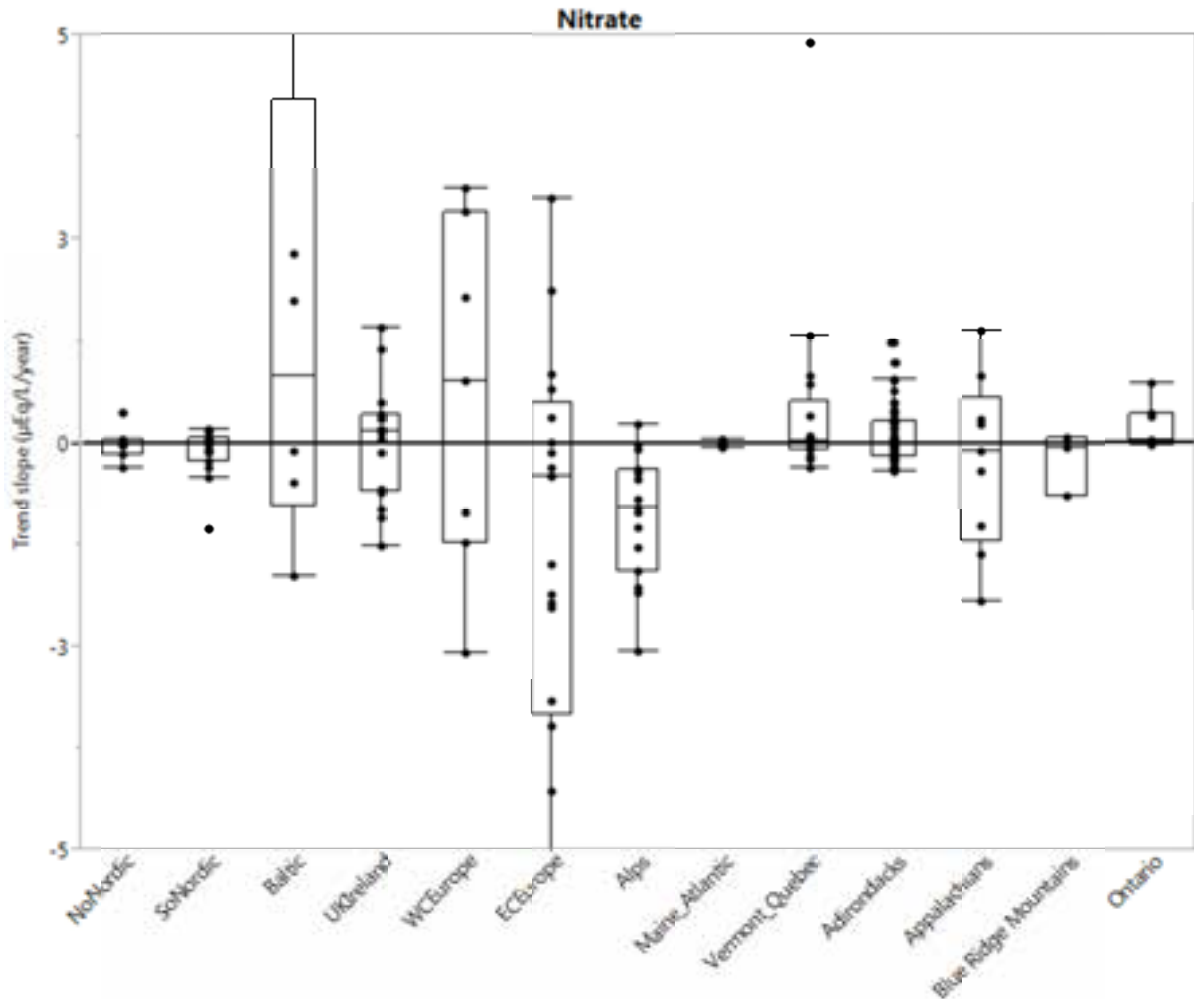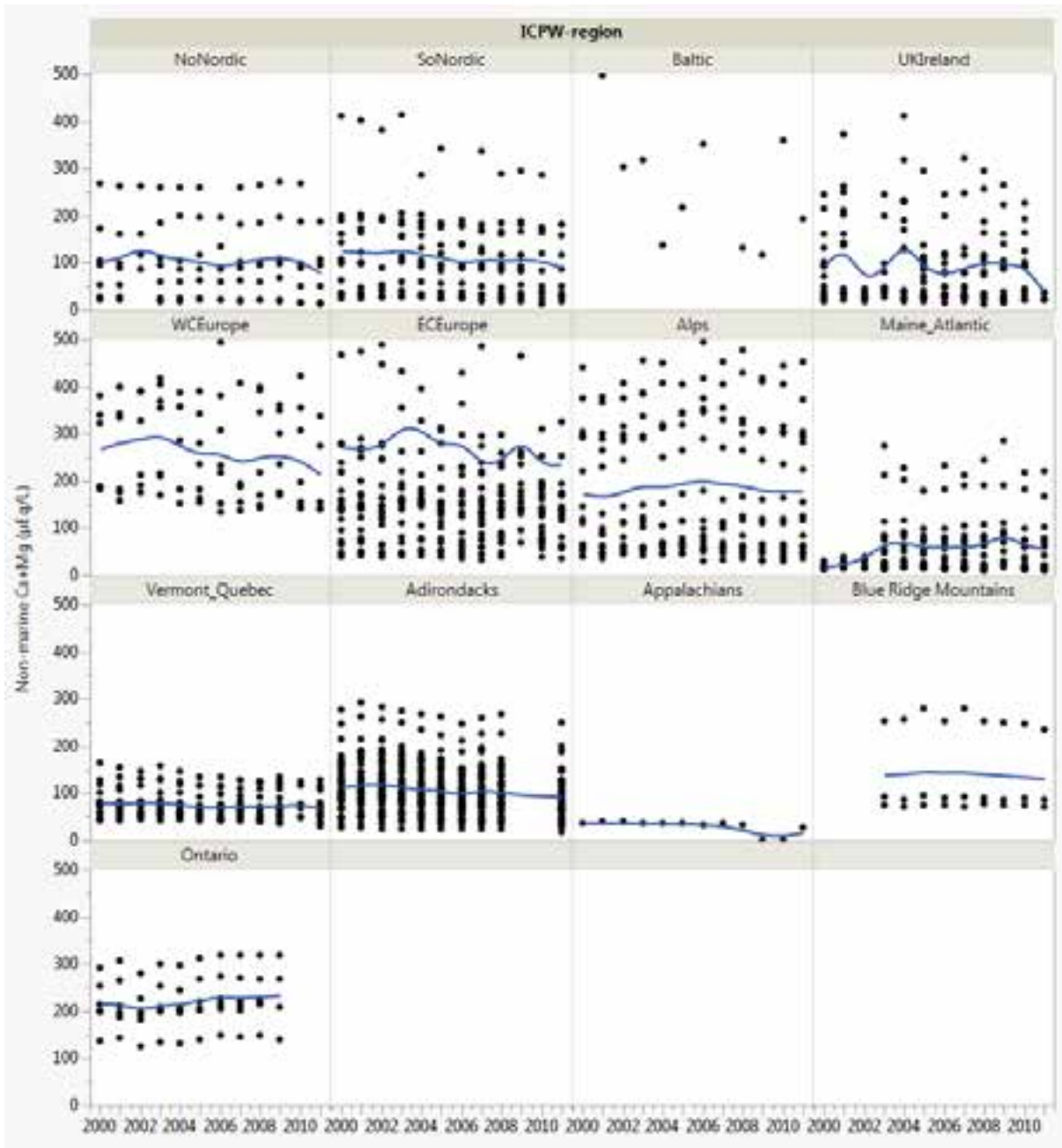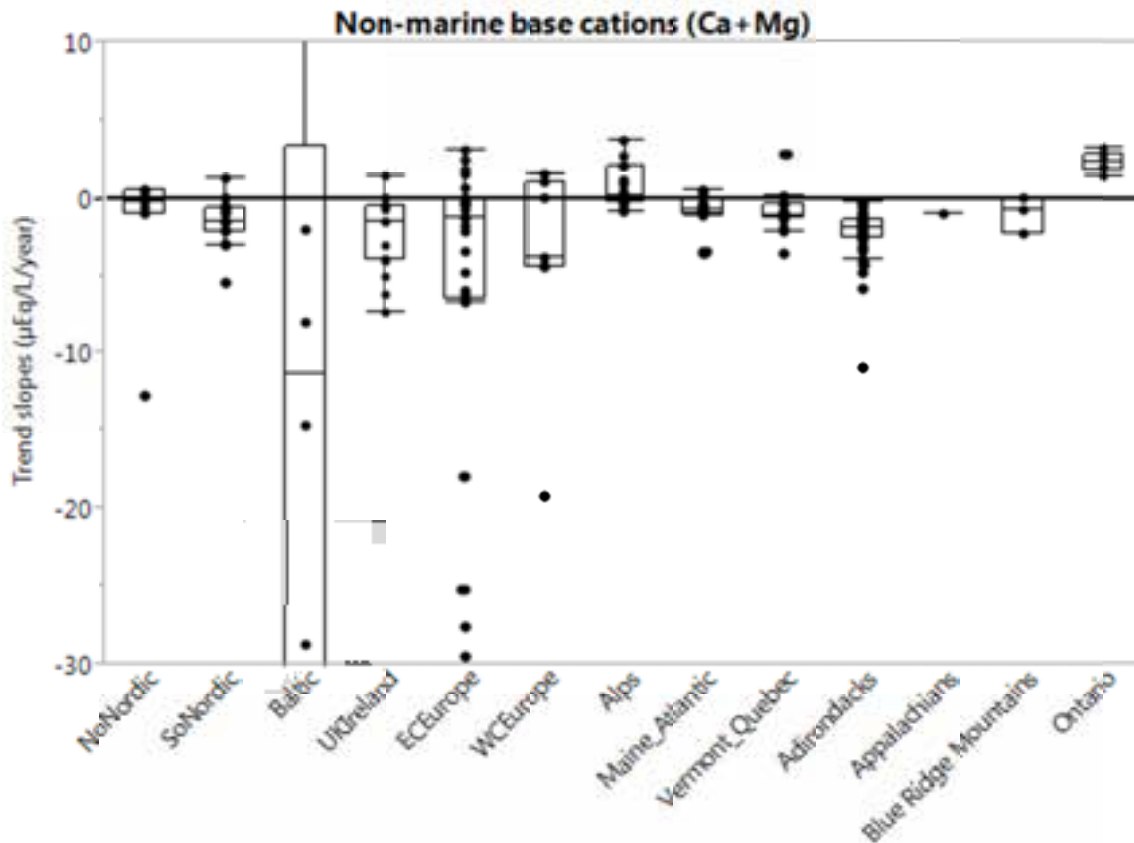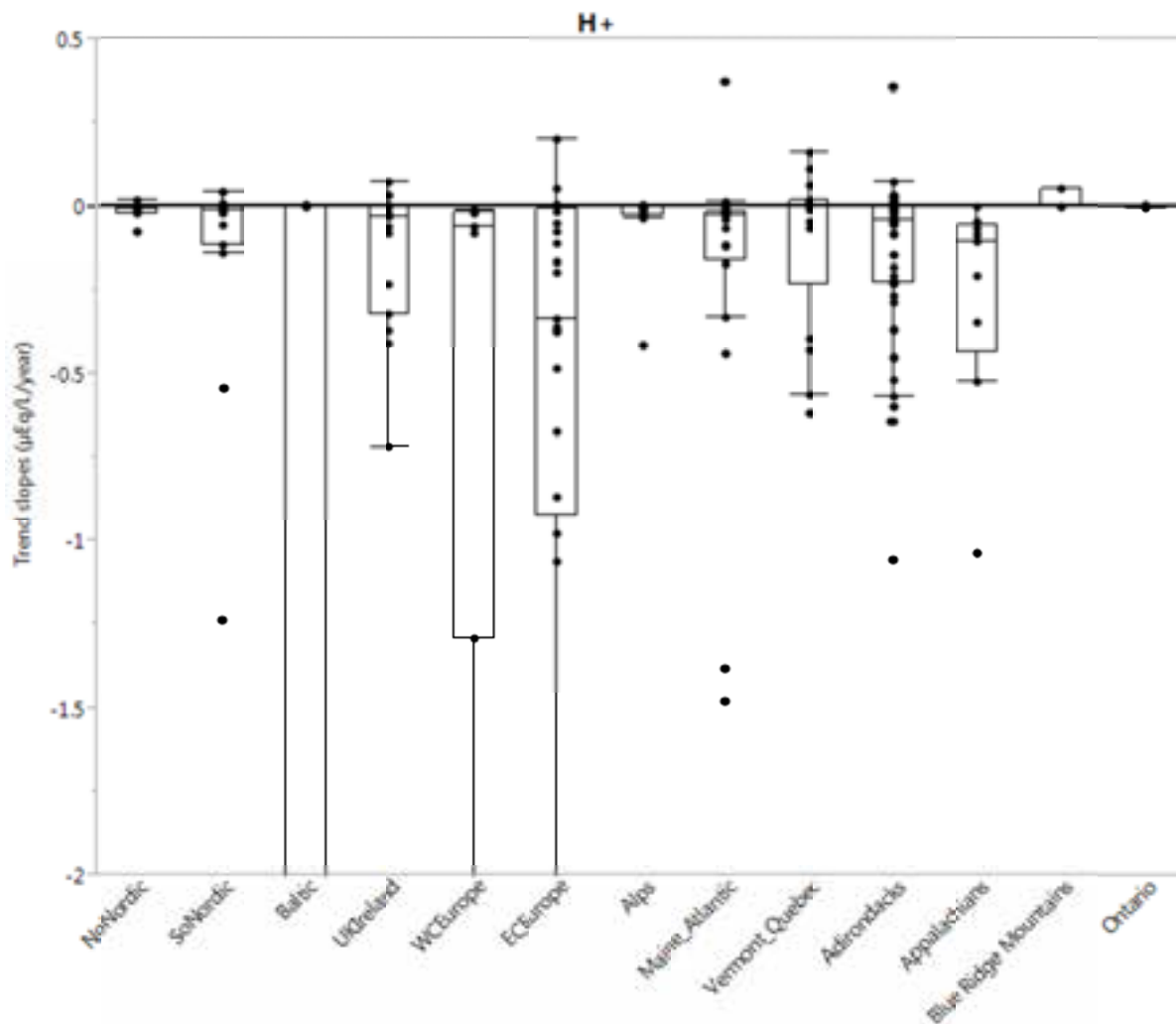
Figure 16. Distribution of H⁺ Sen slopes in regions of Europe and North America between year 2000 and 2011. Statistical significance is not indicated in the plot. Boxes and whiskers cover the 25th to 75th and 10th to 90th percentiles of slopes, respectively, while the median values are indicated with lines.

2.2.2 Prognosis of water chemistry in Europe in 2020

Validation of approach. The use of the F-factor and empirical relationships to calculate background concentrations of sulphate (i.e., a level unaffected by human activities) is central for estimating the critical load of sulphur deposition in catchments (see manuals of ICP Modelling and mapping, http://www.icpmapping.org/Mapping_Manual). The critical load is a theoretical quantity valid for steady-state conditions. The procedures/models used for calculating critical loads are therefore not necessarily useful for predicting annual to decadal variations in water chemistry. A key assumption underpinning the procedure described in chapter 2.1.4 is that the ratio between annual deposition of oxidised sulphur in the catchment and runoff of non-marine sulphate from the catchment is considered constant (i.e. there is no significant delay between change in sulphur deposition and change in SO_4^* concentrations in runoff). Such a direct link between sulphur input and output has been demonstrated in UK lakes (Cooper and Jenkins, 2003), but it is not true in all circumstances. Release of stored sulphate can, for example, result in a delay of acidification reversal owing to desorption of sulphate from soil particles, especially in deeply weathered soil rich in iron and aluminium oxides, and mineralisation of organic sulphur stored through years of high sulphate deposition. Increased weathering rates because of warmer temperature and less snow cover could also affect the release of SO_4^* in some catchments (Rogora et al., 2003). These are processes that can only be simulated with dynamic models such as MAGIC (Cosby et al., 2001; Helliwell et al., 2014) and require information not registered in the database of ICP Waters such as catchment soil properties.

The validity of our F-factor model approach was tested with data from 12 acid sensitive sites in the ICP Integrated monitoring network (four of these are also ICP Waters stations). In contrast to ICP Waters, ICP IM includes measurements of sulphur deposition, runoff volume as well as hydrochemistry. Measured values for deposition, runoff and water chemistry could then be compared to estimated values. Measured sulphur deposition is clearly different from EMEP estimates for some of the sites, but there is apparently no overall bias (Figure 17). Use of average runoff instead of measured runoff is not expected to introduce bias, but much of the year-to-year variations in water chemistry will not be captured when average runoff is used as input to the F-factor model. More relevant for testing the validity of the F-factor approach is the comparison of measured water chemistry variables (sulphate and base cation concentrations, and ANC) with predicted water chemistry, using measured sulphur deposition and runoff volume as inputs for each site (Figure 17 - Figure 20). This is a test of the steady-state assumption mentioned above. The results show that there is a difference between measured and estimated sulphate concentrations for most of the stations (Figure 18) (note that the scaling factor was not used here, i.e. no calibration). If the results from all 12 stations are pooled (not shown) there is little difference between mean estimates and mean measurements, suggesting that there is no systematic bias associated with our approach and that individual differences are caused by catchment specific conditions. These catchment specific causes could be of dynamic nature, but apparently not on the time scales considered here, as trend slopes in estimated and measured sulphate concentrations are similar. Differences at individual stations are much smaller for base cation concentration and ANC because the catchment specific F-factor is calculated from measured water chemistry. The apparently good agreement between trends in measured and estimated ANC (Figure 20) indicates that our method for estimating hydrochemistry in year 2020 is suitable.
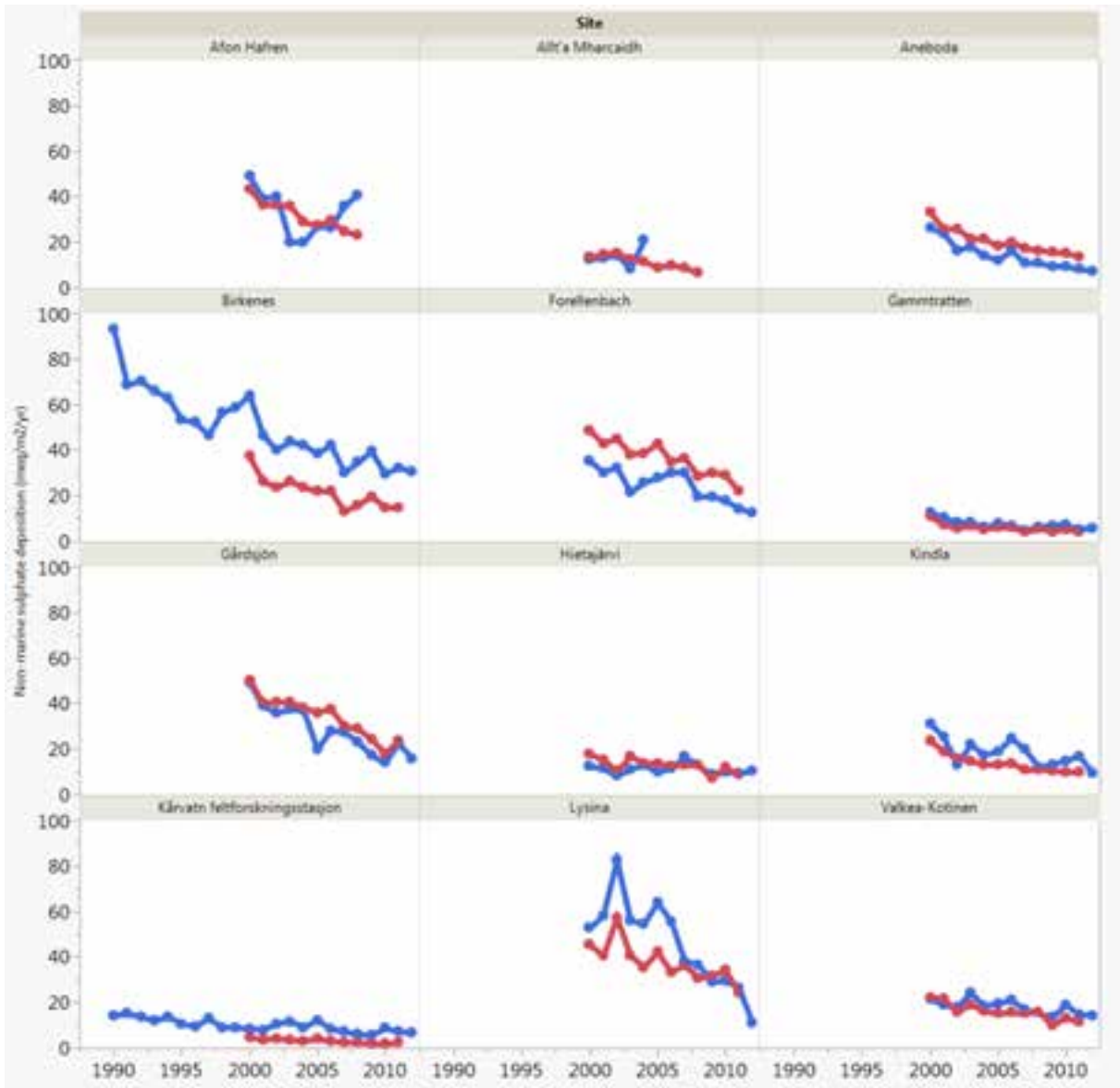
Figure 17. Measured (blue) and EMEP modelled (red) (model version rv4.3beta,13 April 2013) annual deposition of non-marine sulphate at 12 ICP IM stations.
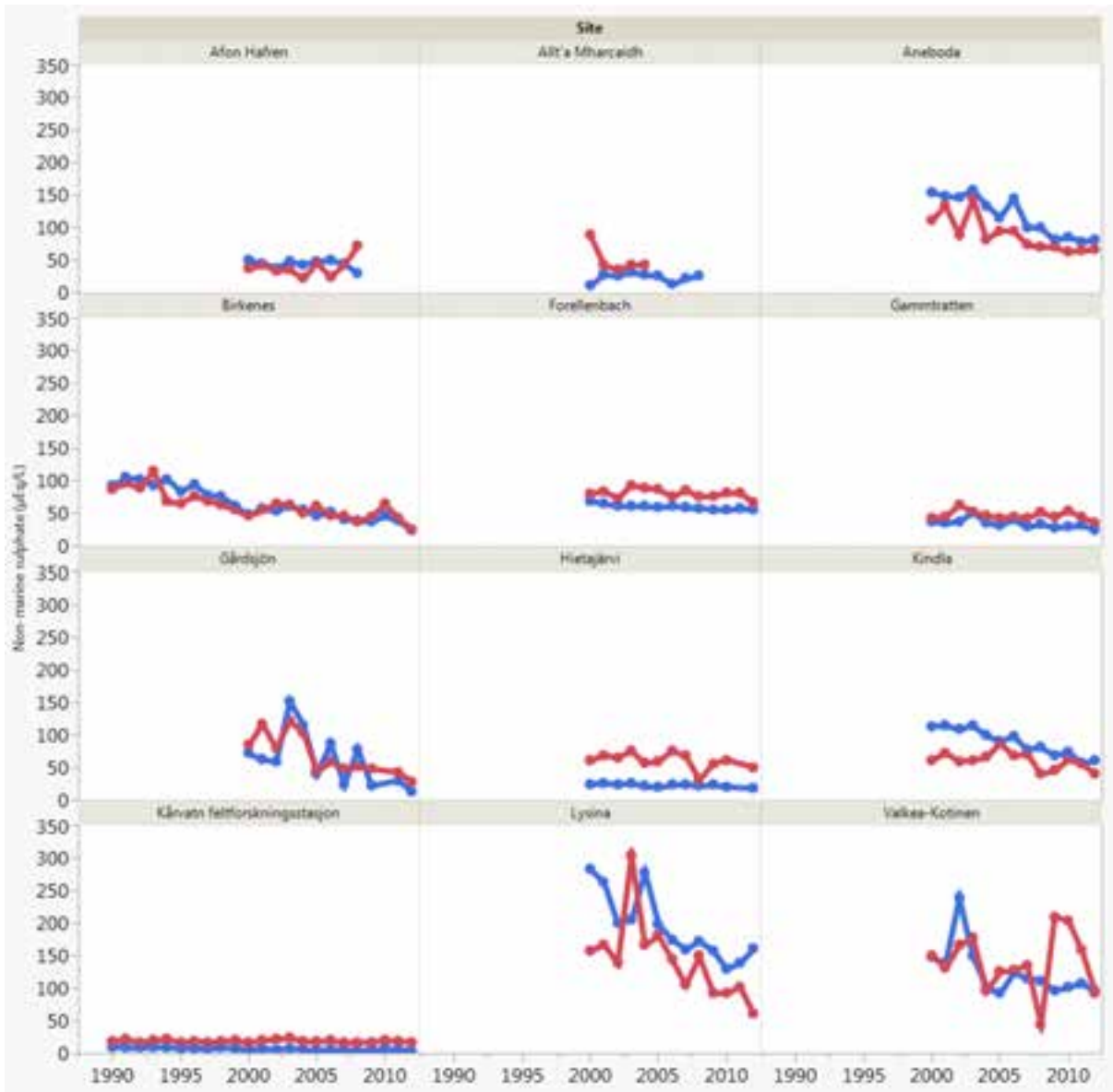
Figure 18. Annual average measured (blue) and estimated (red) concentration of non-marine sulphate in surface waters at 12 ICP IM stations.
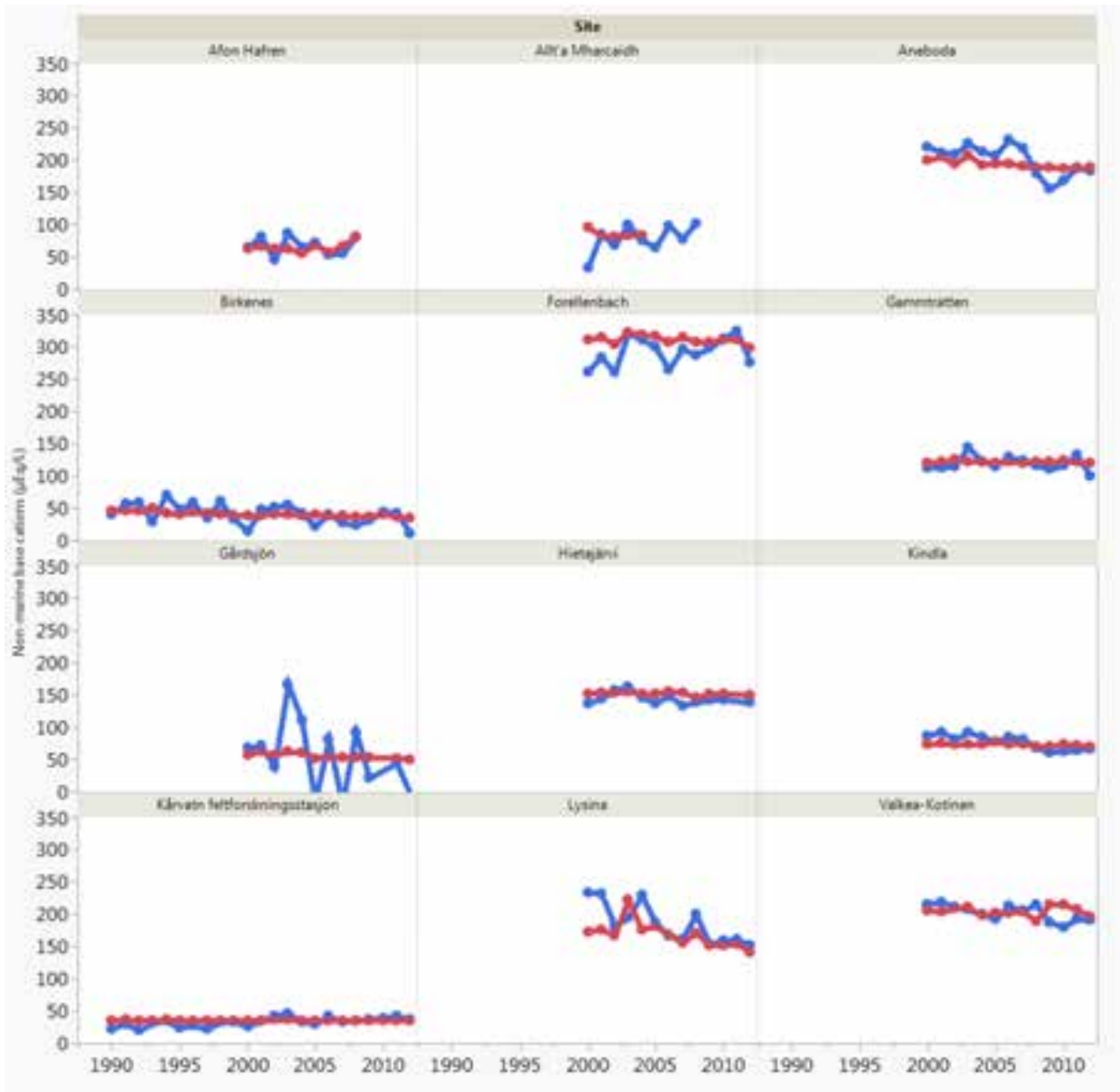
Figure 19. Annual average measured (blue) and estimated (red) concentration of non-marine base cations in surface waters at 12 ICP IM stations.
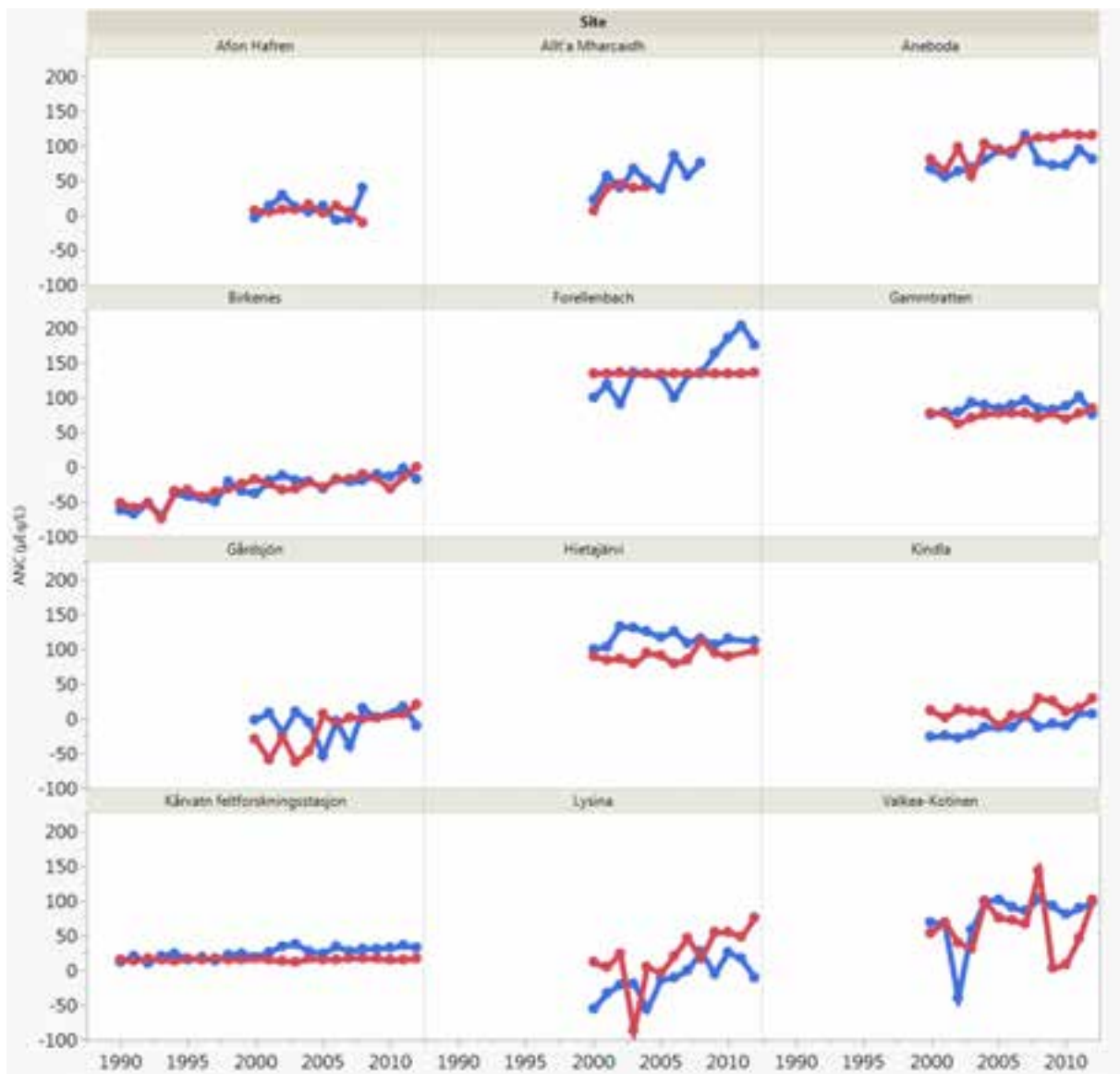
Figure 20. Annual average measured (blue) and estimated (red) ANC in surface waters at 12 ICP IM stations.

Sulphur deposition estimated by EMEP. Estimated sulphur deposition has varied greatly between regions but has decreased everywhere since year 2000 (Figure 21). Sulphur deposition will decrease further up to 2020. However, the changes are expected to be small compared to those observed between 2000 and 2011, and in the preceding decade (see case studies below).
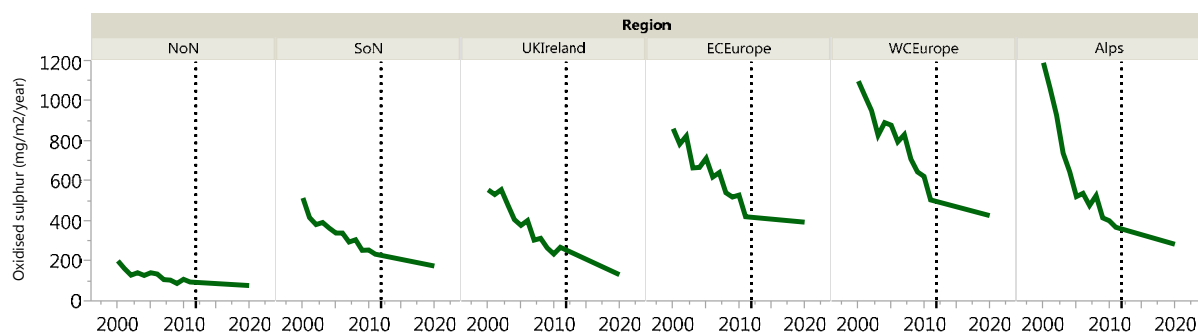
Figure 21. Sulphur deposition between 2000 and 2011 modelled by EMEP (model version rv4.3beta,13 April 2013). The lines represent average values for all the stations in each region. The scenario for 2020 is based on emission ceilings defined by the revised Gothenburg protocol 2012.

Regional prognoses for water chemistry up to 2020. The model exercise was performed for 61 acid sensitive European stations that fulfilled the data requirements (Figure 22). The concentration of non-marine sulphate has decreased in all regions between 2000 and 2012 (see also section 2.2.1), and concentrations in 2020 are expected to be lower than they were in 2005 (Figure 23, upper panel). In 4 of 6 regions the concentration of non-marine sulphate has decreased by approximately the same rate as predicted by a linear interpolation between 2005 and 2020 scenarios. In the South Nordic region and the East Central European region the sulphate concentration has dropped faster and to a lower level than estimated. The sulphate concentration is still higher than estimated background (i.e., pre-industrial) levels in all regions, and this is also expected to be the case in 2020.

The deviations between observations and model estimates described for sulphate above are propagated for non-marine base cations (Figure 23, mid panel) because of the way base cation concentration is estimated (see section 2.1.4). In 4 of 6 regions there is good agreement between observed and estimated base cation concentrations, assuming linear trends between 2005 and 2020. The same is true for ANC (Figure 23, lower panel). Note that in all regions the expected increase in ANCis small compared to the year-to-year variations in measured values caused by fluctuations in deposition, climate and, for some catchments, seasalts. This was also the conclusion in a so-called ex-post analysis of deposition scenarios for 2020, where the dynamic MAGIC model was used on 8 case studies (Wright et al., 2011). Other studies also emphasize the increased importance of deposition, climate and seasalts for future chemical recovery (Clark et al., 2010; Helliwell and Simpson, 2010; Moldan et al., 2012; Wright and Dillon, 2008 and other papers in that special issue on effects of climate change on recovery).

An annual mean ANC of 20 µEq/L is often used as a threshold for protecting sensitive aquatic organisms from negative effects of acidification. This threshold is based on Lien et al. (1992) who reported that for ANC >20, brown trout populations were not impacted at 90 % of the study sites (1095 lakes and 30 rivers in Norway) , and Atlantic salmon were not impacted for 100 % of the sites. There are more nuanced ways to estimate the critical ANC limit than the "one for all" limit of 20 µEq/L (Henriksen et al., 1995; Lydersen et al., 2004; Malcolm et al., 2014), but it can still be used as an indicator of the potential for recovery. According to our calculations, 20 of the 61 studied sites will still have ANC < 20 µEq/L in 2020 (Appendix C), indicating that full biological recovery throughout Europe cannot be expected in the near future.

The next section will supplement the presentation of regional average results given above with four individual cases where data from the 1990s also are included.
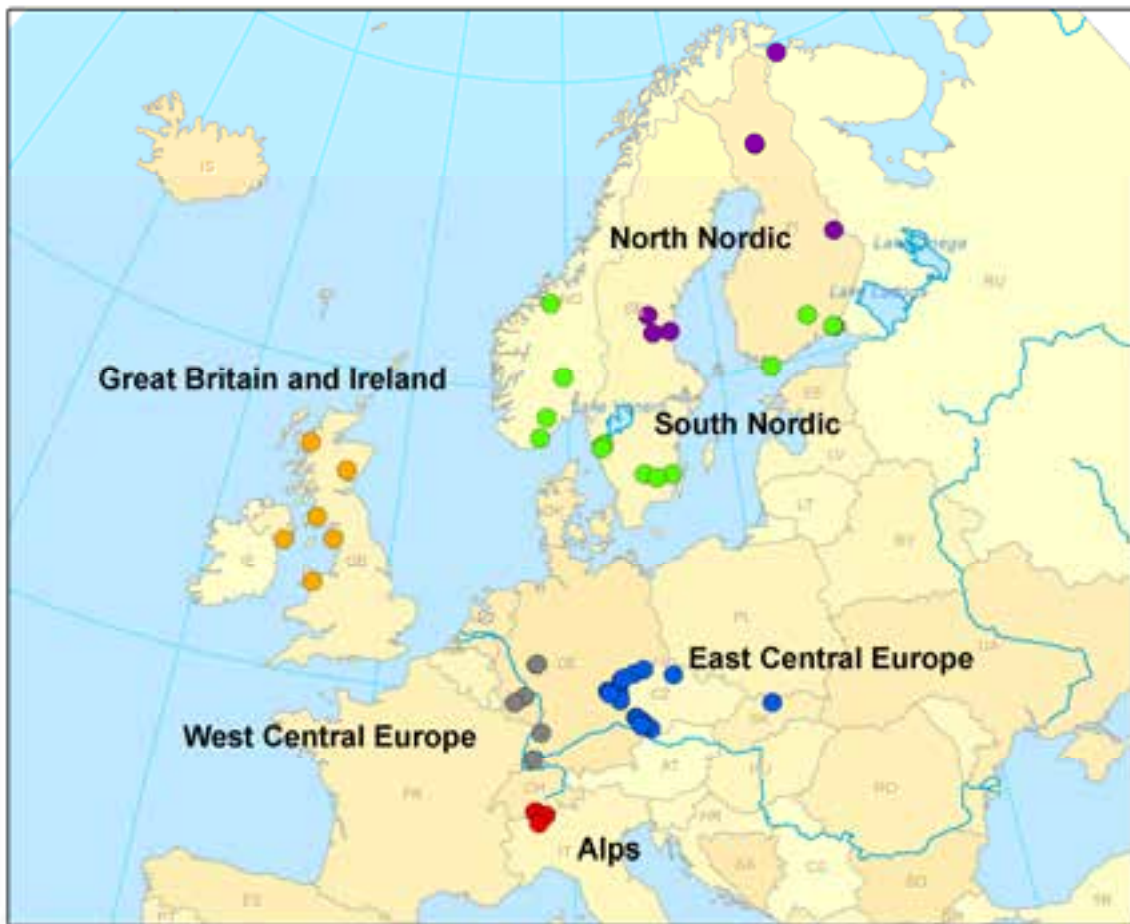
Figure 22. Map showing the 61 stations included in the modelling exercise. The different colours of the markers indicate grouping of the stations into geographical regions.
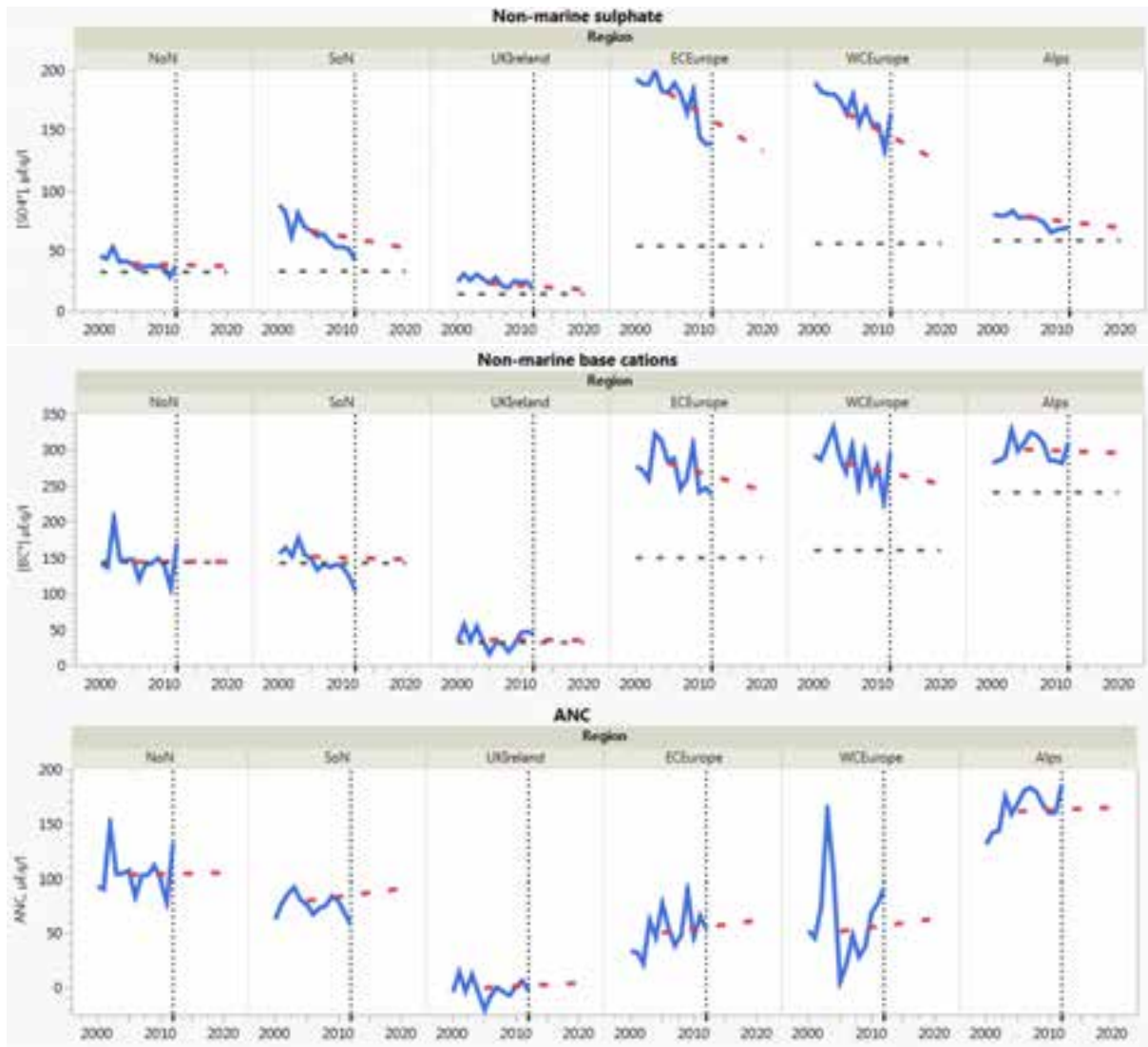
Figure 23. Measured (blue lines) and modelled (red lines) average concentration of non-marine sulphate (upper panel), non-marine base cations (mid panel) and ANC. The grey horizontal lines represent the estimated background concentrations (regional average).

Examples from different regions

The four examples below have been selected not because they provide the best fit with the model, but rather because they differ with respect to deposition of sulphur and seasalts in the catchment, and climatic factors such as temperature and amount of precipitation. They also have delivered good quality data since 1990 (and before).

Birkenes (South Nordic region) (Figure 24) is a stream water station draining a small forested catchment dominated by old Norway spruce. It is about 20 km from the coast and receives moderate amounts of seasalt. The average runoff (1960-1990) is 1020 mm. There is good agreement between measured and estimated values in the time spans 1990-2005-2020 if we interpolate linearly. The reductions in sulphur emissions in the 1990s resulted in a large increase of ANC. According to the predictions, ANC will continue to increase but at a slower rate. Sulphur deposition in 2020 is expected to still be high enough to keep sulphate concentration above background levels.

Cerné lake (East Central Europe) (Figure 25) is a small forest lake located at 1008 m.a.s.l. in the Bohemian forest. The average annual runoff is 1157 mm. Here, the decrease in sulphate concentration has been

almost linear since 1990, even if the EMEP model results indicate much higher deposition in 1990. Moreover, the sulphate concentration is already at the level predicted for 2020, suggesting that the relative changes in EMEP modelled deposition are too high and too low, respectively, for the timespans 1990-2005 and 2005-2020.

Vasikkajärvi (North Nordic region) (Figure 26) is a lake in Lapland with low runoff (330 mm/year). The sulphur deposition has been much lower here than in Birkenes and Cerné, but there is still a clear trend in aqueous sulphate that agrees well with model predictions. The drop in base cations has been somewhat steeper than simulated and actually balances the drop in strong acid anions. There has therefore been no clear trend in ANC. A possible explanation is dilution caused by high rainfall in summer and autumns in the period 2004-2008 (see also section 3.3).

Round Loch of Glenhead (United Kingdom) (
Figure 27) is a lake located at 298 m.a.s.l. in the Galloway region of south-west Scotland. The average annual runoff is 2001 mm. This site has higher runoff and larger variation in base cation concentrations than the other three stations. There is good agreement between measured and estimated values in the time spans 1990-2005. The concentration of non-marine sulphate will decrease further up to 2020, and this will increase ANC (see also Helliwell et al., 2014). Non-marine sulphate will in 2020 be close to the calculated background concentration.
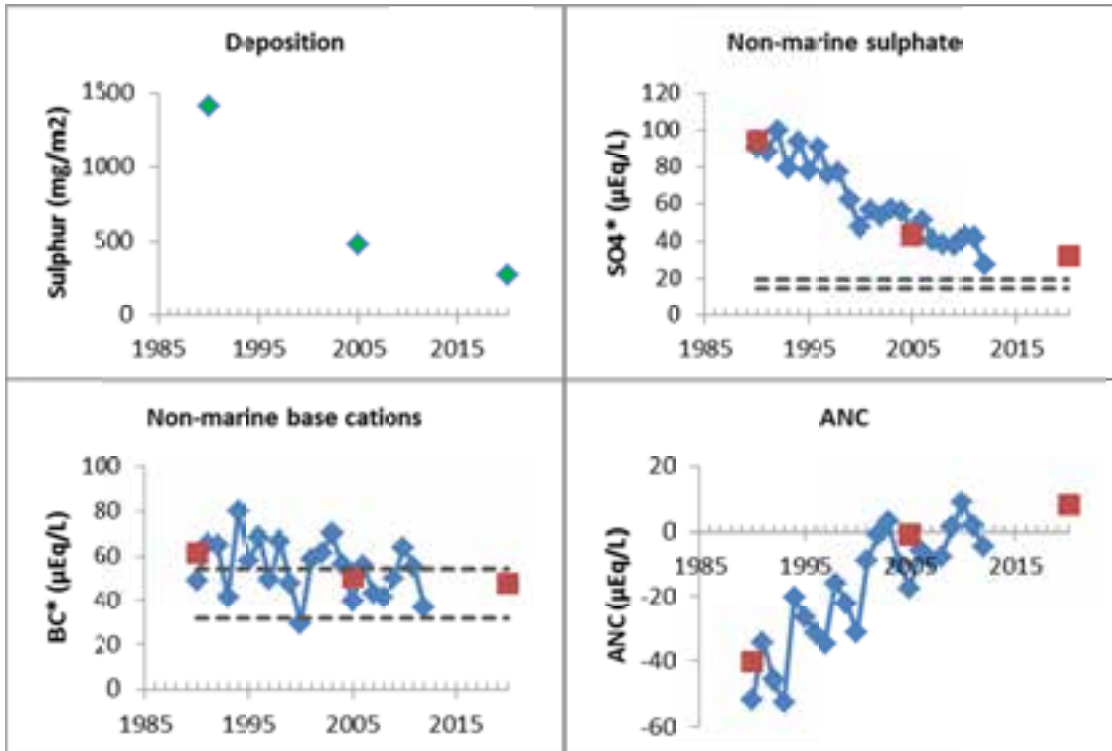
Figure 24. Deposition of oxidised sulphur estimated by EMEP (green), mean annual concentration of non-marine sulphate, base cations and ANC (blue) at Birkenes. Red points represent model estimates. Grey lines are the average +/- 1 standard deviation of estimated background concentrations.
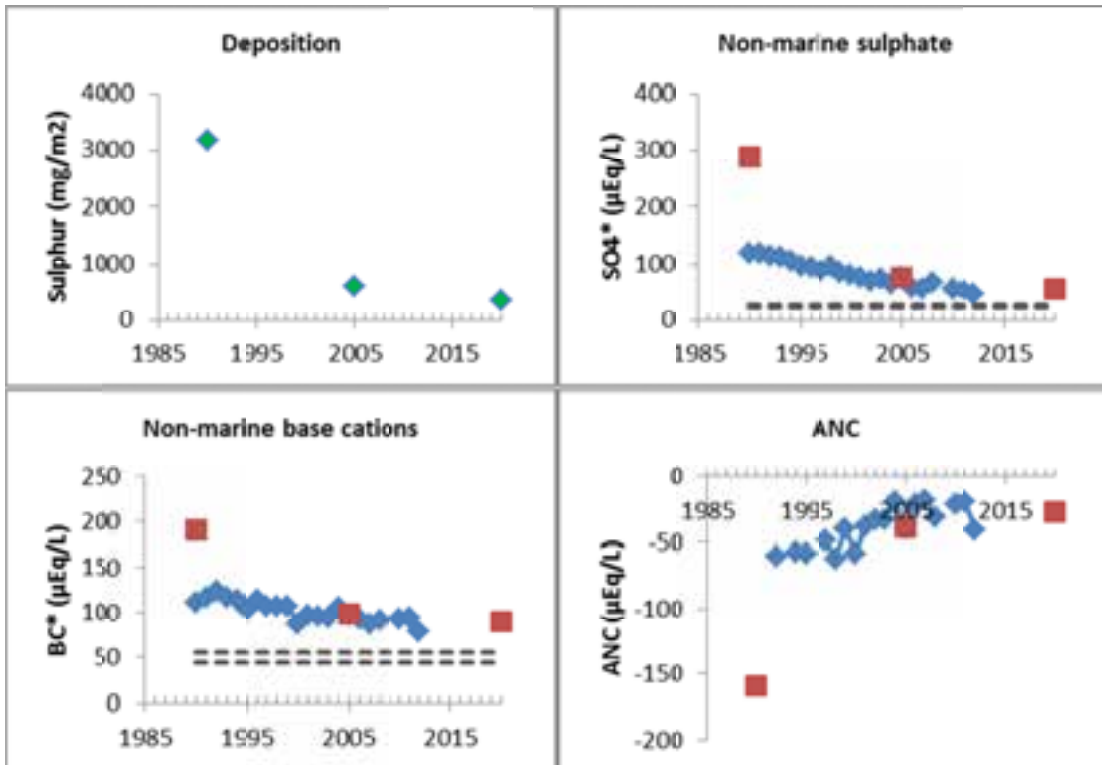
Figure 25. Deposition of oxidised sulphur estimated by EMEP (green), mean annual concentration of non-marine sulphate, base cations and ANC (blue) at Bohemian Forest, Cerné. Red points represent model estimates. Grey lines are the average +/- 1 standard deviation of estimated background concentrations
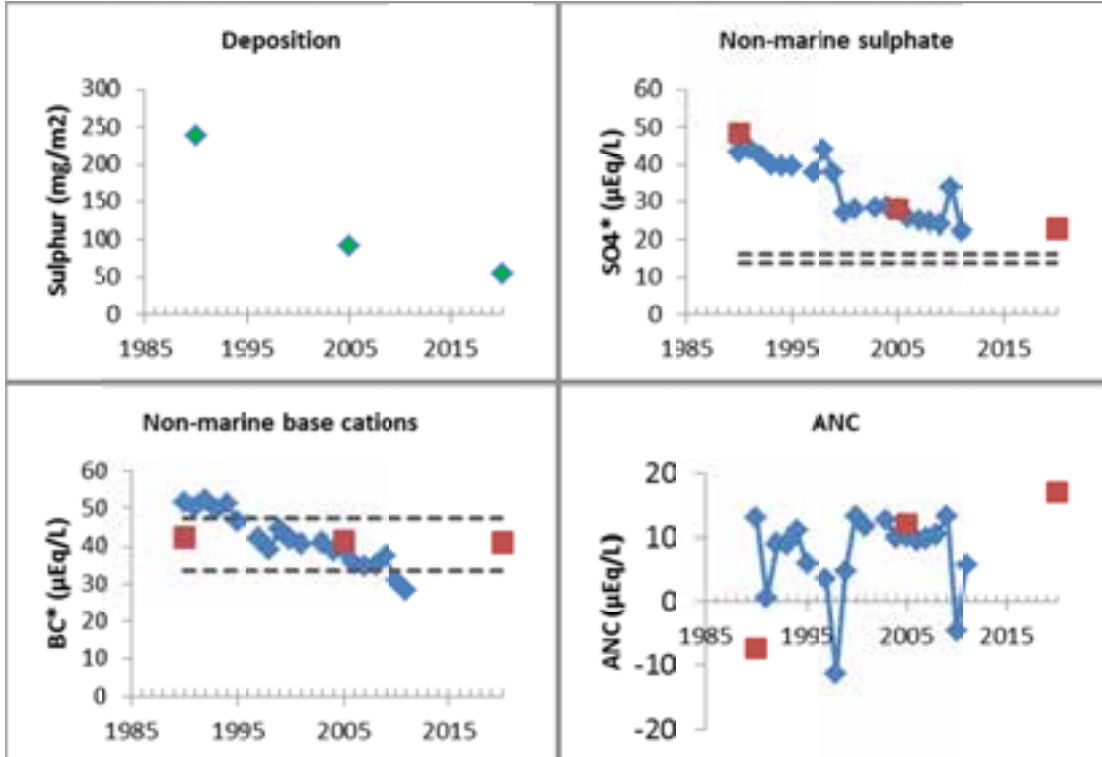
Figure 26. Deposition of oxidised sulphur estimated by EMEP (green), mean annual concentration of non-marine sulphate, base cations and ANC (blue) at Lapland, Vasikkajärvi. Red points represent model estimates. Grey lines are the average +/- 1 standard deviation of estimated background concentrations.
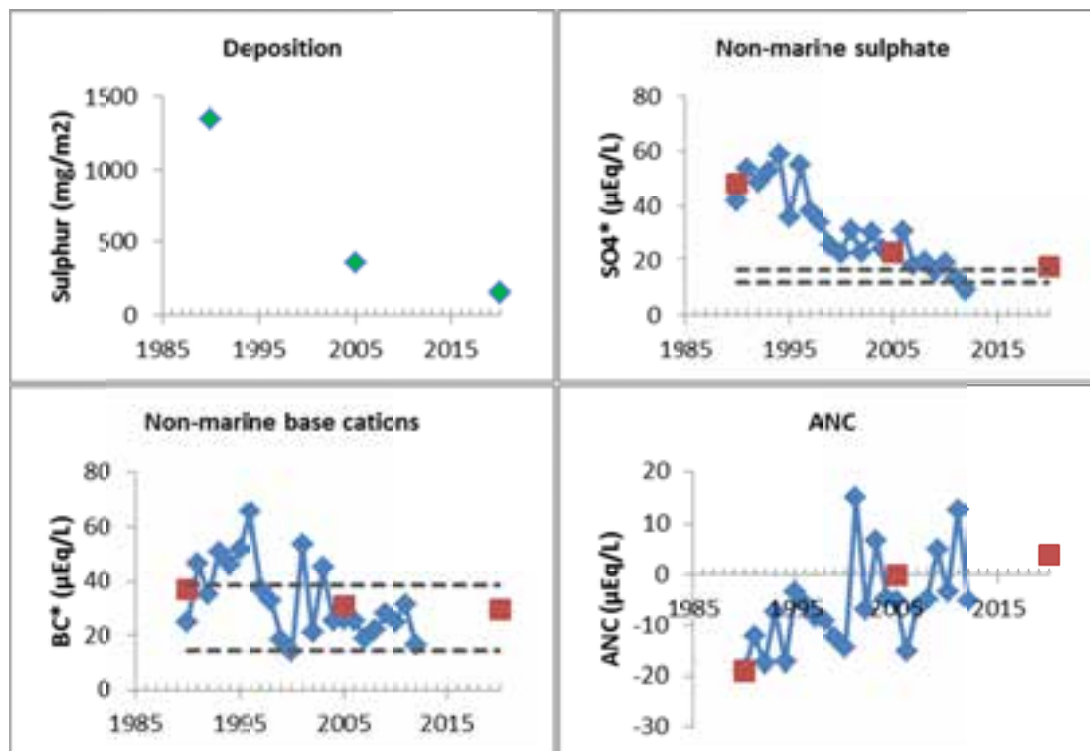
Figure 27. Deposition of oxidised sulphur estimated by EMEP (green), mean annual concentration of non-marine sulphate, base cations and ANC (blue) at Round Loch of Glenhead. Red points represent model estimates. Grey lines are the average +/- 1 standard deviation of estimated background concentrations.

2.3 Conclusions

Trends in water chemistry. Sulphate concentrations in headwaters have declined between 2000 and 2011 as a result of reduced sulphur emissions. Nitrate has been decreasing in the Alps over the whole period. Elsewhere the pattern for nitrate is mixed, and clear changes on a regional scale are not apparent in any of the regions. Consistently increasing trends in both alkalinity and ANC are not evident in most regions, despite the decrease in sulphate, but positive trends still dominate. The clearest indication that water chemistry is recovering from acidification is a widespread increase of pH.

Prognosis water chemistry in Europe up to 2020. Sulphur deposition will decrease further up to 2020, but changes are expected to be smaller than those observed between years 2000 and 2010. Increases in ANC are predicted, but changes will be small and subtle, as they also were between 2000 and 2011. In many acid-sensitive headwaters, estimated annual mean ANC in 2020 will still be lower than levels required to achieve full biological recovery. Year-to-year variations in ANC due to fluctuating deposition, climate and, for some regions, seasalts will be larger than the difference between 2010 and 2020 levels. These climate-related variations in water chemistry may hamper biological recovery. Further improvements in surface water chemistry beyond 2020 will require additional reductions in S emissions. The calculated S deposition in 2020 under the Current Legislation scenario is very similar to that under the Maximum Feasible Reduction scenario (Wright et al 2011), but both are still above the estimated background S deposition levels.

3. Trends in biological recovery of acidified surface waters

3.1 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. Documentation of chemical recovery of acid-sensitive waters is considerable (Chapter 2) based on an extensive regionally distributed database with time series that date back to the 1980s and 1990s (Stoddard et al., 1999; Skjelkvåle et al., 2005; Garmo et al. 2014). For documentation of biological recovery, data collection is more resource-demanding than for water chemistry and often started later than for data collection of water chemistry. Still, data on biological communities in acid-sensitive waters have been collected in many countries with time series varying between 10-30 years. Many of these sites are in the ICP Waters monitoring programme. The data allow for analysis of the effects of acid deposition on freshwater ecosystems with respect to water chemistry, biology 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 comparing trends in regions across national boundaries is challenging. Each country supplies a report of data relevant for assessing biological recovery in acid-sensitive regions. In 2014, the ICP Waters subcentre received contributions from the Czech Republic, Finland, Norway and Switzerland. 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 phytoplankton (photosynthesizing microscopic organisms), ciliates (one-celled organisms), zooplankton (small animals living in the free water bodies), water bugs and fish populations are also given. Part of the data records presented in the current report are included in previous trend reports (Raddum et al. 2004, 2007, Fjellheim & de Wit, 2011) evaluated against trends in water chemistry.

Biological responses to acidification and to improved water chemistry are different
There are fundamental differences in the processes leading to biological damage under acidification, and the processes of biological recovery when the critical load is no longer exceeded. The main driver for biological change during acidification is the toxic effect of water chemical components, such as labile aluminium and low pH. When the critical load is exceeded, concentrations of toxic components exceed critical thresholds, which lead to damage and disappearance of acid-sensitive species (Raddum and Fjellheim, 2002, Posch et al., 2003). During the acidification process, the community structure is generally predictable at different acidic levels. Biotic interactions occur due to the disappearance of key sensitive organisms. An example is the effect of loss of fish species and the immediate increase of invertebrate species sensitive to fish predation. If all fish species are extirpated, invertebrate species become 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, gradually declines in importance. Now the main structuring drivers 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 exact opposite path of the changes in community structure during acidification. Due to this, predicting the biological target and

'end-point' of the biological recovery is difficult. Recovery from acidification is thus not likely to result in the pre-acidification biological community.

An example of a recovery process is the development of the caddisfly *Hydropsyche siltalai* in rivers in the Farsund area in southwest Norway during 1981 – 2013 (Figure 28). The species was missing from the samples in the first eleven years and appeared at the locality in 1992, when the critical limits were no longer exceeded. This corresponds to stage t5 of Posch et al. (2003). During the 1990s the populations were small, as reflected by low densities. In 2000 - 2001, *H. siltalai* exceeded the critical abundance (stage t6) and has since then been found regularly in good numbers. We assume that the population of the species now has reached the stage 4 of the MIRACLE conceptual model (Raddum & Fjellheim 2002), i.e. natural population fluctuations. The time to reach stage 4 depends mostly on biotic factors (Raddum & Fjellheim 2002, Yan et al., 2003).
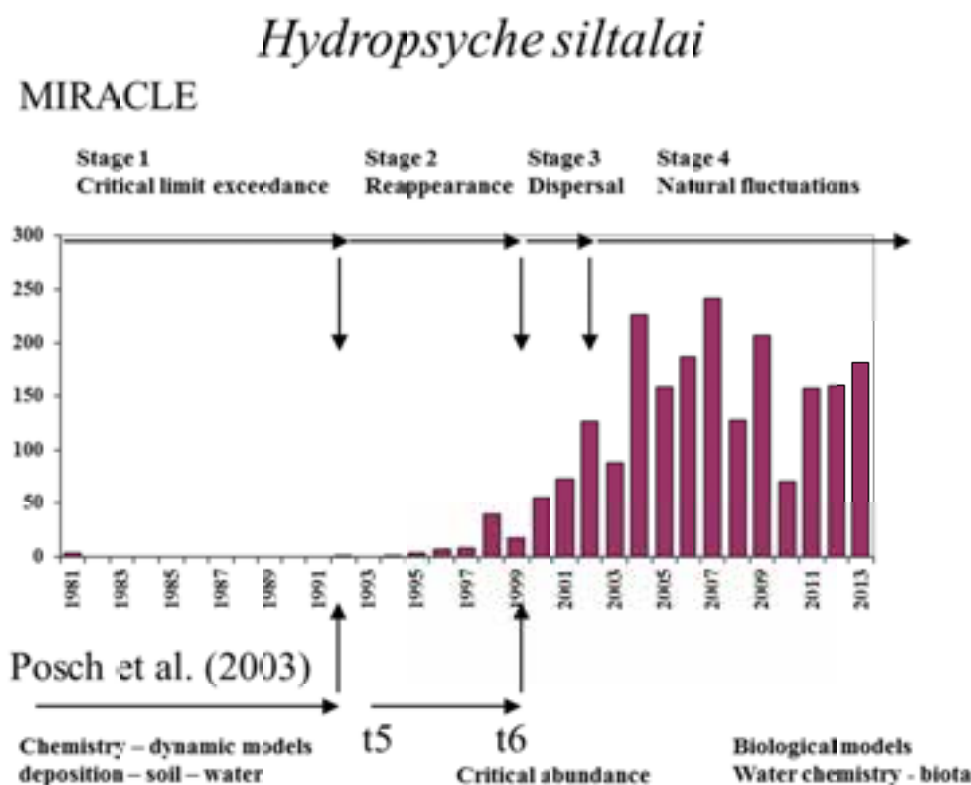
Figure 28. Colonisation of the caddisfly Hydropsyche siltalai in the Farsund area, within the frames of conceptual models suggested by Raddum and Fjellheim (2002) and Posch et al. (2003). Modified after Fjellheim and Raddum (2005). H. siltalai figure taken from this report.

Summary of the national contributions on biological recovery
The data presented in this report show that biological recovery after reduced emissions of acidic components has been documented to a variable extent in the contributing countries (Table 1). Most regions document chemical recovery, accompanied with trends in biological recovery that generally agree 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, some of them related to climate change. Also, in some countries the times series are still rather short.

The Czech contribution consists of phytoplankton, ciliates, zooplankton, and aquatic insects in Bohemian Forest lakes. The data indicate that high inorganic aluminium concentrations are a serious bottleneck for survival and biological recovery. This, in combination with nutrient status and high DOC concentrations, form major constraints for biological recovery in the Bohemian Forest lakes. Biotic interactions control the structure and abundance of plankton, especially predation. Highly unfavourable plankton stoichiometry (high C:P ratio) and limited dispersion due to the lack of potential source localities may represent additional constraints to recovery in the region.

In Finland, the integrated monitoring of fish populations and water chemistry in acidified lakes indicates that chemical recovery was followed by a clear biological recovery, especially in perch populations. Perch and roach populations were monitored, and perch, as an acid-tolerant species, has responded rapidly to improved water quality conditions, while for more acid-sensitive species such as roach little if any recovery of affected populations was recorded. However, the biological recovery or biological changes due to declined acid deposition and expected improvement in water quality are difficult to predict because of the complex interaction between the many climatic, chemical and biological variables involved. The findings of the present study emphasize the importance and value of the integrated monitoring approach which includes both physical, chemical and biological variables, and the suitability of small headwater lakes for such monitoring.

Biological recovery in Norwegian stream localities has been documented 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 increased biodiversity. The response in Norway is clearer than results from other regions in Europe. 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. Second, the data are based on running waters, which generally gives a quicker response than the fauna from better buffered lakes. Third, the Norwegian catchments are characterised by surface waters with very low concentrations of electrolytes and DOC and a thin layer of sediments making the localities both more susceptible to acidification and easier to restore.

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

Conclusions
The national contributions (Czech Republic, Finland, Norway, Switzerland) 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. All contributions report evidence of chemical and biological recovery. The best documentation of improved water quality is where the longest time series (over 30 years) were available (Norway, Finland) or where acid deposition has been reduced most strongly (Czech republic). Biological time series are usually shorter than chemical time series and do not show an equally consistent recovery as for water quality.

The only example of a fully recovered biological population in the data presented here, stems from Finland, from a fish species with a relatively high acid-tolerance (perch). Populations from a more sensitive fish species (roach) showed only a little recovery and were still damaged. Complex interactions between climate, water quality and fish populations make it difficult to predict the endpoint of fully recovered populations.
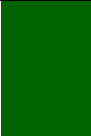
In Norway, all investigated invertebrate populations showed strong signs of recovery, which was related to improved water quality. However, climate-related seasalt events were an occasional setback for recovery. Water quality is still acidic and poorly buffered, and invertebrate populations remain vulnerable and still have potential for further recovery.

In the Czech Republic, all investigated lakes showed recovery of invertebrates, but most clearly in the lakes that were less acidified. High aluminium concentrations, biotic interactions (presence of predators) and limited dispersion potential (absence of nearby undamaged populations) appeared to be the most important bottlenecks for biological recovery.

The lakes in Switzerland did not show any biological recovery, despite improvements in water quality in the most acidified lakes, but rather the contrary: loss of sensitive species. This was possibly related to a higher frequency of acidic episodes after 2005, related to more intense precipitation.

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 (Fjellheim & Raddum, 1995).

Table 1. Summary of finding from national reporting on biological recovery. Colour coding for trends: ████ , only positive trends. ████ , mixture of positive and no trends. ▭ , no trends.

Region	Country	Water body	Biota	Biological parameter	period	Trends	recovery potential reached?	Cause of recovery	other relevant factors
Nordic	Norway	5 rivers	Zoobenthos	Acidification index, Biodiversity, Acid-sensitive organisms	1982-2013		no	improved water quality	Seasalt episodes. Slow recovery
	Finland	21-30 lakes	Fish	Abundance, Population structure	1985-2012		No (in some locations)	improved water quality	Climate, Sensitivity of fish species, biotic interactions
East Central Europe	Czech Republic	8 lakes	Phytoplankton	Species number, abundancel	1999-2011		no	improved water quality	Climate, lake acidity status; biotic interactions
			Zooplankton	Species number, abundance	1999-2011		no	improved water quality (especially reduced aluminium concentrations)	,
			Zoobenthos, Nepomorpha	Species number, abundance	1999-2011		no	improved water quality	Lake morphometry (presence of littoral zone), biotic interactions
Alps	Switzerland	4 lakes	Zoobenthos	Species number, abundance, Acid-sensitive organisms	2002-2013		no information	increase of acidic episodes	Increased precipitation, short time series, ecosystems with poor invertebrate fauna especially

3.2 Czech Republic: Recent biological recovery of the Bohemian Forest lakes from acid stress

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Jiří Kopáček, Miroslav Macek, Tomáš Soldán, Biology Centre ASCR, České Budějovice, Czech Republic
Jan Fott, Linda Nedbalová, Veronika Sacherová, Charles University, Prague, Czech Republic
Pavel Chvojka, National Museum, Prague, Czech Republic

Since the late 1980s the Bohemian Forest (Šumava in Czech, Böhmerwald in German) has experienced the largest changes in acidic deposition in central Europe, as documented by a 86%, 53%, and 35% reduction in sulphur dioxide, nitrogen oxides, and ammonia emissions, respectively, (Kopáček *et al.* 2012). In small and geologically sensitive catchments covered with coniferous forests, eight Bohemian Forest lakes along the Czech–German border have rapidly shown substantial recovery of water chemistry (Kopáček *et al.* 1998, Vrba *et al.* 2003). Biological recovery, however, has lagged behind for a decade or even longer (Nedbalová *et al.* 2006, Soldán *et al.* 2012, see also ICP Waters report 106/2011). This is attributed to continuing adverse effects of aluminium (Vrba *et al.* 2006, Stockdale *et al.* 2014).

We evaluated biotic responses to environmental changes at the Bohemian Forest lakes based on data from four years (1999, 2003, 2007 and 2011) covering the 12-year period 1999-2011. During this period chemical recovery was ongoing in response to decreases in the extremely high regional atmospheric depositions of sulphur and nitrogen. Several ecologically different groups of organisms were studied to cover as many aspects of biological recovery as possible: phytoplankton, zooplankton (Rotifera, Cladocera, and Copepoda), ciliates, and four groups of aquatic insects (Ephemeroptera, Plecoptera, Trichoptera, and Nepomorpha).

We used surface water chemistry to characterise the environmental conditions in the epilimnion, the habitat of both planktonic and littoral biota during the growing season. The Bohemian Forest lakes formed two distinct groups during this study: chronically acidified and slightly acidified lakes (further referred as acidic and recovering according to their chemistry; (Table 2). Lakes in both groups showed an exceptional amelioration in September 2003 due to warm and dry summer and, in contrast, a certain regress of chemical recovery caused by cold and rainy weather at the beginning of September 2007. Nevertheless, chemical recovery progressed gradually in all lakes over the period 1999 to 2011, with higher pH, positive ANC, lower aluminium concentrations, or higher P_t and DOC concentrations in the recovering lakes compared to the acidic lakes (Table 2). The most pronounced biotic response in all the lakes was the gradual disappearance of microbial filaments resulting in a one order of magnitude decrease in heterotrophic biomass during the studied period (see HB in Table 2).

In total, 28, 51, 33, 27, 30, and 21 taxa of phytoplankton, ciliates, rotifers, crustaceans, EPT, and Nepomorpha, respectively, were recorded in Bohemian Forest lakes during the study. Despite wide ranges, average numbers of taxa of all taxonomic groups, except for phytoplankton, were generally lower in acidic than in recovering lakes in all sampling years (Table 2). When comparing by years, average numbers of taxa increased in time in the majority of groups, showing more variation of less species in recovering than in acidic lakes. In contrast, phytoplankton exhibited an inverse pattern and species richness of Nepomorpha remained the same until 2003 and then markedly increased (Table 2). No change in species richness, however, was significant in pairwise comparisons of consecutive years indicating rather gradual changes. A comparison of the first and last sampling years showed a significant ($p < 0.03$) decrease in phytoplankton and marginally significant ($p = 0.05$) increase in EPT and Nepomorpha.

Species richness of phytoplankton partly differed between acidic and recovering lakes (cf. numbers of taxa in Table 2). When comparing sampling years, phytoplankton assemblages were more similar in acidic lakes than in recovering lakes (Figure 29A). While the abundance of some species changed, species composition remained rather stable in acidic lakes. Lakes that showed evidence of chemical recovery displayed a variation in biological responses: a certain retreat, decrease in abundances or even species loss of acid-tolerant species, as well as increasing top-down control by zooplankton (see
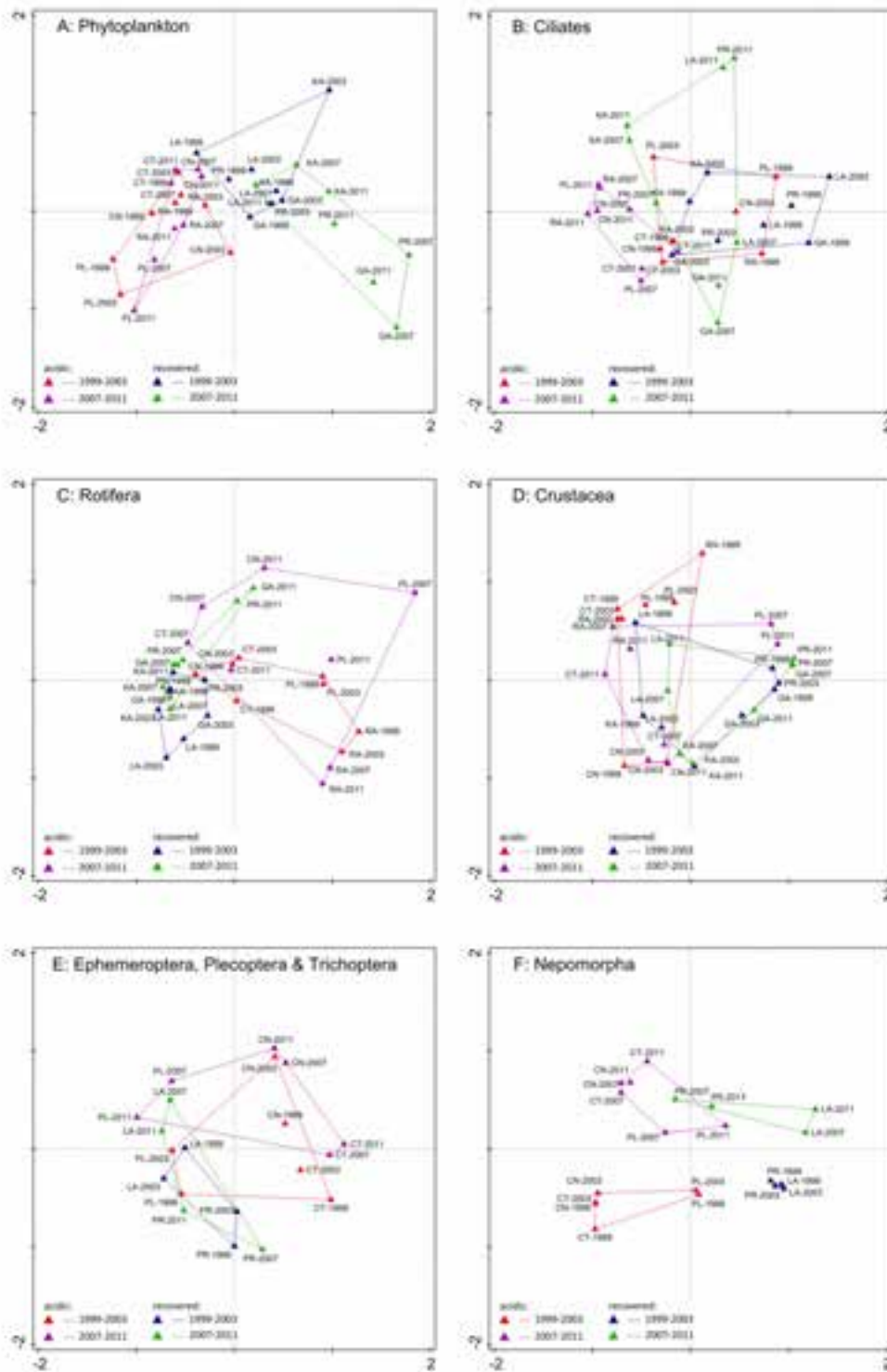
Figure 29. Changes in community composition of eight acidified lakes in the Bohemian Forest (ICP Waters codes in parentheses). CT – Čertovo Lake (CZ02), CN – Černé Lake (CZ01), PL – Plešné Lake (CZ03), RA – Rachelsee (DE17), PR – Prášilské Lake (CZ04), GA – Großer Arbersee, KA – Kleiner Arbersee, and LA – Laka Lake (CZ05) sampled during late-summer (August/September) campaigns in 1999, 2003, 2007, and 2011. Comparison of particular taxocoenoses by NMDS model is based on Bray-Curtis dissimilarity of lakes and sampling campaigns; acidic vs. recovering lakes, as well as two periods (1999–2003 vs. 2007–2011) of the study are distinguished by colours of symbols and envelopes.

below) gradually appeared in recovering lakes (cf. KA, PR, and GA in Figure 29A). Ciliates showed high variability in species richness (Table 2) and composition of assemblages among both years and lakes (Figure 29B), however, without clear trends or significant changes.

Gradual recovery of zooplankton and benthos has been affected by the overall improvement of chemical conditions in the Bohemian Forest lakes and, consequently, by food resources and trophic interactions. Rotifers generally have responded by an order-of-magnitude increase in their abundance, unless there was competition from herbivores (*Ceriodaphnia quadrangula* or *Daphnia longispina*) or predation by invertebrates (*Cyclops abyssorum*, *Heterocope saliens*, Nepomorha, *Chaoborus obscuripes*, etc.) (cf. PL and PR in Figure 29C). For instance, an acid-tolerant rotifer, *Microcodon clavus* recently formed a dense pelagic population in Rachelsee (for details, see Vrba *et al.* 2014; cf. RA in Figure 29C). In addition, some sensitive rotifers have appeared in some lakes (e.g. *Keratella hiemalis* in CN and GA) during the study period. Unlike in the recovering lakes (and PL with successful re-introduction of *C. abyssorum*), crustacean zooplankton have remained scarce and poor in species in the acidic lakes, where *C. quadrangula* seems to be a single indicator of zooplankton recovery (Figure 29D).

Recent increases in both species richness and abundance of littoral insect larvae in all eight Bohemian Forest lakes has been reviewed by Soldán *et al.* (2012); however, only five Czech lakes could be evaluated in detail. In Ephemeroptera, Plecoptera and Trichoptera (EPT), samples showed the highest similarity of all studied groups of organisms. The composition of EPT assemblages moved along the first axis from heavily acidified lakes without vegetated littoral zones (CN and CT in Figure 29E) via recovered lakes with limited littoral vegetation (PR) towards both acidified and recovering lakes with large and rich sedge and moss littorals (LA and PL in Figure 29E). This suggests the important role of littoral conditions in lakes, which could dominate any impact of acidification. Nevertheless, some temporal changes in the composition of assemblages were apparent. Samples taken sequentially were ordered along the second axis, particularly in acidic lakes (cf. CN, CT, and PL in Figure 29E). It was connected with an increase in species richness (Table 2) and re-colonisation of some species originally known from lakes in the past (Soldán *et al.* 2012).

The most pronounced changes in the composition and species richness of assemblages were found in Nepomorpha. Species-poor assemblages found in lakes in 1999–2003 were conspicuously dissimilar to species-rich and diversified assemblages found in 2007–2011; moreover, in both periods, acidic and recovering lakes hosted dissimilar assemblages (Figure 29F). This suggests a superior role of prey availability in the recovery of mostly acid-tolerant top predators.

We also used multivariate analyses RDA and CCA using the forward selection to evalauate the role of main environmental factors in structuring of assemblages. Aluminium (Al) concentration was the most important factor determining the assemblages of three studied groups of organisms: total Al (Al_t) explained 24.9% and 27.4% of variability in phytoplankton and rotifers, respectively, while inorganic Al (Al_i) explained 25.9% of variability in Nepomorpha. Seston stoichiometry (C:P) was the only significant factor that influenced cladocerans and copepods, and pH was the only factor significant in EPT. No factor was significant for ciliates.

In summary, the Bohemian Forest lakes differ in Al concentrations and speciation. In the acidic lakes, considerable seasonal variations in Al_i are controlled by pH, resulting in either summer epilimnetic minima or early spring peaks. Hence, aluminium may be considered as a serious bottleneck for survival and biological recovery of the lakes, including macrophytes and fish (Vrba *et al.* 2006, Čtvrtlíková *et al.* 2009, 2012, 2014, Stockdale *et al.* 2014, Matěna *et al.* in prep.).
Our data suggest that this environmental framework, together with nutrient status and DOC concentrations, form major constraints of biological recovery and are responsible for different trajectories in individual lakes. Besides the environmental factors, biotic interactions (such as competition for resources, their availability or quality, predation by invertebrates or re-stocked fish) control the structure and abundance of plankton. Highly unfavourable plankton stoichiometry, namely the high C:P ratio, and limited dispersion due to the lack of potential source localities may represent additional constraints to plankton recovery in the region. By contrast, our preliminary data from Malaise traps rather suggest that recovery of aquatic insects is not strongly limited by species dispersal.

Acknowledgements

Table 2. Mean epilimnetic values (± SD) of selected environmental variables of chronically acidified (acidic: Čertovo, Černé, Plešné, and Rachelsee) and slightly acidified (recovering: Prášilské, Großer Arbersee, Kleiner Arbersee, and Laka) lakes and mean (min–max) number of taxa of the studied assemblages in either group of lakes in the four sampling campaigns.

Sampling year	1999		2003		2007		2011	
Lake group	acidic	recovering	acidic	recovering	acidic	recovering	acidic	recovering
pH	4.9 ± 0.3	5.6 ± 0.4	5.0 ± 0.3	5.9 ± 0.5	4.9 ± 0.3	5.4 ± 0.4	5.2 ± 0.4	5.7 ± 0.4
ANC (mmol L^{-1})	−23.5 ± 19.4	4.5 ± 12.8	−7.5 ± 7.0	30.0 ± 28.0	−16.0 ± 11.1	5.3 ± 11.9	−1.3 ± 11.6	19.5 ± 12.6
P_t (µg L^{-1})	5.3 ± 2.7	6.3 ± 0.7	4.0 ± 3.1	7.1 ± 3.3	6.9 ± 5.7	5.9 ± 1.0	7.1 ± 5.6	7.9 ± 1.9
NO_3-N (µg L^{-1})	495 ± 325	276 ± 105	593 ± 485	152 ± 85	597 ± 241	331 ± 240	614 ± 404	520 ± 360
SO_4^{2-} (mg L^{-1})	4.0 ± 0.8	2.6 ± 0.7	3.2 ± 0.5	1.9 ± 0.6	3.0 ± 0.3	2.1 ± 0.5	2.2 ± 0.2	1.5 ± 0.6
Al_t (µg L^{-1})	320 ± 70	138 ± 41	266 ± 91	136 ± 31.9	324 ± 113	165 ± 17	302 ± 84	182 ± 25
Al_p (µg L^{-1})	101 ± 97	52 ± 19	138 ± 116	44 ± 18	102 ± 94	29 ± 16	136 ± 157	46 ± 40
Al_i (µg L^{-1})	204 ± 153	24 ± 19	111 ± 69	9 ± 41	152 ± 78	29 ± 21	133 ± 157	50 ± 21
DOC (mg L^{-1})	2.2 ± 0.9	3.4 ± 0.9	1.6 ± 0.6	3.2 ± 1.3	3.0 ± 1.0	4.8 ± 1.1	3.1 ± 1.0	5.0 ± 0.7
C:P (molar)	661 ± 130	422 ± 158	1483 ± 704	542 ± 838	747 ± 106	430 ± 152	1062 ± 266	301 ± 97
HB (µg C L^{-1})	269 ± 70	105 ± 30	108 ± 80	42 ± 29	22 ± 14	20 ± 8	39 ± 14	14 ± 2
Chl$a$ (µg L^{-1})	11.2 ± 5.7	4.7 ± 1.5	4.6 ± 6.5	8.4 ± 7.5	8.8 ± 5.9	3.7 ± 1.5	10.3 ± 11.1	5.2 ± 2.4
Phytoplankton	19.5 (17–22)	16.3 (14–19)	17.3 (15–22)	14.5 (9–20)	18.3 (18–19)	11.5 (9–14)	18.3 (16–21)	11.8 (11–13)
Ciliates	4.5 (1–7)	11.8 (5–19)	9.0 (4–12)	7.8 (5–12)	9.0 (6–12)	9.3 (3–17)	7.0 (3–9)	11.5 (5–27)
Rotifera	6.8 (6–8)	6.8 (3–10)	6.3 (3–8)	9.8 (6–15)	3.0 (1–5)	6.8 (4–11)	5.8 (5–6)	7.0 (6–9)
Crustacea	2.8 (1–6)	7.5 (6–9)	3.8 (2–5)	8.3 (7–10)	3.5 (2–5)	8.3 (5–11)	5.0 (4–7)	7.5 (4–8)
EPT	9.7 (8–11)	12.0 (9–15)	11.3 (10–13)	12.0 (9–15)	10.7 (9–13)	11.5 (7–16)	13.7 (9–17)	15.5 (13–18)
Nepomorpha	2.7 (2–4)	7.0 (6–8)	2.7 (2–4)	6.0 (6–6)	5.7 (4–7)	8.0 (8–8)	7.3 (6–9)	11.5 (11–12)

ANC – acid neutralising capacity; P_t – total phosphorus; Al_t, Al_p, and Al_i – total reactive, particulate, and ionic aluminium, respectively; DOC – dissolved organic carbon; C:P – carbon to phosphorus ratio in seston; HB – heterotrophic microbial biomass in plankton; Chl$a$ – concentration of chlorophyll a; EPT – sum of Ephemeroptera, Plecoptera, and Trichoptera.

3.3 Finland: Trends in chemical recovery of acidified lakes and subsequent responses of perch and roach populations

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Introduction

The Finnish lake survey and Finnish fish-status surveys indicated that 4900 small headwater lakes suffered from acidic deposition and 1600–3200 roach (*Rutilus rutilus*) and perch (*Perca fluviatilis*) populations were affected or extinct by the end of 1980s (Forsius *et al.* 1990, Rask *et al.* 1995a). Sulphate deposition has been the major driving force to the anthropogenic acidification of acid-sensitive surface waters in Finland and elsewhere in Europe (e.g. Prechtel *et al.* 2001, Skjelkvåle *et al.* 2001, Wright *et al.* 2005, Garmo *et al.* 2014). Since the late 1980s, successful sulphur emission reduction measures in Europe have resulted in decrease of acid sulphate deposition and induced a chemical recovery of acidified lakes. The regional recovery of acid-sensitive Finnish lakes, indicated by decreasing concentrations of sulphate and increasing alkalinity, was first observed in the early 1990s (Mannio and Vuorenmaa 1995). At the same time, the first signs of recovery in affected perch populations were recorded (Nyberg *et al.* 1995, Rask *et al.* 1995b). The chemical and biological recovery of lakes from acidification has been most evident in southern Finland, where lakes were both exposed to the highest sulphate deposition and showed the strongest responses to emission reduction (Vuorenmaa 2004, Vuorenmaa and Forsius 2008). Sulphur deposition in southern Finland has declined by about 60–70% since the late 1980s, while the decline in the nitrogen deposition was about 30–40% (Vuorenmaa 2007, Ruoho-Airola *et al.* 2014).

In this report, we summarize trend patterns in water quality recovery from acidification and subsequent responses of perch and roach populations in selected acid-sensitive monitoring lakes of southern Finland during an almost 30 year period, 1985–2012 (Rask *et al.* 2014). Because the period of our study covers the years of strongest acidification pressure on lakes, we had the opportunity to record the decline of fish populations, some of which became extinct. Further, after the onset of the chemical recovery of the lakes, we could detect the start and follow the progress of the recovery of fish populations, including changes in fish abundance and growth. Attention was also paid to a comparison in responses of an acid tolerant (perch) and a sensitive (roach) species, including the potential effects of increased organic carbon concentrations on the two species in the later part of the study period.

Monitoring of lake acidification

Water chemistry

The chemical monitoring of air pollution effects in reference lakes in Finland is presently being carried out in 27 lakes (Table 3). In most of the lakes, the monitoring started in 1987 in connection with national Finnish lake acidification survey, conducted as a part of the Finnish Acidification Research Programme (HAPRO, 1985-1990), and since 1990 the lakes have been monitored regularly using a seasonal sampling strategy (samples in winter, spring, summer and autumn). Some lakes were monitored once a year during autumn overturn phase in 1990–1999, and the seasonal sampling strategy has been applied in these lakes since 2000. The monitoring lakes are small, headwater or seepage forest lakes, and are distributed throughout the country thus reflecting gradients in deposition, climate and landscape. The lakes are acid-sensitive with low alkalinity and low base cation concentrations, and both clear water lakes and humic lakes are represented in the network.

Table 3. Basic catchment characteristics and mean (1990–2012) alkalinity, pH, non-marine sulphate (xSO_4), sum of non-marine base cations (xBC= Ca+Mg+Na+K) and total organic carbon (TOC) in the 27 Finnish monitoring lakes. The monitoring lakes in Table 3 are sorted by latitude.

Lake	Lat.	Long.	Lake area	Catchment area	Max. depth	Alkalinity	pH	xSO_4	xBC	TOC
			km^2	km^2	m	$\mu eq\ l^{-1}$		$\mu eq\ l^{-1}$	$\mu eq\ l^{-1}$	$mg\ l^{-1}$
Vitsjön	59.9634	23.3158	0.29	0.88	13	56	6.40	118	208	5.5
Simijärvi	60.1677	23.5702	2.06	9.21	43	14	6.08	131	171	3.3
Kattilajärvi	60.3014	24.6160	0.34	2.00	10	33	6.17	123	181	4.7
Iso Lehmälampi	60.3444	24.5990	0.05	0.28	8	-6	5.25	68	76	4.9
Luomijärvi	60.6981	27.1471	1.04	4.86	10	87	6.59	138	280	5.2
Hirvilampi	60.7054	27.9181	0.06	0.37	10	12	5.57	125	146	3.9
Vuorilampi	60.7315	27.9185	0.03	0.30	2	40	5.92	111	181	6.9
Valkea-Kotinen	61.2423	25.0634	0.04	0.22	7	22	5.47	119	197	11.8
Sonnanen	61.2944	26.4147	1.63	4.36	14	75	6.75	78	195	2.5
Kangastakunen	61.6127	25.1221	0.42	1.74	15	47	6.14	89	181	8.0
Iso-Hanhijärvi	61.7991	24.2811	0.31	4.88	12	16	5.27	56	140	17.2
Siikajärvi	61.8102	24.2072	0.90	4.01	22	20	5.85	75	116	5.0
Koverojärvi	62.0178	23.9304	1.39	22.3	51	61	6.25	88	195	7.2
Kangasjärvi	62.1662	22.0038	0.47	1.82	7	6	5.70	70	74	2.4
Iso-Soukka	62.3810	25.4449	0.56	8.60	5	79	6.37	45	191	11.1
Kiminginjärvi	62.6104	24.7529	1.74	9.12	2	59	6.38	25	140	9.2
Pieni Hietajärvi	63.1638	30.6929	0.02	0.76	7	85	6.10	12	159	11.8
Iso Hietajärvi	63.1615	30.7103	0.83	4.64	9	90	6.80	26	144	4.4
Kakkisenlampi	63.6565	29.9466	0.19	0.79	18	-6	5.31	36	31	2.6
Älänne	63.7268	28.2352	4.55	17.1	12	41	6.44	40	124	6.6
Takkajärvi	64.9526	28.2152	1.13	7.49	23	93	6.53	32	175	6.3
Suopalampi	67.0655	26.0946	0.39	4.20	5	44	6.31	20	98	6.9
Vasikkajärvi	67.1170	26.0844	0.24	1.56	17	7	5.97	31	38	1.6
Sierramjärvi	69.1847	26.8978	1.08	5.32	19	128	7.08	44	187	2.3
Lampi 3/88	69.4486	29.1382	0.05	0.20	4	11	6.14	38	54	1.7
Lampi 222	69.4471	29.1036	0.24	2.22	22	64	6.82	43	118	1.9
Vuoskojärvi	69.7426	26.9499	0.17	1.78	7	146	7.08	30	197	5.2

Lakes can be characterized as pristine or semi-pristine with no or only minimum direct human disturbances in the catchment. In this report, the main emphasis is on parameters reflecting changes in acidification status and chemical recovery: alkalinity (measured, Gran method) and pH, charge-balance Acid Neutralizing Capacity (calculated, ANC_{CB}), non-marine sulphate (xSO_4), non-marine base cations xBC = (xCa + xMg + xNa + xK), nitrate-nitrogen (NO_3-N) and total organic carbon (TOC). For comparability between the study lakes and between the decades, samples taken only in the autumn overturn phase were used in the statistical trend analysis for water chemistry. The results of chemical monitoring given in this report refer to samples taken at 1 meter depth.

Fish populations
The Finnish Game and Fisheries Research Institute and the Finnish Environment Institute started an integrated monitoring of water chemistry and fish populations in acidified lakes in 1990 (Rask *et al.* 1995b, Nyberg *et al.* 2001, Tammi *et al.* 2004, Rask *et al.* 2014). The regular fish monitoring in these acidified lakes was started in 1985, at the beginning of the HAPRO programme. Twelve of these lakes were included in the present biological assessment representing the following four levels (three study lakes in each level) of fish community response to acidification in small Finnish lakes according to Rask *et al.* (1995a): (i) Perch extinct (pH<5; Al_{lab} 80–280 $\mu g\ l^{-1}$), (ii) Perch affected (pH 4.8–5.5; Al_{lab} 50–160 $\mu g\ l^{-1}$), (iii) Roach extinct (pH 5.2–6.0; Al_{lab} 25–135 $\mu g\ l^{-1}$) and (iv) Roach affected (pH 5.3–6.4; Al_{lab} 5–40 $\mu g\ l^{-1}$).

In lake group 1 "perch extinct", according to sediment diatom analyses rapid acidification took place in the 1960s (Tolonen and Jaakkola 1983, Tulonen 1985). In lake group 2 "perch affected", rapid acidification started between the 1950s and 1960s (Tolonen and Jaakkola 1983, Tolonen *et al.* 1986). In lake group 3 "roach extinct", no paleolimnological records on the acidification history are available, but

according to calculations on pre-acidification alkalinity (Kämäri 1985), these lakes were also acidified. In L. Kattilajärvi of group 4, a half pH unit decrease to level of 5.5 was set to the 1960s (Liukkonen 1989). Fish were sampled at a three-year intervals with gill net series of eight 1.8 x 30 m nets of mesh sizes 12–60 mm in 1985–1992 (Raitaniemi et al. 1988). NORDIC multimesh survey nets (1.5 x 30 m; 12 panels with mesh sizes 5-55 mm, CEN 2005) were used since 1995. For comparability, net panel area and selectivity corrections were calculated according to Tammi et al. (2004) and the catches from mesh sizes 5, 6.25, 8 and 10 mm of NORDIC nets were excluded in the analyses. The detailed descriptions of the methods are presented in Rask et al. (2014). CPUE (number of fish in one net in one night), length frequency distribution of fish, mean weight, and growth patterns of fish were the key parameters in illustrating the responses of perch and roach population to the recovery of the lakes from acidification.

The monitoring of fish populations and water chemistry were carried out until 2007. In four of the lakes belonged to this monitoring group, chemical monitoring has continued after 2007 including L. Iso Lehmälampi (group 1), , L. Simijärvi (group 3), L. Kattilajärvi and L. Vitsjön (group 4) (Tables 3 and 4).

Results and discussion

Chemical recovery

The observations of the present study provide good evidence that emission reduction measures led to regional-scale recovery from acidification in sensitive ecosystems in Finland. A significant ($p < 0.05$) decreasing trend in lake water sulphate (xSO_4) concentrations was observed in all of the study lakes in 1990–2012, and significant increasing trend in alkalinity (measured) and ANC_{CB} (calculated) occurred in 90% and 70% of the monitoring lakes, respectively (Table 4). Along with increasing buffering capacity, there was in general a decrease in acidity from the early 1990s, indicated by decreasing hydrogen-ion (H^+) concentrations (increase in pH), being significant in ca. 40% of the lakes. Base cation (xBC) concentrations are still declining in the lakes, but presumably to a lesser extent than sulphate. The less steep decline of lake water base cation concentrations compared to that of sulphate resulted in improved acid-base status of soils and has led to increased buffering capacity of the lakes. Low pH is associated with high concentrations of labile aluminium, and a decrease in acidity is also reflected in decreasing labile aluminium concentrations in recovering lakes (Vuorenmaa and Forsius 2008, Rask et al. 2014). Nitrate (NO_3-N) concentrations have also decreased in most of the study lakes, but trends were rarely statistically significant. According to the earlier assessments, NO_3-N plays only a minor role in the acidity status and as an acidifying agent in Finnish lakes (Mannio 2001a, b, Vuorenmaa and Forsius 2008, Rask et al. 2014), and inorganic nitrogen concentrations and exports in boreal Finnish forest streams did not change, or showed a slight decrease in concentrations during the 15–28 years (Sarkkola et al. 2012). This is also the case elsewhere in Europe and North America where only a few significant trends in NO_3-N concentrations were detected, but concentrations were decreasing rather than increasing (Garmo et al. 2014).

In conditions of decreasing minerogenic acidification, increased catchment-derived organic acidity has become proportionally more important in affecting recovery process of sensitive lakes in Finland (Vuorenmaa and Forsius 2008). Increasing total organic carbon (TOC) concentrations throughout the 1990s and the 2000s, as observed in Norway, Sweden, UK and North America can also be recorded in small forest lakes and streams in Finland (Vuorenmaa et al. 2006, Monteith et al. 2007, Sarkkola et al. 2009) and also in the present study in 1990–2012. A significant ($p < 0.05$) increasing trend in TOC concentrations occurred in 81% of the monitoring lakes. Several studies have suggested that the principal driver for the regional increase in TOC (DOC) concentration was the recovery from acidification, i.e. a decrease of sulphate deposition and an increase of pH in the soil, and a consequent increase of the solubility of DOC in the soil (e.g. Monteith et al. 2007). However, both production and transport of DOC are strongly influenced by climate, and the increases are also linked, in part, e.g. to variations in runoff (e.g. Erlandsson et al. 2008). Elevated TOC concentrations in the study lakes were observed particularly between 2004 and 2008, due to rainy summers and autumns. For example in the ICP Waters lake L. Vitsjön, located in south Finland, the extreme hydrological conditions resulted in 5–6 years decline of buffering capacity (Figure 30). Organic acid surges, together with diluted and low-buffered runoff water, can be an important factor suppressing recovery of pH and alkalinity in sensitive Finnish lakes (Wright

et al. 2006, Vuorenmaa and Forsius 2008, Nyberg et al. 2010, Rask et al. 2014). This highlights the importance of climatic and hydrological variability in affecting the recovery process.

Table 4. Trends of key chemical acidification variables during 1990–2012 in the 27 Finnish monitoring lakes.   For the annual change (µeq l-1 yr-1 or mg l-1 yr-1), a statistically significant trend (Mann-Kendall test, Theil-Sen slope) are indicated with asterisks (*** p < 0.001, ** p < 0.01, * p < 0.05).

Lake	Alkalinity	ANC_{CB}	H^+	xSO_4	xBC	NO_3-N	TOC
	µeq l^{-1} yr^{-1}	µeq l^{-1} yr^{-1}	µeq l^{-1} yr^{-1}	µeq l^{-1} yr^{-1}	µeq l^{-1} yr^{-1}	µeq l^{-1} yr^{-1}	mg l^{-1} yr^{-1}
Vitsjön	2.33***	2.67***	-0.11**	-5.26***	-2.67***	-0.01	0.12***
Simijärvi	1.29***	0.83	-0.05**	-2.69***	-2.31***	-0.45***	0.07***
Kattilajärvi	0.83***	1.00	-0.02*	-4.24***	-3.57***	-0.00	0.08***
Iso Lehmälampi	1.07***	0.56	-0.26*	-2.75***	-2.29***	-0.04	0.10**
Luomijärvi	1.20**	1.88**	-0.00	-4.19***	-2.58***	0.06**	0.12***
Hirvilampi	1.42***	2.79***	-0.15**	-5.00***	-2.47***	0.03	0.17***
Vuorilampi	1.45*	2.33**	-0.03	-4.50***	-2.00**	0.01	0.29***
Valkea-Kotinen	1.64***	1.50**	-0.14**	-2.91***	-1.20***	-0.02	0.15**
Sonnanen	1.53***	1.59***	-0.00	-2.33***	-0.86**	0.00	0.04*
Kangastakunen	1.92***	1.75***	-0.01	-2.00***	-0.24	-0.01	0.12**
Iso-Hanhijärvi	1.00**	1.90***	-0.10	-1.86***	-0.00	-0.06*	0.31*
Siikajärvi	1.50***	1.30***	-0.08***	-2.18***	-0.90***	0.03	0.08***
Koverojärvi	1.60***	2.00***	-0.01	-2.42***	-0.50*	0.00	0.08**
Kangasjärvi	0.60***	1.36***	-0.13**	-2.33***	-0.86***	-0.05*	0.06***
Iso-Soukka	3.80**	3.13***	-0.19*	-1.50***	1.29**	0.00	0.04
Kiminginjärvi	0.60	0.47	0.00	-0.57**	-0.07	0.00	0.10*
Pieni Hietajärvi	0.13	1.14	-0.01	-0.73***	0.36	-0.01**	0.18
Iso Hietajärvi	0.94*	1.67***	-0.01	-0.08***	0.53	-0.01*	0.07**
Kakkisenlampi	1.00***	1.00**	-0.27*	-1.38***	-0.65**	-0.02	0.12***
Älänne	0.64***	1.11**	-0.01	-1.29***	-0.31	-0.00	0.06*
Takkajärvi	0.60**	1.07**	-0.00	-1.06***	0.00	0.01	0.06**
Suopalampi	0.67**	0.50*	-0.01	-0.63***	-0.17	-0.00	0.12
Vasikkajärvi	0.52**	0.56*	-0.02	-1.00***	-0.43**	-0.00	0.04**
Sierramjärvi	0.94***	0.80*	0.00	-0.56***	0.22	-0.00	0.00
Lampi 3/88	0.80***	0.75**	-0.00	-1.20***	-0.71*	-0.00	0.04***
Lampi 222	0.77***	-0.22	0.00	-0.78***	-1.00*	-0.00	0.04**
Vuoskojärvi	0.38	0.33	-0.00	-0.75***	-0.38	-0.00	0.03
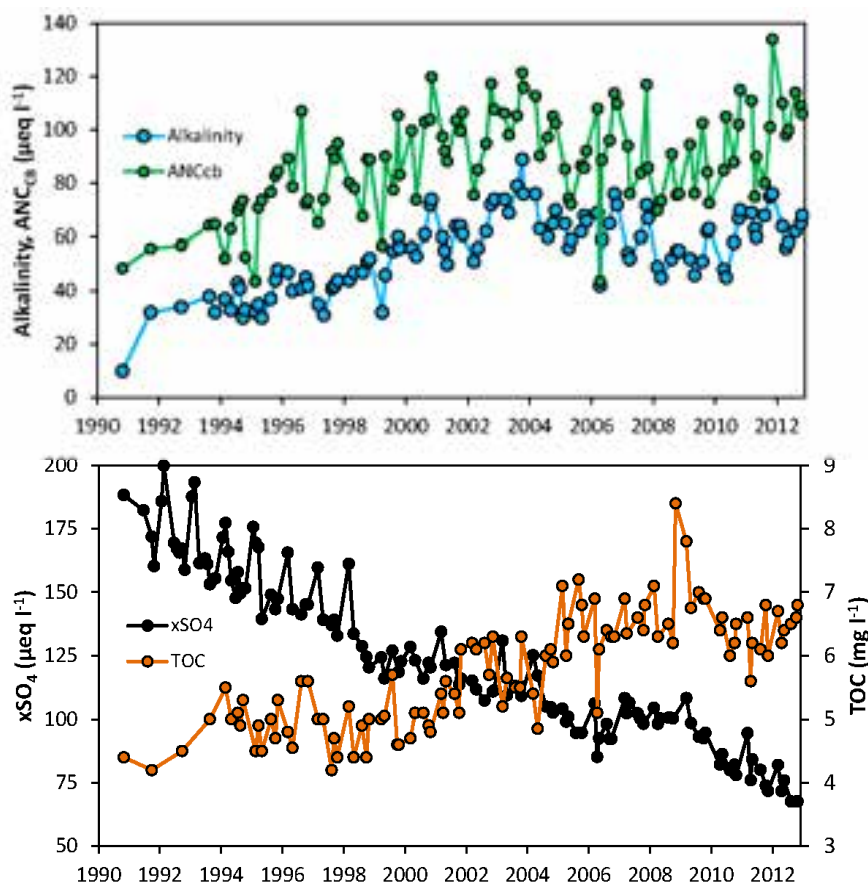
Figure 30. Alkalinity (measured) and charge-balance Acid Neutralizing Capacity (ANCCB, calculated) (upper)   and non-marine sulphate (xSO4) and total organic carbon (TOC) concentrations (lower) in L. Vitsjön in 1990–2012.

Recovery of perch and roach populations

Perch, an acid-tolerant species, was responding rapidly to improved water quality conditions, and the first signs of population recoveries were detected in the early 1990s as appearance of new strong year-classes of perch (Nyberg *et al.* 1995, Rask *et al.* 1995b). According to the observations and trends of the present study, the structure of affected perch populations has become normal during the monitoring period. Successful re-establishment of disappeared perch populations into previously heavily acidified lakes (lake group 1) further emphasizes the importance of the chemical recovery of the lakes (Nyberg *et al.* 2010). Our growth data of perch from L. Orajärvi, including samples from year-classes 1964–2006, a time period of more than 40 years, illustrates the entire history of acid rain impacts on the fish population of a single lake (Nyberg *et al.* 1995, Rask *et al.* 2014). Although we had data only from a few perch born in the 1960s, their slow growth suggests that the population density in those years was high or normal with no impacts of acidification on reproduction. This is also supported by paleolimnological records, suggesting that a rapid acidification of the lake started in the 1960s (Tolonen and Jaakkola 1983). The striking increase of growth towards the late 1980s is an excellent indication of flexibility of the perch in changing circumstances, in this case to declining population density due to acidity-induced reproduction failures in the 1970s and the 1980s. It is worth attention that the fast growth rate was possible with an invertebrate diet, for example *Asellus aquaticus*, as no fish were available (Raitaniemi *et al.* 1988). The sharp decrease of growth after the onset of recovery and new strong year-classes further emphasizes the density dependent growth pattern of perch.

For the more acid-sensitive species roach, little if any recovery of affected populations was recorded. Neither the abundance nor the growth of roach showed significant responses to the chemical recovery of

the study lakes. This may indicate that water chemistry is still critical for the success of roach populations, and evidently more time and more suitable water quality are needed for a distinct recovery. The appearance of some stronger year-classes of roach in L. Vitsjön in the late 1990s, following the weak or missing ones between mid-1980s and mid-1990s, looked like an onset of recovery, but the year-classes of the 2000s remained scarce again. Compared to other studies from small Finnish lakes (Raitaniemi and Rask 1990, Rask and Tuunainen 1990, Estlander et al. 2010), the growth of roach in L. Vitsjön has been faster throughout the study period, suggesting that there has been lower intraspecific or interspecific food competition.

Perch is a generalist carnivore feeder, usually shifting from zooplankton to zoobenthos and further to fish during its life span (e.g. Horppila *et al.* 2000). Roach is an omnivore feeding on zooplankton and zoobenthos and later turning to detritus and macrophytes (Horppila *et al.* 2000, Estlander *et al.* 2010). Thus, these species have a competitive interaction when feeding on zooplankton in their first years. As perch is a visual feeder, the light conditions are of importance, and clear water favours perch (Estlander *et al.* 2012). Moreover, large perch can prey on roach, which may delay the recovery of a roach population in conditions of critical water quality like in the case of L. Vitsjön. In poor light conditions, decreased feeding efficiency of perch has been shown (e.g. Estlander *et al.* 2012), as well as dominance of roach over perch in competition for zooplankton food in the turbid water of eutrophicated lakes (e.g. Olin *et al.* 2002) or in highly humic lakes (Estlander *et al.* 2010, Olin *et al.* 2010). Consequently, the recent increasing trend in organic carbon load, resulting in increased TOC and water colour of lakes, could favour roach if the acidity – either minerogenic or organic – did not disturb the reproduction of roach.

To conclude, the chemical recovery of the study lakes was clearly followed by biological recovery, especially of perch populations. Thus, the present findings indicate success in the ultimate intention of the emission abatement policy. However, the biological recovery or biological changes due to declined acid deposition and expected improvement in water quality is difficult to predict because of the complex interaction between the many climatic, chemical and biological variables involved. The findings of the present study emphasize the importance and value of integrated monitoring approaches including physical, chemical and biological variables, and the suitability of small headwater lakes for such monitoring.

3.4 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 dates back to the early 1980s. During the period of study, the concentrations of sulphate in precipitation at the various sites decreased with 72 to 90 % in south Norway (Aas et al. 2014). Similarly, the nitrate and ammonium concentrations in precipitation show significant decreases 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.

The regional benthic surveys in running waters include the monitoring of five running water localities. This monitoring was started in 1981 with the monitoring of two adjacent catchments in the Farsund area in southern Norway. During the next three years, four other rivers were added to the monitoring network. The rivers have been sampled 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). These data represent some of the longest time-series of benthic invertebrate data in the world. They make

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 (un-acidified).

Results and discussion
The development of the acidification score in the different rivers (Figure 31) shows that the acidification damage at all sites was significantly reduced compared to the state at the start of the monitoring. In 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 is a consequence of the fact that reductions in sulphur emissions and deposition in Europe show the same trends.
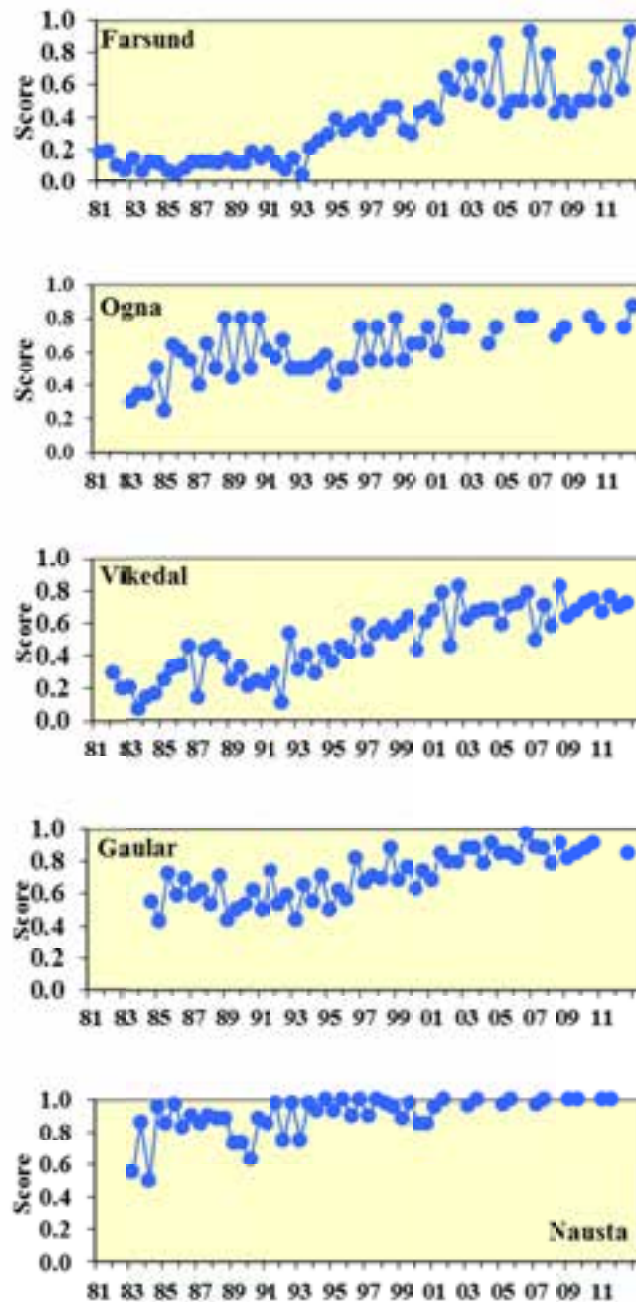
Figure 31. Acidification score for invertebrates in the monitored rivers during 1981 − 2013 ranged from south (top) to north (bottom). The score is described by Fjellheim and Raddum (1990).

The most pronounced recovery was observed in the southernmost sites, for example, Farsund and Vikedal. This is further outlined by Fjellheim and Raddum (2001) and Hesthagen et al. (2011). Populations of benthic invertebrates in these areas were heavily damaged at the start of the monitoring. Benthic invertebrates in river Nausta, the northernmost site, were initially less damaged. However, a significant recovery has also taken place here (Halvorsen et al. 2003).

The fauna in the localities of this study show increased species diversity in response to the improved water quality. The succession is mainly due to colonisation of acid sensitive species. In the following we will give some examples.

The development of sensitive taxa in the Farsund area has been very positive since 1990. In 2010 we documented nine sensitive taxa compared to three in 1990. The increased biological diversity is documented by an increased EPT diversity (Figure 32, for explanation of EPT se caption of Figure 32).
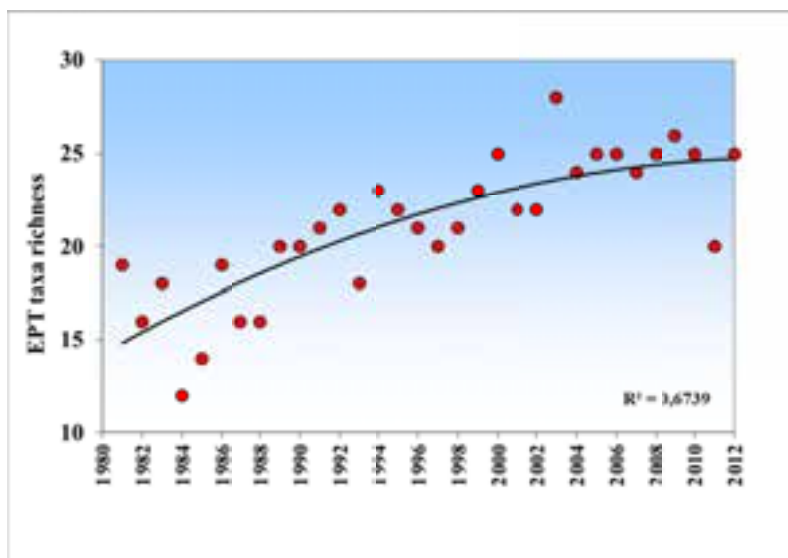
Figure 32. Development of the EPT taxa richness in the Farsund area 1981 - 2012. This index is calculated by the sum   of species within the insect groups Ephemeroptera (mayflies), Plecoptera (stoneflies) and Trichoptera (caddisflies).

The caddisflies *Hydropsyche siltalai* and *Wormaldia sp.* are examples of sensitive species that colonised running water localities at Farsund (Figure 33). Their response seems to be closely related to the substantial improvement in water quality after 1996, with increased lake 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 strongly 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 years with unusually high deposition as well as confounding factors such as sea salt episodes (Raddum et al. 2007). The water quality in Farsund is still too unstable to permit a permanent establishment of this species. Moderately acid-sensitive species, however, show stable populations.
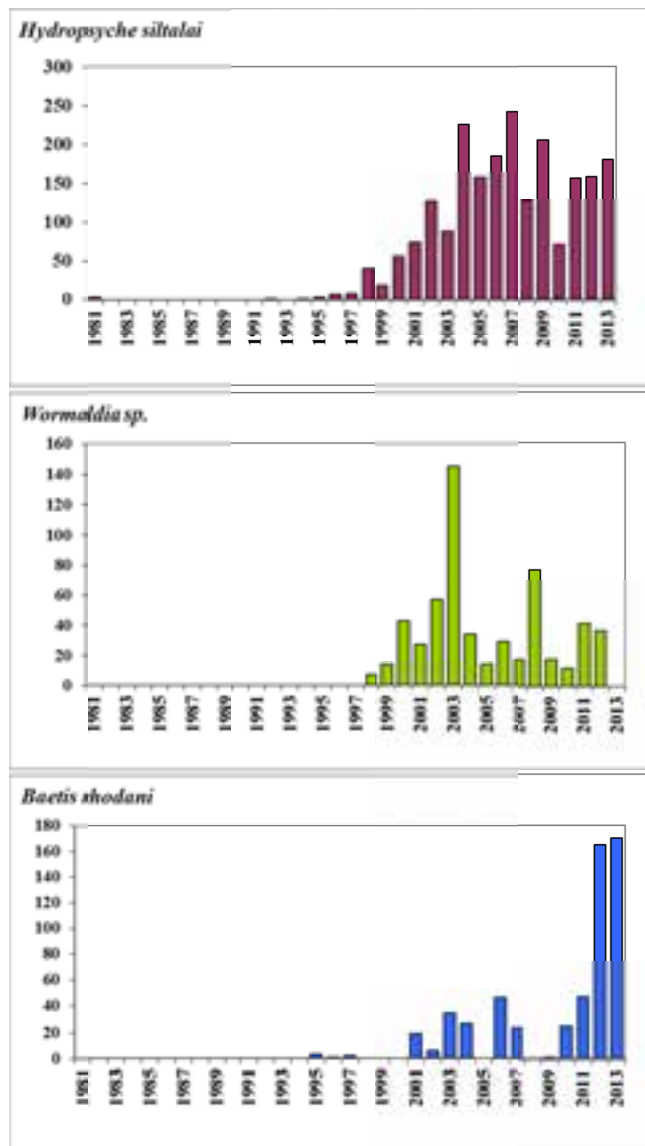
Figure 33. Total number of the caddisflies Hydropsyche siltalai and Wormaldia sp. and the mayfly Baetis rhodani in the Farsund area during 1981-2012.

In River Vikedal lime dosers have been operated since 1987, to secure acceptable water qualities for Atlantic salmon and sea trout in the lower, anadromous, part. The ICP Waters monitoring of benthic fauna is performed in the upper, unlimed river. Results from this monitoring show that the fauna has recovered to some extent. The area must still be considered affected by acidification. Several normally abundant benthic animal species are still missing at many localities. The biological diversity of the localities will increase if water quality improves. The situation in recent years shows a positive development with reduced acidification damage and increased biological diversity (Figure 34). The development of the strongly sensitive mayfly *B. rhodani* in the lowermost unlimed section is an example of this (Figure 35). 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.

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.
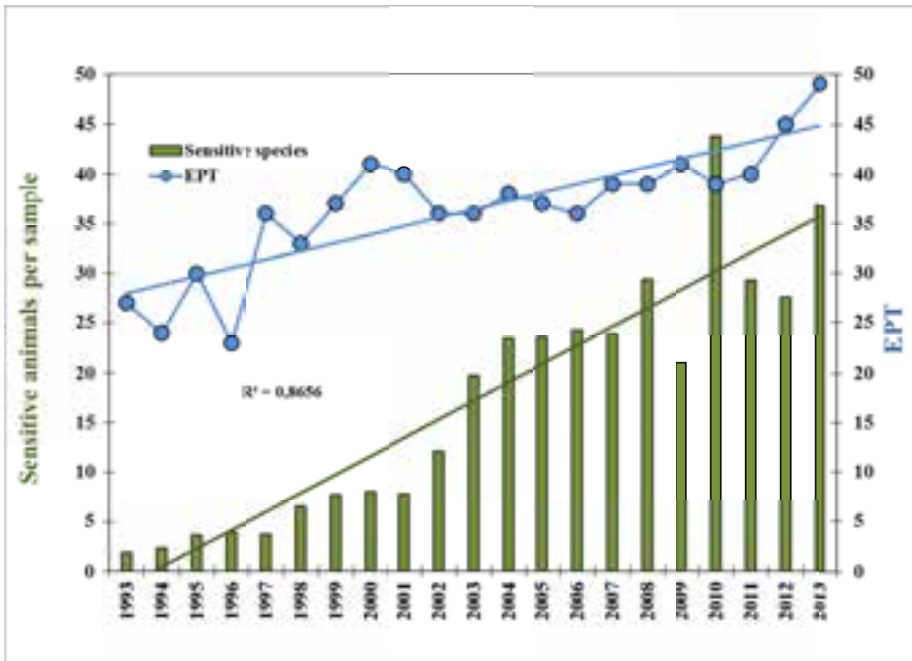
Figure 34. Number of sensitive animals per sample and EPT taxa richness in the upper unlimed part of River Vikedal in the period 1993 - 2010.
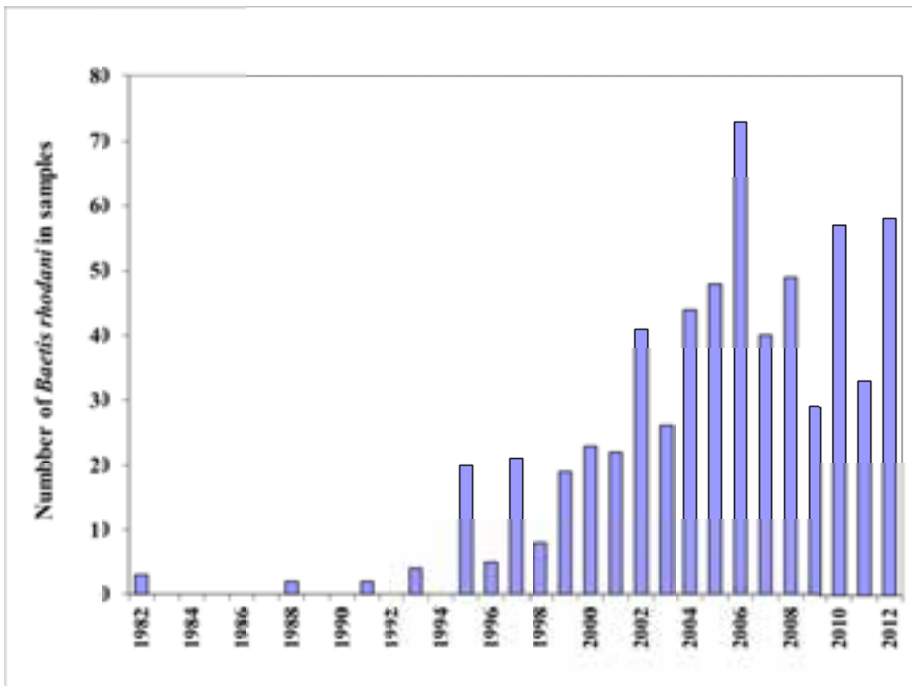
Figure 35. Numbers of the acid-sensitive mayfly Baetis rhodani in an unlimed locality in the main River Vikedalselva during 1982-2010.

The positive development of the communities of acid-sensitive animals is related to reduced air pollution resulting from the LRTAP international emission protocols from 1979 and onwards. Thus the changed

benthic invertebrate communities in formerly strongly acidified regions of South Norway represents one of many positive signals that reduced emissions of pollutants are favourable to nature.

3.5 Switzerland: Benthic invertebrate trends in alpine acid-sensitive lakes

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Introduction
The subalpine and alpine areas of north-western Italy and southern Switzerland (Ticino region), in the Central Alps are affected by long-range transport of pollutants from the plain of the River Po, in Italy, one of the most urbanized and industrialised areas of Europe (Rogora et al. 2006). Because of the presence of slowly-weathering rocks in their catchments and of the limited buffer capacity of their waters many high-altitude lakes in southern Switzerland are sensitive to acidification (Mosello et al. 1992; Marchetto et al. 1994, 1995). During the last 30 years the area has been affected by a substantial decrease in sulphate deposition, while deposition of oxidized nitrogen and to a minor extent reduced nitrogen started to decrease only recently (Steingruber and Colombo 2010a; Steingruber and Colombo, 2007-2014). In response to decreasing acid deposition, lakes in the study area have shown signs of chemical recovery from acidification since the mid-1990s (Steingruber and Colombo 2010b; Steingruber and Colombo 2007-2014; Rogora et al. 2013). Because the ultimate goal of emission control programmes is biological recovery, sampling of invertebrates in 4 acid-sensitive lakes was added to the monitoring programme in the year 2000. Temporal trends in community composition are presented here.

Methods
Benthic invertebrates samples were collected by "kicksampling" according to the ICP Waters Manual (ICP Waters Programme Centre, 2010) from the littoral and the emissary of Laghetto Inferiore (INF), Laghetto Superiore (SUP), Lago di Tomè (TOM) and Lago del Starlaresc da Sgiof (STA).
Before 2000 the 4 lakes were sampled irregularly. From 2000 to 2005 they were sampled twice a year (once at beginning of summer, once in autumn), and after 2006 three times a year (once at beginning of summer, twice in autumn). Geographic and morphologic lake characteristics, measured parameters and analytical methods are described in Steingruber and Colombo (2007-2014).

Temporal trends are shown for the relative abundances of the most important taxonomic groups (oligochaeta, chironomidae, EPT taxa). In addition, the total number of taxa, the number of EPT taxa and the number of acid sensitive taxa (according to the literature) are presented. In order to avoid differences in the taxa number caused by different identification levels used through time, for each taxonomic group a taxonomic identification level was defined and the results filtered through. The identification levels were the following: Annelida -> class, Arachnida -> subcohort, Coleoptera -> genus, Diptera -> family, Ephemeroptera -> genus, Heteroptera -> genus, Megaloptera -> genus, Odonata -> genus, Trichoptera -> genus, Mollusca -> class, Plathelminthes -> family. Moreover, since the sample sizes varied greatly from year-to-year and the number of taxa/species increases with the number of individuals, the yearly number of taxa were standardized. For each sampling site a potential regression was calculated between the annual total number of taxa and the annual number of sampled individuals. With this function for each year the number of taxa were standardized to a sample size of 1000 individuals.

Results and Discussion

Chemical trends

All 4 lakes showed signs of recovery during the last 30 years with decreasing concentrations of sulphate and base cations and increasing alkalinity, pH and concentrations of labile aluminium (*Figure 1*). However, in the period since the start of the invertebrate monitoring in 2002, autumn pH and alkalinity have increased significantly only in STA and SUP. In STA pH increased from 5.2 to 5.9 and alkalinity from -11 μeq L^{-1} to 8 μeq L^{-1}. In SUP autumn pH increased from 6.3 to 6.7 and alkalinity from 29 μeq L^{-1} to 33 μeq L^{-1}. At present, in the other 2 lakes autumn pH and alkalinity are 6.7 and 32 μeq L^{-1} in INF and 5.7 and 4 μeq L^{-1} in TOM. However, during spring pH and alkalinity can drop to lower values (data not shown). During the last 3 years minimum spring pH and alkalinity were 6.1 and 13 μeq L^{-1} in INF, 6.5 and 10 μeq L^{-1} in SUP, 5.7 and 3 μeq L^{-1} in STA and 5.4 and -3 μeq L^{-1} in TOM. Interestingly, although TOM is more acidic than STA, its concentrations of labile aluminium are lower (STA: 50 μg L^{-1}, TOM: 20 μg L^{-1}).

Invertebrate trends

Chironomidae are at present the most important taxonomic group in the studied lakes both in the littoral and in the outlet. Their relative importance increased during the monitoring period in INF, SUP and TOM and decreased in STA (Figure 37). In the outlets of INF and SUP the increase of Chironomidae was compensated by a decrease of Oligochaeta and EPT taxa (mainly *Nemoura sp.*) and in TOM only by EPT taxa (mainly *Leuctra sp.*). In the littoral the relative abundance of EPT taxa is extremely low and the increase in Chironomidae was compensated only by a decrease of Oligochaeta. In contrast, in the outlet of STA the decrease in Chironomidae seemed to be occasionally slightly compensated by an increase of EPT taxa (mainly the plecopterian *Nemoura sp.*). However, more important is the recent increase in Simuliidae (data not shown). In the littoral of STA Chironomidae were compensated by Ceratopogonidae.

Numbers of total, EPT and acid sensitive taxa in the outlets were higher in the less acid sensitive lakes INF and SUP compared to TOM and STA (Figure 38). In particular, acid sensitive taxa were only found in INF and SUP, which is not surprising since pH of TOM has always been below 6 and of STA only recently occasionally reached values slightly above 6. In the littoral, number of total, EPT and acid sensitive taxa are not significantly different between the lakes. The near-absence of acid sensitive taxa in the littoral is also not surprising since most known acid sensitive taxa prefer lotic waters. The total number of taxa slightly decreased in most sampled lake outlets and littoral. The number of EPT and acid sensitive taxa also decreased in the less acid sensitive outlets of INF and SUP, while they remained fairly constant at the other sites, where they were low from the beginning.

In summary, acid sensitive indicators like the relative abundance of EPT and acid sensitive taxa (data not shown) and the standardized number of total, EPT and acid sensitive taxa generally have not increased since the monitoring of invertebrates began in 2002. Only the relative abundance of EPT taxa in the outlet of STA increased, but this was caused by an increase of the relative abundance of the acid tolerant *Nemoura sp.* This means that the observed chemical recovery in SUP and STA did not influence their biology. In SUP pH at the beginning of the monitoring period was probably already too high (6.3) and the increase in pH too small to influence significantly the population of invertebrates. In STA biological recovery may be delayed in time with respect to chemical recovery and first signs may appear soon or pH has first to clearly pass the pH 6 limit. However, it must also be added that the generally poor invertebrate fauna of alpine mountain lakes inhabited mainly by chironomids and oligochates makes it not easy to identify changes in population without determining these taxa to deeper taxonomic levels.

Instead of biological recovery, most of the trends here observed seem even to suggest a worsening of lake water quality (increasing %Chironomidae and decreasing %Oligochaeta, %EPT and number of total and EPT taxa). It is also true that the most important indicators of acidity here studied are the relative abundance and the number of acid sensitive taxa. The first was very small in all lakes both in the littoral and in the outlets and did not change over time (data not shown). The second was relevant only in the outlets of the less acid sensitive lakes INF and SUP, where it decreased. However, the lake water chemistry does not show a worsening of water quality. Interestingly, we noticed that during the monitoring period, between 2002 and 2013, there was first a 6-years period with relatively low

precipitation (mean*2: 1953 mm) followed by a 6-years long period with more abundant precipitation (mean*: 2420 mm). It is therefore probable that during the second period episodic acidification events were more frequent. In fact, although autumn pH during the invertebrate monitoring period slightly increased in all lakes, pH after snow melt at the beginning of summer increased only at STA, while at INF and SUP mean spring pH was higher during the first than during the second period. It is therefore possible that the decrease of the number of acid sensitive taxa in the outlets of INF and SUP was caused by an increase of the number of acidic episodes. The relatively dry period with fewer rain events at the beginning of the monitoring period could have also favoured the diffusion of the oligochaets Naidinae in the outlets of INF and SUP (most important oligochaete family in these samples during 2003 and 2007). Their absence in acidic waters has been reported in Smith et al. (2000), Steingruber et al. (2013) and Dumnicka et al. (2014).

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[2] *Mean precipitation during the period corresponds to the average of annual mean precipitation, calculated over the hydrological year (October to September).
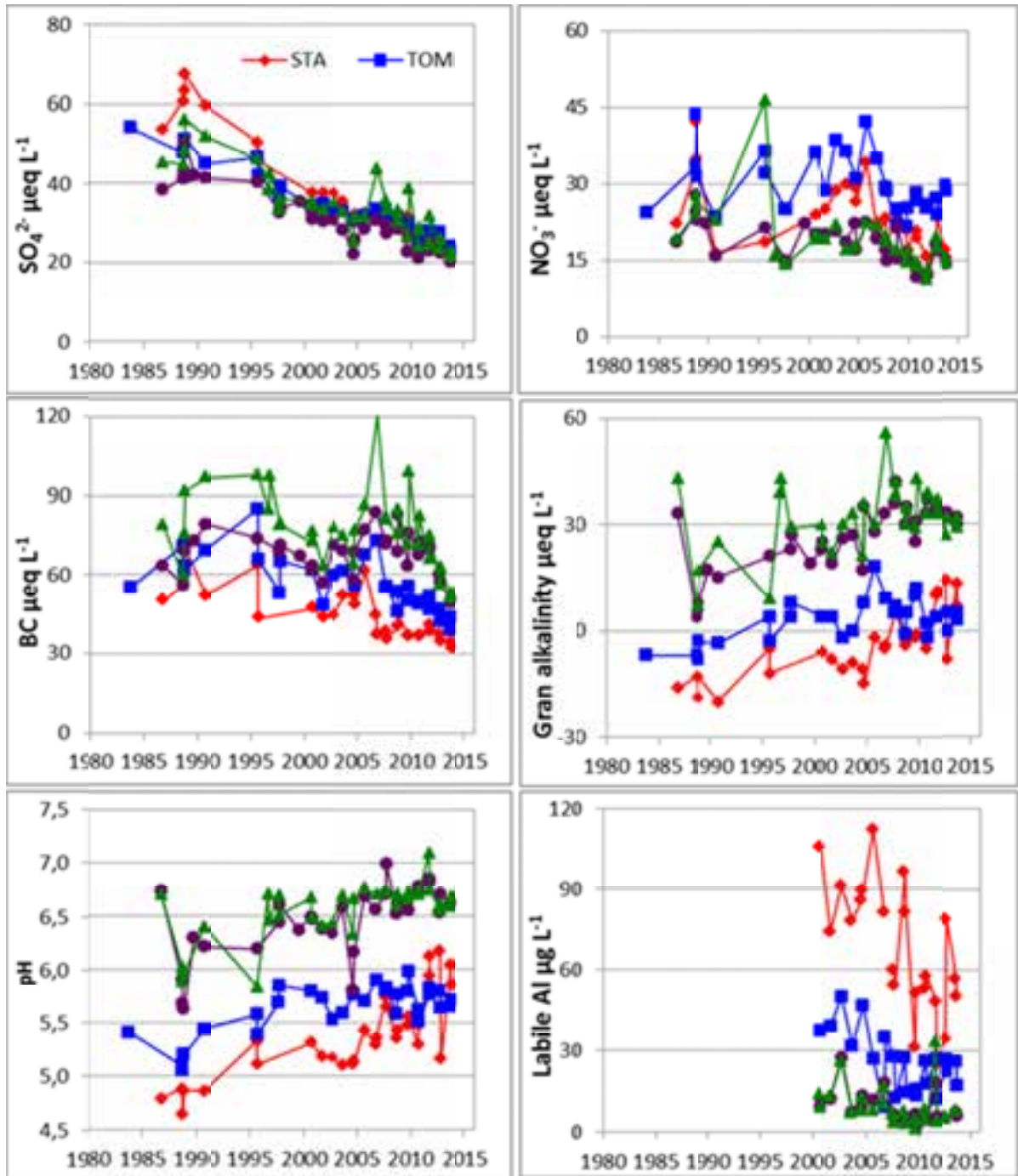
Figure 36. Long-term trends of key chemical parameters in 4 high-altitude acid sensitive lakes: average autumn values are shown.
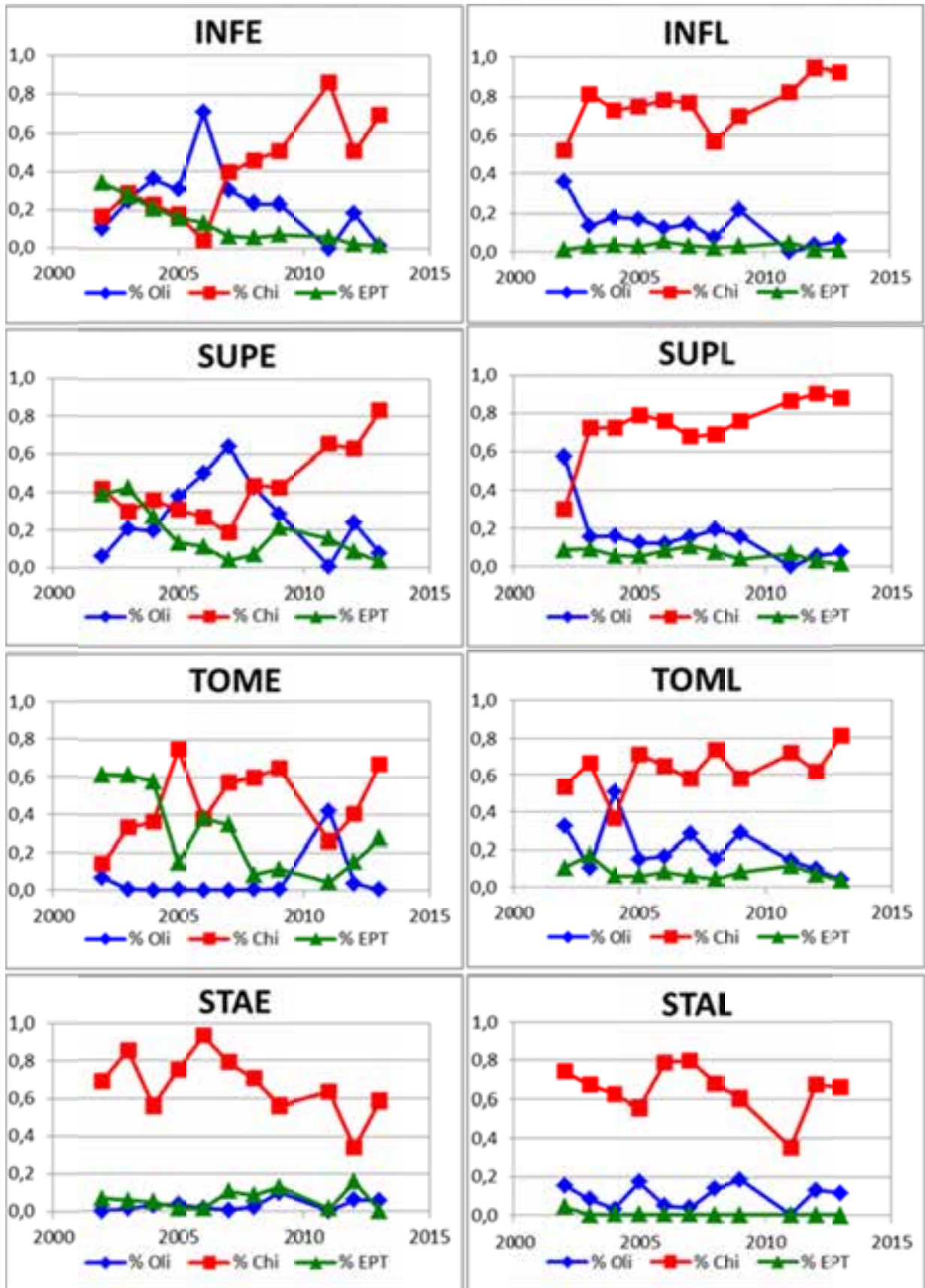
Figure 37. Relative abundances of oligochaeta, chironomidae and EPT taxa in the outlet (INFE, SUPE, TOME, STAE) and the littoral (INFE, SUPE, TOME, STAE) of 4 high altitude Swiss lakes .
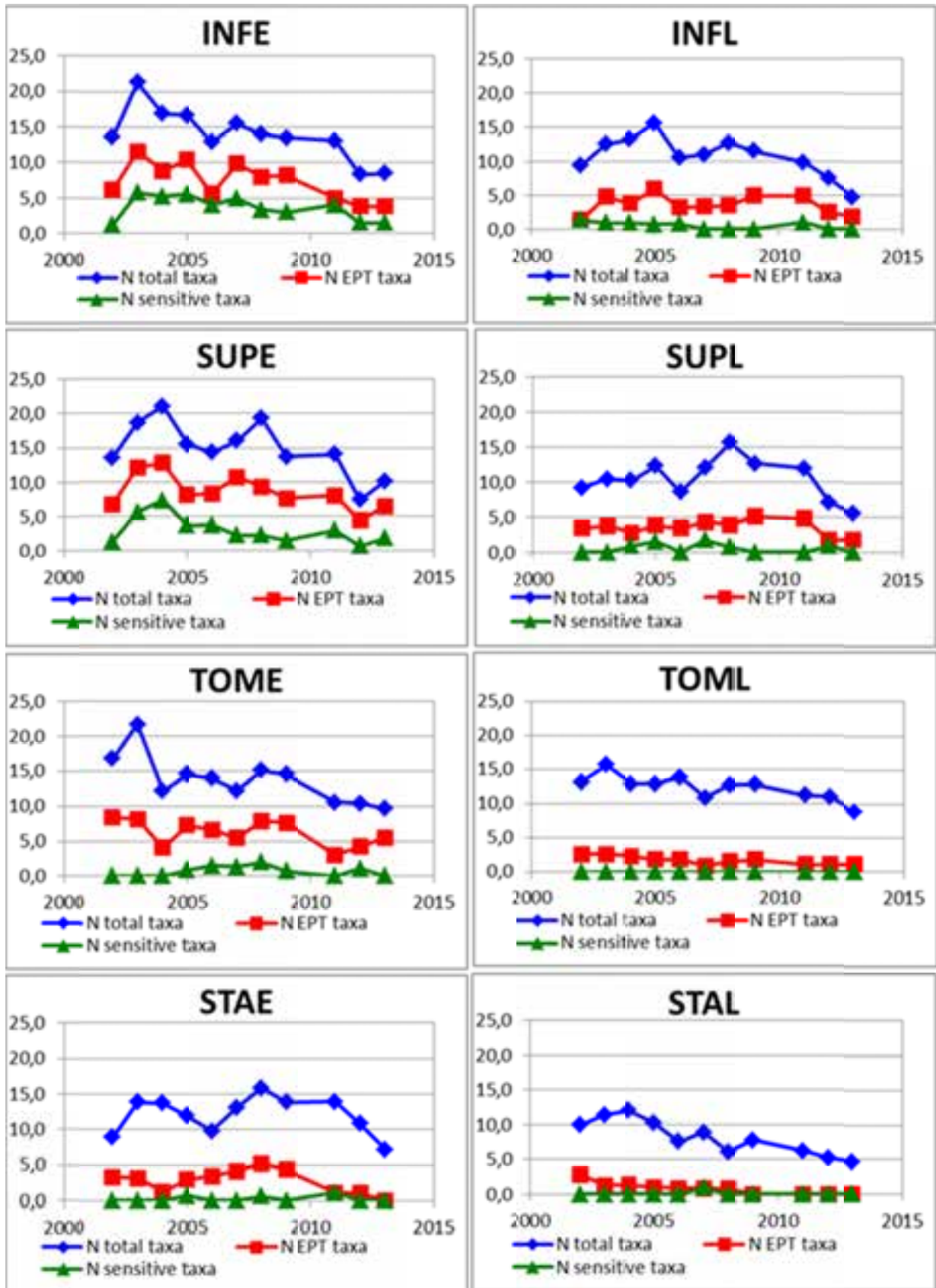
Figure 38. Number of standardized total, EPT and acid sensitive taxa in the outlet (INFE, SUPE, TOME, STAE) and the littoral (INFE, SUPE, TOME, STAE) of 4 high altitude Swiss lakes .

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5. Reports and publications from the ICP Waters Programme since 2000

All reports from the ICP Waters programme from 2000 up to present are listed below. Reports before year 2000 can be listed on request. All reports are available from the Programme Centre. Reports and recent publications are also accessible through the ICP-Waters website; http://www.icp-waters.no/

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Appendix A. Water chemistry data from ICP Waters

Table A. Mean results for samples collected from January 1 2009 to December 31 2011 (n is the number of samples in this time span).

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	TOT N µg/l N	TOT P µg/l P	DOC mg/l C	H+ µEq/l
Belarus	BY01	Berezinsky Biosphere Reserve	14	38.08	7.57	47.36	14.06	3.67	1.93	6.93	20.36	2982		925	1794	2734	40		0.03
Canada	CA01	Ontario, Algoma Region, Batchawana Lake	72	2.04	6.02	2.25	0.36	0.56	0.20	0.21	3.50	77	49	44	211	479	5	4.7	1.23
Canada	CA02	Ontario, Algoma Region, Wishart Lake	72	2.73	6.60	3.56	0.40	0.61	0.19	0.19	3.99	124	93	27	425	571	4	3.8	0.28
Canada	CA03	Ontario, Algoma Region, Little Turkey Lake	72	3.37	6.76	4.65	0.46	0.65	0.21	0.20	4.40	174	146	37	460	622	4	3.7	0.22
Canada	CA04	Ontario, Algoma Region, Turkey Lake	72	3.88	6.83	5.61	0.49	0.68	0.23	0.23	4.67	222	196	25	421	571	5	3.5	0.18
Canada	CA05	Quebec, Lac Veilleux	5	1.11	6.48	1.18	0.22	0.48	0.16	0.22	1.95	46	47	5	40			3.3	0.34
Canada	CA06	Quebec, Lac Josselin	5	1.05	6.24	1.03	0.18	0.47	0.13	0.26	2.00	42	31	8	30			3.5	0.57
Canada	CA07	Quebec, Lac Bonneville	5	0.93	5.56	0.70	0.20	0.44	0.07	0.36	2.24		13	8				6.1	3.21
Canada	CA08	Quebec, Lac Laflamme	24		6.63	1.84	0.42	0.96	0.18	0.17	2.41	117	95	14	16			4.4	0.28
Canada	CA09	Quebec, Lac Macleod	5	0.95	5.70	0.74	0.20	0.45	0.06	0.32	2.34		16	5				5.7	2.81
Canada	CA10	Nova Scotia, Mount Tom Lake	3	2.42	4.75	0.27	0.27	2.40	0.19	3.47	0.94		-1			263	7	10.1	17.82
Canada	CA11	Nova Scotia, Mountain Lake	3	2.01	5.37	0.28	0.27	2.63	0.18	3.71	1.18		13			147	5	4.6	4.59
Canada	CA12	Nova Scotia, Little Red Lake	3	3.52	4.37	0.29	0.32	2.91	0.26	3.64	1.16		-30			260	14	17.4	43.62
Canada	CA13	Nova Scotia, Kejimkujik Lake	3	2.64	5.10	0.57	0.35	3.19	0.32	4.45	1.44		9			223	11	9.0	8.08
Canada	CA14	Nova Scotia, Beaverskin Lake	3	1.96	5.79	0.29	0.29	2.58	0.22	3.82	1.33		15			183	6	3.4	1.63
Czech Republic	CZ01	Bohemian Forest, Cerné	4	2.23	5.06	0.66	0.42	0.70	0.47	0.52	2.68	-21	-12	35	844		13	1.9	9.60
Czech Republic	CZ02	Bohemian Forest, Certovo	4	2.22	4.75	0.32	0.29	0.56	0.28	0.40	2.26	-24	-24	43	513		16	3.8	18.19
Czech Republic	CZ03	Bohemian Forest, Ple¿né	4	2.60	5.14	1.16	0.29	0.93	1.06	0.42	2.04	-4	-6	82	1378		21	6.8	7.47
Czech Republic	CZ04	Bohemian Forest, Prá¿ilské	4	1.78	5.17	0.60	0.37	0.63	0.54	0.48	1.24	21	-2	54	574		23	5.8	7.23
Czech Republic	CZ05	Bohemian Forest, Laka	4	2.62	5.57	1.27	0.59	1.04	0.92	0.51	1.04	34	10	30	1539		16	4.9	3.29
Czech Republic	CZ06	Bohemian Forest, Zd´árské	4	2.76	6.39	1.80	0.49	2.43	0.45	0.63	4.58	131	71	77	47		37	8.1	0.49
Czech Republic	CZ07	Lysina	13	4.42	4.43	1.76	0.38	2.23	0.49	1.04	8.19	25	-33	13	56	319	85		43.15
Czech Republic	CZ08	Uhlirska	36	86.80	6.00	4.45	1.02	3.10	0.86	1.14	12.05	155	79	31	239	391	19		3.85

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	TOT N µg/l N	TOT P µg/l P	DOC mg/l C	H+ µEq/l
Finland	FI01	Hirvilampi	5	2.36	5.50	1.42	0.37	1.22	0.55	1.22	4.56	35	15	39	50	312	6	4.9	3.43
Finland	FI02	Vuorilampi	5	2.46	5.86	1.72	0.43	1.38	0.44	0.62	4.20	86	44	75	24	484	8	7.9	1.96
Finland	FI03	Mäkilampi	5	2.00	5.52	1.32	0.30	0.93	0.35	1.08	4.04	22	10	33	42	372	5	4.7	3.30
Finland	FI05	Lapland, Suopalampi	16	1.24	6.32	0.71	0.26	1.04	0.21	0.42	0.72	79	54	9	22	351	14	7.7	0.62
Finland	FI06	Lapland, Vasikkajärvi	17	0.74	5.96	0.24	0.08	0.51	0.16	0.41	1.34	4	11	7	11	149	7	1.9	1.25
Finland	FI07	Vusimaa, Vitsjön	17	3.27	6.55	1.58	0.74	2.81	0.51	3.28	4.12	94	60	21	35	347	6	6.5	0.36
Finland	FI08	N-Karelia,Kakkisenlampi	18	0.50	5.31	0.26	0.05	0.32	0.10	0.26	1.21	1	1	5	6	142	3	3.0	5.23
Finland	FI09	Sonnanen	18	3.02	6.73	2.76	0.48	1.45	0.56	2.13	3.25	125	86	9	18	214	3	2.7	0.19
Germany	DE01	Schwarzwald, Dürreychbach	36	3.07	6.21	2.36	0.64	1.33	2.21	2.63	3.44	64		13	1042			3.7	0.84
Germany	DE02	Fichtelgebirge, Eger	34		6.13	2.71	1.28	5.82	1.02	10.21	4.59	72			905				1.27
Germany	DE03	Rothaargebirge, Elberndorfer Bach	22	6.00	6.55	3.73	2.91	1.40	0.34	5.35	10.50	25		52	1243			2.1	0.62
Germany	DE05	Schwarzwald, Goldersbach	34	2.94	6.77	2.52	0.52	1.62	1.61	2.22	2.82	125		14	432			5.1	0.21
Germany	DE06	Hunsrück, Gräfenbach	24	7.71	5.09	3.77	2.24	4.11	1.01	5.76	17.20	17		29	581		12	7.0	9.17
Germany	DE07	Erzgebirge, Grosse Pyra	22	23.44	4.87	3.32	0.75	2.09	0.93	1.53	12.09	1		21	630	577	28	4.8	18.90
Germany	DE08	Bayerischer Wald, Grosse Ohe	63		6.69	1.95	0.66	1.56	0.53	0.63	2.27	98		15	992		12		0.34
Germany	DE10	Bayerischer Wald, Hinterer Schachtenbach	36		6.52	1.91	0.53	1.43	0.45	0.55	2.53	79		11	926				0.44
Germany	DE13	Erzgebirge, Talsperre Neunzehnhain	66	16.61	6.70	12.79	5.18	8.92	1.56	15.98	35.45	150		43	2147			3.3	0.27
Germany	DE17	Bayerischer Wald, Rachelsee	17		5.42	0.76	0.39	0.65	0.37	0.48	2.51	5		57	524		5		4.30
Germany	DE18	Fichtelgebirge, Röslau	34		5.78	2.11	0.55	3.08	0.52	1.39	8.12	46			450				3.90
Germany	DE21	Erzgebirge, Rote Pockau	23	43.57	5.87	6.91	2.60	4.50	0.93	4.71	25.22	82		23	543	626	33	9.2	1.88
Germany	DE23	Bayerischer Wald, Seebach	36		6.45	1.74	0.66	1.35	0.53	0.61	2.08	75		12	1085				0.65
Germany	DE24	Erzgebirge, Talsperre Sosa	48	5.03	5.75	3.74	1.02	2.63	1.20	2.67	14.34	10		74	443				1.97
Germany	DE26	Hunsrück, Traunbach 1	32	6.55	5.42	1.93	1.55	5.45	1.01	10.44	5.86	36		46	504		16	10.2	7.69
Germany	DE27	Bayerischer Wald, Vorderer Schachtenbach	35		6.71	2.34	0.65	1.91	0.55	0.63	2.75	131		11	853				0.29
Germany	DE28	Oberpfälzer Wald. Waldnaab 2	0		6.88	1.98	1.40	2.80	1.33	5.00				20	900		27		0.14
Germany	DE29	Oberpfälzer Wald, Waldnaab 8	0		5.89	2.91	1.04	3.96	1.31	5.14				21	600		53		2.69
Germany	DE30	Erzgebirge, Wilde Weisseritz	25	44.30	7.24	7.48	1.72	3.37	1.42	4.14	17.36	171		27	689	786	21	4.4	0.20
Germany	DE31	Erzgebirge, Wolfsbach	22	82.95	7.06	13.09	4.35	10.35	1.90	16.95	21.32	462		21	1771	1459	22	5.6	0.10
Germany	DE32	Rothaargebirge, Zinse	22	5.35	6.49	3.51	2.34	1.38	0.32	5.01	8.80	26		52	1043			2.7	0.72
Germany	DE33	Fichtelgebirge, Zinnbach	34		4.42	2.86	0.65	2.45	0.93	1.46	13.53	-61			887				39.25
Germany	DE35	Taunus, Rombach 4	36	5.99	6.99	3.68	1.16	2.83	2.35	6.45	3.19	177		11	622			1.0	0.12
Ireland	IE01	Wicklow, Glendalough, Lake Upper, Mid Lake	11	3.53	6.77	1.45	0.66	3.15	0.21	4.65	2.98	49		34	296		10	3.9	0.37

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	TOT N µg/l N	TOT P µg/l P	DOC mg/l C	H+ µEq/l
Ireland	IE02	Wicklow, Glendalough, Lake Upper, Inflow 1	3	4.23	6.53	2.50	0.63	3.33	0.50	4.33	3.93	123		10	100		8		0.54
Ireland	IE03	Wicklow, Glendalough, Lake Upper, Inflow 2	2	5.74	6.35	3.30	1.20	4.10	0.50	6.05	4.90	125		10	800		5		0.45
Ireland	IE04	Wicklow, Glendalough, Lake Upper, Inflow 3	3	4.44	6.03	0.60	0.60	4.63	0.50	7.53	3.23	2		10	167		5		2.96
Ireland	IE05	Galway, Lough Maumwee, Mid Lake	18	6.30	7.12	4.09	0.88	7.07	0.63	12.48	2.32	176		23	308		8	6.4	0.16
Ireland	IE06	Galway, Lough Maumwee, Inflow 1	3	9.89	6.63	4.20	1.50	8.67	0.50	14.10	3.00	255		13	100		5		0.35
Ireland	IE07	Galway, Lough Maumwee, Inflow 2	3	8.34	7.00	2.67	1.20	9.00	0.50	14.47	2.73	164		10	100		5		0.20
Ireland	IE08	Donegal, Lough Veagh, Mid Lake	20	6.46	6.62	1.98	0.92	7.31	0.61	13.43	2.44	87		25	65		6	5.8	0.35
Ireland	IE09	Donegal, Lough Veagh, Inflow 1	3	8.38	6.53	1.67	1.33	8.60	0.50	14.17	2.27	126		10	100		5		0.40
Ireland	IE10	Donegal, Lough Veagh, Inflow 2	3	6.84	6.70	1.23	1.10	8.10	0.50	13.20	2.20	92		10	100		5		0.29
Italy	IT01	Piemonte, Lake Paione Inferiore	4	1.20	6.63	1.25	0.12	0.31	0.35	0.09	1.40	47	39	4	221	294	3	0.3	0.24
Italy	IT02	Piemonte, Lake di Mergozzo	4	5.92	7.19	5.79	1.47	2.36	0.99	1.75	7.02	297	275	9	642	809	4	1.2	0.10
Italy	IT03	Piemonte, Lake Paione Superiore	4	0.77	6.22	0.65	0.07	0.16	0.28	0.09	0.92	17	15	12	188	311	4	0.6	0.64
Italy	IT04	Piemonte, River Cannobino	36	4.61	7.30	3.92	1.41	1.92	0.72	1.61	4.83	226	217	7	595	662	6	1.0	0.05
Italy	IT05	Piemonte, River Pellino	36	5.42	7.28	4.44	1.13	3.47	0.57	2.67	3.96	220	214	7	1424	1504	13	0.8	0.06
Italy	IT06	Piemonte, River Pellesino	36	5.87	7.19	3.91	0.87	4.93	0.68	5.36	3.06	180	175	28	1458	1579	28	1.2	0.07
Latvia	LV03	Liela Jugla, Zaki	15	41.81	8.05	61.17	14.85	3.69	2.26	5.03	24.02	3808	3856	67	561	1234	52	13.4	0.01
Latvia	LV04	Tulija, Zoseni	15	34.06	7.92	52.46	12.44	3.12	2.11	3.86	11.39	3454	3461	67	414	994	59	13.4	0.01
Latvia	LV05	Zvirbuli stream, hydrosite	11	7.89	3.84	3.67	0.75	1.76	0.46	1.73	1.30	255	20	101	39	1313	22	54.1	191.29
Latvia	LV07	Amula mouth	15	45.27	7.84	62.03	20.54	4.83	3.35	5.60	13.08	4605	4465	57	606	1199	65	14.2	0.03
Norway	MO10	Storgama v. dam	152	1.18	5.09	0.47	0.08	0.69	0.04	0.80	0.90	18	2	19	31	282	4	6.2	9.26
Norway	NO01	Birkenes	157	3.01	4.93	0.74	0.24	2.86	0.10	4.19	2.54	4	2	22	118	344	5	6.8	13.80
Norway	NO03	Langtjern, utløp	153	1.26	5.08	0.88	0.13	0.53	0.07	0.38	0.86	50	3	12	12	272	5	11.7	9.55
Norway	NO04	Dalelv	156	3.58	6.40	1.50	0.80	3.56	0.28	5.51	3.64	70	45	5	12	141	3	3.6	0.46
Norway	NO11	Kårvatn feltforskningsstasjon	138	1.14	6.45	0.63	0.17	1.12	0.13	1.54	0.57	41	33	3	19	72	2	1.0	0.39
Poland	PL01	Tatra Mountains, Dlugi Staw Gasienicowy	49	15.62	6.10	1.70	0.10	0.42	0.11	0.17	1.68	45	44	14	413				1.15
Poland	PL02	Tatra Mountains, Zielony Staw Gasienicowy	46	18.60	6.21	2.31	0.18	0.45	0.16	0.16	1.62	99	106	42	226				0.76
Spain	ES01	Lac Redon	38	1.13	6.78	1.40	0.07	0.27	0.05	0.22	0.83	59	62	30	87			0.7	0.18
Sweden	SE01	Delångersån Iggersund	16	4.46	6.94	3.79	1.20	2.76	0.80	2.63	3.71	270	204	6	102	341	8	6.8	0.12
Sweden	SE02	Alsterån Getebro	16	5.55	6.74	4.24	1.43	4.12	1.23	5.58	3.63	297	192	18	127	509	37	12.8	0.19
Sweden	SE05	Tväringen	17	2.61	6.76	2.81	0.68	1.33	0.53	0.83	1.63	209	133	10	13	271	5	9.4	0.18

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	TOT N µg/l N	TOT P µg/l P	DOC mg/l C	H+ µEq/l
Sweden	SE06	Stensjön	34	1.64	6.33	1.35	0.40	1.23	0.26	0.75	1.47	107	46	13	21	256	7	8.4	0.49
Sweden	SE09	Fiolen	32	4.64	6.67	2.73	1.01	3.61	1.53	5.57	4.62	159	91	22	49	504	13	8.8	0.23
Sweden	SE10	Storasjö	17	2.88	5.48	1.26	0.54	2.79	0.41	3.63	2.48	83	1	36	39	470	17	16.6	3.96
Sweden	SE11	Fräcksjön	34	5.86	6.47	3.04	1.09	6.04	0.71	9.68	3.92	164	78	16	59	429	10	11.7	0.41
Sweden	SE12	Härsvatten	34	4.65	5.09	0.50	0.63	5.62	0.40	9.45	3.08	-5	-15	44	80	356	5	4.3	9.32
Switzerland	CH03	Lago di Tomè	9	3.51	5.75	0.83	0.07	0.27	0.14	0.10	1.30	7	6	7	357			0.3	1.85
Switzerland	CH05	Laghetto Inferiore	9	4.04	6.66	1.14	0.09	0.27	0.32	0.08	1.30	40	32	4	214			0.5	0.25
Switzerland	CH06	Laghetto Superiore	9	3.44	6.59	1.03	0.09	0.24	0.28	0.09	1.08	37	30	5	199			0.5	0.29
Switzerland	CH09	Lago Nero	8	6.76	6.83	2.04	0.16	0.33	0.39	0.09	2.55	72	59	6	158			0.4	0.18
Switzerland	CH19	Lago d'Alzasca	9	6.90	6.92	2.03	0.22	0.46	0.45	0.14	1.96	88	76	7	253			0.5	0.12
Switzerland	CH20	Lago del Starlaresc da Sgiof	9	3.40	5.62	0.56	0.10	0.26	0.15	0.13	1.28	1	3	17	284			0.7	2.74
Switzerland	CH26	Maggia	36	22.99	7.38	7.89	0.64	1.61	1.40	1.24	8.73	296	268	7	546			0.6	0.05
Switzerland	CH27	Vedeggio	36	17.07	7.10	4.78	0.91	1.58	0.51	0.96	5.82	178	155	5	965			0.7	0.08
Switzerland	CH28	Verzasca	36	9.44	6.81	2.87	0.23	0.68	0.57	0.20	3.60	80	69	6	640			0.4	0.16
UK	UK04	Scotland, Lochnagar	7	1.72	5.51	0.36	0.28	1.80	0.27	2.96	1.71	-6	1		186	311		2.4	3.36
UK	UK07	Scotland, Round Loch of Glenhead	8	2.56	5.22	0.44	0.39	2.83	0.24	4.91	1.41	7	-5		113	311		4.9	6.40
UK	UK10	England, Scoat Tarn	8	2.67	5.28	0.41	0.43	2.99	0.23	5.21	1.99	-8	-4		168	263		2.2	5.40
UK	UK15	Wales, Llyn Llagi	8	2.86	5.72	0.81	0.48	3.40	0.24	6.13	1.66	20	12		91	193		3.3	2.18
UK	UK21	N.Ireland, Blue Lough	8	4.56	5.03	0.54	0.57	5.18	0.72	8.40	3.10	-16	-6		459	855		5.6	9.99
US	US05	Maine, Little Long Pond	3	1.75	6.18	0.75	0.28	1.89	0.26	2.48	2.21	33	13		0	239	3	2.7	0.67
US	US06	Maine, Tilden Pond	3	1.87	6.69	1.09	0.33	2.00	0.24	2.09	1.77	79	51		1	95	3	3.1	0.22
US	US100	Howe, Vermont	12	1.45	5.94	1.26	0.29	0.56	0.31	0.47	2.55	44	36		87		17	4.4	1.31
US	US102	Forester, Vermont	6	8.09	5.24	1.14	0.21	12.20	0.39	20.18	2.17	-7	6		11		9	4.8	6.54
US	US103	Paine Run, Virginia	181	2.03	5.74	0.59	0.60	0.56	1.78	0.85	5.21	15	10	2	12				2.03
US	US104	Piney River, Virginia	190	3.54	7.07	2.81	1.35	1.84	0.26	1.06	3.02	242	235	8	27				0.09
US	US105	Staunton River, Virginia	206	1.86	6.66	1.30	0.33	1.43	0.42	0.81	2.25	95	86	15	9				0.26
US	US11	New York, Adirondack Mnt., Arbutus	12	2.05	6.89	2.32	0.39	0.65	0.20	0.26	3.45	84	68	14	248		5	5.6	0.13
US	US12	New York, Adirondack Mnt., Constable	12	1.62	5.26	1.11	0.20	0.51	0.21	0.22	3.16	-16	7	27	594		5	6.4	6.81
US	US123	Haystack, Vermont	9	1.12	5.01	0.48	0.14	0.29	0.09	0.35	1.72	-6	-8		147		6	2.1	11.59
US	US14	New York, Adirondack Mnt., Heart Lake	9	1.42	6.83	1.57	0.22	0.51	0.07	0.20	2.39	47	49	24	246		5	2.6	0.15

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US	US15	New York, Adirondack Mnt., Lake Rondaxe	10	1.82	6.67	1.67	0.33	0.73	0.27	0.34	3.09	27	50	35	661		5	4.7	0.25
US	US16	New York, Adirondack Mnt., Moss Lake	10	2.30	6.94	2.32	0.45	0.89	0.32	0.39	3.49	60	85	19	777		5	4.4	0.13
US	US17	New York, Adirondack Mnt., Otter Lake	10	1.45	5.75	1.00	0.25	0.59	0.18	0.29	3.28	-13	12	13	516		4	3.5	1.92
US	US23	New York, Catskill Mnt., E. Branch Neversink, Head	75	1.59	5.31	0.85	0.39	0.28	0.51	0.37	3.32	5	2	39	216			2.7	5.91
US	US24	New York, Catskill Mnt., Rondout Creek	51	1.57	5.74	1.25	0.41	0.33	0.73	0.37	3.69	29	12	21	187			1.7	2.44
US	US25	W Br Neversink R At Winnisook, Catskills	74	1.71	4.99	0.70	0.31	0.23	0.52	0.33	3.45	-14	-5	23	238			2.4	12.04
US	US26	Biscuit Brook, Catskills	124	1.63	6.12	1.59	0.35	0.27	0.15	0.38	3.27	33		25	168			1.8	1.01
US	US27	Little Hope Pond, Adirondacks	34	12.35	6.00	0.54	0.14	0.19	0.08	0.09	2.14	-5	50	36	93		7	10.6	2.71
US	US28	Big Hope Pond, Adirondacks	35	13.89	6.20	0.61	0.16	0.29	0.09	0.32	2.32	-4	60	21	70		6	8.3	1.60
US	US29	East Copperas Pond, Adirondacks	35	15.07	5.05	0.19	0.04	0.06	0.11	0.09	1.20	-13	-15	158	68		21	13.9	28.32
US	US30	Sunday Pond, Adirondacks	35	9.96	5.39	0.13	0.05	0.02	0.09	0.06	1.83	-29	-1	32	40		7	1.8	5.61
US	US31	Sochia Pond, Adirondacks	34	9.78	5.25	0.08	0.03	0.03	0.04	0.05	1.20	-23	-6	65	64		6	4.5	10.52
US	US32	Marcy Dam Pond, Adirondacks	36	11.59	5.98	0.48	0.08	0.22	0.02	0.07	2.84	-83	37	37	881		6	3.4	2.01
US	US33	Grass Pond, Adirondacks	35	12.36	4.96	0.14	0.04	0.03	0.09	0.07	0.92	-12	-17	112	67		14	9.2	18.56
US	US34	Little Clear Pond, Adirondacks	35	12.44	6.26	0.49	0.12	0.03	0.13	0.05	0.90	18	83	155	17		17	6.8	1.71
US	US35	Loon Hollow Pond, Adirondacks	35	12.42	5.15	0.15	0.03	0.12	0.06	0.09	2.48	-67	-8	38	417		3	5.6	13.90
US	US36	Willys Lake, Adirondacks	34	12.18	5.22	0.26	0.04	0.14	0.08	0.08	2.98	-81	-2	52	574		3	3.7	9.12
US	US37	Woods Lake, Adirondacks	35	12.21	5.90	0.55	0.06	0.13	0.06	0.08	3.05	-100	25	59	1037		4	4.2	3.78
US	US38	Middle Settlement Lake, Adirondacks	36	10.37	5.83	0.32	0.07	0.23	0.08	0.08	2.73	-44	31	89	267		6	3.5	4.22
US	US39	Grass Pond, Adirondacks	36	12.57	5.90	0.46	0.10	0.30	0.10	0.09	3.68	-124	29	21	1276		5	4.2	3.16
US	US40	Middle Branch Lake, Adirondacks	36	11.68	6.19	0.54	0.12	0.30	0.11	0.08	3.14	-30	53	13	203		7	4.9	1.82
US	US41	Limekiln Lake, Adirondacks	35	11.63	6.27	0.55	0.09	0.24	0.07	0.22	2.93	-48	47	14	395		4	3.4	2.24
US	US42	Squaw Lake, Adirondacks	36	9.59	6.18	0.42	0.11	0.14	0.06	0.07	2.96	-44	26	11	244		5	3.3	1.16
US	US43	Indian Lake, Adirondacks	36	11.34	5.58	0.30	0.08	0.14	0.05	0.07	2.59	-60	6	16	470		6	6.3	6.12
US	US44	Brook Trout Lake, Adirondacks	36	9.82	5.83	0.27	0.07	0.14	0.06	0.07	2.85	-57	11	39	323		6	3.0	2.80
US	US45	Lost Pond, Adirondacks	27	13.37	5.27	0.23	0.06	0.16	0.04	0.07	3.29	-83	25	36	521		9	6.1	12.77
US	US46	South Lake, Adirondacks	36	10.05	5.87	0.38	0.08	0.17	0.06	0.08	2.79	-90	15	17	897		8	3.2	3.20
US	US47	North Lake, Adirondacks	35	11.27	5.73	0.38	0.08	0.17	0.07	0.08	2.88	-91	15	13	903		5	5.8	4.08
US	US48	Willis Lake, Adirondacks	34	13.39	6.23	0.65	0.11	0.29	0.03	0.31	2.58	-12	71	10	60		7	7.3	1.54
US	US49	Long Pond, Adirondacks	35	12.38	5.08	0.30	0.10	0.14	0.10	0.08	1.95	-18	-2	53	101		8	15.0	14.12
US	US50	Carry Pond, Adirondacks	34	9.57	5.63	0.20	0.05	0.07	0.09	0.07	1.81	-30	5	61	135		13	3.6	3.68
US	US51	Lake Colden, Adirondacks	36	13.21	5.65	0.35	0.06	0.15	0.03	0.07	2.41	-103	14	19	1129		5	5.1	6.15

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	TOT N µg/l N	TOT P µg/l P	DOC mg/l C	H+ µEq/l
US	US52	Avalanche Lake, Adirondacks	36	10.93	5.34	0.35	0.05	0.14	0.05	0.08	2.07	-124	11	36	1510		5	6.6	7.06
US	US53	Little Simon Pond, Adirondacks	36	13.55	6.50	0.91	0.09	0.21	0.06	0.09	3.90	-79	71	23	813		5	3.9	0.63
US	US54	Raquette Lake Reservoir, Adirondacks	33	13.86	5.77	0.49	0.11	0.22	0.08	0.09	3.66	-78	39	33	624		7	8.6	5.04
US	US55	G Lake, Adirondacks	35	11.61	5.94	0.31	0.08	0.17	0.05	0.08	2.76	-71	17	12	590		4	3.1	2.60
US	US57	Sagamore Lake, Adirondacks	11	1.97	6.17	1.81	0.44	0.73	0.23	0.25	3.36	31	41	11	793		7	9.0	1.17
US	US58	Black Pond Outlet, Adirondacks	11	3.28	7.47	3.40	1.00	1.01	0.34	0.25	3.57	202	198	36	299		9	4.2	0.03
US	US59	Windfall Pond Outle, Adirondacks	12	2.42	7.02	3.11	0.40	0.41	0.20	0.23	3.27	25	95	40	1557		5	4.3	0.11
US	US60	Queer Lake, Adirondacks	11	1.56	6.16	1.41	0.25	0.43	0.22	0.23	3.22	-1	23	34	611		4	4.0	1.51
US	US61	Big Moose Lake, Adirondacks	10	1.50	5.97	1.29	0.22	0.55	0.22	0.28	2.81	6	23	31	561		4	5.2	1.36
US	US62	Cascade Lake Outlet, Adirondacks	10	1.95	6.86	1.95	0.37	0.73	0.29	0.23	3.35	53	63	15	534		7	4.1	0.16
US	US63	Little Echo Pond, Adirondacks	9	2.43	4.40	0.53	0.31	0.27	0.21	0.44	1.28	26	-32	148	53		12	15.8	40.52
US	US64	Squash Pond Outlet, Adirondacks	12	1.87	4.64	0.57	0.12	0.40	0.20	0.22	2.08	-54	-14	81	913		6	9.2	23.53
US	US65	West Pond Outlet, Adirondacks	12	1.45	5.33	1.01	0.20	0.48	0.22	0.20	2.01	18	13	63	393		6	9.0	5.60
US	US66	Bubb Lake Outlet, Adirondacks	12	1.82	6.77	1.69	0.36	0.72	0.27	0.22	3.23	25	53	19	757		5	3.4	0.19
US	US67	Owen Pond, Adirondacks	8	2.69	7.11	3.12	0.56	0.76	0.17	0.29	3.66	77	119	28	1087		6	5.6	0.09
US	US68	Jockeybush Lake, Adirondacks	11	1.44	5.89	1.07	0.27	0.47	0.13	0.23	2.99	-45	11	29	1055		4	2.7	1.63
US	US69	Clear Pond, Adirondacks	11	2.13	7.15	2.56	0.30	0.86	0.08	0.22	3.02	107	106	15	230		4	3.8	0.07
US	US70	Nate Pond, Adirondacks	11	1.94	6.70	1.82	0.46	0.72	0.24	0.24	3.62	62	56	18	322		7	5.8	0.21
US	US71	Bean Pond, Maine	8	1.54	6.67	1.46	0.46	0.87	0.27	0.43	1.82	105	61		1	228	8	7.9	0.22
US	US72	Bracey Pond, Maine	8	1.96	7.08	2.21	0.36	1.30	0.27	1.27	0.98	148	111		0	179	4	5.6	0.08
US	US73	Anderson Pond, Maine	4	1.58	6.26	0.63	0.25	1.78	0.21	2.26	1.91	31	15		2	94	4	2.1	0.55
US	US74	Mud Pond, Maine	8	2.29	4.77	0.43	0.22	1.89	0.17	2.72	2.41	-1	-14		1	118	2	5.7	17.44
US	US75	Salmon Pond, Maine	8	1.88	6.81	1.27	0.34	1.84	0.26	2.12	1.73	82	60		1	126	5	2.9	0.17
US	US76	Wiley Pond, Maine	8	2.37	7.19	3.77	0.62	0.72	0.16	0.44	1.63	228	154		1	287	12	10.7	0.07
US	US77	Second Pond, Maine	8	1.93	6.86	1.58	0.39	1.61	0.24	1.38	1.98	107	71		1	156	5	4.8	0.15
US	US78	Abol Pond, Maine	8	2.68	7.34	3.02	0.38	1.57	0.86	0.51	2.11	213	183		2	93	3	2.4	0.05
US	US79	Duck Pond, Maine	8	1.68	4.61	0.26	0.16	0.67	0.17	1.15	0.93	8	-23		2	202	9	7.2	24.43
US	US80	Jellison Hl Pd, Maine	8	1.96	6.58	1.24	0.37	1.93	0.23	1.92	2.23	81	44		0	145	4	5.1	0.27
US	US81	Crystal Pond, Maine	8	0.80	5.53	0.30	0.13	0.58	0.15	0.81	1.10	9	-1		3	172	4	2.7	3.09
US	US82	Newbert Pond, Maine	8	2.25	4.86	1.30	0.38	1.41	0.34	1.61	1.17	96	1		7	431	36	18.8	14.07
US	US83	Partridge Pond, Maine	8	1.47	6.59	0.96	0.25	1.47	0.21	1.18	1.76	68	38		0	155	3	3.5	0.26
US	US84	Benner Run, Mid-Apps	36	2.76	6.08	1.63	0.62	1.43	0.77	3.36	4.54	-17	24	3	580	0		1.3	1.11
US	US85	Linn Run, Mid-Apps	36	3.25	6.33	3.49	0.63	0.60	0.38	1.00	8.47	12	56	3	632	0		1.0	0.78
US	US86	Roberts Run, Mid-Apps	36	2.58	5.35	1.64	0.73	0.43	0.52	0.78	7.42	-15	2	2	161	0		1.6	7.08

Country	ID	Station name	n	K25	pH	Ca	Mg	Na	K	Cl	SO4	ANC	Alk	NH4-N	NO3-N	TOT N	TOT P	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
US	US87	Stone Run, Mid-Apps	36	2.57	5.28	1.71	0.65	0.48	0.43	0.95	7.64	-20	-2	1	73	0		1.0	7.32
US	US88	Baldwin Creek, Mid-Apps	36	3.46	6.44	3.28	0.93	0.46	0.65	1.12	8.81	-11	54	2	1027	0		0.8	0.43
US	US89	Bourn, Vermont	6	1.03	5.82	0.66	0.29	0.48	0.30	0.25	1.57	44	21		23		13	5.9	1.60
US	US90	Grout, Vermont	11	1.25	5.97	1.04	0.29	0.49	0.40	0.36	1.87	52	32		65		11	4.4	1.27
US	US91	Hardwood, Vermont	13	1.66	5.98	1.61	0.44	0.47	0.15	0.28	2.29	79	58		58		12	5.3	1.30
US	US92	Little ¿ Woodford, Vermont	12	1.48	5.20	0.76	0.19	0.53	0.38	0.34	2.89	-8	-1		318		9	2.1	8.19
US	US93	Stamford, Vermont	6	1.43	5.70	1.03	0.22	0.69	0.27	0.36	2.98	18	19		196		10	3.0	2.62
US	US95	Sunset, Vermont	11	1.33	5.92	0.99	0.27	0.70	0.17	0.70	2.72	27	25		27		13	2.1	1.41
US	US96	Big Mud, Vermont	147	1.60	6.09	1.69	0.37	0.29	3.91	0.36	3.17	97	28	32	760		15	2.1	1.21
US	US97	Branch, Vermont	6	1.13	5.06	0.49	0.18	0.43	0.28	0.26	1.80	18	0		33		15	6.4	9.15
US	US98	Beaver Pond, Vermont	12	1.79	6.19	2.20	0.28	0.58	0.22	0.34	2.29	95	66		134		8	6.2	0.88
Estonia	EE01	River Ahja, Kiidjärve	6	40.25	8.02	63.83	15.30	3.68	1.87	7.05	13.30	4094		46	1157	1452	28	8.9	0.01

Appendix B. Mann-Kendall trend analysis results

Table B. Results of Mann-Kendall trend analysis for the time span 2000-2011. Slope is annual change in the unit given for each variable, 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.

		SO4* (μeqv/l)			NO3 (μeqv/l)			Ca*+Mg* (μeqv/l)			Alkalinity (μeqv/l)			ANC (μeqv/l)			H+ (μeqv/l)			TOC (mg/l)		
ICPW-region	Station	n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
Adirondacks	US11	**10**	**0.00**	**-4.32**	9	0.53	-0.18	**9**	**0.01**	**-4.27**	10	0.06	-1.06	9	0.14	-1.45	10	0.18	0.03	10	0.18	0.08
Adirondacks	US12	**10**	**0.00**	**-3.38**	9	0.53	-0.06	**9**	**0.02**	**-3.06**	10	0.79	0.15	9	0.68	0.57	10	0.18	-0.22	**10**	**0.04**	**0.12**
Adirondacks	US13	**9**	**0.01**	**-2.90**	9	0.53	-0.29	**9**	**0.01**	**-1.81**	**9**	**0.04**	**0.52**	9	0.10	0.86	9	1.00	0.01	9	0.21	0.07
Adirondacks	US14	**10**	**0.00**	**-1.97**	9	0.68	0.06	**9**	**0.00**	**-2.36**	10	0.24	-0.36	9	0.30	-0.60	10	0.79	0.01	10	0.18	-0.03
Adirondacks	US15	**10**	**0.00**	**-2.74**	9	0.30	-0.18	9	0.06	-2.63	10	0.53	0.51	9	0.83	0.63	10	0.33	-0.04	10	0.07	0.07
Adirondacks	US16	**10**	**0.00**	**-3.12**	9	0.53	0.25	**9**	**0.01**	**-2.38**	10	0.65	0.30	9	0.68	-0.03	10	0.65	0.01	10	0.33	0.02
Adirondacks	US17	**10**	**0.00**	**-3.27**	**9**	**0.02**	**0.27**	**9**	**0.04**	**-1.49**	10	0.65	0.59	9	0.30	0.91	10	0.24	-0.09	10	0.18	0.03
Adirondacks	US23	**12**	**0.00**	**-2.53**	9	0.53	0.41				**12**	**0.00**	**1.14**				**12**	**0.00**	**-1.06**	12	0.07	-0.04
Adirondacks	US27	**12**	**0.00**	**-3.23**	9	0.06	-0.34	**9**	**0.02**	**-2.41**	10	0.79	0.42	9	1.00	-0.11	12	0.68	-0.05	12	0.41	-0.07
Adirondacks	US28	**12**	**0.00**	**-3.29**	9	0.06	-0.16	9	0.33	-1.20	10	0.18	1.16	9	0.40	0.81	12	0.89	0.00	12	0.22	0.06
Adirondacks	US29	**12**	**0.00**	**-2.65**	9	0.30	-0.16	**9**	**0.00**	**-1.64**	10	0.93	0.04	9	0.06	2.30	12	0.41	0.35	12	0.05	0.29
Adirondacks	US30	12	0.13	-0.75	9	0.83	0.02	**9**	**0.00**	**-1.07**	10	0.53	-0.07	**9**	**0.00**	**-1.21**	12	0.41	-0.23	12	0.10	-0.07
Adirondacks	US31	**12**	**0.00**	**-1.80**	9	0.40	-0.13	**9**	**0.00**	**-0.50**	10	0.24	0.49	**9**	**0.01**	**1.43**	12	0.17	-0.46	12	0.58	0.01
Adirondacks	US32	**12**	**0.00**	**-2.58**	9	0.10	1.23	**9**	**0.04**	**-1.60**	10	0.93	-0.15	9	0.40	-1.36	12	0.27	-0.04	12	0.13	0.03
Adirondacks	US33	**12**	**0.00**	**-1.14**	9	0.68	-0.04	9	0.24	-0.34	10	0.42	-0.41	9	0.10	0.56	12	0.22	-0.23	12	0.58	0.04
Adirondacks	US34	**12**	**0.00**	**-1.61**	9	0.40	0.04	**9**	**0.00**	**-11.02**	**10**	**0.00**	**-9.02**	**9**	**0.00**	**-10.38**	**12**	**0.01**	**0.03**	12	0.07	-0.06
Adirondacks	US36	**12**	**0.00**	**-3.69**	9	0.21	-0.30	**9**	**0.00**	**-1.63**	10	0.13	0.53	9	0.06	1.20	**12**	**0.01**	**-0.52**	**12**	**0.03**	**0.14**
Adirondacks	US37	**12**	**0.00**	**-2.91**	9	0.68	-0.03	**9**	**0.02**	**-2.69**	10	0.24	0.31	9	0.06	0.88	12	0.22	0.07	12	0.78	0.01
Adirondacks	US38	**12**	**0.00**	**-2.35**	9	0.68	-0.01	9	0.33	-0.31	**10**	**0.01**	**1.34**	**9**	**0.01**	**1.34**	12	0.34	-0.21	**12**	**0.00**	**0.05**
Adirondacks	US39	**12**	**0.00**	**-2.01**	9	0.53	0.18	**9**	**0.04**	**-1.93**	10	0.65	-0.10	9	0.83	-0.24	12	0.68	-0.05	12	0.68	0.01
Adirondacks	US40	**12**	**0.00**	**-1.91**	9	0.68	0.05	**9**	**0.04**	**-1.49**	10	0.93	0.00	9	0.53	-0.51	12	0.41	0.03	12	0.17	0.05
Adirondacks	US41	**12**	**0.00**	**-3.34**	9	0.14	-0.33	**9**	**0.00**	**-2.24**	10	0.09	0.99	9	0.83	0.40	12	0.78	-0.01	12	0.58	0.00

ICPW-region	Station	SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
		n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
Adirondacks	US42	12	0.00	-2.33	9	0.30	0.26	9	0.00	-1.86	10	0.33	0.38	9	1.00	0.03	12	0.41	-0.04	12	0.05	-0.05
Adirondacks	US43	12	0.00	-3.22	9	0.40	0.27	9	0.01	-1.92	10	0.03	0.60	9	0.04	1.16	12	0.04	-0.45	12	0.03	0.09
Adirondacks	US44	12	0.00	-2.44	9	0.02	0.98	9	0.00	-1.91	10	0.93	0.00	9	0.14	-0.72	12	0.27	-0.15	12	0.13	-0.05
Adirondacks	US45	12	0.00	-1.78	9	0.10	0.63	9	0.04	-1.40	10	0.18	1.07	9	1.00	-0.07	12	1.00	0.03	12	0.22	0.03
Adirondacks	US46	12	0.00	-2.70	9	0.40	-0.23	9	0.00	-2.74	10	0.53	-0.35	9	0.04	-1.13	12	1.00	0.03	12	0.05	0.03
Adirondacks	US47	12	0.00	-3.30	9	0.40	0.31	9	0.00	-2.00	10	0.09	0.75	9	0.53	0.43	12	0.04	-0.37	12	0.09	0.08
Adirondacks	US48	11	0.00	-4.11	8	0.46	0.03		0.01	-3.99	9	1.00	-0.13				11	0.82	0.02	11	0.02	-0.27
Adirondacks	US49	12	0.00	-4.11	9	0.02	-0.14	9	0.01	-2.00	10	0.93	0.12	9	0.53	0.74	12	0.27	-0.60	12	0.22	0.14
Adirondacks	US50	12	0.01	-3.40	9	0.30	-0.11	9	0.01	-1.49	10	0.04	2.15	9	0.01	4.36	12	0.01	-0.65	12	0.02	0.16
Adirondacks	US51	12	0.00	-3.70	9	0.83	-0.22	9	0.00	-1.85	10	0.09	0.69	9	0.68	0.26	12	0.22	-0.27	12	0.04	0.08
Adirondacks	US52	12	0.00	-3.74	9	0.83	0.24	9	0.01	-2.17	10	0.04	0.44	9	0.83	0.30	12	0.07	-0.37	12	0.27	0.08
Adirondacks	US53	12	0.00	-3.07	9	0.53	-0.35	9	0.33	-2.58	10	0.65	1.48	9	0.21	3.16	12	0.49	-0.02	12	0.10	0.04
Adirondacks	US54	12	0.00	-3.99	9	0.83	-0.14	9	0.01	-2.32	10	0.42	0.69	9	0.68	0.79	12	0.13	-0.29	12	0.78	0.01
Adirondacks	US55	12	0.00	-3.07	9	0.40	0.79	9	0.01	-1.85	10	0.79	0.13	9	0.68	-0.48	12	0.41	-0.15	12	0.34	0.04
Adirondacks	US57	10	0.00	-3.85	9	0.14	0.49	9	0.01	-3.53	10	0.53	-0.44	9	0.83	-0.34	10	0.93	-0.01	10	0.33	0.06
Adirondacks	US58	10	0.00	-2.71	9	0.83	-0.04	9	0.01	-3.37	10	0.09	-1.35	9	0.83	-0.25	10	0.33	0.00	10	0.13	0.04
Adirondacks	US59	10	0.01	-3.04	9	1.00	0.01	9	0.65	-0.64	10	0.18	1.43	9	0.40	1.74	10	0.13	-0.06	10	0.53	-0.08
Adirondacks	US60	10	0.00	-2.50	9	0.06	-0.23	9	0.00	-2.38	10	0.03	0.75	9	0.10	0.89	10	0.79	-0.05	10	0.02	0.10
Adirondacks	US61	10	0.00	-3.23	9	0.53	-0.31	9	0.00	-2.10	10	0.04	0.82	9	0.04	0.86	10	0.65	-0.08	10	0.15	0.09
Adirondacks	US62	10	0.00	-2.89	9	0.06	0.76	9	0.18	-2.46	10	0.53	1.47	9	0.40	0.95	10	0.42	-0.02	10	0.65	-0.04
Adirondacks	US63	10	0.42	-0.40	9	0.68	-0.02	9	0.18	-0.92	10	0.79	0.12	9	0.83	0.16	10	0.79	-0.01	10	0.33	-0.06
Adirondacks	US64	10	0.01	-2.27	9	0.30	0.48	9	0.01	-1.14	10	0.04	0.88	9	0.53	0.16	10	0.13	-0.57	10	0.24	0.14
Adirondacks	US65	10	0.09	-1.55	9	0.68	0.09	9	0.03	-1.58	10	0.33	0.70	9	0.83	-0.01	10	0.42	-0.23	10	0.42	-0.05
Adirondacks	US66	10	0.02	-1.35	9	1.00	-0.03	9	0.00	-2.50	10	0.09	-1.31	9	0.01	-2.06	10	0.53	0.01	10	0.24	-0.06
Adirondacks	US67	10	0.00	-5.12	9	0.68	0.36	9	0.01	-5.86	10	0.24	-1.18	9	0.40	-0.98	10	0.33	0.00	10	0.65	0.03
Adirondacks	US68	10	0.00	-2.78	9	0.53	0.36	9	0.01	-1.64	10	0.02	0.65	9	0.68	0.19	10	0.24	-0.18	10	0.65	0.01
Adirondacks	US69	10	0.00	-3.40	9	0.14	0.16	9	0.00	-3.24	10	0.33	-0.40	9	0.40	-0.70	10	0.53	0.00	10	0.18	0.03

ICPW-region	Station	SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
		n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
Adirondacks	US70	**10**	**0.00**	**-3.40**	9	0.53	-0.08	**9**	**0.02**	**-4.74**	10	0.33	-1.95	9	0.30	-1.15	10	0.79	-0.01	10	0.33	0.04
Alps	CH03	**12**	**0.00**	**-0.65**	**12**	**0.04**	**-1.13**	12	0.17	-0.96	12	0.73	0.17	12	0.13	0.64	12	0.68	-0.03	10	0.79	0.00
Alps	CH05	**12**	**0.00**	**-0.89**	**12**	**0.05**	**-0.69**	12	0.34	0.31	12	0.27	0.49	**12**	**0.03**	**1.55**	12	0.07	-0.02	12	0.58	0.00
Alps	CH06	**12**	**0.00**	**-1.15**	**12**	**0.01**	**-0.84**	12	0.78	-0.10	12	0.06	0.74	**12**	**0.00**	**1.98**	**12**	**0.01**	**-0.03**	12	0.49	0.01
Alps	CH09	12	0.58	0.24	11	0.48	-0.06	12	0.22	0.85	12	1.00	-0.01	12	0.10	1.18	12	0.89	0.00	11	0.39	0.02
Alps	CH19	**12**	**0.04**	**-0.66**	12	0.34	-0.36	12	0.22	1.19	12	0.17	0.81	**12**	**0.04**	**2.13**	12	0.49	0.00	11	0.59	0.02
Alps	CH20	**12**	**0.00**	**-1.04**	12	0.05	-0.87	12	0.34	-0.64	12	0.07	0.00	**12**	**0.04**	**0.99**	**12**	**0.00**	**-0.42**	12	0.89	0.00
Alps	CH26	12	0.78	-0.39	12	0.49	0.22	12	0.34	1.97	12	0.78	-0.14	12	0.34	2.65	12	0.34	0.00	12	0.68	0.01
Alps	CH27	12	0.27	-0.53	12	0.49	-0.33	12	0.22	2.65	**12**	**0.01**	**1.85**	12	0.07	3.58	12	0.07	0.00	**12**	**0.04**	**0.01**
Alps	CH28	12	0.68	0.30	12	0.34	-0.46	**12**	**0.04**	**2.07**	**12**	**0.00**	**1.24**	**12**	**0.00**	**3.30**	12	0.10	-0.01	12	0.49	0.00
Alps	IT01	**12**	**0.01**	**-1.16**	**12**	**0.00**	**-1.59**	12	0.68	-0.32	9	0.59	0.13	12	0.05	2.78	12	0.13	-0.01			
Alps	IT02	**12**	**0.00**	**-2.58**	12	1.00	-0.02	12	0.17	3.76	**9**	**0.00**	**3.93**	**12**	**0.01**	**7.76**	12	0.33	0.00			
Alps	IT03	**12**	**0.00**	**-1.18**	**12**	**0.00**	**-1.29**	12	0.89	0.10	**9**	**0.02**	**1.27**	**12**	**0.01**	**2.56**	12	0.17	-0.04			
Alps	IT04	12	0.22	-1.01	**12**	**0.00**	**-1.79**	12	0.78	-0.41	9	0.35	2.11	12	0.34	1.29	12	0.27	0.00			
Alps	IT05	12	0.05	-0.73	**12**	**0.00**	**-2.57**	12	1.00	-0.16	9	0.21	3.25	12	0.41	1.52	**12**	**0.03**	**0.00**			
Alps	IT06	**12**	**0.01**	**-1.15**	**12**	**0.02**	**-1.85**	12	0.89	0.08	9	0.40	2.64	12	0.78	0.53	12	0.49	0.00			
Appalachians	US24	**12**	**0.00**	**-2.14**	11	0.14	-1.03				**12**	**0.02**	**1.27**				**12**	**0.01**	**-0.53**	12	0.41	-0.02
Appalachians	US25	**12**	**0.00**	**-2.50**	9	0.14	0.82				**12**	**0.00**	**1.29**				**12**	**0.02**	**-1.04**	12	0.41	-0.02
Appalachians	US26	**12**	**0.00**	**-3.09**	9	0.40	1.37				9	0.21	-1.30				12	1.00	0.00	12	0.49	-0.01
Appalachians	US35	**12**	**0.00**	**-2.57**	9	0.68	-0.34	**9**	**0.00**	**-1.11**	10	0.24	0.45	**9**	**0.01**	**1.43**	12	0.10	-0.35	**12**	**0.01**	**0.15**
Appalachians	US84	12	0.05	-1.20	9	0.40	-0.11	9			**12**	**0.04**	**0.86**	9	0.68	0.25	12	0.49	-0.09	12	0.41	-0.02
Appalachians	US85	**12**	**0.00**	**-2.99**	9	0.14	-1.37				**12**	**0.02**	**2.89**				12	0.10	-0.21	**12**	**0.00**	**-0.02**
Appalachians	US86	**12**	**0.04**	**-1.14**	9	0.06	0.28				12	0.07	0.59				12	0.49	-0.05	**12**	**0.03**	**-0.07**
Appalachians	US87	**12**	**0.01**	**-2.31**	9	0.14	0.24				12	0.49	0.10				12	0.41	-0.11	**12**	**0.03**	**-0.03**
Appalachians	US88	12	0.22	-0.83	**9**	**0.02**	**-1.94**				12	0.68	0.34				12	0.07	-0.07	12	0.27	-0.01
Baltic	BY01	**11**	**0.00**	**-21.30**	**11**	**0.00**	**9.90**	11	0.59	-14.75	9	0.14	19.73				11	0.70	0.00			
Baltic	EE01	10	0.33	-2.40	10	0.24	1.74	11	0.93	-2.14				11	0.82	4.04	10	0.42	0.00			

		SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
ICPW-region	Station	n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
Baltic	LV03	11	0.14	-42.47	**11**	**0.02**	**-1.63**	10	0.10	-54.24	10	0.18	33.50	10	0.93	-3.76	11	0.59	0.00			
Baltic	LV04	11	0.24	-15.35	11	0.94	-0.11	11	0.48	19.82	10	0.13	57.91	11	0.70	16.51	11	0.24	0.00			
Baltic	LV05	**11**	**0.01**	**-12.77**	**11**	**0.02**	**-0.49**	11	0.07	-28.77	**10**	**0.01**	**-10.57**	11	0.31	35.23	11	0.24	-9.95			
Baltic	LV07	9	0.06	-8.44	9	0.30	2.31	11	1.00	-7.98				11	0.82	-0.64	9	0.53	0.00			
Blue Ridge Mountains	US103	**12**	**0.04**	**-0.22**	12	0.27	-0.06	9	0.83	-0.07	12	0.89	0.06	9	0.68	-32.89	12	0.10	0.05			
Blue Ridge Mountains	US104	12	0.27	-0.43	**12**	**0.00**	**-0.66**	**9**	**0.04**	**-2.35**	12	0.89	-0.21	9	1.00	0.02	12	0.17	0.00			
Blue Ridge Mountains	US105	12	0.07	0.17	12	0.13	0.06	9	0.06	-0.74	12	0.58	0.31	9	0.21	-0.68	12	0.34	-0.01			
ECEurope	CZ01	**11**	**0.00**	**-2.75**	11	0.48	-0.40	11	0.19	-0.42	10	0.42	0.50	**11**	**0.02**	**2.74**	11	0.05	-0.87	11	0.14	0.08
ECEurope	CZ02	**11**	**0.00**	**-4.00**	11	0.31	-0.31	11	0.05	-0.41	10	0.33	0.86	**11**	**0.01**	**4.13**	**11**	**0.01**	**-2.06**	11	0.14	0.12
ECEurope	CZ03	**11**	**0.00**	**-3.70**	**11**	**0.00**	**8.53**	**11**	**0.00**	**3.11**	10	0.79	0.32	11	0.94	-0.53	11	0.39	-0.16	**11**	**0.03**	**0.29**
ECEurope	CZ04	**11**	**0.04**	**-1.90**	11	0.31	0.84	11	0.31	0.66	10	0.42	0.45	11	0.24	1.61	11	0.14	-0.67	**11**	**0.02**	**0.21**
ECEurope	CZ05	11	0.39	-1.31	**11**	**0.01**	**2.99**	11	0.05	2.38	10	0.33	-0.61	11	0.70	-0.33	11	0.39	-0.08	11	0.24	0.18
ECEurope	CZ06	11	0.14	-2.56	11	0.23	0.00	11	0.31	-1.29	10	0.18	2.35	11	0.07	2.51	11	0.14	-0.06	11	0.82	-0.04
ECEurope	CZ07	**11**	**0.01**	**-12.93**	11	0.07	0.31	**11**	**0.04**	**-4.77**	11	0.07	2.18	**11**	**0.00**	**5.24**	11	0.07	-2.02	11	0.14	0.52
ECEurope	CZ08	12	0.10	-3.44	**12**	**0.02**	**1.87**	12	0.49	1.75	12	0.07	4.36	12	0.41	2.98	12	0.17	-0.38			
ECEurope	DE02	10	0.33	-0.21				12	0.68	-0.26							**12**	**0.04**	**-0.36**			
ECEurope	DE07	**11**	**0.00**	**-15.49**	**11**	**0.00**	**-1.96**	**11**	**0.00**	**-6.70**				**11**	**0.00**	**9.00**	11	0.10	-1.07	11	0.27	0.09
ECEurope	DE08	**11**	**0.02**	**-0.99**	**12**	**0.00**	**-2.02**	11	0.49	-0.43				11	0.14	2.64	**12**	**0.00**	**-0.20**			
ECEurope	DE10	11	0.48	-0.15	**12**	**0.00**	**-5.81**	11	0.13	-1.32				**11**	**0.02**	**4.51**	**12**	**0.00**	**-0.48**			
ECEurope	DE13	**9**	**0.00**	**-13.31**				**9**	**0.00**	**-25.30**							11	0.82	0.00			
ECEurope	DE17	12	0.13	-0.69	**12**	**0.00**	**-10.41**	12	0.10	-6.14				12	0.78	-1.65	**12**	**0.00**	**-3.71**			
ECEurope	DE18	**10**	**0.00**	**-3.27**				12	0.13	-0.84							**12**	**0.04**	**-0.98**			
ECEurope	DE21	**11**	**0.00**	**-17.20**	**11**	**0.00**	**-4.28**	**11**	**0.00**	**-17.89**				11	0.39	3.25	11	0.82	-0.02	11	0.24	0.28
ECEurope	DE23	**10**	**0.00**	**-0.96**	12	0.78	-0.12	9	0.24	1.48				9	0.06	2.92	**12**	**0.01**	**-0.34**			
ECEurope	DE24	**10**	**0.00**	**-14.05**				**10**	**0.00**	**-27.56**							**12**	**0.00**	**-0.37**			
ECEurope	DE27	**10**	**0.00**	**-0.69**	**12**	**0.00**	**-3.49**	10	0.94	0.00				**10**	**0.03**	**4.10**	**12**	**0.00**	**-0.11**			
ECEurope	DE28	9	0.53	1.92	9	0.14	-1.49	10	0.42	-5.99							10	0.93	0.00	9	0.53	0.12

		SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
ICPW-region	Station	n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
ECEurope	DE29	9	0.06	-7.98	9	0.30	0.66	10	0.53	-2.22							10	0.33	0.20	9	0.14	0.38
ECEurope	DE30	**11**	**0.00**	**-13.49**	**11**	**0.00**	**-3.18**	11	0.05	-6.65				**11**	**0.02**	**7.55**	11	0.70	0.00	11	0.59	-0.02
ECEurope	DE31	**11**	**0.01**	**-27.33**	**11**	**0.02**	**-5.73**	**11**	**0.00**	**-29.50**				11	0.94	0.18	11	0.24	0.00	11	0.07	0.13
ECEurope	DE33	**10**	**0.00**	**-9.01**	9	0.10	-1.86	**12**	**0.04**	**-3.54**				**12**	**0.00**	**-2.79**						
ECEurope	PL01	**9**	**0.00**	**-1.79**	**9**	**0.04**		9	0.30	-0.70				**9**	**0.02**	**3.14**	9		-0.02			
ECEurope	PL02	**9**	**0.00**	**-1.52**	9	0.21	-0.40	**9**	**0.04**	**-1.80**				**9**	**0.01**	**1.53**	9	0.06	0.05			
Maine/Atlantic	CA10	**11**	**0.02**	**-1.14**	9			9	0.42	0.14				9	0.68	0.27	11	0.05	-0.33	11	0.05	0.22
Maine/Atlantic	CA11	**11**	**0.00**	**-1.45**	9	1.00	0.00	**9**	**0.00**	**-1.17**				9	0.40	-0.81	11	0.82	0.01	11	0.05	0.15
Maine/Atlantic	CA12	**11**	**0.00**	**-1.37**	9			9	0.94	-0.03				9	0.68	0.78	11	0.59	-0.12	11	0.14	0.26
Maine/Atlantic	CA13	**11**	**0.02**	**-1.03**	9	1.00	0.00	9	0.05	-0.99				9	0.40	-1.67	11	0.07	-0.44	11	0.10	0.12
Maine/Atlantic	CA14	**11**	**0.00**	**-1.53**	9	0.80	0.00	**9**	**0.01**	**-0.96**				9	0.30	-1.35	**11**	**0.01**	**-0.17**	**11**	**0.01**	**0.11**
Maine/Atlantic	US05	**12**	**0.00**	**-1.95**	12	0.80	0.00	9	0.14	-0.59	12	0.84	0.02	9	0.40	1.20	11	0.43	-0.07	11	0.12	0.08
Maine/Atlantic	US06	**12**	**0.00**	**-1.67**	12	0.11	-0.03	9	0.30	-1.11	12	0.54	-0.71	9	0.83	0.22	11	0.59	0.00	11	0.88	-0.01
Maine/Atlantic	US71	**11**	**0.00**	**-3.44**	11	0.49	0.00				11	0.82	0.26				10	0.33	-0.02	11	0.59	0.11
Maine/Atlantic	US72	**12**	**0.04**	**-1.62**	12	0.33	-0.03	9	0.10	-3.59	12	0.07	-1.03	9	0.10	-0.90	**11**	**0.02**	**-0.01**	12	0.68	0.04
Maine/Atlantic	US73	**11**	**0.00**	**-1.56**	11	0.18	-0.03				11	0.09	0.26				10	0.72	-0.04	10	0.37	-0.02
Maine/Atlantic	US74	**12**	**0.00**	**-3.59**	**12**	**0.01**	**-0.03**	9	0.30	-0.88	12	0.13	-0.52	9	0.21	0.65	11	0.39	0.37	12	0.07	0.12
Maine/Atlantic	US75	**12**	**0.00**	**-0.90**	12	0.07	-0.03	9	0.53	-0.37	12	0.19	-0.63	9	0.83	-0.16	11	0.07	-0.02	**12**	**0.00**	**-0.10**
Maine/Atlantic	US76	**12**	**0.03**	**-2.10**	12	0.49	-0.01	9	1.00	0.39	12	0.41	1.07	9	1.00	0.38	11	0.10	-0.02	12	0.84	0.00
Maine/Atlantic	US77	**12**	**0.00**	**-1.59**	12	0.06	-0.02	9	0.40	-0.90	12	0.73	0.21	9	0.68	-0.07	11	0.05	-0.02	12	0.78	-0.03
Maine/Atlantic	US78	**12**	**0.01**	**-1.23**	12	0.53	0.00	9	0.14	-3.46	**12**	**0.04**	**-3.01**	9	0.21	-2.50	11	0.59	0.00	12	0.83	0.00
Maine/Atlantic	US79	**11**	**0.00**	**-2.56**	**11**	**0.02**	**-0.06**	9	0.68	0.27	**11**	**0.00**	**1.63**	**9**	**0.01**	**1.81**	**10**	**0.01**	**-1.38**	11	0.09	0.17
Maine/Atlantic	US80	**12**	**0.00**	**-2.02**	12	0.19	0.00	9	0.40	-0.74	12	0.27	0.36	9	0.21	0.98	11	0.10	-0.02	12	0.05	0.13
Maine/Atlantic	US81	**12**	**0.00**	**-1.33**	12	0.09	0.03	9	0.14	-0.73	12	0.24	-0.20	9	0.68	0.18	**11**	**0.02**	**-0.12**	12	0.21	-0.02
Maine/Atlantic	US82	**11**	**0.02**	**-5.86**	11	0.59	-0.01				**11**	**0.01**	**2.29**				**10**	**0.01**	**-1.48**	11	0.94	-0.06
Maine/Atlantic	US83	**12**	**0.00**	**-1.29**	**12**	**0.01**	**-0.02**	9	0.10	0.53	12	0.68	0.21	**9**	**0.01**	**2.30**	11	0.19	-0.03	12	0.84	0.02
NoNordic	FI05	11	0.05	-0.53	11	0.82	0.04	11	0.59	0.47	11	0.10	0.75	11	0.19	0.90	11	0.70	-0.01	**11**	**0.01**	**0.20**

ICPW-region	Station	SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
		n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
NoNordic	FI06	11	0.07	-0.52	11	0.64	-0.02	**11**	**0.00**	**-1.00**	11	0.14	0.53	11	0.07	-0.53	11	0.48	0.02	11	0.12	0.02
NoNordic	FI08	**11**	**0.00**	**-1.02**	**11**	**0.01**	**-0.15**	**11**	**0.02**	**-0.65**	**11**	**0.02**	**0.75**	11	0.59	0.20	11	0.48	-0.08	11	0.48	0.05
NoNordic	NO04	**12**	**0.01**	**-1.30**	12	0.34	-0.02	12	0.58	-0.30	**12**	**0.01**	**1.18**	12	0.27	1.17	**12**	**0.00**	**-0.02**	12	0.10	0.02
NoNordic	SE01	**10**	**0.02**	**-1.01**	10	0.33	-0.30	10			**10**	**0.00**	**3.43**	10	0.04	1.76	10	0.33	0.00	10	0.42	0.02
NoNordic	SE02	**10**	**0.00**	**-13.12**	10	0.13	0.37	**10**	**0.01**	**-12.78**				10	0.42	-2.18	10	0.09	-0.01	10	0.79	0.02
NoNordic	SE06	**12**	**0.00**	**-1.18**	12	0.58	-0.04	12	0.34	0.45	12	0.13	0.80	**12**	**0.00**	**1.85**	12	0.27	-0.02	12	0.17	0.16
Ontario	CA01	**10**	**0.00**	**-2.70**	10	0.65	-0.04	10	0.13	1.37				**10**	**0.01**	**5.09**	10	0.93	0.00	10	0.53	-0.02
Ontario	CA02	**10**	**0.02**	**-1.41**	10	0.24	0.35	10	0.13	2.76				**10**	**0.04**	**4.41**	10	0.42	0.00	10	0.53	-0.03
Ontario	CA03	**10**	**0.02**	**-1.37**	10	0.42	0.30	10	0.09	1.93				**10**	**0.03**	**3.81**	10	0.24	0.00	10	0.42	-0.01
Ontario	CA04	10	0.06	-0.93	10	0.24	0.72	**10**	**0.00**	**3.14**				**10**	**0.02**	**4.31**	10	0.13	0.00	10	0.42	0.02
Ontario	CA16	**9**	**0.00**	**-0.87**	9	0.68	-0.01				9	1.00	-0.11				9	1.00	0.00			
Ontario	CA17	**9**	**0.04**	**-0.73**	9	1.00	0.00	9	0.30	2.05	9	0.21	-0.70	9	0.06	1.56	9	0.06	-0.01			
Ontario	CA20	**9**	**0.01**	**-0.51**	9	0.21	0.03	**9**	**0.00**	**2.53**	9	0.53	0.57	**9**	**0.02**	**3.08**	9	0.14	0.00			
SoNordic	FI01	**9**	**0.01**	**-4.17**	9	0.53	0.14	**9**	**0.01**	**-1.06**	9	0.53	-0.20	9	0.21	0.96	9	0.83	-0.02	**9**	**0.01**	**0.14**
SoNordic	FI02	9	0.14	-2.61	**9**	**0.00**	**-0.43**	9	0.06	-3.00	9	0.40	-1.82	9	1.00	-0.12	9	0.83	0.04	9	0.30	0.16
SoNordic	FI03	**9**	**0.00**	**-2.80**	**9**	**0.02**	**0.16**	**9**	**0.00**	**-3.17**	9	0.25	-0.64	9	0.21	-2.59	9	0.53	-0.06	9	0.40	-0.02
SoNordic	FI07	**11**	**0.00**	**-4.82**	11	0.48	0.04	**11**	**0.00**	**-5.37**	11	0.39	-0.94	11	0.48	-1.10	11	0.94	0.00	**11**	**0.02**	**0.18**
SoNordic	FI09	**11**	**0.00**	**-2.10**	11	0.39	0.04	11	0.10	-1.54	11	0.31	0.52	11	0.48	0.16	11	0.59	0.00	11	0.21	0.03
SoNordic	NO01	**12**	**0.01**	**-1.49**	12	0.68	-0.13	12	0.22	-0.64	12	0.34	0.12	**12**	**0.04**	**1.80**	12	0.78	-0.12	12	0.07	0.16
SoNordic	NO03	**12**	**0.00**	**-1.09**	12	0.05	-0.02	12	0.13	-0.94	12	0.49	-0.15	12	0.58	0.15	12	0.68	0.04	**12**	**0.01**	**0.14**
SoNordic	NO10	**12**	**0.01**	**-1.05**	**12**	**0.00**	**-0.22**	12	0.89	-0.03	**12**	**0.00**	**0.18**	**12**	**0.00**	**1.39**	**12**	**0.00**	**-0.55**	**12**	**0.00**	**0.08**
SoNordic	NO11	12	0.05	-0.33	**12**	**0.00**	**-0.11**	12	0.17	-0.49	12	0.07	0.26	12	0.68	0.30	12	0.05	-0.01	12	0.41	0.01
SoNordic	SE05	**12**	**0.00**	**-0.85**	12	0.45	-0.01	12	0.22	1.28				12	0.34	2.70	12	0.17	0.00	12	0.05	0.21
SoNordic	SE09	**12**	**0.00**	**-4.70**	12	0.68	0.13	**12**	**0.00**	**-2.07**	**12**	**0.00**	**3.10**	**12**	**0.04**	**2.87**	12	0.78	0.00	**12**	**0.00**	**0.24**
SoNordic	SE10	**12**	**0.00**	**-3.54**	12	0.41	0.06	**12**	**0.00**	**-1.61**	12	0.05	0.70	12	0.22	1.39	12	0.17	-0.14	**12**	**0.04**	**0.44**
SoNordic	SE11	**12**	**0.00**	**-3.24**	**12**	**0.02**	**-0.31**	**12**	**0.03**	**-2.17**	12	0.41	1.40	12	0.68	0.70	12	0.27	0.01	**12**	**0.00**	**0.26**
SoNordic	SE12	**12**	**0.00**	**-3.62**	12	0.58	-0.07	**12**	**0.00**	**-1.61**	**12**	**0.00**	**2.20**	12	0.17	1.67	**12**	**0.00**	**-1.24**	**12**	**0.04**	**0.12**

ICPW-region	Station	SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
		n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
UKIreland	IE01	10	0.65	0.23	10	0.47	0.29	10	0.37	-0.75				10	0.18	-8.58	10	0.21	-0.06			
UKIreland	IE02	10	0.93	1.27	10	0.07	-0.57	10	0.53	-5.10				10	0.53	-3.04	10	0.86	0.00			
UKIreland	IE03	9	0.68	1.86	9	0.83	1.41	9	0.53	-3.08				9	0.21	-10.03	9	0.10	0.07			
UKIreland	IE04	10	0.18	-1.09	**10**	**0.00**	**-1.28**	**10**	**0.03**	**-7.36**				10	0.18	-6.35	10	0.24	-0.41			
UKIreland	IE05	9	0.68	0.27	**9**	**0.05**	**1.14**	9	1.00	-0.09				9	0.30	-5.78	9	0.53	-0.02			
UKIreland	IE06	9	0.53	-0.82	9	0.92	-0.12	9	0.68	1.32				9	1.00	-0.78	9	1.00	0.00			
UKIreland	IE07	9	0.30	0.29	9	0.20	0.36	9	0.30	-6.24				9	0.14	-13.10	9	0.40	-0.03			
UKIreland	IE08	9	0.53	0.22	9	0.83	0.04	9	0.53	-4.07				**9**	**0.04**	**-10.20**	9	0.14	-0.03			
UKIreland	IE09	9	0.67	-0.29	9	0.60	0.17	9	0.68	-4.06				9	0.21	-8.61	9	0.68	0.03			
UKIreland	IE10	9	0.68	-0.08	9	0.38	0.30										9	0.14	-0.32			
UKIreland	UK01	9	**0.05**	**-0.99**	9	**0.04**	**0.17**	9	0.21	-1.59				9	0.06	-3.48						
UKIreland	UK04	**11**	**0.00**	**-1.16**	**12**	**0.04**	**-0.61**	**11**	**0.00**	**-1.64**	11	0.07	0.43	11	0.82	-0.11	12	0.22	-0.08	12	0.10	0.08
UKIreland	UK07	**12**	**0.02**	**-1.10**	12	0.34	0.12	12	0.17	-0.49	11	0.58	-0.15	12	0.34	0.65	12	0.17	-0.23	12	0.41	0.06
UKIreland	UK10	**12**	**0.03**	**-0.77**	**12**	**0.00**	**-0.83**	12	0.13	-0.59	11	0.53	0.23	12	0.17	0.82	**12**	**0.01**	**-0.37**	12	0.11	0.08
UKIreland	UK15	**12**	**0.01**	**-0.79**	**12**	**0.02**	**0.49**	12	1.00	-0.01	**11**	**0.04**	**1.17**	12	1.00	0.08	12	0.58	0.03	12	0.78	0.01
UKIreland	UK21	12	0.41	-0.73	12	0.34	-0.93	12	0.27	-0.56	11	0.94	0.25	12	0.22	1.33	**12**	**0.00**	**-0.72**	12	0.68	0.08
Verm./Quebec	CA05	12	0.05	-1.25	10	0.05	-0.21	10	0.10	-0.45	**12**	**0.01**	**1.77**	10	0.79	0.08	12	0.58	0.00	**12**	**0.01**	**0.06**
Verm./Quebec	CA06	**12**	**0.00**	**-1.37**	**11**	**0.01**	**-0.16**	**11**	**0.01**	**-0.89**	12	0.27	0.50	11	0.24	0.36	12	0.89	0.00	**12**	**0.00**	**0.08**
Verm./Quebec	CA07	**12**	**0.00**	**-2.50**	**9**	**0.00**	**-0.30**	**9**	**0.00**	**-2.24**	12	0.68	0.28	9	0.10	-1.73	**12**	**0.04**	**0.16**	**12**	**0.01**	**0.16**
Verm./Quebec	CA08	**11**	**0.00**	**-1.26**	11	0.48	-0.07	11	0.24	-0.43	11	0.39	1.79	11	0.48	0.61	11	0.59	-0.01	11	0.24	0.04
Verm./Quebec	CA09	**12**	**0.01**	**-1.44**	**9**	**0.02**	**-0.09**	**9**	**0.04**	**-1.13**	12	0.06	0.61	9	0.30	-0.63	12	0.78	-0.05	**12**	**0.01**	**0.09**
Verm./Quebec	US100	11	**0.02**	**-0.59**	**11**	**0.01**	**0.72**	**11**	**0.00**	**-2.00**	11	0.14	-1.58	**11**	**0.01**	**-2.98**	11	0.70	0.01	11	0.12	-0.07
Verm./Quebec	US102	11	0.19	-1.09	11	0.64	0.04	11	0.24	-1.21	**11**	**0.01**	**1.51**	11	0.70	-1.32	11	0.07	-0.62	**11**	**0.03**	**0.25**
Verm./Quebec	US123	**11**	**0.00**	**-1.61**	11	0.70	0.33	11	0.10	-0.48	11	0.70	-0.10	11	0.48	0.37	11	0.39	-0.07	11	0.07	0.09
Verm./Quebec	US89	**11**	**0.00**	**-2.35**	11	1.00	0.00	11	0.70	0.07	**11**	**0.00**	**1.77**	**11**	**0.01**	**1.99**	**11**	**0.00**	**-0.40**	11	0.64	0.04
Verm./Quebec	US90	**11**	**0.00**	**-1.59**	11	0.48	0.06	**11**	**0.04**	**-1.22**	11	0.94	0.03	11	0.94	0.11	11	0.24	0.02	**11**	**0.01**	**0.10**
Verm./Quebec	US91	**11**	**0.00**	**-2.21**	11	0.75	0.00	**11**	**0.04**	**-1.27**	11	0.48	0.34	11	0.70	0.32	11	0.82	0.01	11	0.75	0.00

ICPW-region	Station	SO4* (µeqv/l)			NO3 (µeqv/l)			Ca*+Mg* (µeqv/l)			Alkalinity (µeqv/l)			ANC (µeqv/l)			H+ (µeqv/l)			TOC (mg/l)		
		n	p	Slope	n	p		n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope	n	p	Slope
Verm./Quebec	US92	**11**	**0.00**	**-2.59**	11	0.06	1.31	**11**	**0.02**	**-0.97**	11	0.39	-0.22	11	0.59	-0.36	11	0.70	0.11	11	0.21	0.04
Verm./Quebec	US93	**11**	**0.00**	**-1.69**	11	0.07	0.83	**11**	**0.01**	**-1.45**	11	0.94	0.08	11	0.19	-1.02	11	0.70	0.06	11	0.35	-0.04
Verm./Quebec	US95	**11**	**0.00**	**-1.13**	11	0.69	0.08	11	0.05	-1.08	11	0.31	0.27	11	0.31	-0.47	11	0.82	0.01	11	0.09	0.03
Verm./Quebec	US96	12	0.89	0.21	**12**	**0.00**	**4.90**	12	0.41	2.82	**12**	**0.00**	**2.28**	12	0.10	5.66	**12**	**0.00**	**-0.57**	12	0.01	-0.60
Verm./Quebec	US97	**11**	**0.00**	**-2.57**	11	0.94	0.00	**11**	**0.00**	**-1.14**	**11**	**0.04**	**0.60**	11	0.19	0.87	**11**	**0.04**	**-0.43**	11	0.48	0.03
Verm./Quebec	US98	**11**	**0.00**	**-3.46**	11	0.81	0.03	**11**	**0.00**	**-3.58**	11	0.48	-0.36	11	0.19	-0.83	11	0.70	0.01	**11**	**0.03**	**0.08**
WCEurope	DE01	12	0.89	-0.09	**12**	**0.01**	**-1.22**	**12**	**0.00**	**-4.35**				12	0.22	-3.45	**12**	**0.00**	**-0.08**	12	0.41	0.08
WCEurope	DE03	**10**	**0.03**	**-3.04**	**10**	**0.02**	**3.12**	10	0.93	0.00							11	0.48	-0.02			
WCEurope	DE05	12	0.07	-0.88	**12**	**0.02**	**-0.86**	12	0.05	-3.95				12	0.17	-3.65	12	0.22	-0.03	**12**	**0.02**	**0.05**
WCEurope	DE06	**12**	**0.02**	**-5.54**	10	0.33	1.79	10	0.89	0.96				10	0.09	6.54	**12**	**0.02**	**-2.09**	12	0.78	-0.04
WCEurope	DE26	**12**	**0.00**	**-4.77**	10	0.65	0.75	9	0.05	-4.36				9	0.83	0.41	12	0.05	-1.29	12	0.27	0.12
WCEurope	DE32	**10**	**0.01**	**-4.00**	**10**	**0.00**	**2.84**	10	0.79	1.52							11	0.70	-0.02			
WCEurope	DE35	**10**	**0.01**	**-5.61**	**10**	**0.03**	**-2.58**	10	0.06	-19.26				10	0.53	2.41	10	0.13	-0.06	**10**	**0.02**	**-0.15**

Appendix C. Appendix C. Estimated water chemistry in 2020

Table C. Estimated deposition of oxidised sulphur, concentration of non-marine sulphate and non-marine base cations, and acid neutralising capacity.

Region	ID	Station	Year	Runoff (mm)	dep oxS mg/m2	F	[SO4*] µEq/l	[BC*] µEq/l	ANC µEq/l
Alps	IT01	Piemonte, Lake Paione Inferiore	2005	1000	378	0.38	36	100	40
Alps	IT01	Piemonte, Lake Paione Inferiore	2020	1000	213	0.38	31	99	43
Alps	IT02	Piemonte, Lake di Mergozzo	2005	1300	635	0.63	154	484	281
Alps	IT02	Piemonte, Lake di Mergozzo	2020	1300	333	0.63	123	465	292
Alps	IT03	Piemonte, Lake Paione Superiore	2005	1000	378	0.23	29	60	11
Alps	IT03	Piemonte, Lake Paione Superiore	2020	1000	213	0.23	24	59	15
Alps	IT04	Piemonte, River Cannobino	2005	1400	493	0.82	115	397	231
Alps	IT04	Piemonte, River Cannobino	2020	1400	288	0.82	98	383	234
Alps	IT05	Piemonte, River Pellino	2005	1300	635	0.84	85	422	225
Alps	IT05	Piemonte, River Pellino	2020	1300	333	0.84	82	420	226
Alps	IT06	Piemonte, River Pellesino	2005	1300	635	0.97	54	346	181
Alps	IT06	Piemonte, River Pellesino	2020	1300	333	0.97	61	352	181
ECEurope	CZ01	Bohemian Forest, Cerné	2005	1157	615	0.43	76	99	-39
ECEurope	CZ01	Bohemian Forest, Cerné	2020	1157	362	0.43	55	90	-27
ECEurope	CZ02	Bohemian Forest, Certovo	2005	1179	615	0.26	77	59	-53
ECEurope	CZ02	Bohemian Forest, Certovo	2020	1179	362	0.26	53	53	-35
ECEurope	CZ03	Bohemian Forest, Ple¿né	2005	1073	512	0.41	72	105	-9
ECEurope	CZ03	Bohemian Forest, Ple¿né	2020	1073	294	0.41	52	97	3
ECEurope	CZ04	Bohemian Forest, Prá¿ilské	2005	1151	615	0.32	43	73	6
ECEurope	CZ04	Bohemian Forest, Prá¿ilské	2020	1151	362	0.32	34	70	12
ECEurope	CZ05	Bohemian Forest, Laka	2005	1255	615	0.58	37	130	37
ECEurope	CZ05	Bohemian Forest, Laka	2020	1255	362	0.58	34	128	38
ECEurope	CZ06	Bohemian Forest, Zd´árské	2005	1012	512	0.80	125	242	112
ECEurope	CZ06	Bohemian Forest, Zd´árské	2020	1012	294	0.80	92	215	119

Region	ID	Station	Year	Runoff (mm)	dep oxS mg/m2	F	[SO4*] µEq/l	[BC*] µEq/l	ANC µEq/l
ECEurope	CZ07	Lysina	2005	450	608	0.37	224	213	-13
ECEurope	CZ07	Lysina	2020	450	373	0.37	154	187	32
ECEurope	CZ08	Uhlirska	2005	986	833	0.99	293	443	142
ECEurope	CZ08	Uhlirska	2020	986	489	0.99	205	356	143
ECEurope	DE02	Fichtelgebirge, Eger	2005	473	618	0.33	74	189	58
ECEurope	DE02	Fichtelgebirge, Eger	2020	473	380	0.33	61	185	67
ECEurope	DE07	Erzgebirge, Grosse Pyra	2005	1200	757	0.91	288	310	-32
ECEurope	DE07	Erzgebirge, Grosse Pyra	2020	1200	479	0.91	204	234	-25
ECEurope	DE08	Bayerischer Wald, Grosse Ohe	2005	956	574	0.71	50	210	82
ECEurope	DE08	Bayerischer Wald, Grosse Ohe	2020	956	343	0.71	48	208	83
ECEurope	DE10	Bayerischer Wald, Hinterer Schachtenbach	2005	1350	574	0.88	53	210	71
ECEurope	DE10	Bayerischer Wald, Hinterer Schachtenbach	2020	1350	343	0.88	49	206	72
ECEurope	DE18	Fichtelgebirge, Röslau	2005	946	618	0.80	187	280	58
ECEurope	DE18	Fichtelgebirge, Röslau	2020	946	380	0.80	134	238	68
ECEurope	DE21	Erzgebirge, Rote Pockau	2005	416	778	0.92	628	750	70
ECEurope	DE21	Erzgebirge, Rote Pockau	2020	416	485	0.92	441	578	86
ECEurope	DE23	Bayerischer Wald, Seebach	2005	1350	574	0.85	45	194	73
ECEurope	DE23	Bayerischer Wald, Seebach	2020	1350	343	0.85	43	192	74
ECEurope	DE24	Erzgebirge, Talsperre Sosa	2005	1050	757	0.80	326	358	-23
ECEurope	DE24	Erzgebirge, Talsperre Sosa	2020	1050	479	0.80	230	281	-4
ECEurope	DE27	Bayerischer Wald, Vorderer Schachtenbach	2005	1350	574	0.97	56	253	121
ECEurope	DE27	Bayerischer Wald, Vorderer Schachtenbach	2020	1350	343	0.97	54	250	121
ECEurope	DE28	Oberpfälzer Wald. Waldnaab 2	2005	750	580	0.82	282	542	154
ECEurope	DE28	Oberpfälzer Wald. Waldnaab 2	2020	750	357	0.82	204	479	168
ECEurope	DE29	Oberpfälzer Wald, Waldnaab 8	2005	750	580	0.79	294	514	158
ECEurope	DE29	Oberpfälzer Wald, Waldnaab 8	2020	750	357	0.79	217	453	174
ECEurope	DE30	Erzgebirge, Wilde Weisseritz	2005	665	1195	0.99	416	615	138
ECEurope	DE30	Erzgebirge, Wilde Weisseritz	2020	665	746	0.99	301	501	139

Region	ID	Station	Year	Runoff (mm)	dep oxS mg/m2	F	[SO4*] µEq/l	[BC*] µEq/l	ANC µEq/l
ECEurope	DE33	Fichtelgebirge, Zinnbach	2005	158	618	0.18	308	301	-62
ECEurope	DE33	Fichtelgebirge, Zinnbach	2020	158	380	0.18	212	283	17
ECEurope	PL02	Tatra Mountains, Zielony Staw Gasienicowy	2005	1544	775	0.79	41	152	94
ECEurope	PL02	Tatra Mountains, Zielony Staw Gasienicowy	2020	1544	402	0.79	38	149	95
NoN	FI05	Lapland, Suopalampi	2005	330	92	0.13	20	100	78
NoN	FI05	Lapland, Suopalampi	2020	330	55	0.13	22	100	77
NoN	FI06	Lapland, Vasikkajärvi	2005	330	92	0.05	28	41	12
NoN	FI06	Lapland, Vasikkajärvi	2020	330	55	0.05	23	41	17
NoN	FI08	Kakkisenlampi	2005	375	200	0.04	28	30	1
NoN	FI08	Kakkisenlampi	2020	375	134	0.04	23	30	6
NoN	NO04	Dalelv	2005	281	101	0.15	64	134	69
NoN	NO04	Dalelv	2020	281	76	0.15	56	132	76
NoN	SE01	Delångersån Iggersund	2005	315	199	0.41	70	339	261
NoN	SE01	Delångersån Iggersund	2020	315	118	0.41	68	338	262
NoN	SE05	Tväringen	2005	330	103	0.30	37	238	201
NoN	SE05	Tväringen	2020	330	57	0.30	42	240	197
NoN	SE06	Stensjön	2005	315	120	0.17	32	134	100
NoN	SE06	Stensjön	2020	315	65	0.17	32	134	101
SoN	FI01	Hirvilampi	2005	330	392	0.18	99	139	38
SoN	FI01	Hirvilampi	2020	330	264	0.18	77	135	56
SoN	FI02	Vuorilampi	2005	330	392	0.25	91	189	95
SoN	FI02	Vuorilampi	2020	330	264	0.25	75	185	107
SoN	FI03	Mäkilampi	2005	330	392	0.16	88	124	35
SoN	FI03	Mäkilampi	2020	330	264	0.16	68	121	51
SoN	FI07	Vitsjön	2005	287	434	0.21	101	192	89
SoN	FI07	Vitsjön	2020	287	238	0.21	74	187	110
SoN	FI09	Sonnanen	2005	297	250	0.23	71	195	122
SoN	FI09	Sonnanen	2020	297	160	0.23	61	193	131

Region	ID	Station	Year	Runoff (mm)	dep oxS mg/m2	F	[SO4*] µEq/l	[BC*] µEq/l	ANC µEq/l
SoN	NO01	Birkenes	2005	1019.7	482	0.22	44	50	-1
SoN	NO01	Birkenes	2020	1019.7	272	0.22	32	48	8
SoN	NO03	Langtjern, utløp	2005	524.16	164	0.14	21	70	48
SoN	NO03	Langtjern, utløp	2020	524.16	82	0.14	20	70	48
SoN	NO10	Storgama v. dam	2005	1219	256	0.17	21	35	11
SoN	NO10	Storgama v. dam	2020	1219	129	0.17	17	34	14
SoN	NO11	Kärvatn feltforskningsstasjon	2005	821	73	0.15	6	48	40
SoN	NO11	Kårvatn feltforskningsstasjon	2020	821	35	0.15	11	49	36
SoN	SE02	Alsterån Getebro	2005	246	318	0.40	138	437	290
SoN	SE02	Alsterån Getebro	2020	246	163	0.40	107	425	309
SoN	SE09	Fiolen	2005	315	307	0.30	102	246	139
SoN	SE09	Fiolen	2020	315	144	0.30	74	238	158
SoN	SE10	Storasjö	2005	250	319	0.13	48	137	87
SoN	SE10	Storasjö	2020	250	155	0.13	39	136	94
SoN	SE11	Fräcksjön	2005	500	367	0.42	64	224	154
SoN	SE11	Fräcksjön	2020	500	159	0.42	54	219	160
SoN	SE12	Härsvatten	2005	632	367	0.10	52	40	-19
SoN	SE12	Härsvatten	2020	632	159	0.10	31	38	1
UKIreland	UK01	Scotland, Loch Coire nan Arr	2005	2632	216	0.23	2	22	18
UKIreland	UK01	Scotland, Loch Coire nan Arr	2020	2632	91	0.23	8	24	14
UKIreland	UK04	Scotland, Lochnagar	2005	835	286	0.16	33	48	-2
UKIreland	UK04	Scotland, Lochnagar	2020	835	119	0.16	23	46	7
UKIreland	UK07	Scotland, Round Loch of Glenhead	2005	2001	348	0.26	22	31	0
UKIreland	UK07	Scotland, Round Loch of Glenhead	2020	2001	145	0.26	17	30	4
UKIreland	UK10	England, Scoat Tarn	2005	1454	466	0.17	28	30	-13
UKIreland	UK10	England, Scoat Tarn	2020	1454	195	0.17	19	28	-6
UKIreland	UK15	Wales, Llyn Llagi	2005	2192	345	0.38	21	45	18
UKIreland	UK15	Wales, Llyn Llagi	2020	2192	137	0.38	18	44	20

Region	ID	Station	Year	Runoff (mm)	dep oxS mg/m2	F	[SO4*] µEq/l	[BC*] µEq/l	ANC µEq/l
UKIreland	UK21	N.Ireland, Blue Lough	2005	934	292	0.16	34	44	-20
UKIreland	UK21	N.Ireland, Blue Lough	2020	934	119	0.16	23	42	-11
WCEurope	DE01	Schwarzwald, Dürreychbach	2005	485	836	0.41	58	220	83
WCEurope	DE01	Schwarzwald, Dürreychbach	2020	485	521	0.41	54	219	85
WCEurope	DE03	Rothaargebirge, Elberndorfer Bach	2005	1191	795	0.87	212	355	60
WCEurope	DE03	Rothaargebirge, Elberndorfer Bach	2020	1191	526	0.87	164	313	66
WCEurope	DE05	Schwarzwald, Goldersbach	2005	1750	480	0.97	59	224	133
WCEurope	DE05	Schwarzwald, Goldersbach	2020	1750	267	0.97	53	218	134
WCEurope	DE06	Hunsrück, Gräfenbach	2005	390	548	0.59	377	413	-70
WCEurope	DE06	Hunsrück, Gräfenbach	2020	390	320	0.59	252	340	-19
WCEurope	DE26	Hunsrück, Traunbach 1	2005	980	642	0.63	86	164	38
WCEurope	DE26	Hunsrück, Traunbach 1	2020	980	367	0.63	65	151	46
WCEurope	DE32	Rothaargebirge, Zinse	2005	1419	843	0.94	193	319	69
WCEurope	DE32	Rothaargebirge, Zinse	2020	1419	570	0.94	151	279	71

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