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Trends and patterns in surface water chemistry in Europe and North America between 1990 and 2016, with particular focus on changes in land use as a confounding factor for recovery

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Norwegian Institute for Water Research

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Summary

The report presents trends in sulphate, nitrate, chloride, base cations, ANC (acid neutralising capacity), pH and DOC at circa 500 ICP Waters sites in Europe and North America for the period 1990-2016. Time series were analysed for trends in annual median values, annual extreme values and change points, that indicate years with sudden changes in trend or level. Also provided is a brief overview of possible implications of land use change for recovery of acidified surface waters.

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CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

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

**Trends and patterns in surface water chemistry in
Europe and North America between 1990 and 2016,
with particular focus on changes in land use as a
confounding factor for recovery**

Prepared at the ICP Waters Programme Centre
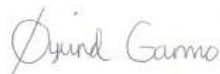Norwegian Institute for Water Research
Oslo, March 2020

Preface

The International Cooperative Programme on Assessment and Monitoring of the Effects 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 (CLRTAP) in July 1985. Since then, ICP Waters has been an important contributor to document the effects of implementing the Protocols under the Convention. ICP Waters has prepared numerous assessments, reports and publications that address the effects of long-range transported air pollution.

ICP Waters and its Programme Centre is chaired and hosted by the Norwegian Institute for Water Research (NIVA). A programme subcentre is established at NORCE (previously known as Uni Research), Bergen. ICP Waters is supported financially by the Norwegian Environment Agency and the Trust Fund of the UNECE LRTAP Convention.

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.

An important basis of the work of the ICP Waters programme is the data from existing surface water monitoring programmes in the participating countries, collected through voluntary contributions. The ICP Waters site network is geographically extensive and includes long-term data series (more than 25 years) for many sites. The programme conducts annual chemical intercomparison and biological intercalibration exercises.

We thank Ingvild Skumlien Furuseth who helped finalise the report.

Øyvind Garmo   and   Heleen de Wit

ICP Waters Programme Centre
Oslo, March 2020

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Summary

The report presents trends in sulphate, nitrate, chloride, base cations, ANC (acid neutralising capacity), pH and dissolved organic carbon (DOC) at circa 500 ICP Waters sites (predominantly headwater lakes and rivers) in Europe and North America for the period 1990-2016. The stations were grouped into 13 regions according to geographic location, acid sensitivity and rates of deposition. Time series were analysed for trends in annual median values, annual extreme values and change points, that indicate years with sudden changes in trend or level for a given component.

Sulphate concentrations have declined at 481 of the 497 sites considered, and 10 of 13 regions show more than 40 % decline for the period 1990-2016. Nitrate has declined in all regions, although many of the individual stations show no trend. Notably, also chloride showed declining trends in 9 of 13 regions, and in Northern Europe the negative slopes were steeper than for nitrate. Chloride levels are affected by deposition of sea salts as well as air pollution. Change points, i.e. the points in the series where the slope changes or where there is a step change in level, were more prevalent in the time series for sulphate and nitrate in the 90s than after year 2000, indicating smoother trends in recent years. On a regional scale, base cation concentrations have declined by approximately half the equivalent change in concentration of sulphate. All regions show increasing ANC between 1990 and 2016. pH has increased in 12 of 13 regions, despite partial replacement of mineral acidity by organic acidity manifested as an increase in DOC. The trajectories of sulphate, ANC and pH indicate that recovery was slowing down in Europe and accelerating in North America since the early 2000s.
This is related to faster declines in emissions of sulphur to the atmosphere in North America and slower emission declines in Europe after 2000. Acidic episodes, i.e. the annual extremes, have become less severe in line with the recovery of average chemistry.

Environmental factors other than acid deposition – so-called 'confounding factors' – can affect chemical and biological recovery of freshwaters in response to reduced acid deposition. Changes in climate and land use can have varied effects on recovery of acidified surface waters and may be viewed as categories of confounding factors. We provide a brief overview of some possible implications of land use change. Disturbances affecting vegetative succession and soil formation can alter the pools of carbon, nitrogen and base cations in soils, which in turn may affect chemical and biological recovery of freshwaters in response to reduced acid deposition. We describe how intensified forestry, reduced summer farming and/or grazing in upland areas, increased vegetation growth/expansion or forest at higher altitudes, and temporary decline or dieback of forest due to drought or disease outbreaks can change the leaching of sea salts, sulphate, nitrate, base cations, DOC, aluminium as well as runoff pH. Effects of land use and climate change on chemical recovery could become more prominent where declines in atmospheric pollutants are levelling off.

1   Introduction

1.1   The ICP Waters programme

Over the past 50 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) was adopted in 1979. Its 40th anniversary was marked at the annual session of the Executive Body in December last year. The convention entered into force in 1983 and was the first step to implement 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 its 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 of air pollution on the environment through monitoring, modelling and scientific review.

The International Cooperative Programme on Assessment and Monitoring of the Effects of Air Pollution on 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, but Norway took over when the Programme Centre was moved in 1987.

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.

One of the responsibilities of ICP Waters, as listed in the mandate[1], is to plan and conduct technical work to assess, using monitoring data and other sources of scientific evidence.

1.2   The ICP Waters database

The ICP Waters database contains surface water data from more than 20 countries across Europe and North America (Table 1). The dataset now comprises 556 stations with long-term monitoring, predominantly located in "acid-sensitive" regions with low acid neutralising capacity and a history of acid deposition (Skjelkvåle et al., 2000). In 2019, much effort was put into completing records available for all stations, with particular emphasis on the period from 1990 to 2016.

Datasets are updated approximately annually and lists of the "required" and "optional" water chemistry parameters are shown in Table 2 and Table 3.

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[1] https://www.unece.org/fileadmin/DAM/env/documents/2019/AIR/EB/ECE_EB.AIR_2019_9-1916525E.pdf

Table 1. ICP Waters stations by country.

Country	Number of stations
Canada	115
Czech Republic	8
Estonia	1
Finland	26
Germany	35
Ireland	21
Italy	12
Latvia	8
Moldova	2
Netherlands	3
Norway	83
Poland	12
Slovakia	12
Sweden	92
Switzerland	9
United Kingdom	22
United States	95
Total	**556**

Table 2. Required parameters

Parameters	Name	Units
Alkalinity	ALK	µeq/L
Sulphate	SULF	mg SO_4^{2-}/L
Nitrate as nitrogen	NO_3-N	µg N/L
Chloride	Cl^-	mg/L
Dissolved carbon, total organic carbon or permanganate	DOC, TOC, PERM	mg C/L mg O/L
pH	pH	pH
Calcium	Ca	mg/L
Magnesium	Mg	mg/L
Sodium	Na	mg/L
Potassium	K	mg/L
Ammonium as nitrogen	NH_4-N	µg N/L
Inorganic (labile) aluminium	LAL	µg/L
Specific conductivity at 25°C	K25	mS/m
Total phosphorus	TOTP	µg P/L

Table 3. Optional parameters

Parameters	Name	Units
Water temperature	TEMP	°C
Flow at time of sampling	RUNOFF	m3/s
Total nitrogen	TOTN	µg N/L
Soluble reactive phosphate	ORTP	µg P/L
Dissolved oxygen	OKS	mg O/L
Silica	SIO_2	mg SiO_2/L
Fluoride	F	mg/V
Colour	COLOUR	mg Pt/L
Turbidity	TURB	FTU
Total aluminium	TAL	µg/L

1.3   The current report

Evaluation of long-term trends in aquatic chemistry that can be linked to atmospheric pollution, is one of the major aims of ICP Waters (see above). The indicators most amenable for such analysis are those that respond to changes in deposition of sulphur and nitrogen compounds. This has therefore been a recurring topic of the scientific reports issued by the Task Force, with a periodicity of about 4 years (see list below). As time series have grown longer and deposition levels have decreased, an additional 3 to 5 years of data does not result in major shifts in regional and temporal patterns. Avid readers of ICP Waters reports might therefore recognize certain elements and phrases. In the most recent reports, we have tried to direct attention to particular aspects of the temporal development. In 2007 the add-on topic was confounding factors, i.e. how some drivers other than acid deposition could affect chemical and biological recovery (Skjelkvåle and de Wit, 2007). In 2011 and 2015 it was biological recovery, or the lack thereof, as well as attempts to compare trends from two different periods in water and precipitation, and to predict future trends (Garmo et al., 2015; Skjelkvåle and De Wit, 2011).

In the current report we use the ICP Waters extended set of water chemistry sites to analyse long-term trends in average chemistry over 26 years (1990-2016). This is by far the largest data set both in terms of number of stations and length of time series that has been subject to this type of analysis directed by ICP Waters. We also use sophisticated statistical techniques to determine when change points have occurred in the time series. Widespread changes in the trends of aquatic chemistry in natural catchments is either an effect of changes in deposition or the result of one or more confounding factors.

Another question which is of relevance for understanding biological recovery, is whether the long-term trends in average chemistry reflect the trends in episode severity. The report contains an analysis of trends in annual extremes and how they compare to average trends. A separate chapter is dedicated to a review of how changes in land use can affect the recovery of acidified waters (Chapter 3). This type of potentially confounding factor was only briefly mentioned in the 2007 report, which focused primarily on climate factors.

2   Trends in surface water chemistry 1990-2016

2.1   Methods

2.1.1 Selection of sites for analysis

The sites with data in the ICP Waters database have been selected by the Focal Centres. They should preferably be sensitive to air pollution (e.g. have inherent low alkalinity) and have short response time. Their catchments should also be relatively undisturbed and be without significant local sources of pollution (ICP Waters Programme Centre, 2010). The sites are dominantly headwaters, many of them located in protected areas.

The objectives of this work were threefold: (i) to analyse trends in annual medians, (ii) to identify change points in time series and (iii) to consider trends in episode severity. The first objective will be familiar to readers of previous ICP Waters reports, since it has been repeated at regular intervals over the last 25 years. This time we included more sites than ever before in our analysis. This was because more data were available, partly because of a large data compilation effort undertaken to investigate changes in DOC since the last extended analysis (Monteith et al., 2007). The period considered was 1990 – 2016 and, to avoid spurious results due to missing data, we applied selection criteria for inclusion of water chemistry time series at each site. The selection of period was a trade-off between length of time series and number of sites meeting certain criteria. For the first objective – analysing trends in annual medians – the following criteria were used:

- Time series had at least one data point within the first 5 years (start of 1990 to end of 1994)
- Time series had at least one data point within the last 5 years (start of 2012 to end of 2016)
- Time series included data from at least of 18 of the 27 years

Water chemistry times series from 497 (out of 556) sites met these requirements (Figure 1).

Objectives (ii) and (iii) considered change points and severe episodes, respectively. For these, it was considered necessary to use stricter criteria for sampling frequency, in order to capture some of the seasonal variation:

- Lakes: At least seasonal frequency and a maximum of 25% missing seasonal values between 1995 and 2011
- Rivers/streams: At least monthly frequency and a maximum of 25% missing monthly values between 1995 and 2011

These criteria are the result of a trade-off between the demands for continuity/sampling frequency and maintaining sufficient spatial coverage across regions. Time series from a total of 231 sites passed these criteria (Figure 2), but note that for some regions the number of stations selected is small.

We report trends for sites that are grouped into regions defined by geographic location, acid sensitivity and rates of deposition. These regions are similar to those used in previous ICP Waters trend reports.
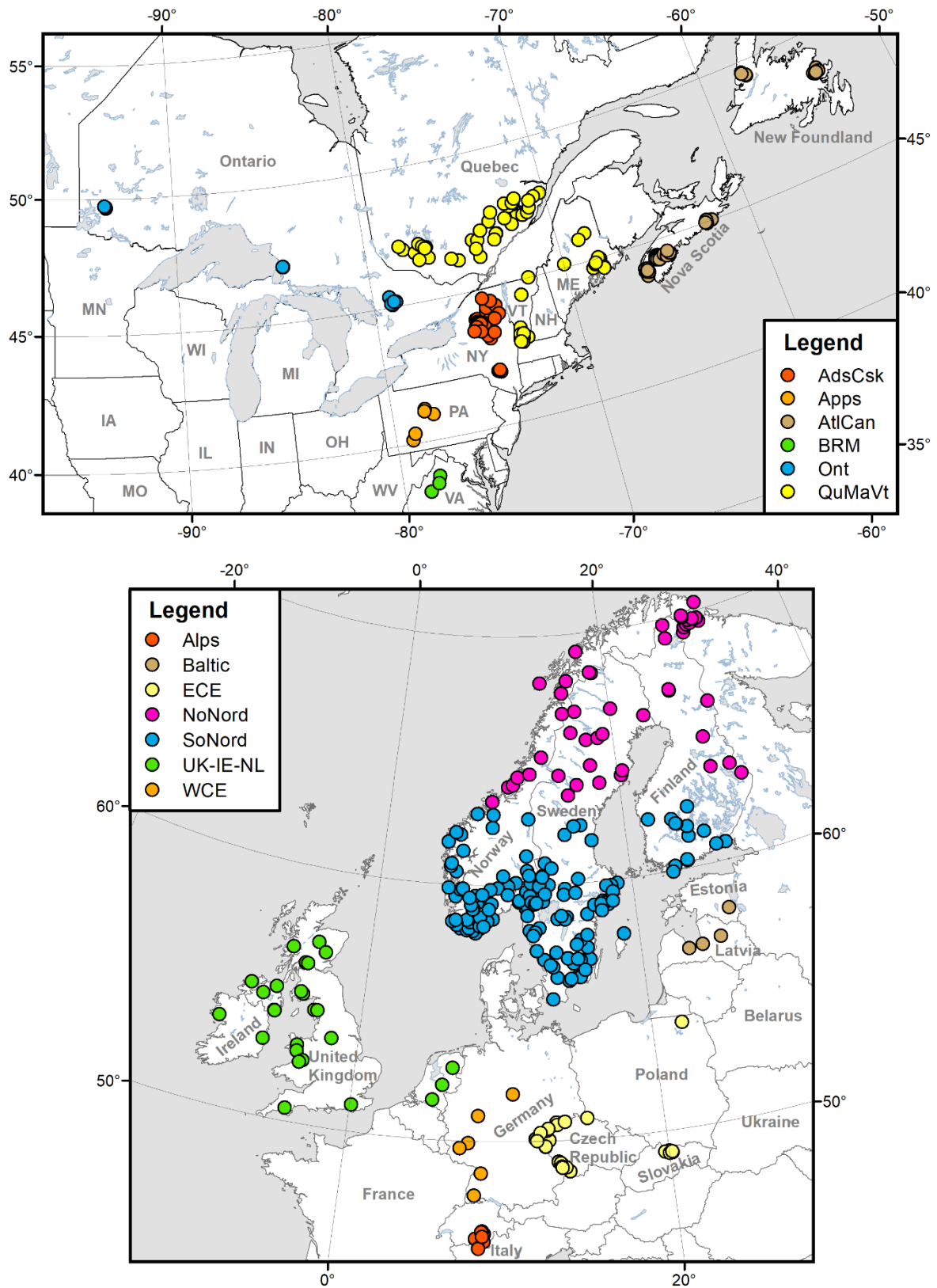
Figure 1. Maps showing the 497 stations that passed the criteria for estimating monotonic trends. Region names are abbreviated as follows: NoNord=North Nordic; SoNord=South Nordic; UK-IE-NL=

Great Britain/Ireland/Netherlands; WCE=West Central Europe; ECE=East Central Europe; Ont=Ontario; AdsCsk=Adirondack/Catskill; QuMaVt=Quebec/Maine/Vermont; AtlCan=Atlantic Canada; Apps=Appalachia; BRM =Blue Ridge Mountains.
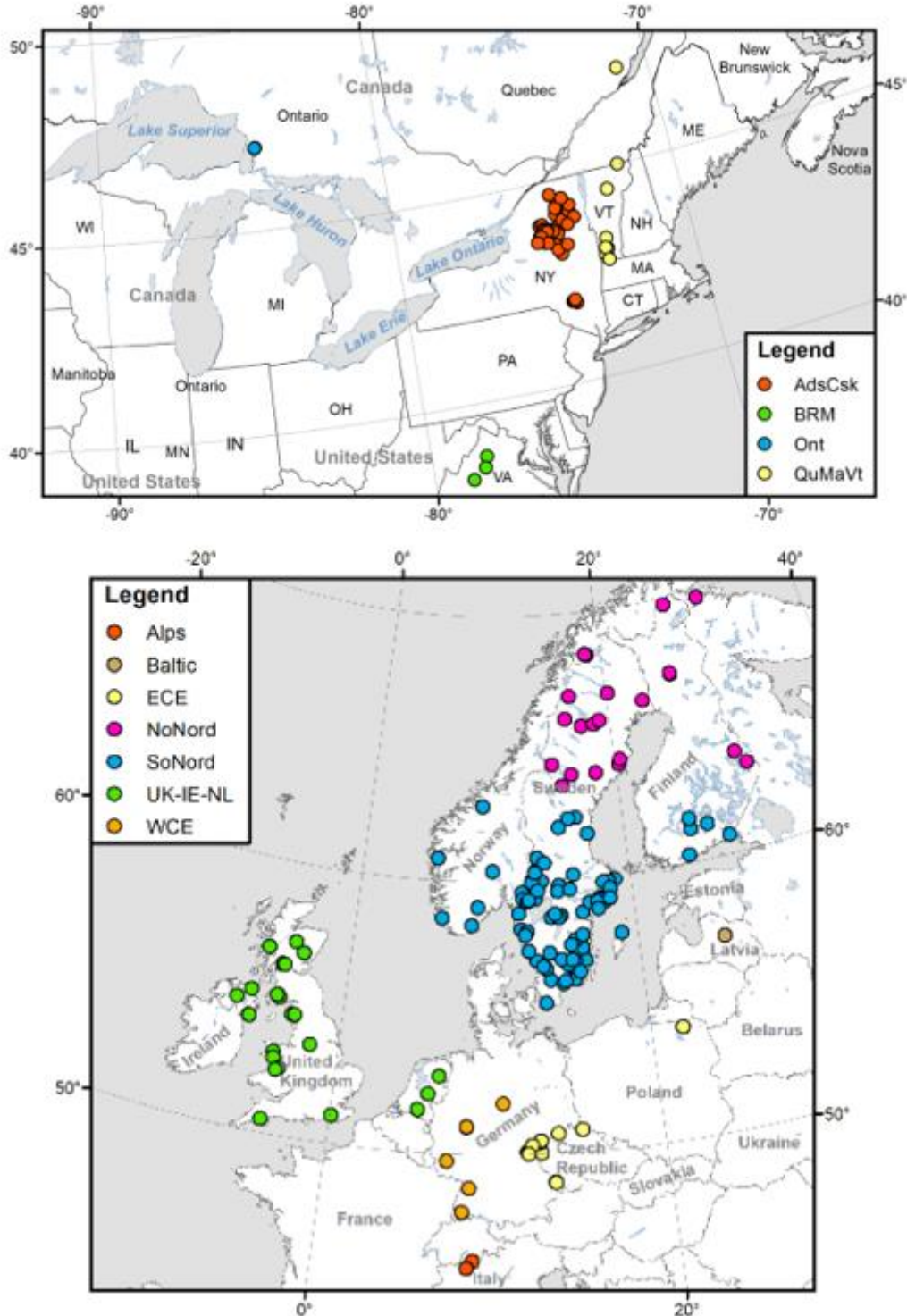
Figure 2. Maps of the 231 stations (subset of sites shown in Figure 1) included in the analysis of time series change points and pH & ANC minima. See caption of **Figure 1** for explanation or abbreviations.

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, 2010).

Three levels of quality control of water chemistry data are distinguished: in-laboratory controls in individual countries, between-laboratory controls (see e.g. Gundersen, 2019) 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 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 methods

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. Garmo et al., 2014 and list of reports from ICP Waters in the appendix).

Our analysis 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 usually 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 NO3-.
3. **pH and ANC[2]** reflect the outcome of changing balances between acid and base.
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.

Analysis of trends in other important variables such as aluminium fractions is hampered by large differences between laboratories due to lack of standardisation (see e.g. Hovind, 1998).

In regions heavily affected by sea salts (SoNord, UK-IE-NL, AtlCan), sulphate 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). All pH values were transformed to H+ concentrations prior to statistical analysis.

Annual medians were used in the statistical analysis of the central tendency of the time series. 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, an annual median was calculated for each variable. Thus, seasonality in the data only influenced the annual value and did not affect the power of the statistical tests. In the analysis of change points, initial attempts included tests on individual data points that would cover seasonal variations. However, it was decided that too few of the time series had the required sampling frequency to pursue this route (i.e. spatial cover would suffer). We therefore ended up using

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[2] ANC was calculated as $[Ca^{2+}]+[Mg^{2+}]+[Na^+]+[K^+]+[NH_4^+]-[SO_4^{2-}]-[NO_3^-]-[Cl^-]$. The brackets refer to concentration in μeq/L. Ammonium concentration was set to zero data were unavailable. ANC was not calculated if data on any of the other ions were missing.

aggregated annual data for analysis of change points as well. Annual medians, maxima and minima were used in the analysis of trends in episode severity.

The Mann Kendall test (MKT) (Hirsch and Slack, 1984) was used to identify individual monotonic trends for the periods 1990-2016, 1990-2004 and 2004-2016 from the value of the test statistic (Z-score). The MKT is robust against outliers, missing data and does not require normal distribution of data. Slopes were calculated using the Sen estimator (Sen, 1968). A paired t-test was used to test for differences between individual trend slopes for the partly overlapping periods. Regional trends in annual median values were assessed using the Regional Kendall Test (RKT) (Helsel and Frans, 2006), which gives median slopes and statistical significance of the trend. The RKT has similar strengths as the MKT and does not require normal distribution of data.

Previous trend reports indicate that changes have not occurred at a constant rate over the last decades. We wanted to take a more systematic approach to find out when time series show distinct change points during the 26-year period. In earlier trend analysis in ICP Waters this has been done "by eye" for whole regions. The selection of an appropriate test is, however, not straightforward. There are two broad approaches: 1) Fit an explicit time series model capable of representing trends and seasonalities (and including break-point detection for the trend component), or 2) fit a piecewise linear model to aggregated annual data (where the aggregation serves as a coarse way of removing seasonalities). The first approach would be more informative but requires more data. The second method is more appropriate in situations where one does not have enough information to accurately characterise seasonalities, but enough to estimate annual averages. We tried both methods on a limited number of frequently sampled sites and found that both provided plausible but not necessarily identical results. We decided to choose option 2, applying the Bayesian analysis of change point algorithm of(Barry and Hartigan, 1993), as implemented in the R package BCP. This maximises the number of sites available in the analysis. Each partition of each time series was assumed to have constant mean and variance, and the prior probability of a change occurring at each point in the series was set to a constant (0.1). A change point i.e. a point in the series where the slope changes or where there is a step change in level, was defined as a point where the posterior probability of change exceeded 75%.

2.2   Results

2.2.1 Regional patterns and temporal trends in annual median values

The strongest evidence that emission control programmes are having their intended effect is a consistent pattern of chemical recovery (decreasing sulphate and nitrate, increasing pH and ANC) across a large number of sites. The surface water chemistry of sensitive headwater catchments has changed markedly across Europe and North America (Figure 3), owing to the reductions in emissions of sulphur and nitrogen achieved under the CLRTAP. Most evident are the sulphate concentrations that have declined by up to 65 % (Table 4). Of 497 individual sites included in this study, 481 show decreasing trends and only 2 show increasing trends (Table 5). In Europe the decline has slowed down somewhat whereas in North America the trend slopes are more negative after 2002 (Table 6). Reductions in nitrate concentrations, especially in absolute values, were smaller and more subtle in most regions than for sulphate. About the same number of individual sites showed either no trend or decrease in nitrate, whereas increase was rare across all regions[3]. Chloride, another acid anion, has also decreased at many sites. These changes, reflecting reduced sulphur, nitrogen and chloride deposition, are the major drivers of reductions in base cation concentrations. Less acidic water draining through the catchment has led to lower amounts of base cations being released from the soil. Overall, base cations have decreased less than the acid anions resulting in widespread increases in ANC and pH. Organic acids, for which DOC serves as a proxy, have increased and partially replaced the mineral acids thus limiting the pH increase.

The changes displayed in Figure 3 are largely driven by reduced long range transport of air pollution. In the next paragraphs we will take a closer look at the magnitude of the trends and how they differ between regions, as well as the change points that seem evident in Figure 3. In Chapter 3 we will describe how factors other than atmospheric deposition can affect trends.

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[3] Many of the sites that showed an increase were located in southern parts of Sweden and Finland (see map in Appendix A).
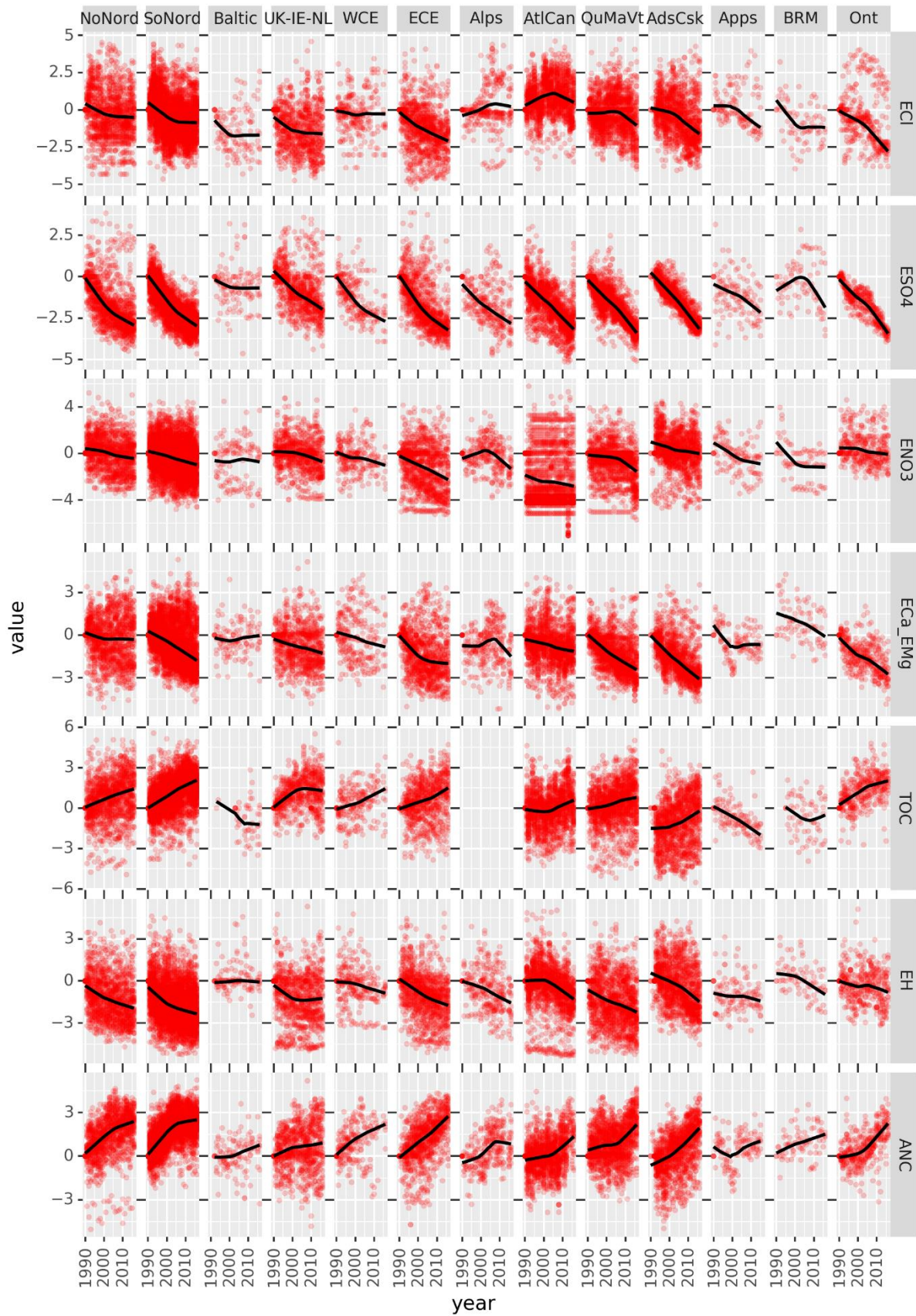
Figure 3. Time series trends between 1990 and 2016 for SO₄, NO₃, sum of Ca and Mg, TOC, H⁺ and

ANC (E refers to Equivalents charge). Each point represents the difference between the annual median value and the annual median of the first year in the time series, divided by the standard deviation. The solid lines indicate the moving average and represent lowess (Locally Weighted Scatterplot Smoothing) smoothers. The plots allow direct comparison across regions and variables. Note that the scales on the y-axis are different for different variables.

Table 4. Regional median levels for the period 2012-2016 and relative change compared to the period 1990-1994.

Region	SO_4^{2-}*	NO_3^-	Cl^-	Ca^{2+}*+Mg^{2+}*	H^+	TOC	SO_4^{2-}*	NO_3^-	Ca^{2+}*+Mg^{2+}*	Cl^-	H^+	TOC
	µEq/L	µEq/L	µEq/L	µEq/L	µEq/L	mg/L	%	%	%		%	%
NoNord	50	1.7	93	169	1.1	6.1	-41	-17	-6	-4	-45	18
SoNord	41	3.9	106	106	4.7	8.7	-61	-21	-26	-15	-55	31
Baltic	333	36	126	3501	36	24	-46	-46	-7	-44	111	-17
UK-IE-NL	33	12	240	79	4.2	6.1	-64	-25	-16	-12	-67	52
WCE	124	39	109	276	4.9	4.6	-51	-29	-23	6	-52	58
ECE	170	31	62	297	8.3	6.4	-42	-58	-10	-36	-58	68
Alps	67	50	47	220	0.20	0.9	-32	-23	-14	51	-31	
Ont	64	18	7	213	0.38	4.1	-42	-12	-8	-15	-24	2
AdsCsk	51	14	9	97	4.6	5.1	-52	-28	-26	-19	-48	26
QuMaVt	42	4.6	37	103	2.0	4.8	-49	-3	-23	59	-4	13
AtlCan	14	1.6	131	51	5.2	8.6	-63	-2	3	1	-28	17
Apps	145	9.8	42	171	2.7	1.1	-18	-49	-13	8	-47	-40
BRM	69	3.9	25	145	0.70	1.5	-3	-78	-3	-6	-24	

Table 5. Number of sites showing positive or negative trends (p<0.05, MK-test) in sulphate, nitrate, base cations, chloride, ANC, hydronium ions or organic carbon. The "+" and "-"signs represent increasing and decreasing trend, respectively. No trend is abbreviated "n.t.".

Region	SO_4^{2-}*			NO_3^-			Cl^-			Ca^{2+}*+Mg^{2+}*			ANC			H^+			TOC		
	-	+	n.t.	-	+	n.t.	-	+	n.t.	-	+	n.t.	-	+	n.t.	-	+	n.t.	-	+	n.t.
NoNord	47	1	3	25	1	25	17	1	33	16	8	27	0	45	6	31	1	19	0	31	18
SoNord	149	0	1	73	11	66	81	4	65	99	11	40	1	138	11	100	0	50	0	120	26
Baltic	3	0	1	2	0	2	3	0	1	0	0	4	0	2	2	0	0	4	0	0	0
UK-IE-NL	26	0	1	14	3	10	15	0	12	12	1	14	1	18	8	16	0	11	0	19	4
WCE	5	0	1	3	0	4	0	0	7	3	0	4	0	4	0	5	0	2	1	4	2
ECE	39	0	2	27	3	10	24	2	13	23	5	13	0	30	1	35	0	7	0	17	12
Alps	10	0	1	5	1	5	4	4	3	2	2	7	0	6	5	5	0	6	0	0	0
Ont	15	0	0	5	0	10	10	1	4	11	0	4	0	13	2	5	0	10	0	11	4
AdsCsk	56	0	0	17	1	38	39	5	12	53	0	3	1	48	7	38	1	17	0	33	23
QuMaVt	63	0	0	37	3	19	11	2	50	48	0	15	0	30	29	42	0	9	5	28	29
AtlCan	63	0	0	17	3	43	3	17	43	29	1	33	0	26	30	42	0	21	0	14	49
Apps	3	0	2	3	0	2	3	1	1	3	0	2	0	1	4	1	0	4	5	0	0
BRM	2	1	0	3	0	0	1	0	2	2	0	1	0	1	2	2	0	1	0	0	0

Table 6. Overview of direction and change in direction of trends 1990-2016. Pluss and minus represent increasing and decreasing trend (RKT), respectively, over the whole period. Blue and red colour indicate whether slopes for the period 2002-2016 are higher (less negative or more positive) or lower (less positive or more negative), respectively, than for the period 1990-2004. Abbreviation n.d. refer to "no data" and n.t. to "no trend". Mean Sen slopes for the two shorter periods are tabulated in Appendix B.

| Variable	Europe							North America						N. Europe	N. America
	NoNord	SoNord	Baltic	UK-IE-NL	WCE	ECE	Alps	Ont	AdsCsk	QaMaVt	AtlCan	Apps	BRM		
SO_4^{2-}*	-	-	-	-	-	-	-	-	-	-	-	-	n.t.	-	-
NO_3^-	-	-	-	-		-	-	-	-	-	-	-	-	-	-
Cl^-	-	-	-	-		-	+	-	-		+	-		-	-
Ca^{2+}*+Mg^{2+}*	-	-	n.t.	-	-	-	n.t.	-	-	-	-	-		-	-
ANC	+	+	+	+	+	+	+	+	+	+	+	+	+	+	+
H	-	-	n.t.	-	-	-	-	-	-	-	-	-	-	-	-
TOC	+	+	n.d.	+	+	+	n.d.	+	+	+	+	-	n.d.	+	+

Trends in sulphate across regions

Sulphate concentrations have declined in all regions, except the Blue Ridge Mountains (Figure 4) where deeply weathered soils with a large capacity to adsorb sulphate delay the response (Church et al., 1990). The regional trends over the whole period were remarkably similar in heavily affected regions across both continents. The lines indicating the trends in Figure 3 and the slopes of the individual trends (Table 6) show that declining trend slopes became less steep in Europe and steeper in North America around 2000. This can be linked to different timing of abatement policies/economic recession that have led to strong reductions in emissions at both continents (Monks et al. 2009; Strock et al. 2014). Emissions reductions in Europe slowed down around the millennium shift and became more distinct in North America around 2005. Retainment of sulphate in the catchments and subsequent release can dampen the effect of changes in deposition on concentration in runoff. Vuorenmaa et al. (2017) found that catchments in Europe retained sulphate in the early and mid-1990s, but this shifted towards a net release in the late 1990s.

The analysis of change points showed that in Europe breaks in the rate of change or changes in level were more frequent in the 90s than after 2000 (Figure 5), indicating smoother trends in recent years. In the North Nordic and central European regions, change points were more frequent before 1995 than after, which is somewhat earlier than the moving averages in Figure 3 suggest. The change point analysis does not consider the direction of change, i.e. it is possible that changes cancel each other out and do not impact the moving average. In the South Nordic region, the maximum density of change points identified occurred around 1998, which is consistent with the observed flattening of declining trends. The distinct peak in change points indicates that the major changes in the individual time series are synchronous across the region and that they are primarily driven by changes in long range transported air pollution, which acts on a regional scale. In the UK-IE-NL region the change points occurred over a wider time span. In North America, where only three of the regions were deemed to

have sufficient data for the analysis, the timing of change points was also more widespread than in most of the European regions, except UK-IE-NL.
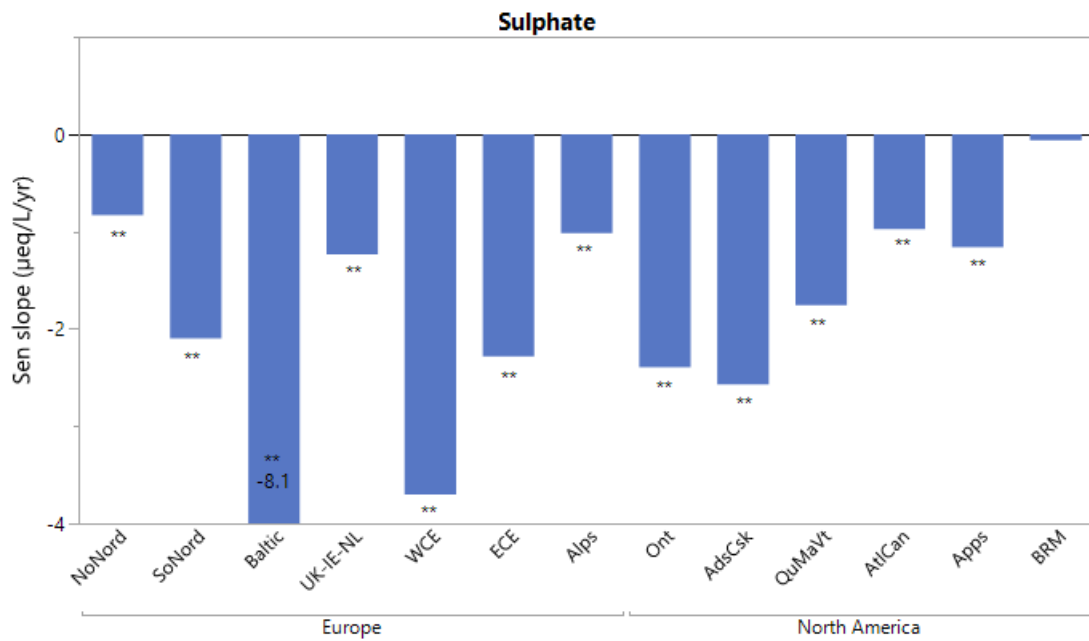
Figure 4. Median regional trend slopes in non-marine sulphate concentration for the period 1990-2016. One and two asterisks indicate statistical significance level of 0.05 and 0.01 according to the Regional Kendall test for trend.
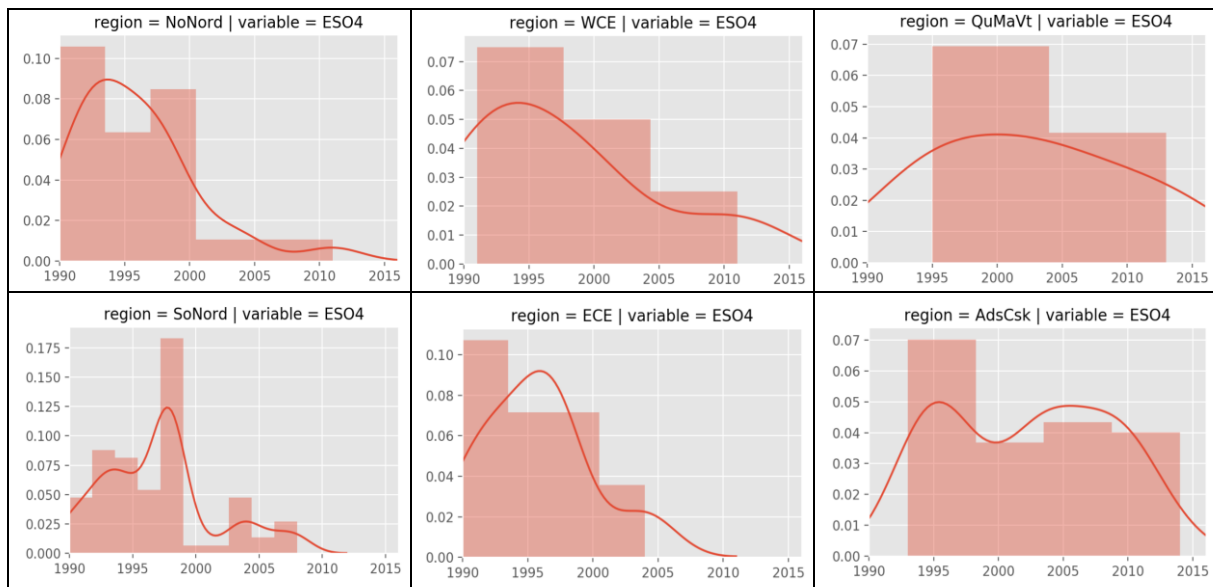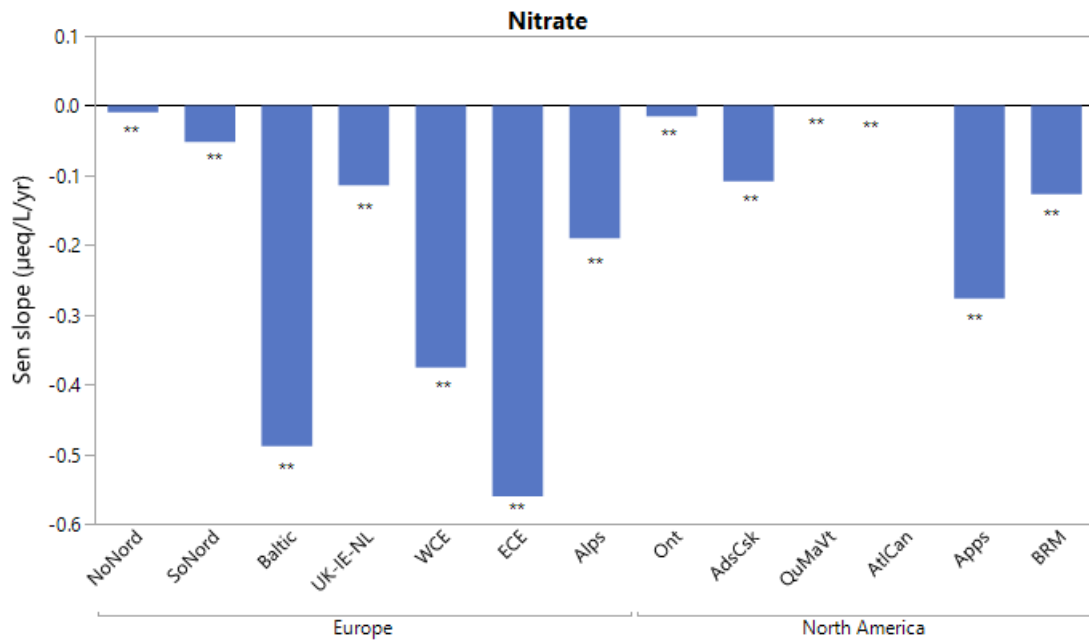
Figure 5. Relative frequency of change points in non-marine sulphate detected with BCP (Bayesian analysis of change point) analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

Trends in nitrate across regions

Nitrate concentrations have decreased in all regions even if the absolute change is small in some of them (Figure 6). The greatest declines were observed in the regions with the highest levels (Table 4). While reduced deposition of sulphur is the main driver behind the extensive decline of freshwater sulphate, the controls of nitrate concentration are affected by the biogeochemical cycling of nitrogen in soils. Factors that can affect soil processes can therefore also affect nitrate leaching. Furthermore, insect attack on vegetation can cause large temporal variation in nitrate concentration as observed in sites from Blue Ridge Mountains, Quebec and the East Central European region (Eshleman et al., 1998; Houle et al. 2009; Oulehle et al., 2019). Despite such confounding factors, the consistent decline of nitrate observed at the regional level strongly suggests that reductions in long-range transported air pollutions are effective and lead to reduced nitrate in surface waters.

Nitrate time series showed more frequent change points in the 90s than in the subsequent years (Figure 7). This pattern was evident for both European and North American regions, but most striking in the UK-IE-NL region. The change points in the time series did not indicate synchronous changes, which is consistent with the mixed pattern of the moving averages in Figure 3 and the lack of differences between individual slopes in the first and second part of the period (Table 6).

Figure 6. Median regional trend slopes in nitrate concentration for the period 1990-2016. One and two asterisks indicate statistical significance level of 0.05 and 0.01 according to the Regional Kendall test for trend.
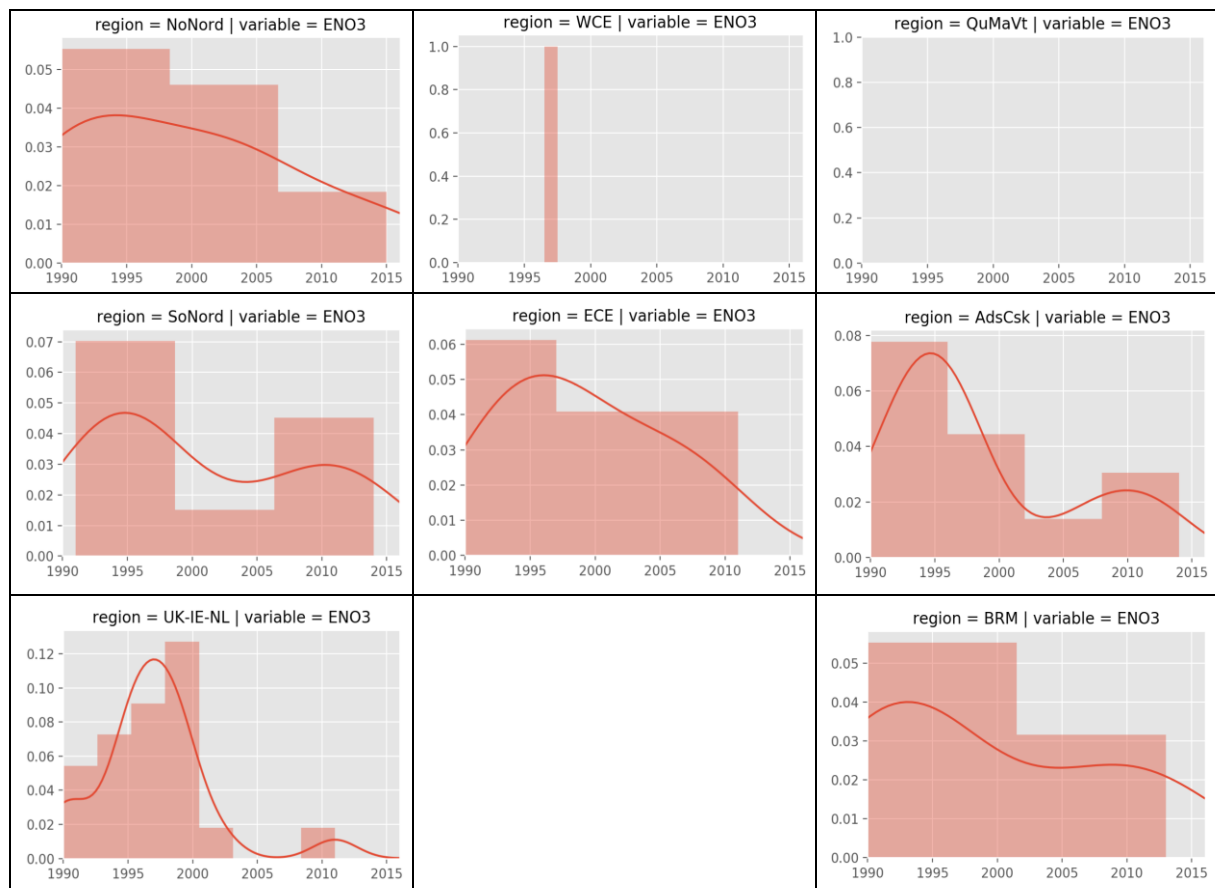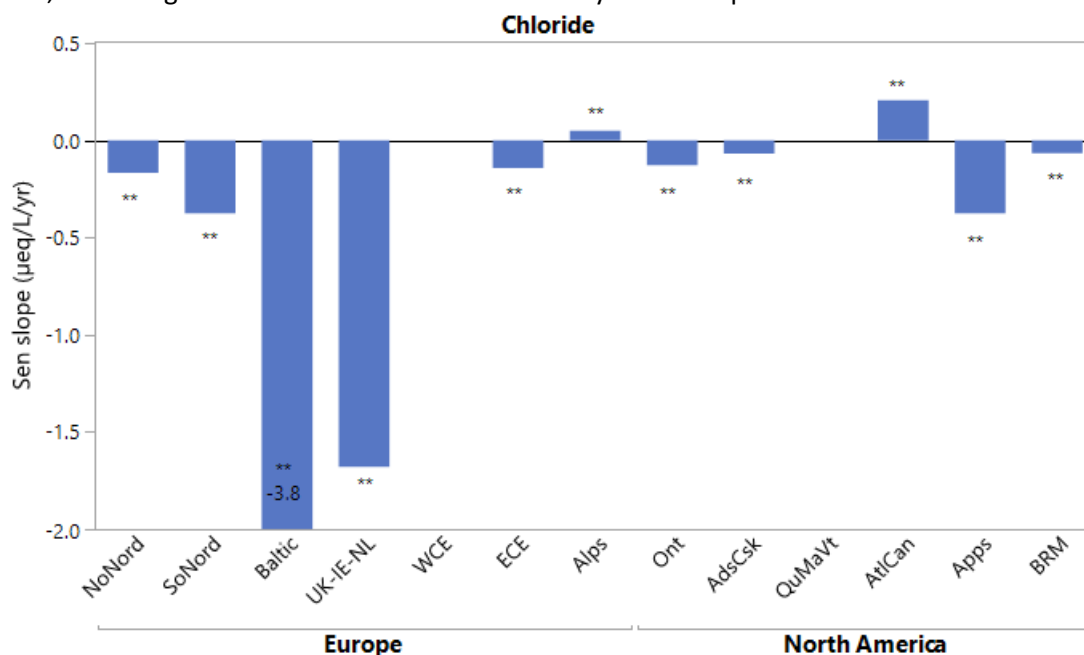

Figure 7. Relative frequency of change points in nitrate detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

Trends in chloride by region

Chloride, the corresponding anion of hydrochloric acid (HCl), is a strong acid like sulphuric and nitric acid. Chloride tends to be more mobile in the soil than sulphate and nitrate, making HCl a very potent acidifier of surface waters. However, it has largely been overlooked as a potential long-range acidifying agent. A possible explanation for the lack of focus on HCl is natural variation caused by sea-salt aerosols blowing across land from the ocean surface. This may have masked trends in regions close to the sea. Dechlorination of sea salt aerosols after interaction with sulphate/nitrate ions from anthropogenic emissions can also complicate the link between chloride emission and chloride deposition (Evans et al., 2011). Measures that have reduced the emission of sulphur, such as substituting coal with gas and rinsing of flue-gas, have also clearly been limiting anthropogenic emissions of chloride. Emissions of HCl have therefore decreased in line with sulphur (Evans et al., 2011). Recently, decreasing trends of chloride in headwaters has been reported in the UK (Evans et al. 2011) as well as Central Europe (Kopáček et al., 2015; Oulehle et al., 2017).

The results of our analysis show decreasing chloride trends in 9 of 13 regions (Figure 8). In the Northern European regions, the negative slopes were steeper than for nitrate. The median slope in e.g. the UK-IE-NL regions may seem surprisingly high compared to the one for sulphate but is within the range of values presented by Evans et al. (2011) for the UK. In the Nordic regions and Appalachia as well, the declines in chloride were substantial compared to sulphate and larger than those for nitrate. The apparent increases in the Alps and Atlantic Canada are not representative for the whole of the regions. The concentrations in Swiss lakes showed no trend or decrease whereas increase was observed in 4 of the 6 Italian rivers. More information about trends and sources of chloride (and sodium) in the Alps can be found in Rogora et al. (2015). In Atlantic Canada the sites with increasing chloride concentrations are the ones found on Newfoundland and eastern parts of Nova Scotia. The sites southwest of Halifax showed no trend or decrease (see Appendix A).

The timing of change points in the chloride time series were in most regions similar to those for sulphate, indicating that the ions have been affected by the same processes.

Figure 8. Median regional trend slopes in chloride concentration for the period 1990-2016. One and two asterisks indicate statistical significance of 0.05 and 0.01, according to the Regional Kendall test for trend.
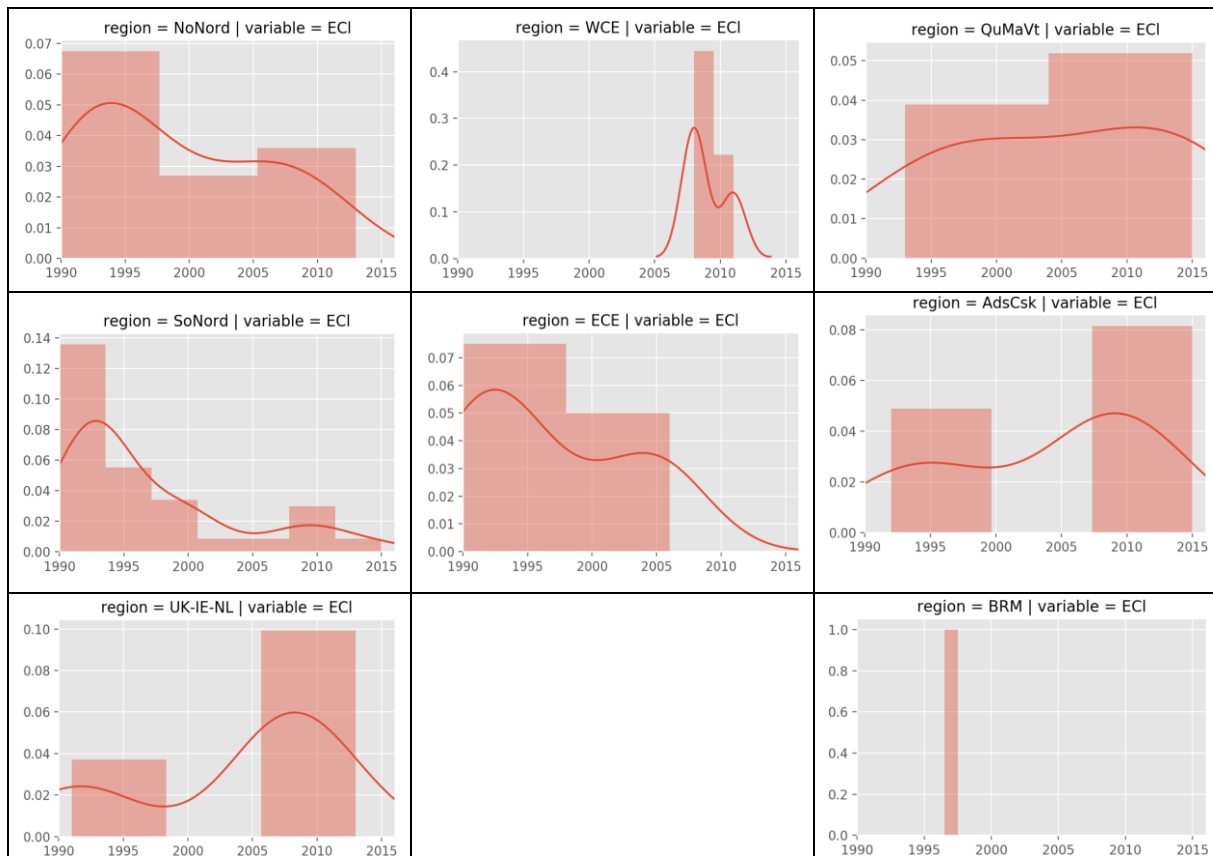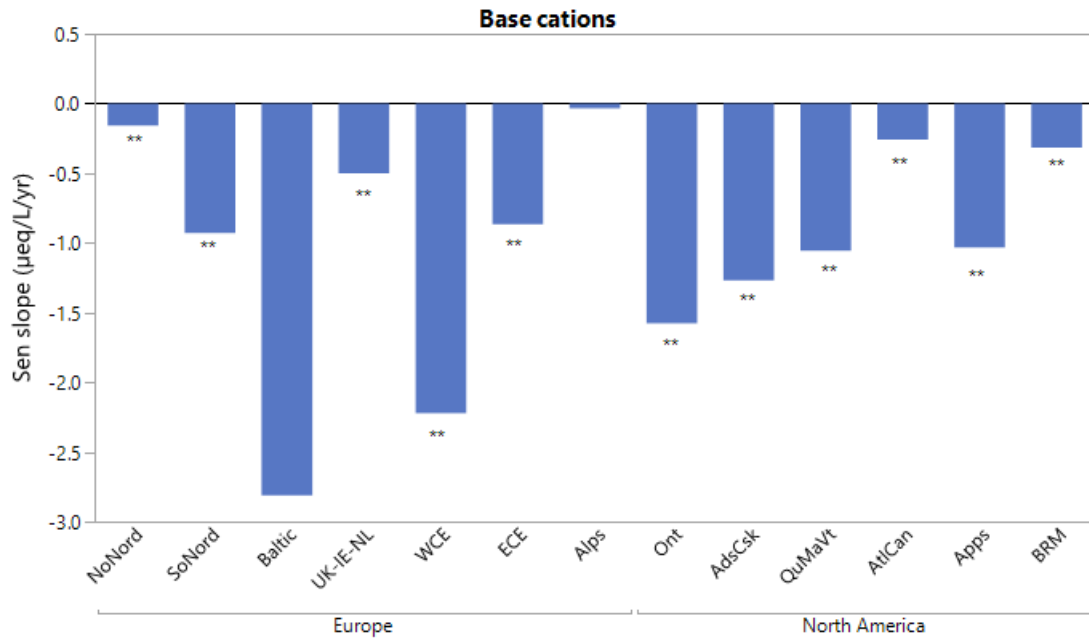
Figure 9. Relative frequency of change points in chloride detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

Trends in base cations across regions

One of the expected responses of catchments to decreasing acid inputs is the reduced leaching of base cations because of increased pH and cation scavenging by cation exchange sites (Galloway et al., 1983). All regions except the Baltic and the Alps showed decreasing concentrations of non-marine base cations between 1990 and 2016 (Figure 10). The regional trends in base cations were correlated to regional trends in sulphate (trend $Ca^{2+}*+Mg^{2+}*= 0.54$ x trend $SO_4^{2-}*$, $r^2=0.80$, $n=13$)[4]. There are smaller areas showing small but consistent increases in non-marine base cations, especially within the North Nordic region. This increase is not driven by acidification, since acid anions were decreasing during the same period, but more likely climatic variations as observed in some alpine waters (Kopáček et al., 2017; Rogora et al., 2020).

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[4] Including nitrate trends in the model did not explain more of the variation.

Figure 10. Median regional trend slopes in base cation concentration for the period 1990-2016. One and two asterisks indicate statistical significance of 0.05 and 0.01, according to the Regional Kendall test for trend.
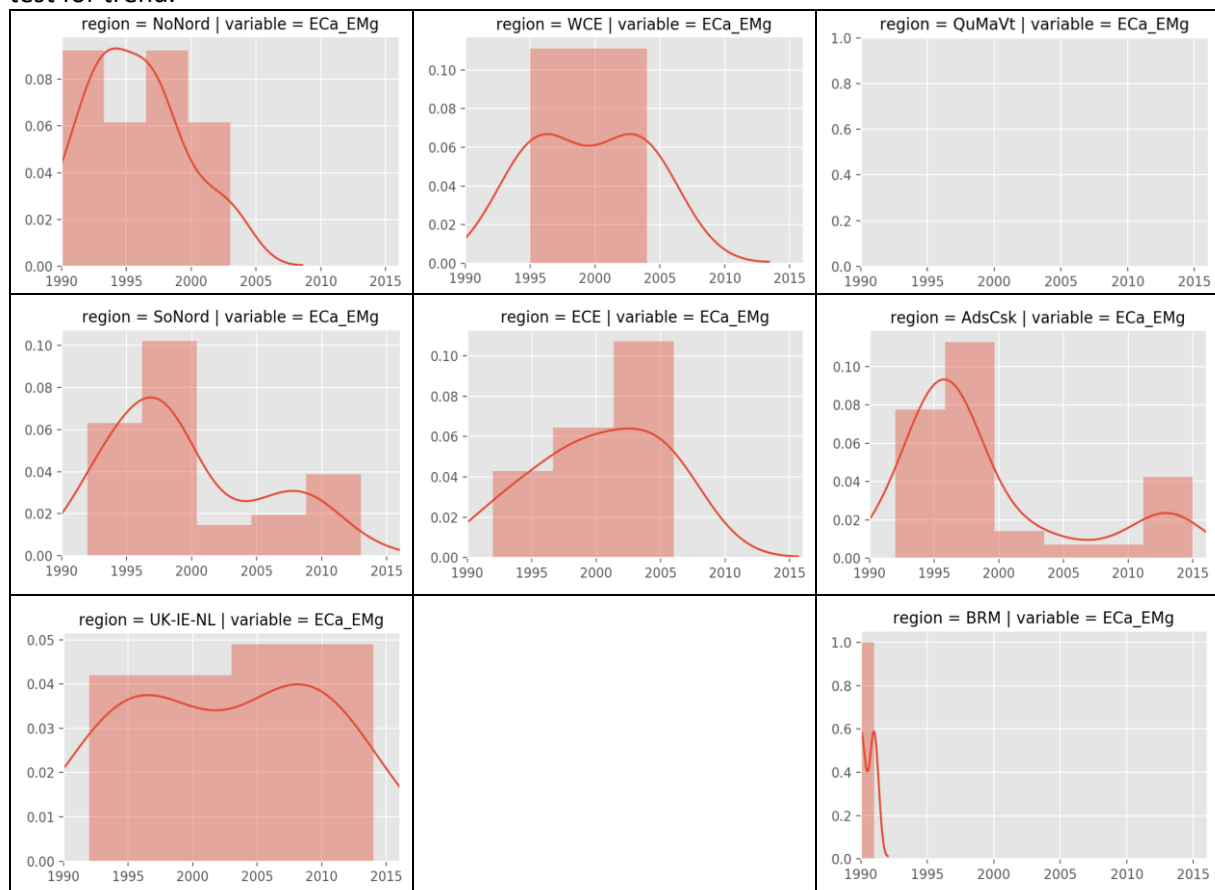

Figure 11. Relative frequency of change points in non-marine base cations detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

For the Nordic regions and UK-IE-NL, the distributions of change points in the base cation time series (Figure 11) resembled those of non-marine sulphate, in that the periods of highest frequency coincided. In the East Central European region, the maximum frequency of change points occurred a few years later than for sulphate. In the Adirondack/Catskill region in North America, change points for base cations occurred more frequently in the 90s than after 2000.

Trends in ANC by region

Calculated charge balance ANC is a widely used indicator of acidification and the susceptibility of natural waters to acidification. The steeper negative regional trend slopes for $SO_4^{2-}*$ relative to those for base cations, combined with the regional signal of decreasing NO_3^- and Cl^-, should result in increasing ANC. Indeed, all regions show positive regional trends in ANC between 1990 and 2016 (Figure 12), indicating chemical recovery from acidification. The greatest improvement was found among the regions that have had the highest acid deposition.
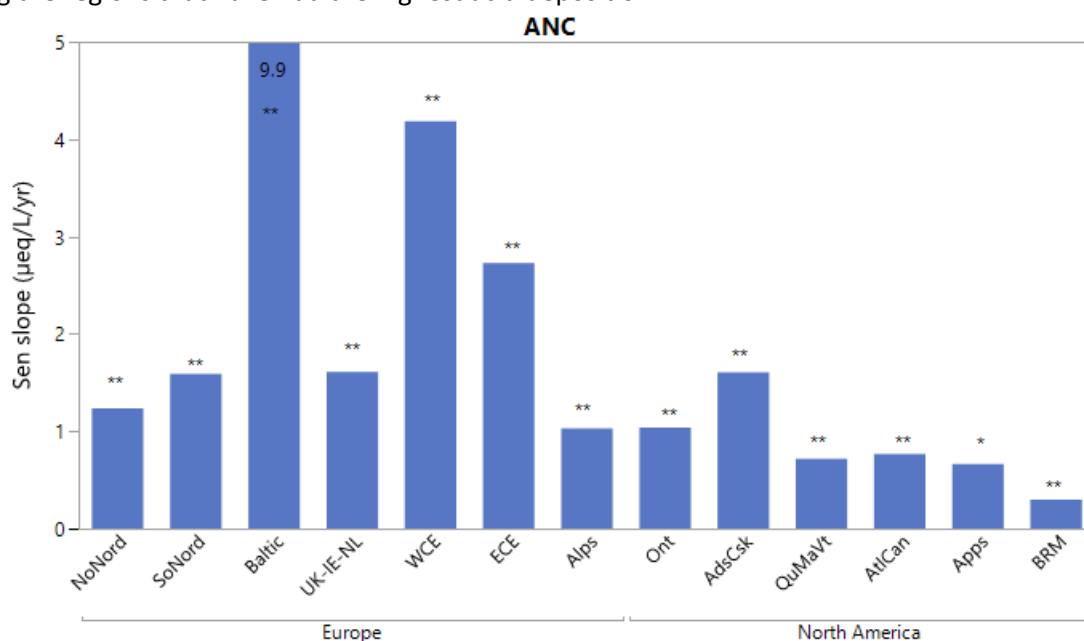
Figure 12. Median regional trend slopes in ANC for the period 1990-2016. One and two asterisks indicate statistical significance of 0.05 and 0.01, according to the Regional Kendall test for trend.

The central tendency in most of the European regions is that the rate of increase in ANC has declined over the last 1.5 decades, whereas ANC increase in the North American regions has accelerated (Figure 3, Table 6). This is not so evident from the change point analysis. The distributions of change points in the ANC time series across the period (Figure 13) were similar to those for non-marine sulphate and base cations.
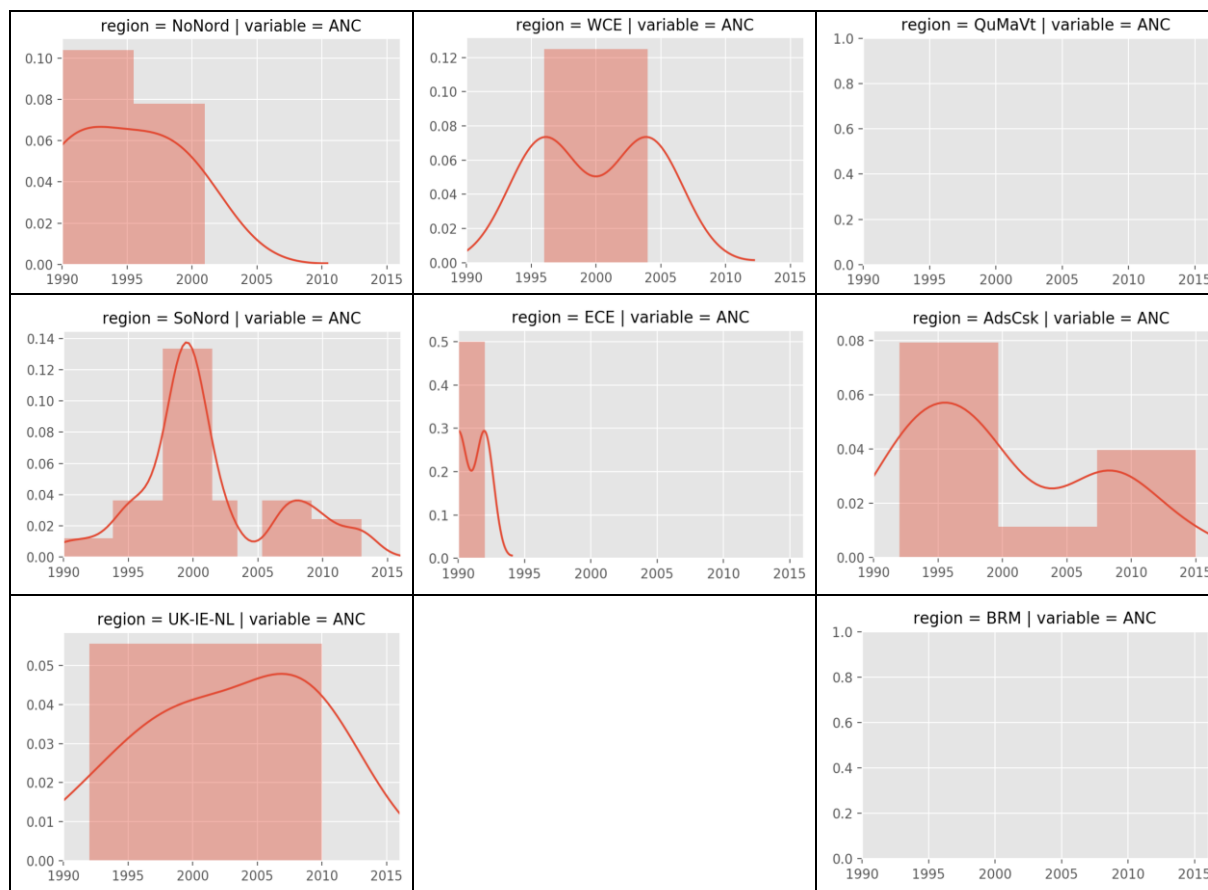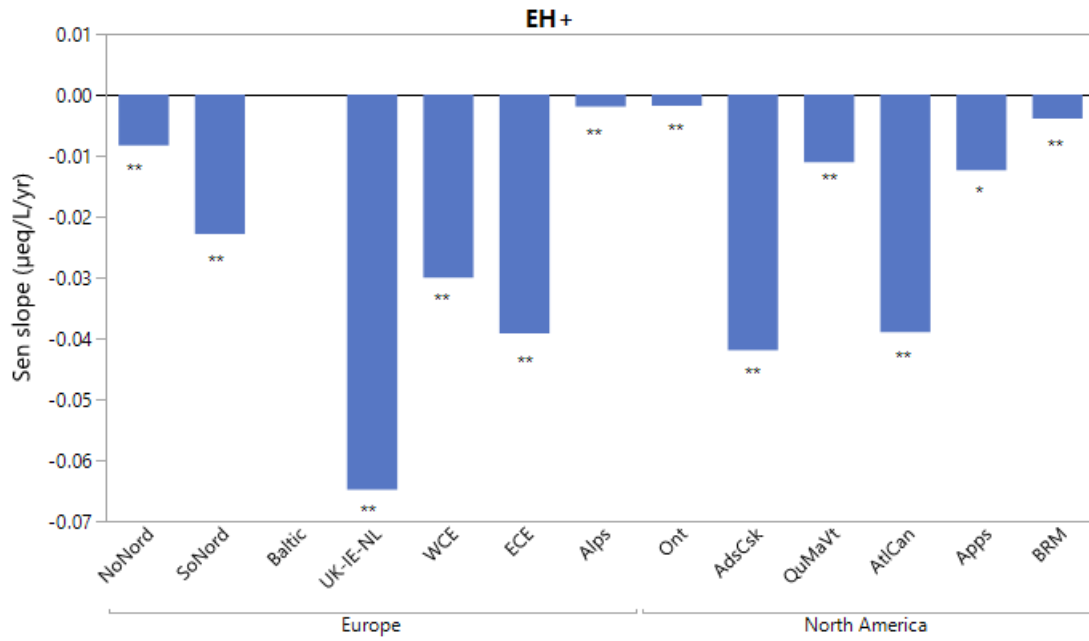
Figure 13. Relative frequency of change points in ANC detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

Trends in pH (H⁺) by region

Most processes in natural aqueous systems are pH-dependent, and pH is an important parameter for understanding biological consequences of acid deposition. Whereas ANC (and alkalinity) is a capacity factor indicating susceptibility or degree of acidification, pH (and inorganic aluminium fractions) is an intensity factor, more directly linked to toxic effects. All regions, except the Baltic, showed decreasing H⁺ concentration between 1990 and 2016 (
Figure **14**). Increasing pH is a clear indicator of chemical recovery from acidification and shows that living conditions for vulnerable biota are improving.

The trends in H⁺ are consistent with those observed for other variables on a regional scale, and so are the changes in trend slopes (Figure 3, Table 6). Also consistent with patterns for the other parameters is the prevalence of change points in the 90s and up to year 2000.

Figure 14. Median regional trend slopes in hydronium ion activity for the period 1990-2016. One and two asterisks indicate statistical significance of 0.05 and 0.01, according to the Regional Kendall test for trend.
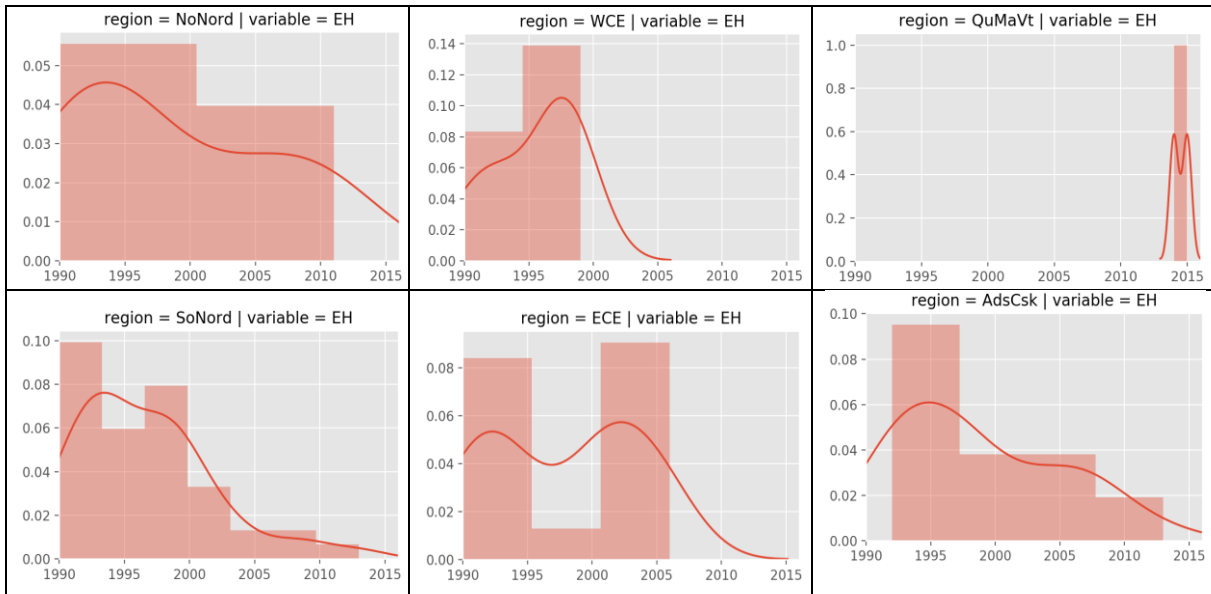
Figure 15. Relative frequency of change points in hydronium ion (H+) detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.
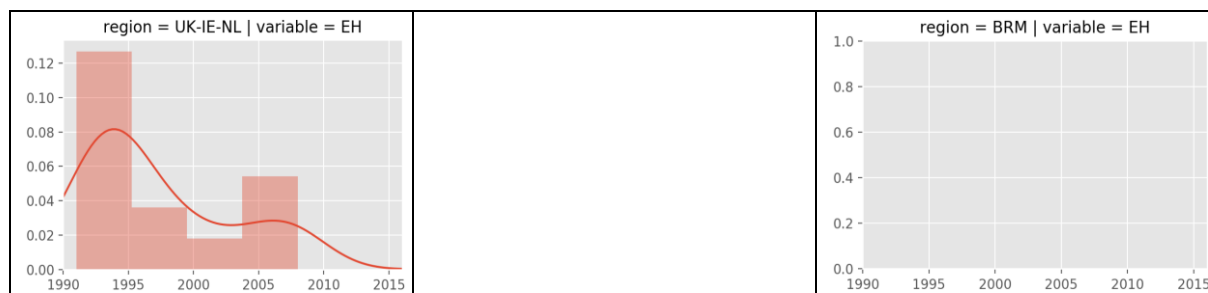
Trends in DOC by region

Dissolved organic carbon (DOC) is a key component of aquatic chemistry, e.g. as an indicator of natural organic acidity. All regions where DOC data were available, except Appalachia, showed increasing DOC concentrations between 1990 and 2016 with median rates varying between 0.03 and 0.11 mg/L/y (Figure 16). There are several drivers of rising DOC, but analysis of ICP Waters data has demonstrated that for the headwaters in the regions considered here, DOC trends are strongly correlated with trends in deposition chemistry and acid sensitivity of the catchment (base cation stores in the soil) (Monteith et al., 2007). Declining ionic strength of soil solution can also drive DOC upwards in non-acidified catchments (Hruška et al. 2009). Recent analysis suggest that climate also plays a role. The link between rising DOC and declining sulphate concentrations indicates that rising DOC concentrations are integral to the process of chemical recovery from acidification. Partial replacement of mineral acidity by organic acidity explains why responses are more muted than had once been anticipated (Battarbee et al., 2005; Erlandsson et al., 2010).

The declining concentrations observed for the five clearwater stream sites in the Appalachians (see Appendix A) are interesting and indicate that drivers other than reduced acidification are important. The cause of the decline is not clear. Climatic factors can exert influence on seasonal and interannual variations (Clark et al., 2010) and may affect DOC trends. "Greening", i.e. increases in terrestrial primary productivity, and afforestation can affect DOC trends over longer time scales (Finstad et al., 2016; Škerlep et al. in press).

The moving averages representing the central tendencies of the DOC trends mirror images of those for H^+ in many regions (Figure 3), demonstrating the importance of recovery from acidification as a driver. The change points, however, were delayed according to the BCP analysis; in the Nordic and UK-IE-NL regions the maximum frequency of change points in the DOC time series occurred around 2005, approximately 10 years later than for sulphate and H^+. This could result from potential non-linear relationships between SO_4^{2-} decline and DOC increase. In the UK-IE-NL region the timing of the change points reflects the point at which DOC concentrations appear to plateau.
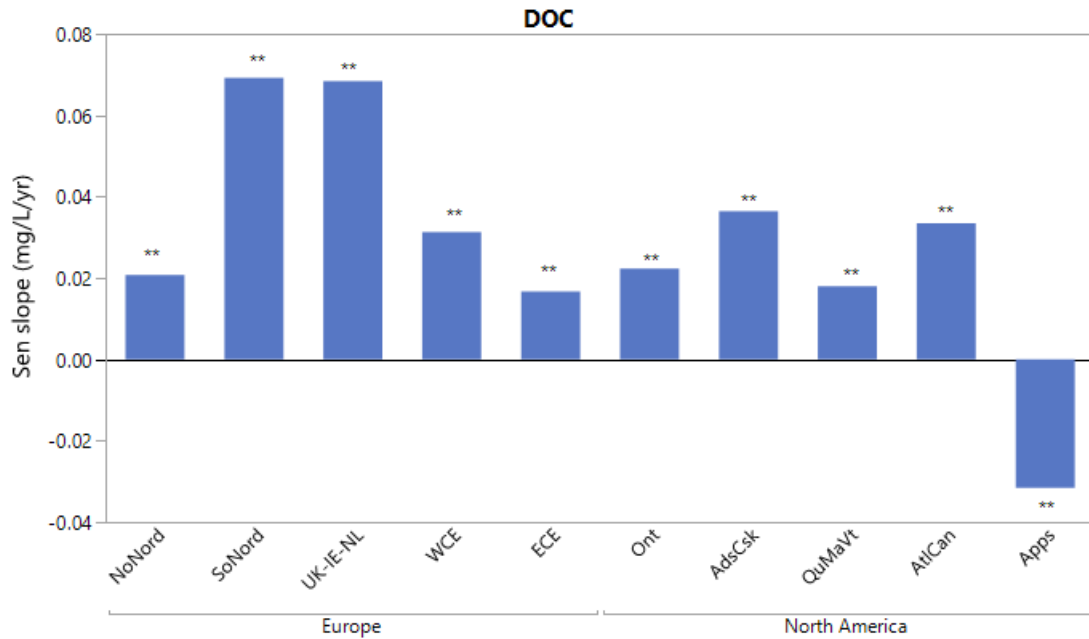
Figure 16. Median regional trend slopes in dissolved organic carbon concentration for the period 1990-2016. One and two asterisks indicate statistical significance of 0.05 and 0.01, according to the Regional Kendall test for trend.
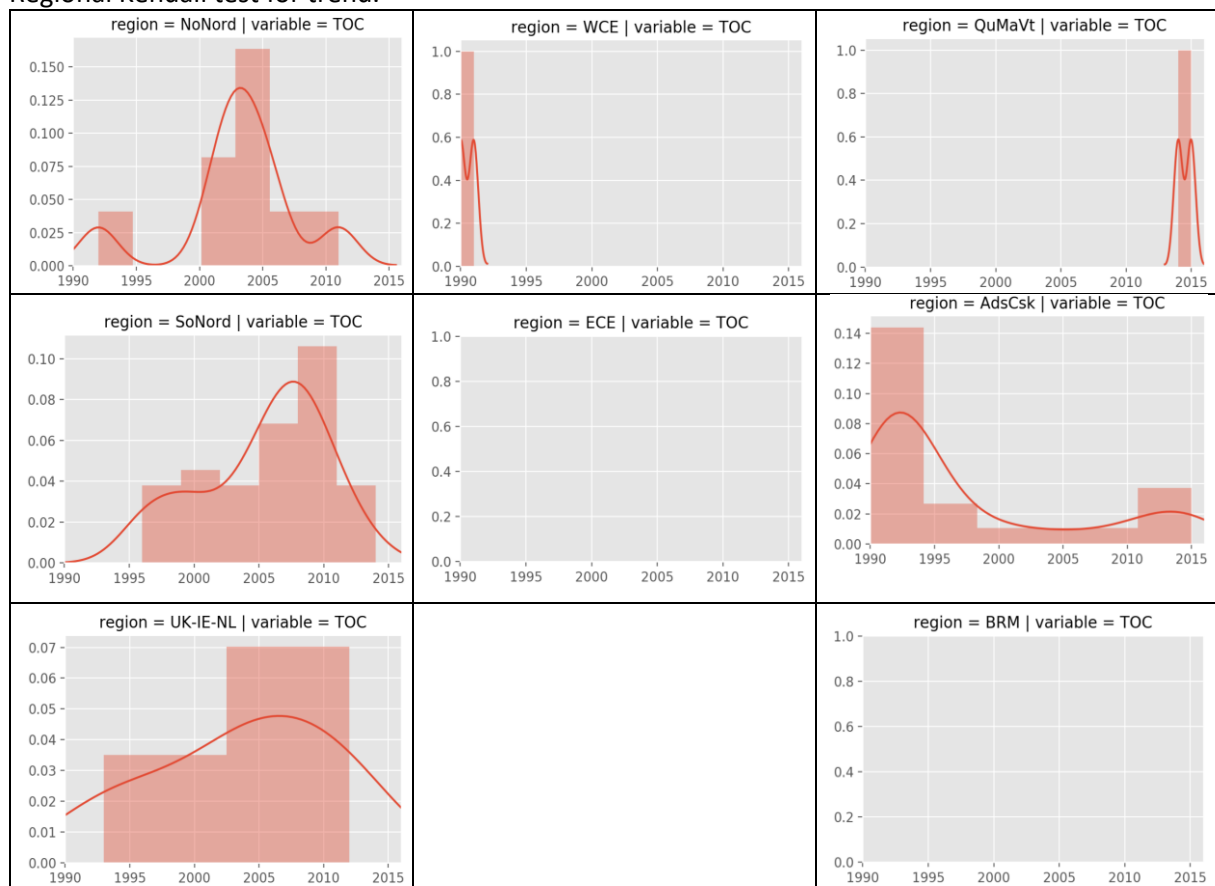

Figure 17. Relative frequency of change points in DOC detected with BCP analysis of individual time series. The pink bars are normalised histogram and the red lines kernel density estimates showing the probability densities of change points between 1990 and 2016.

2.2.2 Trends in episode severity (pH and ANC minima)

The last subchapters as well as earlier trend reports show that average water chemical conditions have improved in all regions. Biological recovery has lagged behind (e.g. Battarbee et al., 2014; Keller et al., 2018; Nedbalová et al., 2006) but is responding (see e.g. Hesthagen et al., 2011; Velle et al., 2016). Several explanations for the delay have been suggested, including dispersal limitations, interactions with other stressors and indirect food web effects (see e.g. Gray et al., 2016). Recurrent acidic episodes, i.e. the transient acidification linked to snowmelt, heavy rain or sea-salt deposition events, is another hypothesis that has been put forward to explain hysteresis of biological recovery (see e.g. Kowalik et al., 2007). A recent study on benthic algae at GEOMON catchments in the Czech Republic showed that acidic episodes were more important for algae biodiversity than annual average chemistry (Schneider et al. 2018). If occasional extreme conditions are limiting biological recovery, the potential for improvement will depend on the extent to which their severity is declining. The question therefore arises if the encouraging trends in annual averages also represent a development towards less severe episodes.

The problem can be illustrated by a brief look at results from 4 Norwegian calibrated catchments that are also ICP Waters sites (Figure 18). The results show that for 3 of the 4 catchments, the annual median pH has improved at approximately the same rate as the minimum pH[5]. At the most sea salt exposed station (OVELV 19 23), however, there has hardly been any increase in the annual minimum pH since year 2000 while annual median pH has increased from about 5.2 to 5.7. Neither is there a trend in the concentration of labile aluminium at the annual minimum pH since 2000 (not shown). It therefore seems likely that the episodes at OVELV 19 23 are equally severe as they were around year 2000, despite considerable decrease in acid deposition.
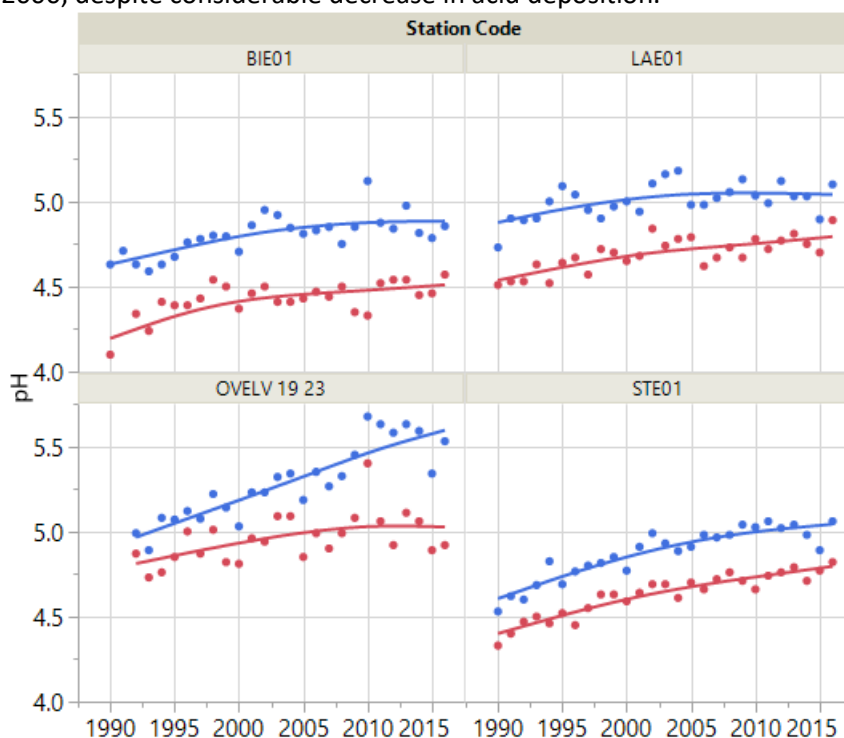
Figure 18. Annual median pH (blue) and annual minimum pH (red) at 4 Norwegian ICP Waters stations sampled weekly.

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[5] Bear in mind that this implies that the annual maximum H^+ activity has decreased by more than the annual median because pH is defined as the negative logarithm of the H^+ activity.

A first comparison of time series in annual extremes versus annual medians shows that the trends are consistent (Figure 19). The tendencies are very similar, especially for ANC[6]. This is perhaps not surprising considering the long period and decrease in acid deposition that has occurred between 1990 and 2016 at the ICP Waters stations. More interesting is how common the diverging trends observed for OVELV 19 23 are at other frequently sampled stations, and if there are differences between regions and between rivers and lakes. The results show that for 84% of the ANC time series and 77 % of the pH time series there is no trend in the difference between annual median and minimum. There are no negative trends for annual medians of these parameters, implying that overall the annual minima tend to increase in line with the median values (as in 3 of 4 of the stations represented in Figure 19). For time series that do show trend in the difference over time, decreasing trend is more common than increase (13 % versus 3 % for ANC and 14 % versus 9 % for pH), i.e. the annual minima increase at a higher rate than the median values. Almost all of the sites that show increasing trend in the differences over time are located in the South Nordic region, the UK-IE-NL region or Adirondack-Catskill (Figure 20). It is not clear if this finding mainly reflects that these regions have a large proportion of the most frequently sampled sites, or if they indeed are more prone to severe episodes. For the UK-IE-NL region there appears to be a higher frequency of diverging trends for pH compared to the other regions. Monteith et al. (2014), however, found that the magnitude of acid pulses at UK sites had declined at a similar rate to mean acidity.
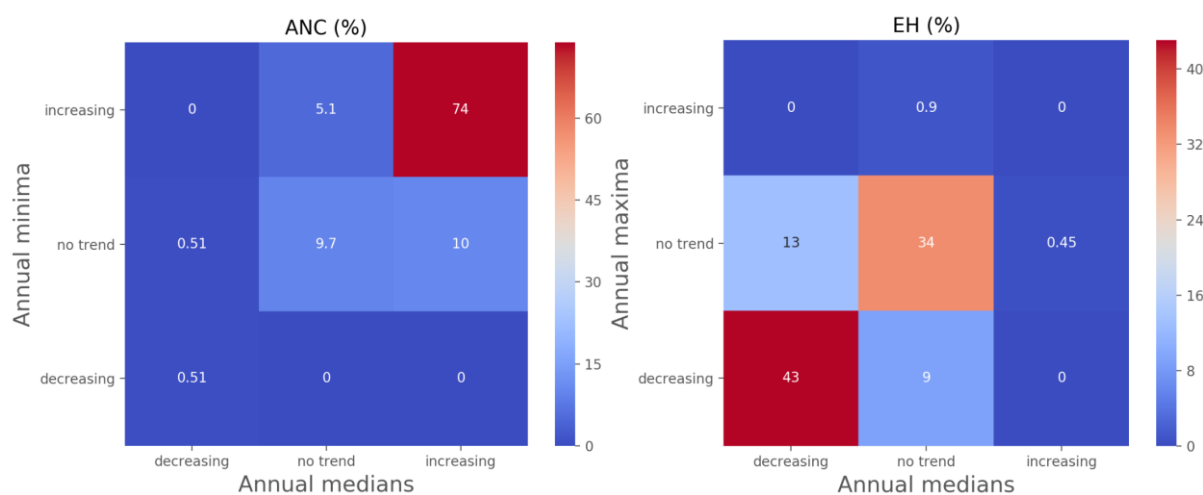
Figure 19. Percentage of stations showing decreasing, increasing or no trend for the period 1990-2016 in annual minimum and annual maximum ANC (left), and annual median and annual maximum hydronium ion activity (right).

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[6]In this analysis we chose to focus on ANC and pH because these are the parameters most closely linked to biological effect of the ones we consider in this report.
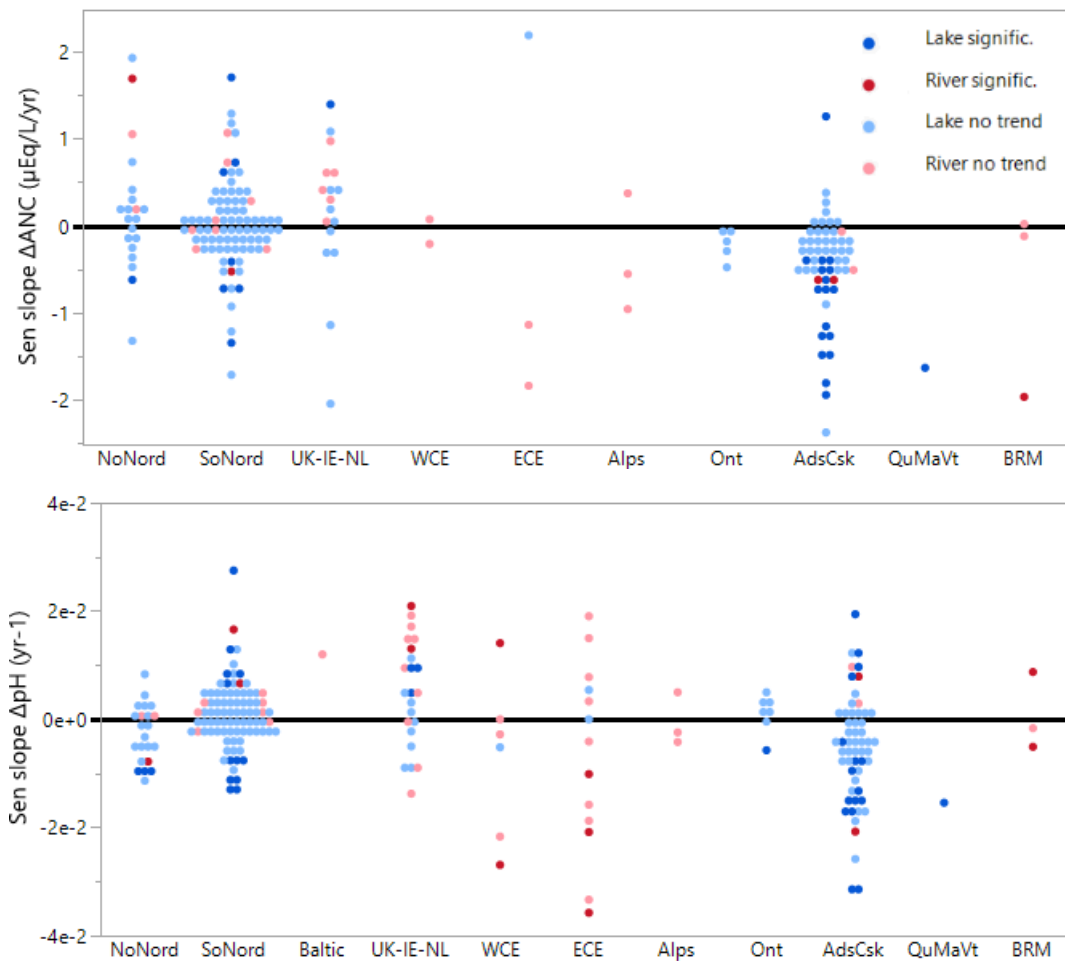
Figure 20. Trends in the difference between annual median and annual minimum pH (bottom) and ANC (top). ANC and pH are increasing. Positive trends in ΔANC and ΔpH is therefore not arising because of increasing difference between two negative slopes.

For pH, there is a higher proportion of river sites than lake sites that showed diverging trends in annual medians and minima over time. This is perhaps to be expected since lakes are larger and more able to dilute spikes of run-off that can create acid episodes. It should also be mentioned that with monthly sampling of rivers, i.e. the minimum criteria used, one is unlikely to capture the truly most severe episode of the year. The main conclusion that can be drawn from this analysis is that for a large majority of sites, the improvement in the central tendency of the water chemistry also represents a lessening in the severity of the episodes.

2.3   Conclusions

Sulphate concentrations have declined at 481 of the 497 sites considered, and 10 of 13 regions show more than 40 % decline for the period 1990-2016. Nitrate has declined in all regions, although many of the individual stations show no trend. Even chloride show declining trends in 9 of 13 regions, and in Northern Europe the negative slopes are steeper than for nitrate. Change points were more prevalent in the time series for sulphate and nitrate in the 90s than after year 2000, indicating smoother trends in recent years. On a regional scale, base cation concentrations have declined by approximately half the equivalent change in concentration of sulphate. All regions show increasing ANC between 1990 and 2016. pH has increased in 12 of 13 regions, despite partial replacement of mineral acidity by organic acidity manifested as an increase in TOC or DOC. The trajectories of sulphate, ANC and pH indicate that recovery has slowed down in Europe and accelerated in North America. This can be linked to different timing of regulatory measures that have led to strong reductions in emissions at both continents.

Acidic episodes, i.e. the annual extremes, have become less severe in line with the recovery of average chemistry. There were only a few exceptions where the change in episode severity did not reflect the improvements observed by increasing median values of ANC and pH.

3 Changes in land use as a confounding factor for recovery

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3.1 Introduction

Environmental factors other than acid deposition – so-called 'confounding factors' – are expected to affect chemical and biological recovery of freshwaters in response to reduced acid deposition. Effects of climate as a confounding factor was described in an earlier trend report (de Wit and Skjelkvåle 2007). In this chapter, we provide a brief overview of some possible implications of land use change for recovery of acidified surface waters.

It is important to note that climate change and land use change are often highly interrelated. Climate change extends the growing season, makes forests grow faster and promotes expansion of the upper forest boundary in many mountainous regions - but reduced grazing pressure will have exactly the same effect. On the other hand, climate change can make forests more vulnerable to drought and insect attacks, which in some cases can lead to forest decline or even dieback in some areas.

To fulfil the obligations related to the Paris Agreement, many countries have identified intensified forestry as an effective way to store more carbon and thereby reduce their net CO_2 emissions. Also, and somewhat contradictory, the 'green shift' – leading to substitution of fossil fuels with biofuel – or more reliance on renewable resources, including organic matter, may lead to lower C storage because of more intensive harvesting. Both increased C binding in forestry and more intensive forestry and harvesting, however, can potentially affect chemical recovery of surface waters. A more intensified forestry might include afforestation on new areas, densification of existing forests, fertilisation prior to harvest, and whole-tree-harvest to produce biofuels that can replace fossil fuels. However, a strong investment in intensified forestry as a climate mitigation measure can also have negative environmental effects, e.g., on surface water quality in sensitive areas.

Land use change or disturbances affecting vegetative succession and soil formation can alter the pools of carbon, nitrogen and base cations in soils, which in turn may affect chemical and biological recovery of freshwaters in response to reduced acid deposition. In the following section, we briefly describe some of the effects that may be related to intensified forestry, reduced summer farming and/or grazing in upland areas, increased vegetation growth / expansion or forest at higher altitudes and temporary decline or dieback of forest due to drought or disease outbreaks.

3.2 Intensified forestry as a climate mitigation measure

New forest planting and intensified forestry are in recent years highlighted as promising ways to move towards a low carbon emission society. In the following section, three sets of measures related to the forestry sector are discussed regarding possible negative effects on surface water quality and recovery from acidification. These are: Afforestation on new areas, forest fertilisation, and new harvesting methods.

3.2.1 Afforestation on new areas

Afforestation is the planting of trees on currently non-forested land. It may involve planting on either scrublands or land previously used for agriculture. Afforestation, especially with spruce, may cause acidification due to: (1) increased dry deposition of acidifying substances and sea-salts due to increased filtering in the canopy (Nisbet et al. 1995), (2) increased base cation uptake and accumulation in tree biomass, and (3) increased acidity of litterfall (Tamm and Hallbäcken 1988).

Effects of afforestation on surface water quality have been extensively studied in the uplands of the UK (Jenkins et al. 1990). Large areas in Europe, e.g. Wales, Scotland and Germany have been planted with spruce, and negative impacts on surface water quality have been well documented (Ormerod et al. 1989; Helliwell et al. 2001). In Norway, the link between afforestation and acidification has mainly been attributed to spruce plantations along the western coast, which are heavily exposed to storms and sea-salts episodes from the North Sea (Wright et al. 1988, Hindar et al. 1995; Larsen and Holme 2006).

Afforested catchments close to the coast are likely to attract higher rates of sea salt deposition as the forests will more effectively scavenge cations, chloride and other anions in sea water. This was clearly demonstrated during a major storm that hit south-western Norway in January 1993 and created an extreme acidification episode in the afforested Svela catchment (Hindar et al. 1995). Stream water chloride concentrations increased by a factor of 8, pH decreased from 4.9 to 4.45 and the concentration of inorganic aluminium increased from 20 to 200 µg/l (Figure 21). Acidification associated with the storm was also observed in nearby catchments with natural (less dense) forest or no forest, but it never reached the levels observed in the afforested catchment.

Large areas in western Norway have been afforested with dense spruce plantations during the past 50-60 years. In a paired catchment approach with afforested and native forest stands along a gradient in acid deposition Larsen and Holme (2006) studied stream water chemistry during a major sea-salt episode in January 2003. pH dropped and inorganic aluminium peaked in all catchments, but the responses were larger in streams draining catchments afforested with dense spruce plantations compared to adjacent and less densely forested catchments with native tree species.

An additional effect of afforestation and fertilization of forests (next chapter) is that increased forest biomass will lead to higher production of organic matter and a larger pool of soil organic matter. This will in turn promote brownification and possible setback of recovery due to increased leaching of organic acids from soil to surface waters (Kritzberg 2017, Kritzberg et al. 2019, Bragée et al. 2015).
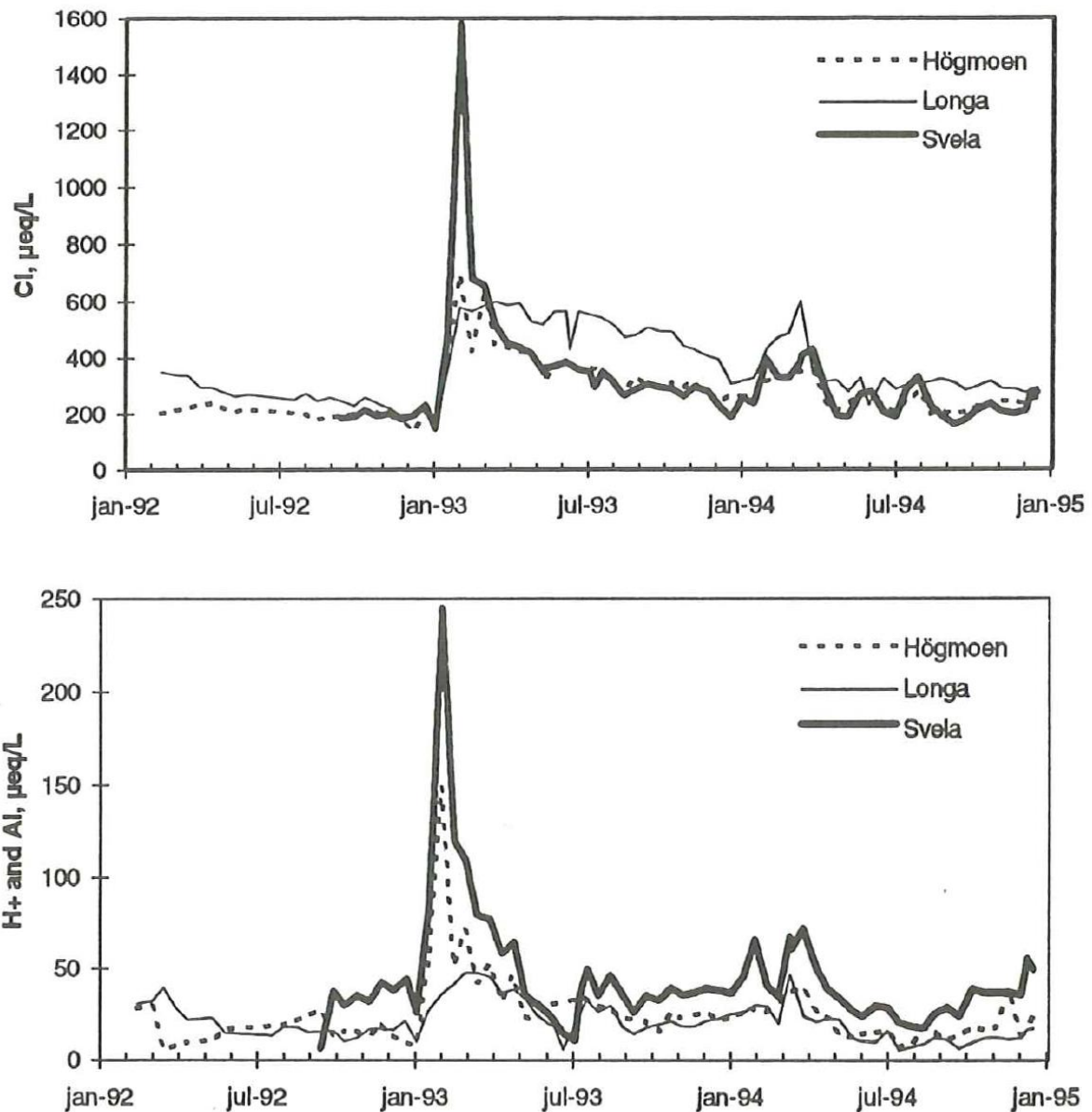
Figure 21. Concentrations of chloride, H+ and aluminium during the sea-salt episode in the three catchments Høgmoen (natural forest), Longa (non-forested) and Svela (afforested). From Hindar et al. (1995).

3.2.2 Forest fertilisation

Nitrogen (N) fertilisation before final harvest is widely used in the Nordic countries as a measure to increase the biomass and quality of trees when harvested (Rytter et al. 2016). The fertilizer dosage applied is typically 150 kg N/ha as ammonium nitrate (Clarke et al. 2018). N leaching associated with forest fertilisation might impact negatively on water bodies that are or have been prone to acidification from long-range atmospheric pollution. Thus, a protective zone has been established for the coastal regions in southern and southwestern parts of Norway (Sundnes et al, submitted).

Nitrogen fertilisation often leads to detectable short-term increases in soil solution N concentrations (Clarke et al. 2018) and can also increase N concentrations in streams draining fertilised areas. However, when done correctly i.e. by leaving unfertilised buffer strips along brooks and streams, the immediate effects on surface waters are usually small. In most Nordic forest surface waters, water quality effects are often transient and rapidly diluted as the water moves downstream in a river network (Schelker et al. 2016).

In a recent study, 150 kg N/ha was applied to a forested area upstream the drinking water reservoir Lake Glitrevann, in SE Norway (Jackson-Blake et al., in prep.). The fertilised area was 0.67 km^2(4% of the total catchment) draining into a stream with a catchment area of 18.6 km^2 and that was monitored one year before – and after the nitrogen fertiliser was added in June 2017. Frequent sampling around the time of the fertilising event showed a temporary increase in the nitrate (NO_3^-) concentration of the stream (from ~50 µg N/l to ~150 µg N/l), but it returned to the initial level a few weeks after (Jackson-Blake et al., in prep.). Thus, the effect appeared to be short-lived although it must be noted that a small signal would be difficult to detect against the background of runoff from the unfertilized part of the catchment.

In a modelling study, where the MAGIC biogeochemical model was used to simulate possible long-term impacts of different forest management scenarios at the long-term monitored site Birkenes in southernmost Norway, N fertilisation of the forest did not result in elevated N leaching until the forest was cut 10 years later, see next chapter (Valinia et al. in prep.). The model was calibrated to weekly water chemistry samples collected from 1974 to 2017, and for the entire record the average simulated concentrations of major ions in stream water differed from the observed by < 1 µeq/l (Valinia et al. in prep.).

The low N leaching level observed after fertilisation of the forest near Lake Glitrevann and the modelling results from Birkenes are largely in line with previous findings elsewhere (Shepard 1994, Lepistö and Saura 1998, Clarke et al. 2018, Moldan et al. 2018).

3.2.3 Forest harvesting

The most pronounced effects of forestry on surface water quality occur at final harvest, i.e., leaching of nitrogen (Kreutzweiser et al. 2008, Futter et al. 2010) and removal of base cations stored in tree biomass (Akselsson et al. 2007). Both processes can have negative consequences for surface water acidification, for years to a decade (N leaching) and longer-term, decades to centuries (depletion of base cations). Felling and removal of trees also lead to wetter soils and greater runoff due to reduced transpiration and water consumption by trees (Likens et al. 1970). In combination with soil disturbance and erosion caused by heavy harvesting equipment, wetter soils can lead to higher dissolved organic carbon (DOC) losses and might also promote mercury methylation (de Wit et al. 2014, Eklöf et al. 2018).

Stem-only harvest (SOH)
During SOH, only the stem is removed, while branches, needles and stumps are left on site. This in contrast to whole-tree harvest (WTH) where both the stem and the logging residues (branches and needles) are removed. Clearcutting (both SOH and WTH) reduces the stand level demand for N and can promote increased N leaching that can last several years after harvest (Likens et al. 1970, Kreutzweiser et al. 2008). de Wit et al. (2014) found moderate NO_3^- and ammonium (NH_4) leaching in stream water following stem-only harvest of about 30% of a forested catchment at Langtjern, Norway. In a modelling study at the forested catchment Birkenes in southernmost Norway, the SOH scenario promoted higher N leaching than the WTH scenario. Due to poorly buffered soils at Birkenes, the NO_3^- leaching resulted in a transient, but significant reduction of the acid neutralising capacity (ANC) and

pH (Valinia et al. in prep.). Even though the effects of N leaching following harvest can be substantial in small brooks close to the harvest, studies from boreal streams show that excess NO_3^- is rapidly retained and diluted in streams further downstream (Futter et al. 2010, Schelker et al. 2016). Thus, downstream detection of a water chemistry signal of harvest will strongly depend on the area that is harvested.

<u>Whole-tree harvest</u>
Greater biomass removal at harvest (incl. stems, needles and branches) offers increased potential for fossil fuel substitution but may have negative consequences for soil and surface water re-acidification as whole-tree harvest removes more base cations than conventional harvest. In a regional study of pine and spruce forests in Sweden, Akselsson et al. (2007) found net losses of Ca and Mg in almost the whole country for both SOH and WTH, whereas WTH in spruce forests led to substantially higher net losses of K and Ca than SOH. Also, other studies have pointed out that WTH to a larger degree than SOH leads to delayed recovery from acidification due to increased rates of base cation removal from slow-weathering sites (Laudon et al. 2011, Iwald et al. 2013, Zetterberg et al. 2013). Because of improved assessment criteria for WTH planning (Akselsson and Belyazid 2018) this harvest method are usually less common on slow-weathering sites.

In a modelling study, where the MAGIC biogeochemical model was used to simulate possible long-term impacts of different forest management scenarios at the long-term monitored site Birkenes in southernmost Norway, the stem-only harvest scenario gave a more pronounced effect on N leaching than the whole-tree harvest scenario, but the NO_3^- peak in the brook decreased a few years after the clear cut in the model simulation. The greater biomass removal with the WTH scenario resulted in a long-term depletion of in soil base cations and a setback in the positive trend in stream ANC by several decades (Valinia et al. in prep.). Also modelling studies from Finland and Sweden have suggested that the increased base cation removal associated with whole-tree harvesting may promote significant surface water re-acidification (Aherne et al. 2012, Zetterberg et al. 2014, Moldan et al. 2017).

3.3   Increased vegetation growth in upland areas

Many northern and alpine areas are experiencing increased vegetation growth, more carbon stored in tree biomass, and expansion of the forested area by regrowth on previously open or cultivated land and increased upper forest altitude limit (Liski et al. 2006, Gehrig-Fasel et al. 2007, Bryn 2008, de Wit et al. 2014, Finstad et al. 2016). The trends are probably not caused by one single factor, but rather a result of complex interactions between several drivers varying both in space and time. The most predominant factors are associated with land use change, climate warming and nitrogen deposition.

Land use has changed dynamically over centuries, but over the past 100 years there have been several structural changes that have promoted increased vegetation growth and larger tree biomass in upland regions. The main underlying factors are related to abandonment of human utilisation of upland areas, i.e. reductions in summer farming, grazing by domestic animals, biomass harvest, heath burning, and abandonment of marginal pastureland (Bryn 2008, Gehrig-Fasel et al. 2007, and references therein).

Climate change is also an important factor, especially temperature rise that may affect snow accumulation at intermediate altitudes, prolong the growing season, and increase the growth rates of terrestrial vegetation in general (Xu et al. 2013). As most terrestrial ecosystems are nitrogen limited, supply of nitrogen from deposition of long-range transported air pollution will also stimulate vegetation growth. Although the emissions of nitrogen oxides peaked around 1990 (Schöpp et al. 2013) the deposition levels still represent a significant contribution to vegetation growth in exposed regions.

Although it is widely accepted that atmospheric deposition is the main driver for the extensive acidification of surface waters in Europe and North America, soil processes such as uptake of base cations by vegetation can also play a role for soil acidification and hence, surface water acidity Changes in land use can affect the buffering capacity of soils and surface water through consumption or release of base cations. From analyses of sediment records from Swedish lakes, Renberg et al. (1993) found that land use changes related to ancient agricultural practices during the iron age (burning, grazing, cultivation) caused pH in lakes to increase from an initial level of 5.5 to around 6.5.

Whereas soil cultivation, grazing by domestic animals and heath burning are alkalinizing processes, regrowth or tree planting (especially with conifers) will remove base cations and promote acidification at poorly buffered sites. In a study of land use influences on acidification and recovery of freshwaters in Galloway, Scotland, Helliwell et al. (2001) found significant acidifying effects of afforestation, by the fact that young trees have a high demand for base cations, which in turn affected surface water acidification. Additionally, if coniferous trees are planted in formerly open areas close to the coast it will increase the acidifying potential of sea-salt episodes.

Expansion of forest in upland regions will also increase the uptake of nitrogen, and potentially reduce nitrogen leaching at sites with elevated levels of nitrogen deposition from long-range transported air pollution (Lucas et al. 2016). Fixed carbon in new forest biomass will also represent an additional source for organic carbon that may be exported from soils to surface waters and cause browning of drinking water sources and possibly also increased acidification from organic acids (de Wit et al. 2006, Monteith et al. 2007, Finstad et al. 2016, Kritzberg et al. 2019). Increased leaching of organic matter from soils to surface water might also be accompanied by heavy metals as e.g. mercury (de Wit et al. 2014).

3.4   Forest decline caused by environmental stress

While forests expand in many areas, there are also examples of forest decline or dieback on various scales – from individual trees, to local areas or even larger regions. There is often more than one factor involved when trees are dying without an obvious cause, but it is usually difficult to clarify the exact causal relationship. One good example was the proposed link between acid rain and forest dieback in Europe during the 1970s and 1980s. In the early 1990s, results from long-term studies showed that the conditions of European forests were close to normal fluctuations, and that climatic factors in combinations with parasite attacks or other stressors appeared more important for the observed fluctuations than air pollution and soil acidification alone (Abrahamsen et al. 2012). It should be noted that in the so-called 'Black Triangle' in central Europe, extensive forest die-back as a consequence of extreme atmospheric pollution did occur during the 1960s until the 1980s. However, such levels of pollution were uncommon outside Central Europe. Examples of drought and heat-related forest stress and dieback are frequently reported from different parts of the world, and with climate change there seems to be an increasing risk of climate-induced dieback of forests (Allen 2009, Seidl et al. 2018). When already exposed to climate stress, trees will be more vulnerable to other threats like insect attacks or air pollution. As an example, bark beetle outbreaks in the northern hemisphere currently affect millions of hectares of forested landscape, and the problem seems to be increasing, possibly as a result of climate warming (Marini et al. 2017).

Oulehle et al. (2019) used the MAGIC biogeochemical model to simulate soil and water chemistry at the Plesne Lake, Czech Republic, which has experienced recovery from anthropogenic acidification but where the surrounding forest has suffered from severe bark beetle attacks. The model simulations

show that (1) N retention in the catchment declined after the bark beetle attack and caused substantially increased NO$_3^-$ leaching and surface water re-acidification for 10 years after the bark beetle disturbance, (2) mineralisation from decaying biomass and uptake in re-growing vegetation reduced the ecosystem losses of N, Ca and Mg, (3) the excess of mineralised base cations in decaying biomass led to a short-term increase of soil base saturation, and (4) the improvement of the catchment soil acid–base status led to a temporary increase of lake water pH and ANC.

3.5   Summary

Environmental factors other than acid deposition – so-called 'confounding factors' – can affect chemical and biological recovery of freshwaters. Land use and climate change, very often closely related, are such factors. Land use change is usually a driver that works on a smaller spatial scale than the global or regional effects of climate change and deposition. Here we considered especially land use change in relation to forest management and forest cover. Forestry interacts with acid-base chemistry by enhancing removal of base-cation rich harvest residues and increasing the scavenging of pollution and sea salts in planted forests, and it can thereby set back surface water acidification status for decades or even longer in acid-sensitive areas.

An additional effect of afforestation and fertilization of forests is that increased forest biomass contributes to a larger pool of soil organic matter, which in turn will promote brownification and possible setback of recovery due to increased leaching of organic acids. Where land use change results in a change in the nitrogen cycle (as in the case with forestry) the effect can go both ways: the systems can tolerate more nitrogen when more N is removed by whole-tree harvest, or it can experience increased N leaching upon harvest in fertilized sites.

Climate change can also lead to strong impacts on vegetation dynamics – and here it might be difficult to differentiate between climate and land use change. A bark beetle attack led to an increase in base-cation status of a very acidic soil (because no biomass was removed), and therefore also to improved water quality. The impact of land use change on chemical recovery of surface waters will thus depend on the extent to which national authorities decide to change forestry practices or increase afforestation, and whether authorities will take catchment acid-sensitivity and critical loads in as a factor in their assessment of benefits and drawbacks of such adaptation measures.

4   Synthesis

Sulphate concentrations have declined everywhere except for sites with deeply weathered soils; 10 of 13 regions show more than 40 % decline for the period 1990-2016. Nitrate has declined in all regions, although many of the individual stations show no trend. Even chloride show declining trends, and in Northern Europe rates of decline have been steeper than for nitrate. Chloride levels are affected by deposition of sea salts as well as air pollution. Change points, i.e. points in the series where the slope changes or where there is a step change in level, were more prevalent in the time series for sulphate and nitrate in the 90s than after year 2000, indicating smoother trends in recent years. On a regional scale, base cation concentrations have declined by approximately half the equivalent change in concentration of sulphate and the two are closely linked. All regions show increasing ANC between 1990 and 2016. pH has increased in 12 of 13 regions, despite partial replacement of mineral acidity by organic acids manifested as an increase in DOC. The trajectories of sulphate, ANC and pH indicate that recovery has slowed down in Europe and accelerated in North America since the early 2000s. This indicates that emission of sulphur to the atmosphere has declined faster in North America and slower in Europe after 2000. Acidic episodes, i.e. the annual extremes, have become less severe in line with the recovery of average chemistry.

A parallel analysis of atmospheric deposition was outside the scope of this report. We can therefore not compare trend slopes and assess delayed responses. Neither could we test if e.g. the fewer time trend change points observed in recent years were a result of less variable deposition compared to the 90s. Even so, the causal link between air pollution, deposition, leaching and acidification/recovery is sufficiently well established to conclude that reduced air pollution is explaining most of the changes presented in Chapter 3. The greatest improvement was found in the areas that historically have had the highest deposition of acid. The consistent pattern of chemical recovery (decreasing sulphate, nitrate, as well as chloride, increasing pH and ANC) across many sites and regions proves that emission control programmes are having their intended effect.

In addition to air pollution, changes in climate and land use, which are often inter-related can also affect the composition of headwaters. Here we considered how intensified forestry, reduced summer farming and grazing in upland areas, increased vegetation growth and expansion or forest at higher altitudes, and temporary decline or dieback of forest can change the leaching of sea salts, sulphate, nitrate, base cations, DOC, aluminium as well as runoff pH. The effect of land use changes on water chemistry on a wider than local spatial scale is not well documented, partly because it is subtle compared to the large effects of reduced air pollution that we have observed at ICP Waters sites. Less extreme pH and ANC during episodes and improved average water quality paves the way for biological recovery. The relative importance of changes in land use and climate change for headwater ecosystems will increase as acid deposition falls to low levels, and in most cases water chemistry is unlikely to ever return completely to its pre acidification state. Prior to acidification, soils had a larger buffering capacity than they do now, and base cation stores will take decades to replenish. Even with the full recovery of water chemistry in the future, biological factors may prevent a full return to pre-acidification ecological reference conditions. Nevertheless, restoration of surface water quality to a state where it can sustain healthy and diverse communities of aquatic species will be worthwhile and the best that we can achieve.

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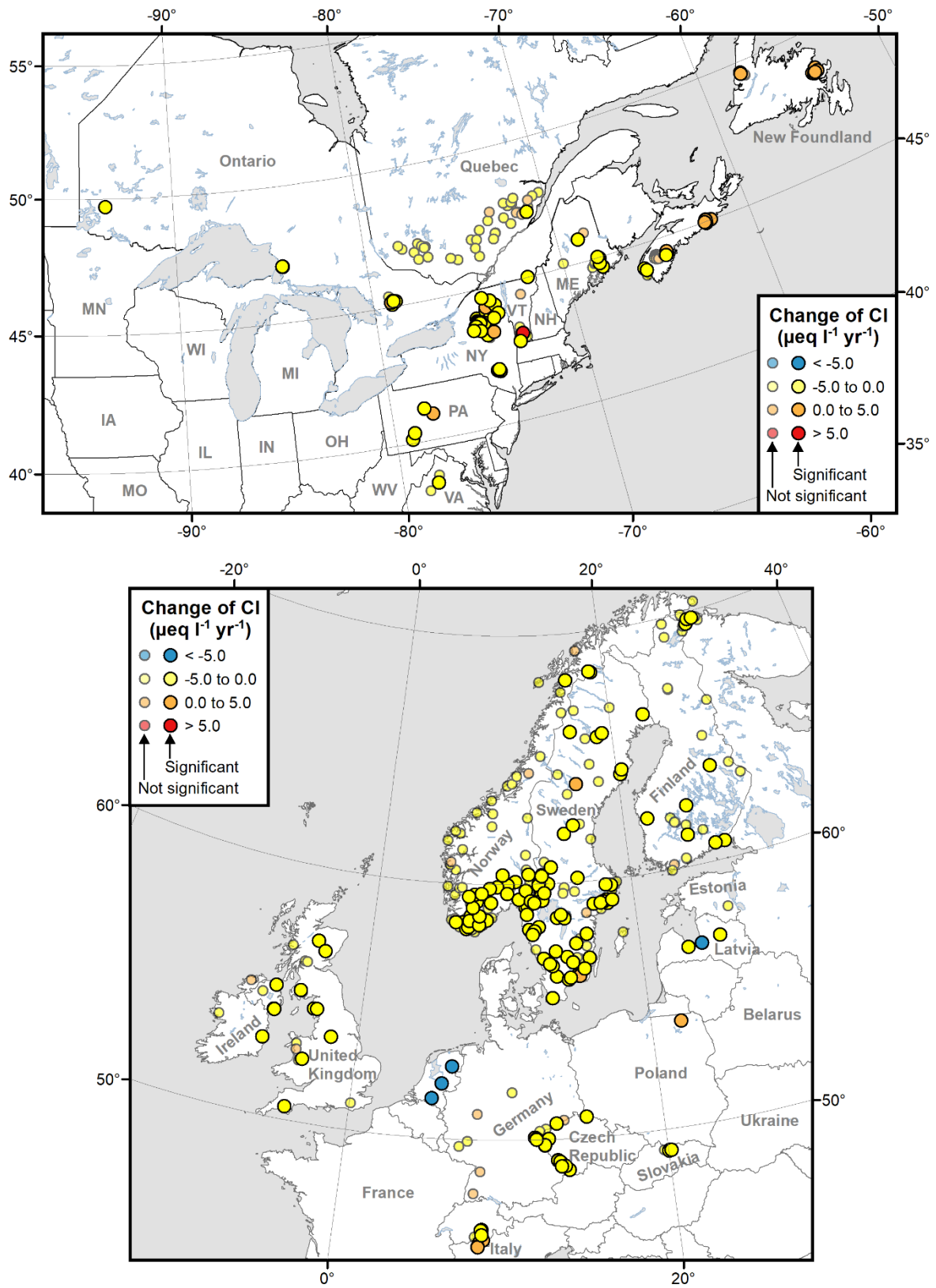# Appendix A. Temporal trends in water chemistry 1990-2016 – maps
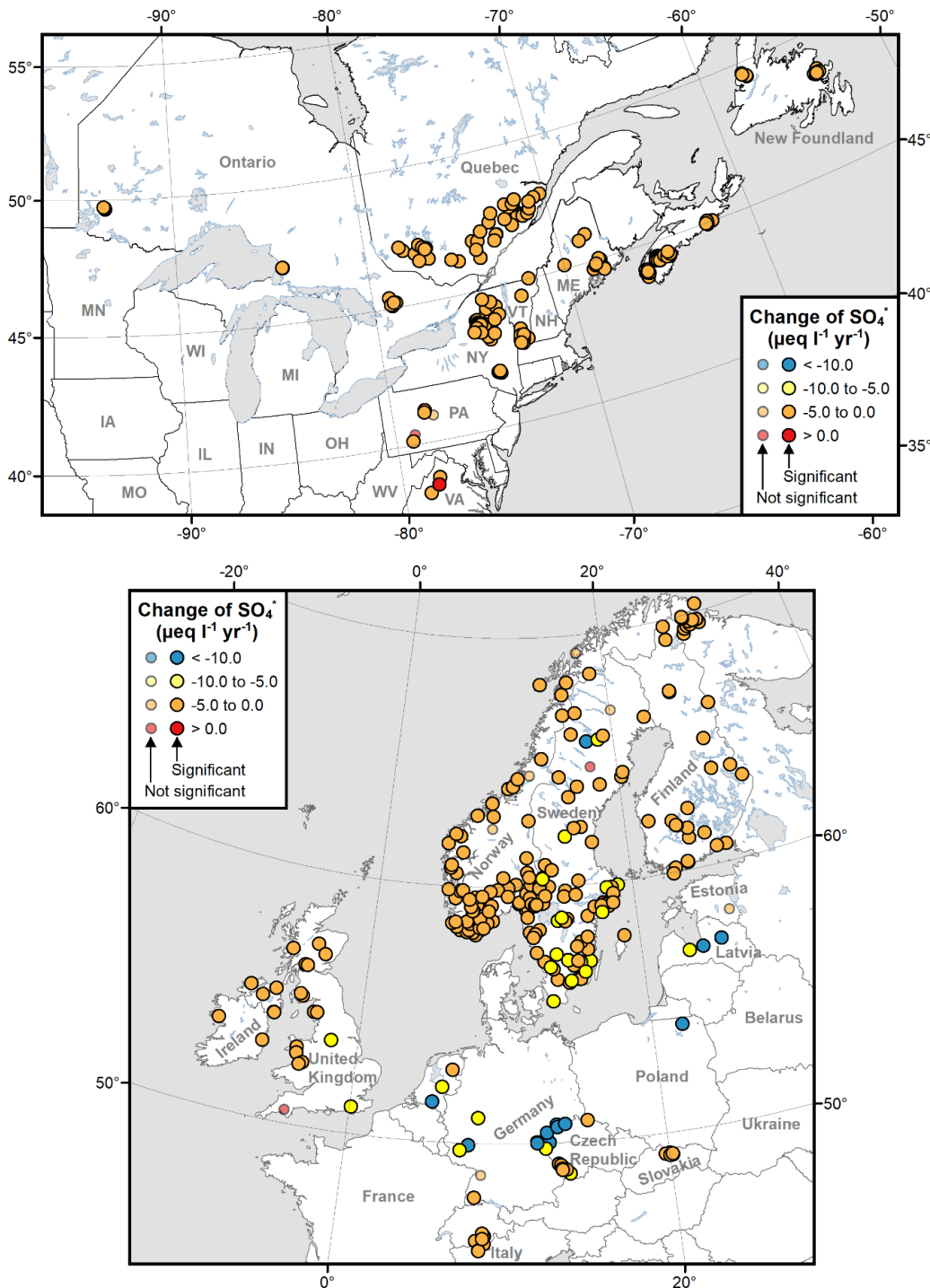
Figure A1. Trends (Mann Kendall, p<0.05) in non-marine sulphate and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.
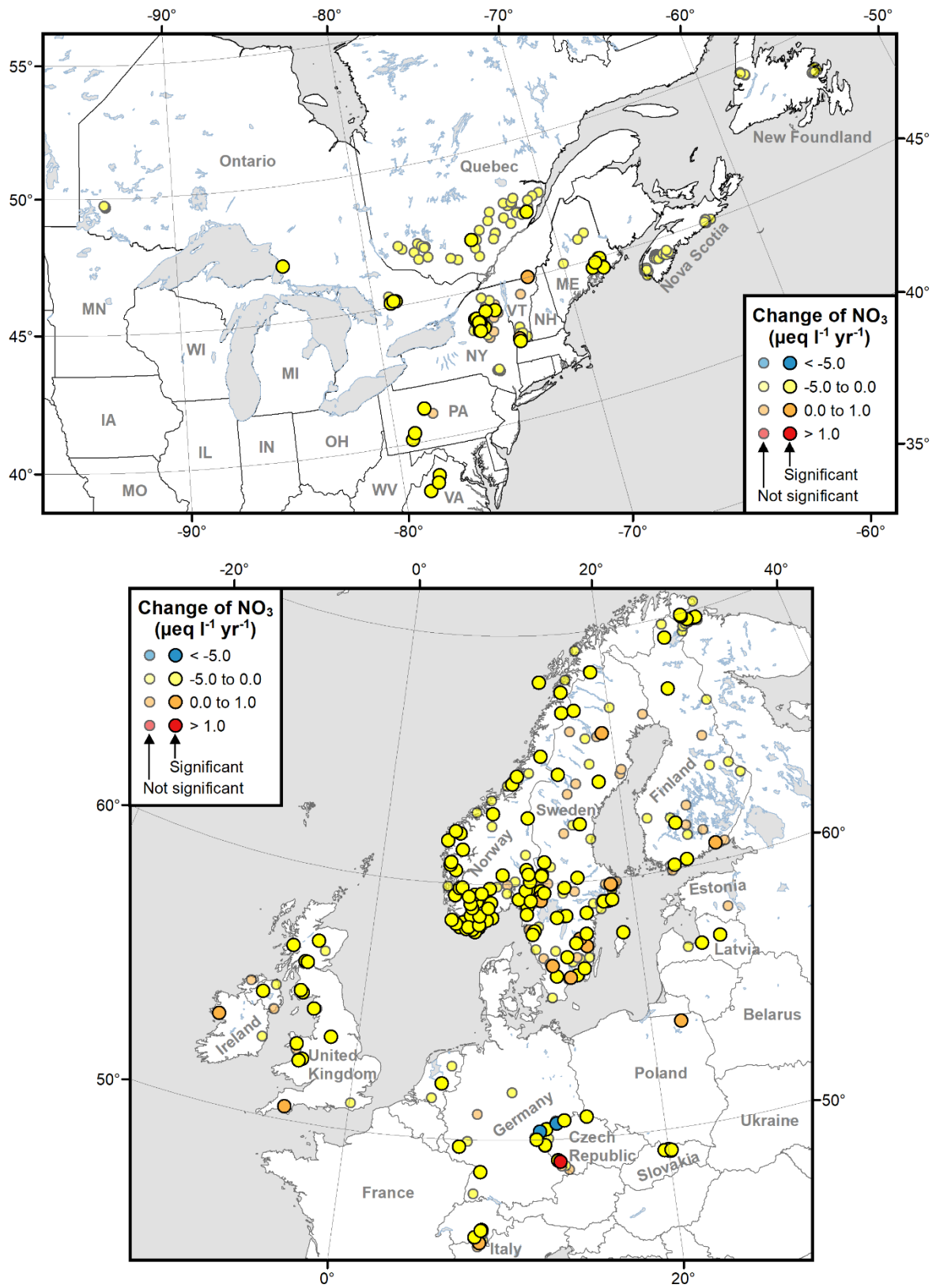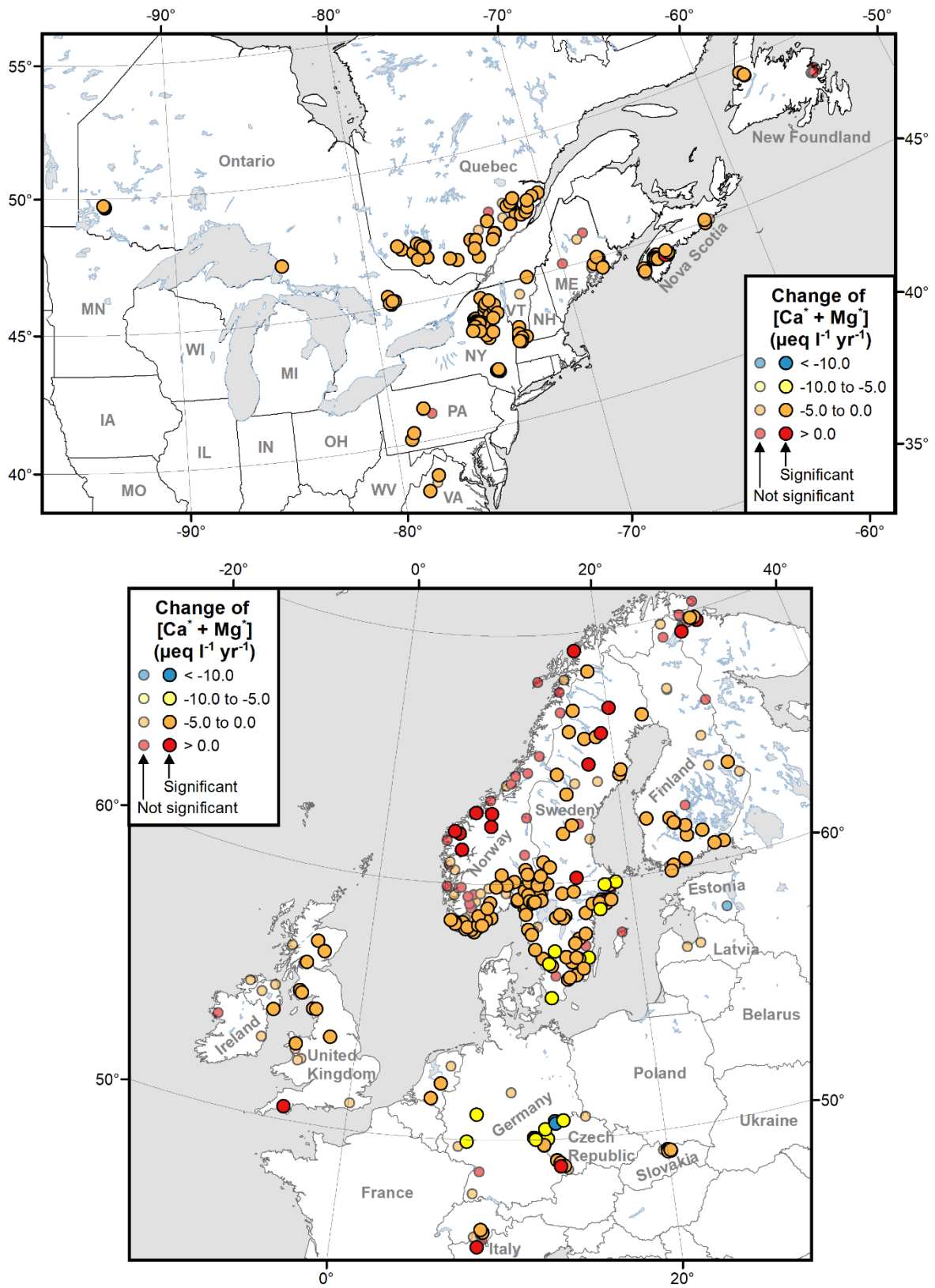
Figure A2. Trends (Mann Kendall, p<0.05) in nitrate and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.

Figure A3. Trends (Mann Kendall, p<0.05) in chloride and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.

Figure A4. Trends (Mann Kendall, p<0.05) in non-marine base cations and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.
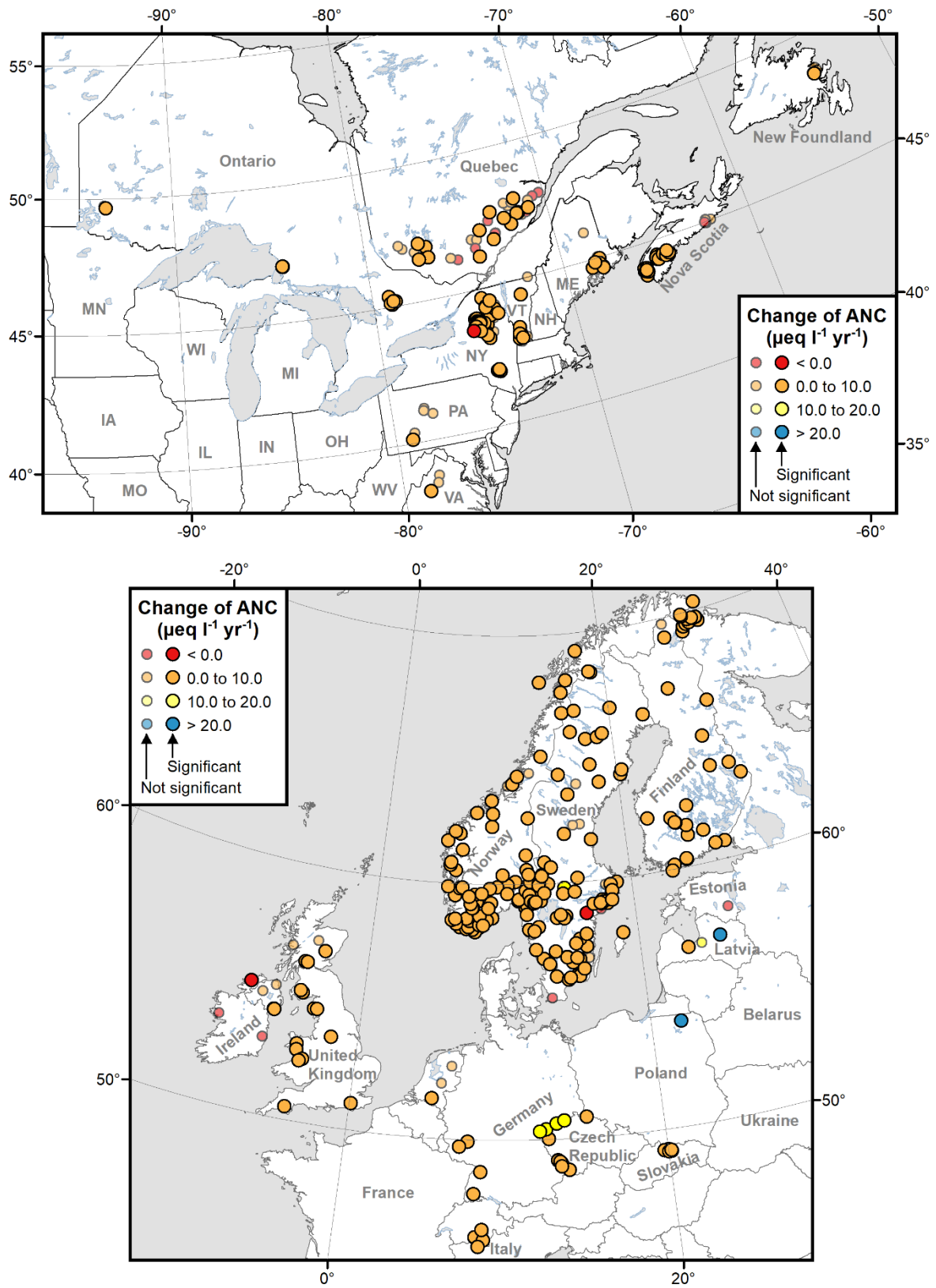
Figure A5. Trends (Mann Kendall, p<0.05) in ANC and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.

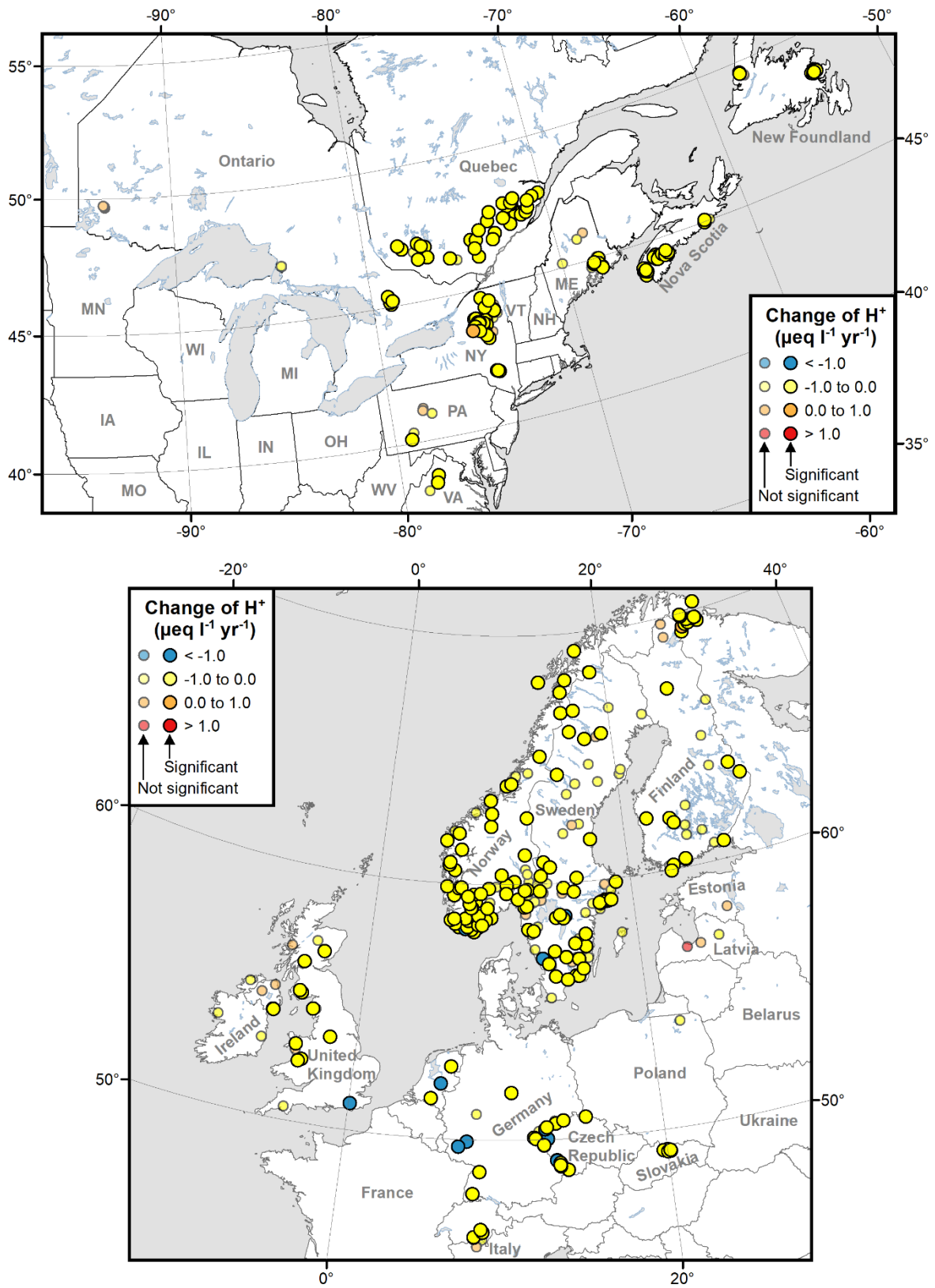**Figure A6.** Trends (Mann Kendall, p<0.05) in hydronium ions and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.

Figure A7. Trends (Mann Kendall, p<0.05) in TOC and distribution of Sen slopes at North American (top) and European (bottom) sites for the period 1990-2016.

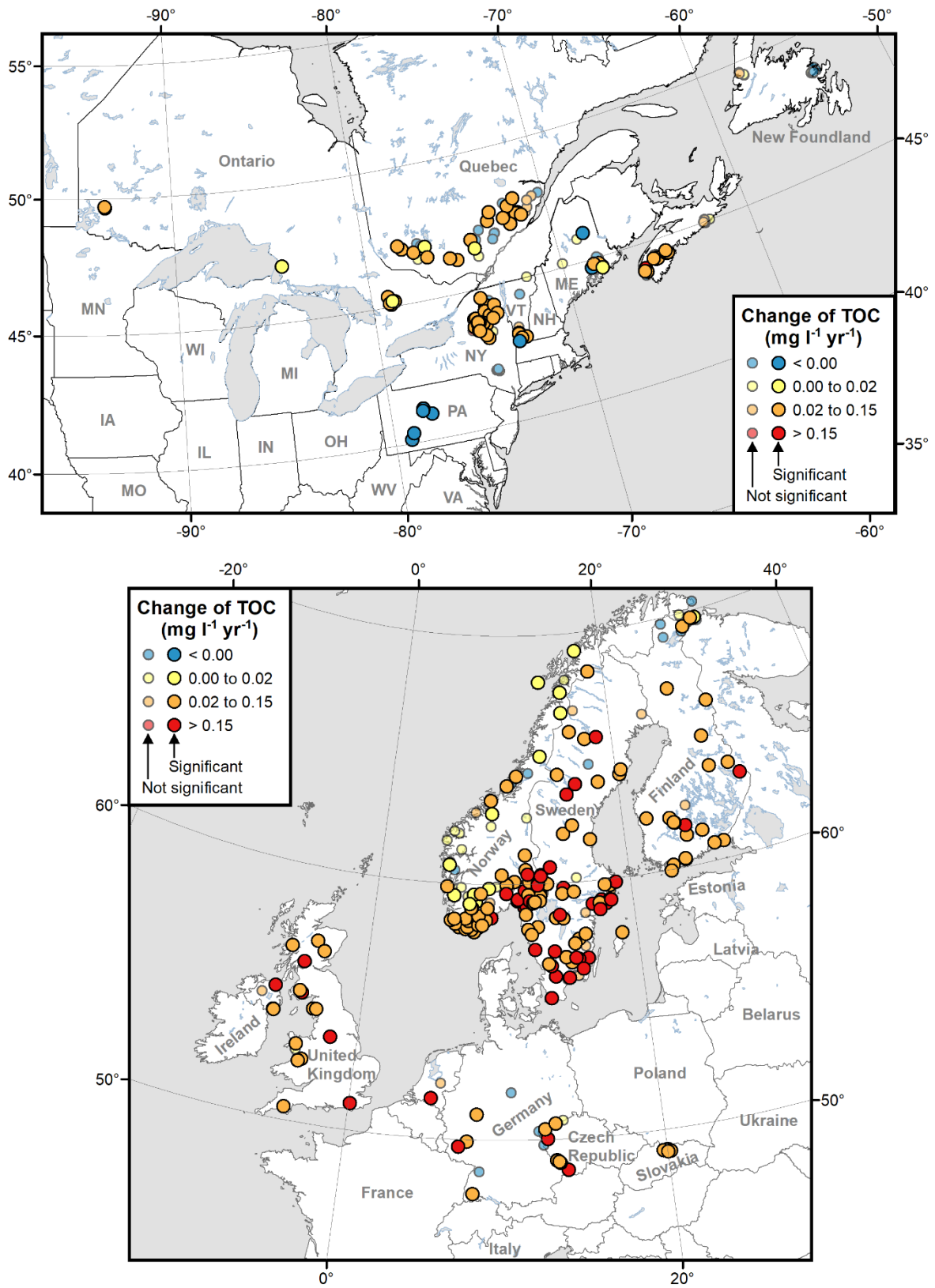# Appendix B. Mean Sen slopes 1990-2004 and 2002-2016

Table B1. Mean Sen slopes for the period 1990-2004 and 2002-2016.The units are µeq/L/yr for all variables except TOC (mg/L/yr).

Period 2002-2016

Variable	Europe							North America						Europe	N. America
	NoNord	SoNord	Baltic	UK-IE-NL	WCE	ECE	Alps	Ont	AdsCsk	QaMaVt	AtlCan	Apps	BRM		
$SO_4^{2-}*$	-1.01	-1.67	-6.44	-1.70	-2.82	-3.52	-0.95	-3.41	-2.99	-2.27	-1.17	-1.64		-1.88	-2.17
NO_3^-	-0.03	-0.06	-0.74	-0.05	-0.96	-1.04	-0.95	-0.03	-0.19	-0.04	0.00	-0.06	-0.03	-0.25	0.07
Cl^-	-0.23	0.15	0.01	-0.88	1.65	-0.27	0.35	-0.63	-0.06	-0.27	-1.26	-0.29	0.03	-0.03	-0.54
$Ca^{2+}*+Mg^{2+}*$	-0.56	-1.38		-0.76	-2.11	-1.37		-1.87	-1.53	-1.00	-0.07	-1.01	-0.51	-1.14	-0.91
ANC	0.12	-0.27	18.44	2.07	3.79	4.67	0.29	1.87	1.79	1.38	1.63	0.39	0.32	0.93	1.56
H	-0.01	-0.07		-0.14	-0.10	-0.23	-0.01	-0.02	-0.22	-0.06	-0.25	-0.07	-0.01	0.08	-0.16
TOC	0.06	0.09		0.06	0.07	0.05		0.01	0.07	0.01	0.13	-0.04		0.08	0.06

Period 1990-2004

Variable	Europe							North America						Europe	N. America
	NoNord	SoNord	Baltic	UK-IE-NL	WCE	ECE	Alps	Ont	AdsCsk	QaMaVt	AtlCan	Apps	BRM		
$SO_4^{2-}*$	-2.26	-3.91	-10.61	-3.18	-6.11	-6.79	-1.39	-2.16	-2.12	-1.73	-0.99	-0.75		-4.00	-1.59
NO_3^-	0.01	-0.08	-1.12	-0.06	-1.26	-1.20	0.52	-0.06	-0.23	-0.02	0.00	-1.09	-1.26	-0.24	-0.12
Cl^-	-1.05	-2.02	-8.40	-4.41	-1.41	-0.93	0.55	0.67	-0.07	0.52	1.70	0.21	-0.25	-1.90	0.71
$Ca^{2+}*+Mg^{2+}*$	-0.52	-1.65		-0.57	-2.84	-3.23		-1.74	-1.13	-1.20	-0.01	-4.20	-0.38	-1.74	-0.92
ANC	2.44	3.50	-5.81	3.11	9.69	6.66	0.66	0.24	1.45	0.78	0.38	-1.97	1.03	3.46	0.75
H	-0.08	-0.28		-0.41	-1.19	-0.55	-0.02	-0.02	-0.15	-0.06	0.09	0.04	0.00	-0.30	-0.03
TOC	0.04	0.12		0.16	0.05	0.05		0.03	0.03	0.01	-0.06	-0.02		0.10	-0.01

Reports and publications from the ICP Waters programme

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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