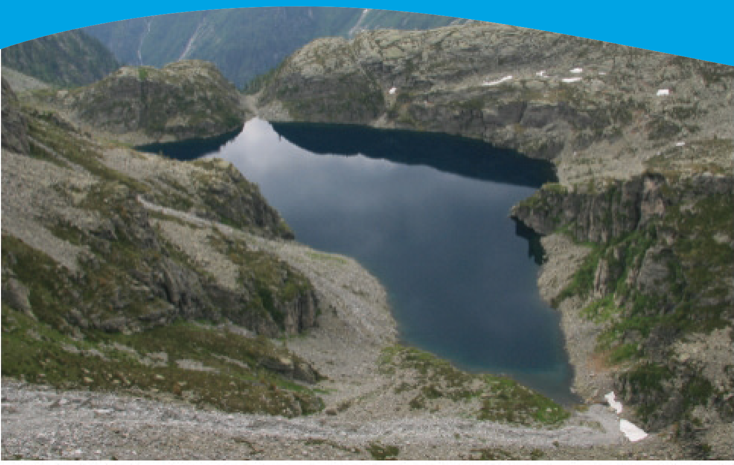
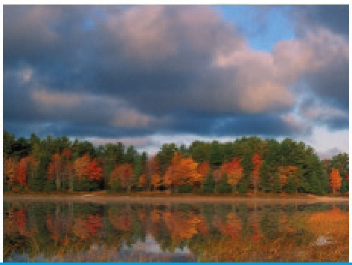
NIVA

NIVA REPORT SNO 7268-2018

ICP Waters Report 135/2018
Regional assessment of the current extent of acidification of surface waters in Europe and North America




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

Convention on Long-Range Transboundary Air Pollution

LRTAP
Long-range Transboundary Air Pollution

Norwegian Institute for Water Research

REPORT

Main Office	NIVA Region South	NIVA Region East	NIVA Region West	NIVA Denmark
Gaustadalléen 21 NO-0349 Oslo, Norway Phone (47) 22 18 51 00 Telefax (47) 22 18 52 00 Internet: www.niva.no	Jon Lilletuns vei 3 NO-4879 Grimstad, Norway Phone (47) 22 18 51 00 Telefax (47) 37 04 45 13	Sandvikaveien 59 NO-2312 Ottestad, Norway Phone (47) 22 18 51 00 Telefax (47) 62 57 66 53	Thormøhlensgate 53 D NO-5006 Bergen Norway Phone (47) 22 18 51 00 Telefax (47) 55 31 22 14	Ørestads Boulevard 73 DK-2300 Copenhagen Phone (45) 8896 9670

Title	Serial number	Date
Regional assessment of the current extent of acidification of surface waters in Europe and North America	7268-2018 ICP Waters report 135/2018	15.10.2018
Author(s)	**Topic group**	**Distribution**
Kari Austnes, Julian Aherne (Trent University), Jens Arle (UBA), Marina Čičendajeva (LEGMC), Suzanne Couture (ECCC), Jens Fölster (SLU), Øyvind Garmo, Jakub Hruška (CGS), Don Monteith (CEH), Max Posch (IIASA), Michela Rogora (CNR ISE), James Sample, Brit Lisa Skjelkvåle (UiO), Sandra Steingruber (TI), John L. Stoddard (US EPA), Rafał Ulańczyk (IEP-NRI), Herman van Dam (WN), Manuel Toro Velasco (CEDEX), Jussi Vuorenmaa (SYKE), Richard F. Wright, Heleen de Wit	Acid rain **Geographical area** Europe, North America	Open **Pages** 134

Client(s)	Client's reference
Norwegian Environment Agency (Miljødirektoratet) United Nations Economic Commission for Europe (UNECE)	
Client's publication: ICP Waters report	Printed NIVA Project number 10300

Summary

The current status of surface water acidification related to air pollution in Europe and North America has been assessed using country reports, monitoring data, critical loads and exceedance data, acid sensitivity and deposition maps, and data reported under the European Commission's Water Framework Directive (WFD). Acidification is still observed in many countries, but the extent and severity vary. Maps of acid sensitivity and deposition suggest that surface water acidification is present in regions and countries for which no data or reports were delivered for the current assessment. Existing national monitoring varies in the ability to assess the spatial extent of acidification and the recovery responses of acidified sites. The monitoring requirements under the European Union's National Emission Ceilings Directive are expected to reverse the recent decline in the number of monitoring sites observed in some countries. The information reported under the WFD is currently of limited value in assessing the extent of acidification of surface waters in Europe. Chemical recovery in response to reductions in acid deposition can be slow, and biological recovery can lag severely behind. Despite large and effective efforts across Europe and North America to reduce surface water acidification, air pollution still constitutes a threat to freshwater ecosystems.

Four keywords	Fire emneord
1. Air pollution	1. Luftforurensning
2. Surface water acidification	2. Forsuring av overflatevann
3. Geographical extent	3. Geografisk utbredelse
4. Europe and North America	4. Europa og Nord-Amerika

This report is quality assured in accordance with NIVA's quality system and approved by:

Kari Austnes
Project Manager

Heleen de Wit
Research Manager

ISBN 978-82-577-7003-7
NIVA-report ISSN 1894-7948

CONVENTION OF LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

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

Regional assessment of the current extent of acidification of surface waters in Europe and North America

Prepared at the ICP Waters Programme Centre
Norwegian Institute for Water Research
Oslo, October 2018

Preface

The International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters) was established under the Executive Body of the *United Nations Economic Commission for Europe* (UNECE) *Convention on Long-range Transboundary Air Pollution* (CLRTAP) at its third session in Helsinki in July 1985. UNECE is a catalyst in the international work aiming at reducing transboundary air pollution. Norway provides facilities for the ICP Waters Programme Centre at the Norwegian Institute for Water Research (NIVA). The Norwegian Environment Agency provides financial support and a representative who is Chair of ICP Waters. ICP Waters receives additional financial support from the UNECE Trust Fund of the CLTRAP. In the long-term strategy for the Convention it is stated that environmental effects of acidifying components, and its potential interaction with climate change and biodiversity, continue to be among the significant remaining problems with regard to air pollution effects on the environment. These can be addressed with the multi-pollutant/multi-effects approach of the Gothenburg Protocol.

The main aim of the ICP Waters is to assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution on surface waters. The objectives of this report were to assess the current extent and severity of surface water acidification related to air pollution in Europe and North America, and to inform policy processes on the need for further emission reductions as well as the need for monitoring of air pollution effects.

This report has been prepared by the ICP Waters Programme Centre and its National Focal Centres and is based on country reports, submitted monitoring data to the Programme Centre and monitoring data submitted under the Water Framework Directive, and other relevant data and information.

We would like to thank all who have contributed, including the co-authors of the country reports. A special thanks to Jennifer Phelan and Jason Lynch for providing critical loads and exceedance maps for the United States, to Anne Probst for information on acidification in France, and to Jens Fölster, Jakub Hruška, Don Monteith, Michela Rogora and John L. Stoddard for reviewing the whole report in addition to providing their separate country reports.

Heleen de Wit

ICP Waters Programme Centre
Oslo, October 2018

Table of contents

Executive summary

Background and key questions

Historically, acidification of surface waters was one of the major environmental impacts of air pollution leading up to the signing of the UNECE Convention on Long-range Transboundary Air Pollution (CLRTAP) in 1979. The resulting reductions in nitrogen and particularly sulphur deposition resulted in a gradual decline in degree and extent of surface water acidification. However, there is a lag in chemical, and particularly biological recovery.

In several countries, monitoring data continue to document recovery. However, a regional assessment of current surface water acidification status is lacking. In this report, regions with a potential risk for surface water acidification are identified, and acidification status is documented where such data are available. Weaknesses in data availability are identified, which are relevant to air pollution policy (CLRTAP, EU National Emission Ceilings (NEC) Directive, Canada-United States Air Quality Agreement).

Identification of acid-sensitive regions

Acidification of surface waters occurs when the chemical buffering capacity of the soils in their catchments is insufficient to balance the level of acid deposition. Mapping of these factors is used to identify regions with potentially acidified surface waters. This is particularly important where local surface water chemical and/or biological data are rare or not available. The critical load is a well-established measure of the acid sensitivity of surface waters, which indicates the amount of acid deposition an ecosystem can tolerate in the long-term without being harmed. However, information on critical loads for surface waters is available only for Norway, Sweden, Finland, Ireland, the UK, the Netherlands, Switzerland, Canada and the US. To provide a consistent overview, maps of potential acid sensitivity for Europe and North America were produced, using available maps on bedrock geology. These maps provide only a crude indication of acid sensitivity, as other factors such as soil characteristics can also be influential. Nevertheless, at a broad regional scale, the geology-based acid sensitivity maps agree reasonably well with the critical loads maps.

Risk of surface water acidification

Where estimates of critical loads are available, the risk of surface water acidification can be expressed by the critical loads exceedance, i.e. where the deposition exceeds the critical load. This occurs in all countries for which critical load estimates are available and indicates that surface water acidification remains an environmental issue in parts of these countries. Acidification may continue after the critical load is no longer exceeded, because the original buffering capacity has been depleted by acid deposition and has yet to be restored. Also, biological recovery may take much longer than chemical recovery.

For the countries where critical load estimates are not available, a comparison of the geology-based map of potential acid sensitivity with atmospheric deposition data suggests that acidification may be an issue particularly in the following regions: the Pyrenees, the border regions of Belgium, Luxembourg, France and Germany, the mountainous regions on the borders of the Czech Republic, Germany and Austria, the Tatra Mountains in Slovakia and Poland, the Italian Alps, northern Croatia, parts of Bosnia and Herzegovina and Serbia, western Albania, parts of western Russia, and central Armenia.

Documentation of the current status of surface water acidification using surveys

While the risk of acidification can be assessed as described above, the actual presence of surface water acidification can be confirmed only by monitoring data. Upon request, thirteen countries submitted monitoring data on current (from 2010 onwards) acidification status. The spatial coverage of the submitted data varies considerably. In general, the data represent only acid-sensitive sites or regions and cannot be extrapolated to entire countries. A few countries collect national or regional data through extensive surveys and can state with some confidence the proportion of acidified lakes and/or streams within their acid-sensitive regions (Sweden, the US, Canada, Norway, Ireland); some countries monitor fewer sites in various parts of their territory (Finland and Germany); while others focus on particularly sensitive or affected regions (the Czech Republic, Switzerland, Italy, Poland). The United Kingdom (UK) survey was based only on sites where critical loads had previously been found to be exceeded and were still expected to be exceeded by 2020, while the data from the Netherlands represent a particularly sensitive habitat type. Care must be taken in comparing the summary results for such different data sets. All thirteen countries, as well as Spain and Latvia, also provided separate country reports with a more detailed overview of acid sensitivity and the acidification status in their countries, frequently including also other monitoring or modelling data representing the entire country.

To decide whether a water body is acidified, a criterion is needed. The most common, and generally accepted, chemical indicator is the Acid Neutralising Capacity (ANC), with a commonly used critical limit of 20 µeq/l. This is based on the level above which fish and invertebrate populations are not likely to be affected by water acidity. The same critical limit is frequently used in critical load calculations.
Based on the submitted data, 0-45% of the water bodies in each country have ANC < 20 µeq/l. 2-33% of the water bodies have ANC between 20 and 50 µeq/l, indicating that they are at risk, and also potentially vulnerable to short-term acidification events driven by sea salt episodes or climate extremes such as drought, heavy rainfall, and snowmelt events. Although these percentages are not directly comparable, the data document that in many countries acidification remains an issue.

In summary, both the submitted data and the information in the country reports show that acidification occurs in large regions in Norway, Sweden, the UK, Canada, and the US. In Finland and Germany acidification is more scattered, while in the Netherlands, the Czech Republic, Ireland, Italy, Switzerland and Poland it is limited to a few smaller areas. The severity of acidification is not necessarily related to the spatial extent of the problem; local hot spots may occur even where acidification is not a major, regional issue, e.g. in the Czech Republic and Ireland. However, based on both extent and severity, surface waters in Italy and Poland and, to a lesser degree, Finland, Germany, Switzerland, Canada and the US seem closer to chemical recovery than waters in the other countries which submitted data. In Spain acidification is a limited issue, and in Latvia it is unlikely to occur at all.

Acidification status for surface waters without submitted data
For several countries in Europe with potentially acidified surface waters, data were not submitted either because there was no National Focal Centre (NFC) or because of the absence of recent, relevant data. Under the Water Framework Directive (WFD) atmospheric deposition is defined as a pressure, and acidification status and water bodies impacted by acidification can be reported. In principle, WFD reporting should give a representative view of pressures and status. In practice, however, WFD reporting has ambiguities in the definitions of acidification status, the pressure type atmospheric deposition and the impact type acidification, as well as in how data are reported. Moreover, the WFD reporting focuses on larger water bodies (lakes > 0.5 km^2 and rivers with catchments > 10 km^2). It does not, therefore, necessarily reflect acidification pressure on smaller headwater lakes and streams that often tend to be the most geologically acid-sensitive systems.

The ambiguities in definitions and reporting means that the national reports on acidification status under the WFD may not be complete, the criteria may not be comparable or relevant, acidification may be due to pressures unrelated to acid deposition (for instance mine drainage), and the cause of the acidification may be unclear. Several countries report water bodies with less than good acidification status. However, far fewer water bodies are reported also with the impact type acidification and/or the pressure type atmospheric deposition. Of the countries which did not submit data for the current study, only Belgium had a significant proportion of water bodies identified as having less than good acidification status, the impact type acidification, and the pressure type atmospheric deposition (3% for rivers, 17% for lakes). Moreover, many countries classify very few of the water bodies with respect to acidification status, and it is often unclear whether this is due to lack of information or because acidification is not a relevant issue.

Comparison of the acid sensitivity and deposition maps suggests that there are potentially additional countries with acidified surface waters than is suggested by the WFD reporting. According to the literature, acidified surface waters have previously been observed in the Vosges and Ardennes mountains in France, the Rila Mountains in Bulgaria and the Retezat Mountains in Romania. In the Slovakian part of the Tatra Mountains the literature confirms that many lakes are still acidified, while in Austria surface water acidification due to atmospheric deposition is no longer considered to be an important issue. Acidified lakes were observed in surveys in European Russia and Western Siberia, but the former survey was more than ten years ago. In Armenia acidified rivers are observed in parts of the country, but relevant data are limited. Summarising the combined information from the literature and the WFD, acidification is observed in Slovakia, Russia and Armenia, and is very likely to occur in Belgium and potentially also in Luxembourg. Deposition-induced acidification has previously been observed in France, Bulgaria and Romania, but present conditions are not known. In the Western Balkans there are indications that acidification may occur, but there are no monitoring studies to confirm this.

Conclusion

The current status of surface water acidification related to air pollution in Europe and North America has been assessed using country reports, monitoring data, critical loads and exceedance data, acid sensitivity and deposition maps, and data reported under the WFD. Acidification is still observed in many countries, despite widespread evidence of a gradual recovery of acid-sensitive waters in response to reduced sulphur and nitrogen deposition. However, the extent and severity of surface water acidification vary. Maps of acid sensitivity and deposition suggest that surface water acidification is present in regions and countries for which no data or reports were delivered for the current assessment. Supporting evidence is in many cases available from previous studies, but recent monitoring data would be necessary for substantiation. The submitted data show that existing national monitoring varies in the ability to assess the spatial extent of acidification and the recovery responses of acidified sites. The monitoring requirements under the European Union’s National Emission Ceilings (NEC) Directive, tailored to assess ecosystem impacts of air pollution, are expected to reverse the recent decline in the number of monitoring sites observed in some countries. Monitoring sites and parameters relevant to acidification are also highly relevant for detecting changes in water quality due to climate change. The information reported under the WFD is currently of limited value in assessing the extent of acidification of surface waters in Europe.

Chemical recovery can be slow and further impaired by factors such as climate change and intensified forestry. Furthermore, biological recovery can lag severely behind chemical recovery, due to wider constraints on species distributions, interactions and re-colonisation. Hence, further emission reductions will be required to enable significant ecological recovery of acidified surface waters in some areas and increase the rates of recovery across wider regions. Even by reaching the emission targets of acidifying compounds set for 2030, critical loads for surface waters will remain exceeded. This implies that the current emission reduction targets may not be sufficiently ambitious for protection of all surface waters from acidification. Despite large and effective efforts across Europe and North America to reduce surface water acidification, air pollution still constitutes a threat to freshwater ecosystems.

Short summary

- The current status of surface water acidification related to air pollution in Europe and North America has been assessed using country reports, monitoring data, critical loads and exceedance data, acid sensitivity and deposition maps and data reported under the European Commission's Water Framework Directive (WFD).
- Acidification is still observed in many countries, despite widespread evidence of a gradual recovery of acid-sensitive waters in response to reduced sulphur and nitrogen deposition. However, the extent and severity of surface water acidification vary.
- Of the countries that submitted data acidification occurs in large regions in Norway, Sweden, the UK, Canada and the US. In Finland and Germany acidification is more scattered, while in the Netherlands, the Czech Republic, Ireland, Italy, Switzerland, and Poland it is limited to a few smaller areas. The severity of acidification is not necessarily related to the spatial extent of the problem; local hot spots may occur even where acidification is not a major, regional issue, e.g. in the Czech Republic and Ireland. However, based on both extent and severity, surface waters in Italy and Poland and, to a lesser degree, Finland, Germany, Switzerland, Canada and the US seem closer to chemical recovery than waters in the other countries which submitted data. In Spain acidification is a limited issue, and in Latvia it is unlikely to occur at all.
- The submitted data show that existing national monitoring varies in the ability to assess the spatial extent of acidification and the recovery responses of acidified sites. The monitoring requirements under the European Union's National Emission Ceilings Directive, tailored to assess ecosystem impacts of air pollution, are expected to reverse the recent decline in monitoring observed in some countries. This monitoring is also highly relevant for detecting changes in water quality due to climate change.
- Maps of acid sensitivity and deposition suggest that surface water acidification is present in regions and countries for which no data or reports were delivered for the current assessment. Summarising the available information, acidification is observed in Slovakia, Russia and Armenia, and is very likely to occur in Belgium and potentially also in Luxembourg. Deposition-induced acidification has previously been observed in France, Bulgaria and Romania, but present conditions are not known, and there are indications that it may occur in the Western Balkans. More documentation is needed to assess the current extent of acidification in these countries.
- Monitoring under the WFD does not fully cover acid-sensitive waters, as these are often small headwater systems, which are not included in the scope of the WFD. Classification systems for acidification status could benefit from intercalibration. The reporting schemes allow for ambiguities. In total, the information reported under the WFD is currently of limited value in assessing the extent of acidification of surface waters in Europe.
- Chemical recovery can be slow and further impaired by factors such as climate change and intensified forestry, and biological recovery can lag severely behind chemical recovery. Hence, further emission reductions will be required to enable significant ecological recovery of acidified surface waters in some areas and increase rates of recovery across wider regions.
- More countries could benefit from submitting critical loads for acidification of surface waters under the LRTAP Convention.
- Even by reaching the emission targets of acidifying compounds set for 2030, critical loads for surface waters will remain exceeded. This implies that the current emission reduction targets may not be sufficiently ambitious for protection of all surface waters from acidification. Despite large and effective efforts across Europe and North America to reduce surface water acidification, air pollution still constitutes a threat to freshwater ecosystems.

1 Background and aims

Acidification of surface waters was one of the major environmental impacts of air pollution leading up to the signing of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP)[1] in 1979. The resulting reductions in nitrogen and, more particularly, sulphur deposition led to a gradual decline in acidification levels and extent of surface water acidification (Skjelkvåle et al., 2005; de Wit et al., 2015; Garmo et al., 2015). However, acidification is still prominent in many parts of Europe and North America. The role of ICP Waters is to continue to address and assess, on a regional basis, the degree and geographical extent of the impact of atmospheric pollution, in particular acidification, on surface waters.

Until now, ICP Waters has focused primarily on investigating trends in water chemistry for Europe and North America, to assess responses of surface waters to air pollution. In most cases, only a subset of available monitoring sites in each participating country have been included in regular reporting exercises. The objective of this report was to investigate the current extent of surface water acidification in Europe and North America (here defined as Canada and the United States (US)). This involved the use of more spatially-extensive water chemistry datasets to establish the current extent and severity of surface water acidification. In addition, identification of potentially acidified regions using sensitivity maps and deposition models would reveal whether the available data are sufficient to fully evaluate the current extent of surface water acidification at a continental scale. The report aims to inform policy processes on the need for further emission reductions, and also the need for monitoring of air pollution effects.

In addition to the LRTAP Convention, other regional legislation also addresses air pollution and effects on surface waters. In the new EU National Emission Ceilings (NEC) Directive[2], the emission reduction commitments for 2020 are in line with the revised Gothenburg protocol[3] of the LRTAP Convention, while the commitments for 2030 are more ambitious. Under the NEC Directive, the Member States are also obliged to monitor and report data on the impacts of air pollution, including the acidification of freshwaters. The Canada-United States Air Quality Agreement[4] is a collaboration framework that builds on national legislation (including the US Clean Air Act), but also covers, for example, emission reduction objectives and exchange of information from monitoring of ecosystem effects. According to the EU Water Framework Directive (WFD)[5] Member States are required to protect, enhance and restore their water bodies to achieve good surface water status by 2015, unless extensions are applied for. This includes good status with respect to acidification.

Acidification of surface waters requires two phenomena to be present: Acid deposition and sensitivity to acid deposition (Gorham, 1961; Wright and Henriksen, 1978). Acid deposition is dominated by oxidised sulphur and oxidised or reduced nitrogen compounds, resulting from emissions from industry and transport (Wright et al., 2018). The emitted compounds can travel long distances before being deposited (OECD, 1977).
Acid sensitivity is related to the buffering capacity of the catchment soils, which is controlled by a range of factors. Bedrock with minerals highly resistant to chemical weathering give rise to poorly-

[1] http://www.unece.org/environmental-policy/conventions/envlrtapwelcome/the-air-convention-and-its-protocols/the-convention-and-its-achievements.html
[2] Directive 2016/2284/EU: http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=uriserv:OJ.L_.2016.344.01.0001.01.ENG&toc=OJ:L:2016:344:TOC
[3] http://www.unece.org/env/lrtap/multi_h1.html
[4] https://www.canada.ca/en/environment-climate-change/services/air-pollution/issues/transboundary/canada-united-states-air-quality-agreement.html#q
[5] Directive 2000/60/EC: http://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX:32000L0060

buffered soils often characterised by low base saturation. With low base saturation the incoming H^+ ions in acid deposition will to a lesser extent be exchanged with base cations in the soil, leaving H^+ or aluminium ions to accompany the mobile acid anions (sulphate and nitrate) from the soil to the surface waters (Reuss and Johnson, 1986). Over time, acid deposition will further deplete the soils of base cations (Kirchner and Lydersen, 1995). Various intrinsic factors also affect the buffering capacity (Gorham, 1961). For example, soil thickness influences the contact time between the percolating water and the soil, as well as the total base cation content of the soil. Barren rock provides hardly any buffering, while thick soils may offer substantial buffering capacity even at low base saturation (Skjelkvåle and Wright, 1998). Areas glaciated during the Pleistocene such as Fenno-Scandia, the European Alps and the Carpathian Mountains, eastern Canada, and the north-eastern US, often have thin and patchy soil overlying bedrock. Geological variability within catchments is essential as well. Small patches of more calcareous bedrock or marine clays within a catchment may be sufficient to avoid surface water acidification. In catchments dominated by laterally-deposited unconsolidated material, the acid sensitivity can be more dependent on the weatherability of this material than the underlying bedrock (Wright, 1983; Clow et al., 1996). Acid sensitivity can thus vary highly between regions and also between individual surface waters in the same region (Skjelkvåle and Wright, 1990, 1998).

The effects of acidification on freshwater ecosystems have been thoroughly studied and documented, in particular since the 1970s. Water acidity is often found to exert a tight control on the health and species composition of biological communities ranging from primary producers, e.g. algae (e.g. Battarbee and Charles, 1986), to top predators, including fish (Baker et al., 1993), and there is widespread evidence that acidification has led to major losses in diversity and shifts in species composition (e.g. Stoner et al., 1984; Hesthagen and Hansen, 1991; Herrmann et al., 1993; Watt et al., 2000; Fjellheim and Raddum, 2001; Lacoul et al., 2011). Several physiological effects are related to toxic effects of elevated hydrogen ion, and particularly inorganic aluminium, concentrations (e.g. Rosseland and Staurnes, 1994; Gensemer and Playle, 1999). Aluminium toxicity in fish is related to effects on the gills, which can be ionoregulatory or respiratory effects, depending on the pH (Exley et al., 1991). The toxic effects are reduced at higher calcium concentration and in the presence of complexing agents such as dissolved organic matter (Howells et al., 1983; Neville, 1985). There is evidence that sensitive species are beginning to recolonise and increases in abundance in chemically recovering waters (Monteith et al., 2005), but these ecosystems are often considered to remain impoverished relative to their pre-acidification state (Battarbee et al., 2014).

The concept of critical loads has been central to the work on emission reductions under the LRTAP Convention (CLRTAP, 2015a). A critical load is defined as “a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge” (Nilsson and Grennfelt, 1988). As such the critical load is also a measure of the acid sensitivity – areas with low critical loads have a higher sensitivity to acid deposition than areas with higher critical loads. Exceedance of the critical loads means that the systems receive a higher deposition load than they can tolerate and depends both on the critical load and the deposition level.

Critical loads are by nature steady-state (CLRTAP, 2017). This means that the exceedance reflects the conditions at the point in time at which the ecosystem has fully adapted to a given level of acid deposition. If deposition levels for a certain system have declined so that the current deposition level no longer exceeds the critical load, it does not necessarily mean that the system is no longer acidified, as a new steady-state may not yet have been reached. Recovery takes time. With respect to surface waters this requires that the base saturation of the soil is restored, through weathering and sea salt deposition. Given that acid deposition has generally been declining over recent decades, currently acidified areas are likely to be more extensive than areas deemed to be exceeded based on current deposition.

Full assessment of the current extent of surface water acidification cannot therefore be achieved solely using exceedance maps. In this report different sources of information have been used to identify regions with surface water acidification or where surface water acidification is likely to occur. This information is structured in two different parts: Chapter 3 provides regional overviews of acid sensitivity and areas with potentially acidified surface waters, overviews of current acidification status based on monitoring data and a summary of data on acidification status as reported under the WFD. Chapter 4 provides several more detailed national assessments. The results from these two parts are combined and put into context in chapter 0, ending in some conclusions and recommendations in chapter 6.

2 Methods

2.1 Mapping of acid-sensitive and potentially acidified surface waters

Regional maps of sensitivity to surface water acidification and potentially acidified areas were produced using two different approaches:

1) Critical loads and exceedance: The critical loads and exceedance maps for Europe in the status reports (e.g. Hettelingh et al., 2017) from the Coordination Centre for Effects (CCE) show results for a combination of habitats. To evaluate critical loads and exceedances only for surface waters, maps were produced using critical loads for acidification for the EUNIS habitat class[6] C only (inland surface water). The latest submitted critical loads were used (approved autumn 2017). Exceedance of critical loads for surface waters were estimated using the standard method (CLRTAP, 2015b) and nitrogen and sulphur deposition for 2015 from the European Monitoring and Evaluation Programme/Meteorological Synthesizing Centre - West (EMEP/MSC-W)[7]. Critical loads for the US were calculated using the Steady-State Water Chemistry (SSWC) model (Lynch et al., 2017), and the deposition used to calculate exceedances was average deposition for 2013-2015. The same model was applied for Canada, and here exceedance was calculated for 2010.
2) Geology and deposition: A map of potential sensitivity to surface water acidification was produced using a global geological map with broad bedrock categories (Hartmann and Moosdorf, 2012). These bedrock categories were assigned to acid sensitivity categories by geological experts at the University of Oslo, based on e.g. potential weathering rates (Table 1).
   Given the range of factors governing acid sensitivity of surface waters (cf. chapter 1), such a map will only give a crude indication of the potential sensitivity. The main purpose was to identify potentially sensitive regions in countries where critical loads were not available. To identify regions with potentially acidified surface waters, the bedrock sensitivity map was compared with deposition maps. The data behind the deposition maps for Europe were from EMEP/MSC-W[7]. Deposition maps for the US (for 2013-2015) were produced by CASTNET/CMAQ/NTN/AMON/SEARCH[8].

[6] https://www.eea.europa.eu/data-and-maps/data/eunis-habitat-classification
[7] http://emep.int/mscw/index_mscw.html
[8] https://www.epa.gov/cmaq/estimating-atmospheric-deposition-cmaq

Table 1 Potential acid sensitivity of surface waters for different categories of underlying bedrock

Geological category	Acid sensitivity category	Confidence
Unconsolidated sediments	Low	Low
Siliciclastic sedimentary rocks	High	High
Pyroclastics	Unknown	
Mixed sedimentary rocks	Medium	Low
Carbonate sedimentary rocks	Low	High
Evaporites	Low	High
Acid volcanic rocks	High	High
Intermediate volcanic rocks	Medium	High
Basic volcanic rocks	Low	High
Acid plutonic rocks	High	High
Intermediate plutonic rocks	High	High
Basic plutonic rocks	Medium	High
Metamorphics	High	High

2.2 Call for national contributions

The ICP Waters database contains water chemical data from the countries taking part in the programme, submitted annually by their National Focal Centres (NFCs). The data collection focuses on small catchments with high frequency of sampling (ICP Waters Programme Centre, 2010). This is ideal for studying temporal trends and responses to short-term events. However, for assessing the spatial extent of surface water acidification, this set of sites is not sufficient. This assessment was thus dependent on the NFCs to submit data from a larger set of sites. The focus was on acid-sensitive regions, where surface water acidification could potentially occur. The knowledge of the NFCs of sensitive areas and the status of surface water acidification in their countries was also essential to the assessment. Thus, an enquiry about data availability was sent to the NFCs in the spring of 2017, followed by a call for contributions in July 2017. The content of the call is summarised in sections 2.2.1 and 2.2.2.

2.2.1 Call for national data

The aim was to get the best possible overview of the acidification status in each country by obtaining recent data that are representative of surface waters in acid-sensitive regions. Both monitoring data, modelling data and extrapolated data could be submitted, and slightly older data could be allowed if this increased the number of sites. In practice the NFCs were asked to make this trade-off between quality, representation of regional conditions, and age of the data, and submit the best possible data set suited for the purpose. Data were requested for acid-sensitive regions only, except for countries where non-sensitive areas are only small and scattered, in which case data from the whole country could be submitted.

An excel template was provided for data delivery and required some basic site information, including coordinates and catchment area. Only four chemical parameters were requested: Dissolved Organic Carbon (DOC) concentration (mg/l) (alternatively total organic carbon (TOC), if DOC constitutes >90%), Acid Neutralising Capacity ($ANC=Ca+Mg+Na+K-Cl-SO_4-NO_3$ (all concentrations, in µeq/l)), ANC adjusted for organic acids ($ANC_{oaa}=ANC-(3.4*DOC)$)[9] and pH. The latter three are all indications of acidification, while DOC is used as a supporting parameter (cf. section 2.3). Only average values were requested, i.e. one value per parameter per site. The number of samples to include in the average

[9] DOC in mg C/l; 3.4 in µeq/mg C is the charge density of the organic acids, assuming a site density of 10.2 µeq/mg C and that 1/3 of the sites are permanently deprotonated (Lydersen et al., 2004)

would depend on the variability and data availability. This was left for the NFCs to decide. The data should, however, represent the current situation, i.e. preferably 2010-2016.

2.2.2 Call for country reports

The country reports were intended to provide a more detailed overview of acid sensitivity and acidification status in the countries. The guidance to NFCs was relatively non-prescriptive, leaving them to decide on the content based on data availability and the acidification situation in their country. The main suggested items to be addressed were:

- Acid sensitivity: The extent and degree of acid sensitivity in the country, acid-sensitive regions
- Monitoring and assessment approach: Acidification monitoring in the country in general, methodological background for the submitted data
- Acidification status: Assessment based on the submitted data and/or other sources of information, including e.g. upscaling, case studies, trends and outlooks

Country reports from countries with no acid-sensitive regions were also encouraged. These could be brief chapters stating/explaining the lack of acid sensitivity and referring to any surveys of acidification confirming this. Data from such countries was not expected.

2.3 Assessment of extent of acidification based on national data

To assess the extent of surface water acidification, a basis for defining surface waters as acidified is needed. However, there is no general, fixed definition. Different countries use different definitions. Thresholds for acceptable levels of acidity (critical limits) can be set based on relationships between chemical and biological variables. These relationships are complex, and in practice different systems will have different critical limits. Systems with naturally higher base cation levels will have ecosystems adapted to these conditions, and a higher ANC is needed to keep these systems intact and vice versa (Henriksen and Posch, 2001). In humic systems, a higher ANC is required to avoid damage to the aquatic organisms due to the acidity caused by the organic acids (Hesthagen et al., 2008). The critical limits also only define conditions with respect to acidity. To assess whether the systems are actually acidified, modelling of pre-industrial water quality must be applied, as in the Swedish assessment system (cf. section 4.12.2).

In the analysis of the national data, both ANC, ANC_{oaa} and pH were used in assessing the degree of acidification. To be able to compare the results in a simple manner, some generally accepted critical limits were used, and sites with data below these critical limits were considered to be acidified. A commonly used critical limit for acidification for ANC is 20 µeq/l, originally based on responses of fish and invertebrate populations to acidification (Lien et al., 1996). This is also frequently used as critical limit in critical loads calculations (CLRTAP, 2017). The ANC_{oaa} takes the strong acid component of the organic acids into account, and can in this sense be considered to be a better indicator. A critical limit of 8 µeq/l was proposed by Lydersen et al. (2004), based on relationships between brown trout population status and ANC_{oaa} in Norwegian lakes. By implication these highly acid-sensitive organisms should be able to tolerate ANC levels below 20 µeq/l in waters where DOC concentrations fall beneath 3.5 mg/l[10] (and as little as 8 µeq/l at DOC = 0 mg/l) but would require an ANC above 20 µeq/l in more humic waters (i.e. to achieve an ANC_{oaa} above the critical limit). The ANC_{oaa} critical limit, however, has not been so widely used, and may not necessarily apply everywhere. More weight was thus given to the ANC results. pH can be difficult to measure accurately, and is usually associated with more uncertainty than ANC (Escudero-Oñate, 2017). pH is also an ambiguous

[10] According to the relationship between ANC and ANC_{oaa} (cf. section 2.2.1), an ANC of 20 µeq/l corresponds to an ANC_{oaa} of 8 µeq/l if the DOC concentration is 3.5 mg C/l (3.4 µeq/mg C*3.5 mg C/l = 12 µeq/l).

indicator, as it reflects both natural and anthropogenic acidity. Many surface waters can have low natural pH due to high concentrations of organic acids, although very low pH levels (below e.g. 4.5) usually indicate acidification. Here pH 5.6 (the pH of pure water in equilibrium with the atmosphere) was used as a cut-off for indicating potential acidification. However, when comparing pH values, DOC concentrations must always be considered. Even at fairly low DOC concentrations (here a threshold was set at 6 mg/l), organic acids will affect the pH.

2.4 Acidification status reported under the WFD

Under the WFD the countries are required to assess and report the ecological status of their water bodies. Lake water bodies are defined as entire lakes or parts of lakes larger than 0.5 km^2, and river water bodies are stretches of rivers or river networks with a catchment area greater than 10 km^2. The countries can define also smaller lakes and river stretches as WFD water bodies if they so wish. Ecological status is assessed based on several quality elements (QEs), which can be biological, hydromorphological or physico-chemical. The latter two can be used only to downgrade the ecological status set based on the biological QEs. Acidification status is one of the physico-chemical QEs and must be at least good for the water body to achieve good ecological status. The normative definition of good acidification status is that “pH, acid neutralizing capacity (…) do not reach levels outside the range established so as to ensure the functioning of the type specific ecosystem and the achievement of the values specified above for the biological quality elements”[11]. How this is defined quantitatively, i.e. the classification system, is decided by the individual countries in line with general WFD principles. While intercalibration of biological QEs is laid down in the directive, there are no such requirements for other QEs.

The countries “shall monitor parameters which are indicative of the status of each relevant quality element”[12]. In surveillance monitoring parameters indicative of all QEs should be monitored, while in the operational monitoring only the parameters that are indicative of the QEs most sensitive to the identified pressures should be monitored. Grouping, modelling and expert judgement can also be used in classification. QE status class should be reported “for each of the relevant QEs that have been assessed for the water body and subsequently used to classify the ecological status or potential of the water body. If the status of QEs is not reported then it is assumed that it is not used in the classification of the ecological status of the water body”[13]. Acidification status should thus be reported whenever acidification has been considered relevant in the classification of ecological status. This should at least include acid-sensitive water bodies in areas that have received or still receive acid deposition, potentially also acid-sensitive water bodies in general. Acidification status can be reported as “High”, “Good”, “Less than good”, “Unknown”, “Not applicable” or “Monitored but not used” (Monitored but no standard has been developed and/or the QE is not used for status assessment). A low proportion of water bodies in the categories “Unknown” means that the classified data to a larger extent can be interpreted as representing water bodies where acidification can be a relevant issue.

In addition to status, the countries are also required to report the significant pressures to which the water bodies are subject and the resulting significant impacts. A significant pressure is one that contributes to an impact that may prevent the water body from achieving good ecological or chemical status. Pressures and impacts should only be reported if they cause a risk that good status is

[11] WFD, Annex V, ch. 1.2
[12] WFD, Annex V, ch. 1.3
[13] WFD Reporting Guidance 2016, Final – version 6.0.6, section 2.4.3.2: https://circabc.europa.eu/sd/a/5b969dc0-6863-4f75-b5d8-8561cec91693/Guidance%20No%2035%20-%20WFD%20Reporting%20Guidance.pdf

not achieved[14]. Several pressure types are defined[15] and a water body can be subject to none, one or several of these. Diffuse pollution from atmospheric deposition is one such pressure type. It may, however, not only comprise deposition of acidifying compounds, but also include, for example mercury deposition, or it may primarily refer to nitrogen deposition, but as a eutrophying, not acidifying agent. Likewise, acidification is one of several impact types[16], but there is no differentiation as to whether the acidification is caused by atmospheric deposition or local pollution. There are also pressure and impact types indicating that the water body is not subject to any significant pressures or impacts, respectively.

Data reported by the countries were extracted from the WFD database on June 5th 2018[17]. Acidification status was analysed for lake and river water bodies that have been classified with respect to ecological status (close to all delineated lake and river water bodies). The proportion of water bodies in each reporting category was presented per country. Relevant pressures and impacts information, as well as ecological status, was analysed for water bodies in less than good acidification status, to identify the cause of this acidification status and examine the consistency in the reporting. Water bodies with less than good acidification status with atmospheric deposition as a pressure are likely to be impacted by air pollution. Other pressure types that may cause acidification are those related to mining or industry, as well as historical pollution. The combination of acidification being identified as an impact and the designation of less than good ecological status would imply that the impaired acidification status is associated with ecological damage.

[14] WFD Reporting Guidance, section 2.3.1
[15] WFD Reporting Guidance, Annex 1a
[16] WFD Reporting Guidance, Annex 1b
[17] https://www.eea.europa.eu/themes/water/water-assessments/quality-elements-of-water-bodies
https://www.eea.europa.eu/themes/water/water-assessments/pressures-and-impacts-of-water-bodies

3 Results

3.1 Acid sensitivity of surface waters in Europe and North America and areas with potentially acidified surface waters

3.1.1 Europe

Critical loads for acidification of surface waters were only available for seven European countries (Figure 1). The map shows that the lowest critical loads (< 200 eq/ha/yr) are found in almost all of Sweden and southern Norway. Low critical loads are also found in eastern and central Finland, Scotland, northern England, parts of Wales, western Ireland, eastern Netherlands, and parts of the Swiss Alps. The exceedance map shows that in these regions the critical loads are still exceeded, meaning that surface waters in these regions are likely to be acidified. For the rest of the countries no information is available.

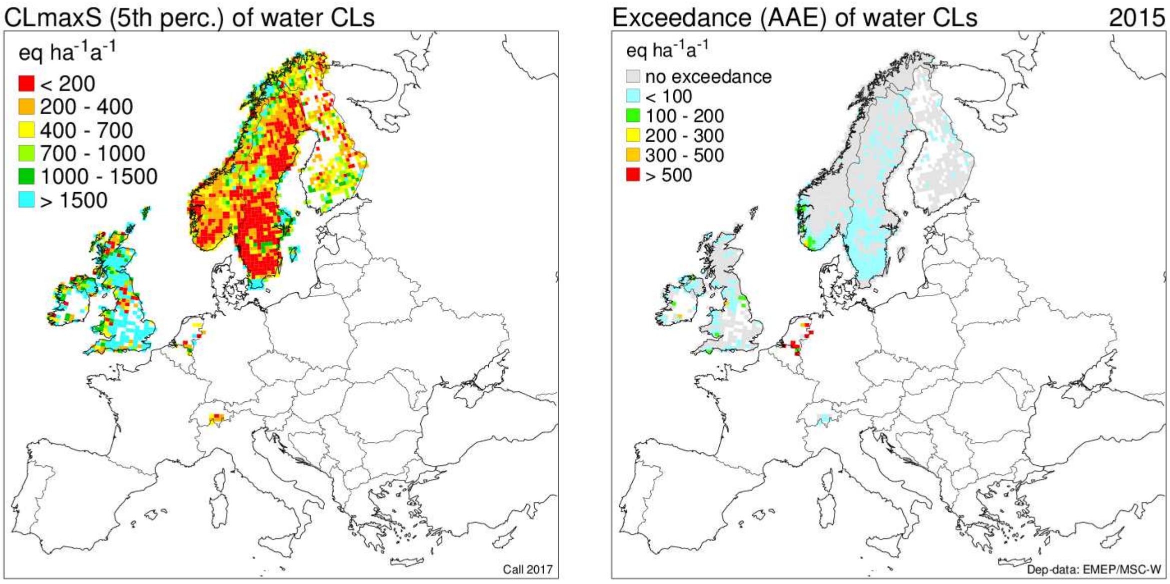


Figure 1 Left: Critical loads (CLs) for acidification of surface waters (sulphur deposition) for some European countries. 5th perc. indicates that the colour of each grid cell represents the 5th percentile of the distribution of critical loads reported for the grid cell. Right: Exceedance of critical loads for acidification of surface waters for some European countries, given the deposition in 2015. AAE = average accumulated exceedance, i.e. area weighted average exceedance for the different reported areas per grid cell (not necessarily adding up to the total area of the grid cell).

The map of potential acid sensitivity for Europe based on bedrock (Figure 2) shows some deviation from the map of critical loads. For example, the low critical loads in the western Netherlands are not reflected as high sensitivity in the bedrock geology map. The critical loads here are calculated for ponds which are poorly buffered not due to the underlying bedrock, but because of sandy soils and that they are largely rain-fed. Likewise, the bedrock geology map indicates that a larger part of Scotland is highly sensitive than what the critical loads map suggests, perhaps reflecting its low spatial resolution in this geologically diverse region. The potential sensitivity map for Norway which was produced using the same principles, but with a higher resolution bedrock map (Figure 37, left), illustrates that the European map provides a broad scale picture only. Still, in most cases there is a fairly good correspondence between the two maps, meaning that the sensitivity map can be used as an indication of acid-sensitive areas for countries not covered by the critical loads map.

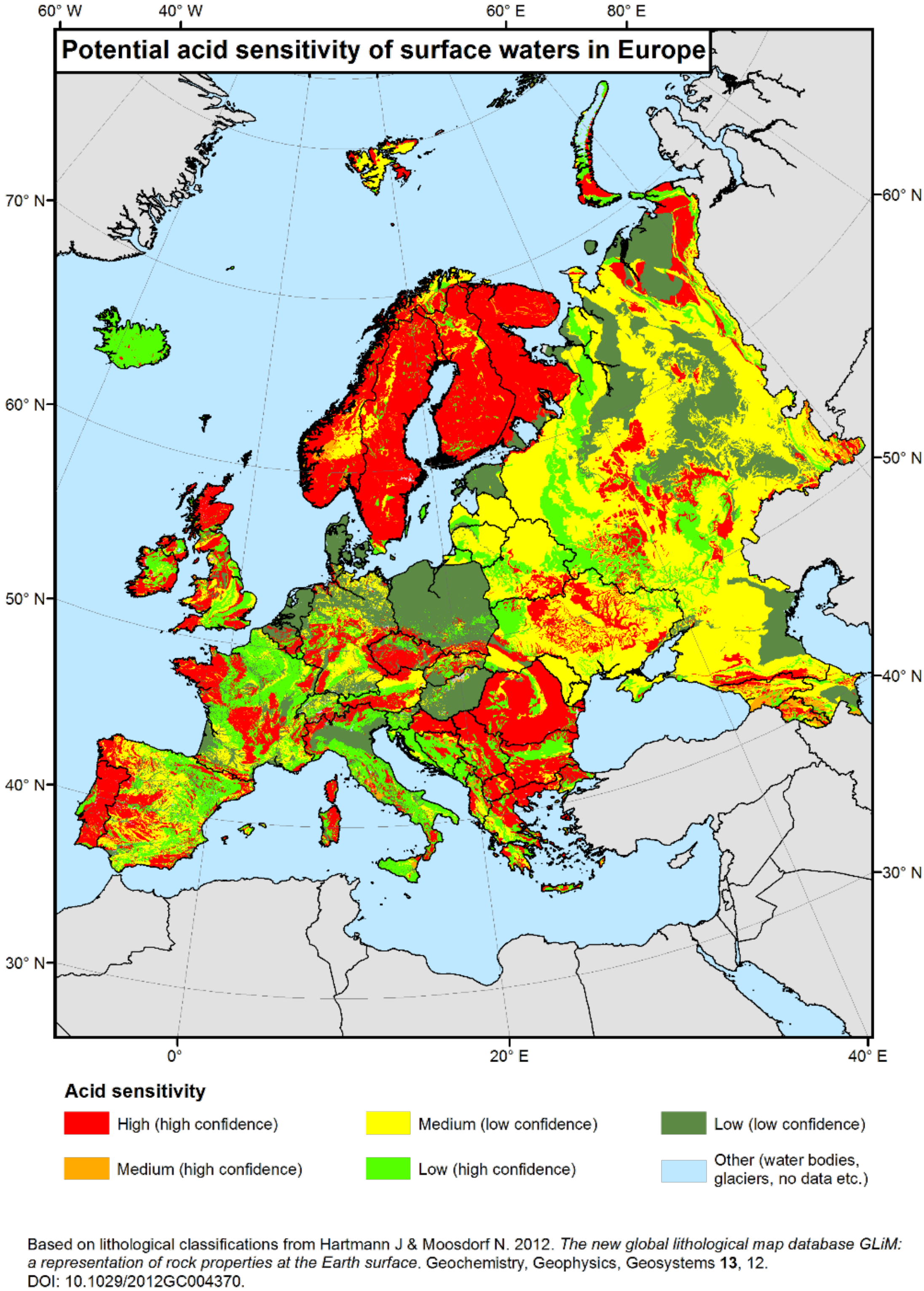


Figure 2 Potential acid sensitivity of surface waters in Europe (and the three Caucasus countries), based on bedrock categories only.

The bedrock geology map indicates that high acid sensitivity may occur in several of the areas for which no information on critical loads is available. However, not all these areas currently receive high levels of deposition (Figure 3). For the countries where critical loads are lacking, a comparison of the potential acid sensitivity with atmospheric deposition suggests that acidified surface waters may occur particularly in the following regions: the Pyrenees, the border regions of Belgium, Luxembourg, France and Germany, the mountainous regions on the borders of the Czech Republic, Germany and Austria, the Tatra Mountains, the Italian Alps, northern Croatia, parts of Bosnia and Herzegovina and Serbia, western Albania, parts of western Russia, and central Armenia. In other regions not covered by the critical loads deposition is low (e.g. Portugal) and/or the potential sensitivity is low (e.g. eastern Ukraine).

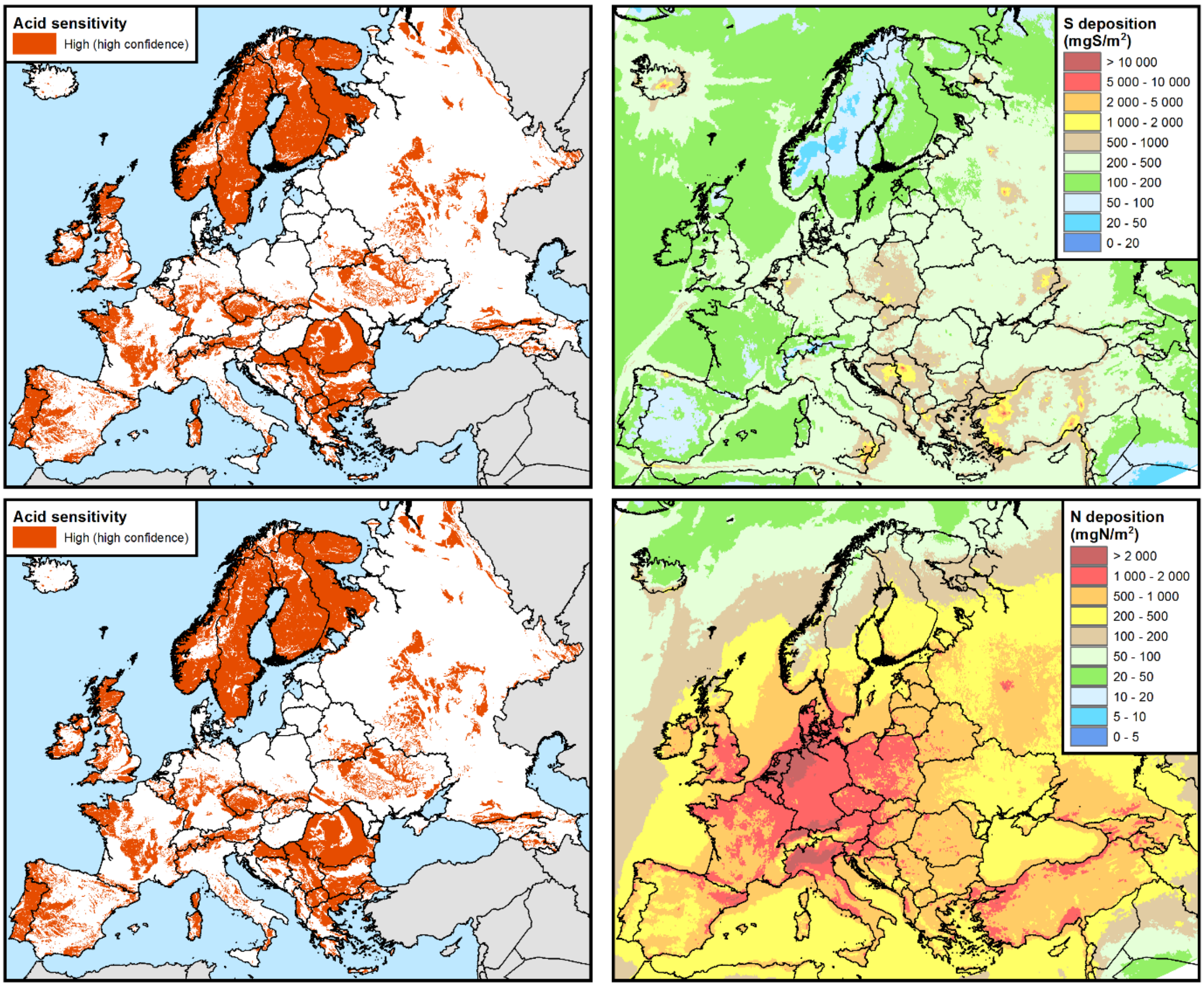


Figure 3 Left: Areas with potentially high acid sensitivity of surface waters. Right: Sulphur deposition (top) and nitrogen deposition (bottom) in 2015. Source: EMEP/MSC-W.

3.1.2 North America

In North America the bedrock geology map shows large areas underlain by bedrock with low weathering rates (Figure 4). Surface waters in these areas may potentially be acid sensitive. Much of the land area in the US and Canada, however, is characterized by well-buffered soils, and thus surface waters are not acid sensitive despite lying in areas with slowly-weathering bedrock. This is confirmed by the critical loads for the US (Figure 59) and Canada (Figure 5), showing that the areas with low critical loads (< 200 eq/ha/yr) are smaller than the areas with high sensitivity in the bedrock based map. Elevated rates of sulphur and nitrogen deposition occur in much of the north-eastern part of the US, as well as scattered parts of the south-east (Figure 6). However, given that not all these areas have sensitive surface waters, the exceedance of critical loads (Figure 6) is limited to the forested and mountainous areas of the North-east and the Mid-Atlantic regions, as well as parts of Florida, Michigan and Wisconsin. In Canada the largest exceedances are found in the east and on the western coast. Exceedances are also observed in northern Saskatchewan. The discrepancy between the distribution of critical loads and exceedances reflects the deposition pattern, with higher deposition in the east.

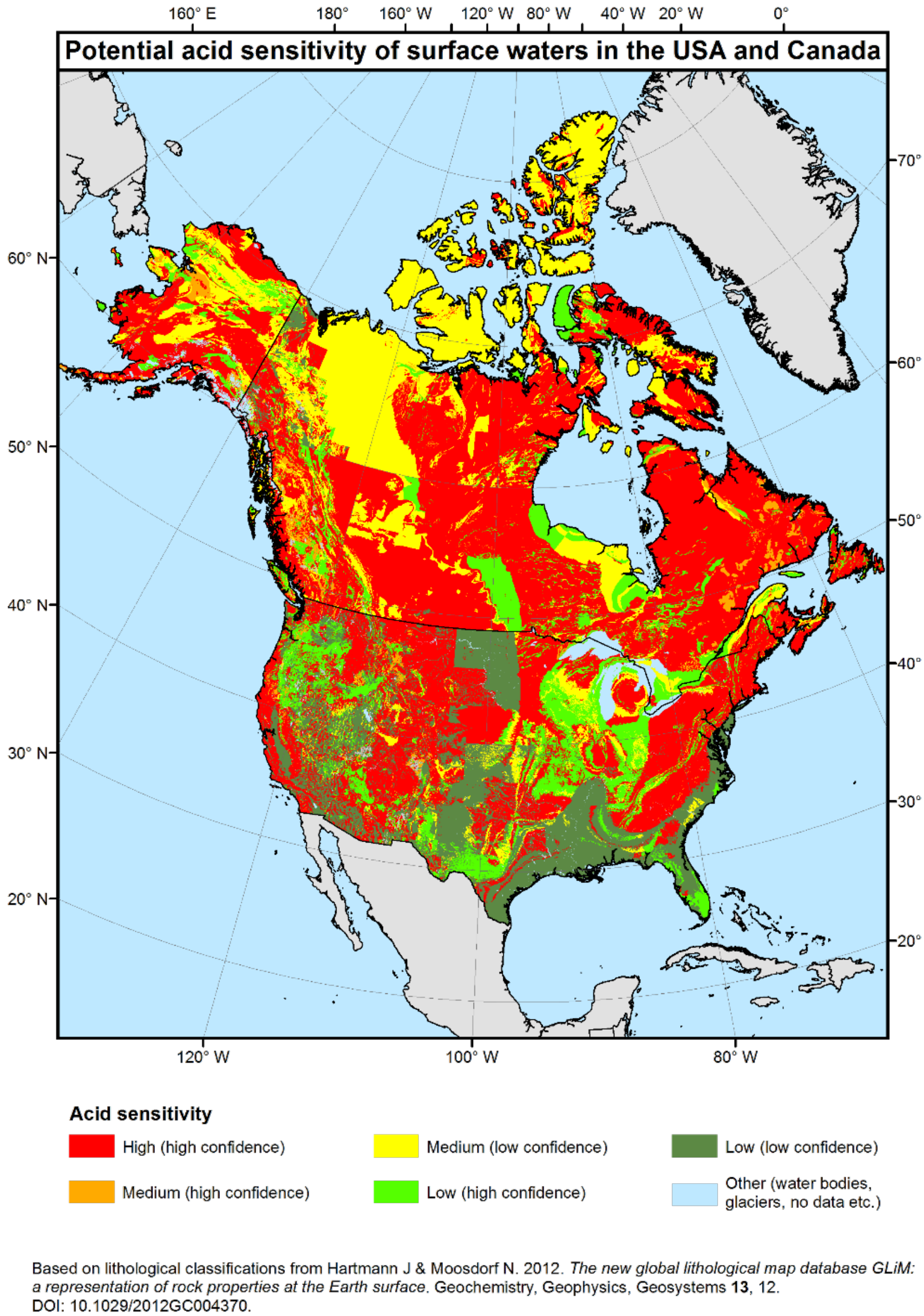


Figure 4 Potential acid sensitivity of surface waters in North America based on bedrock categories only.

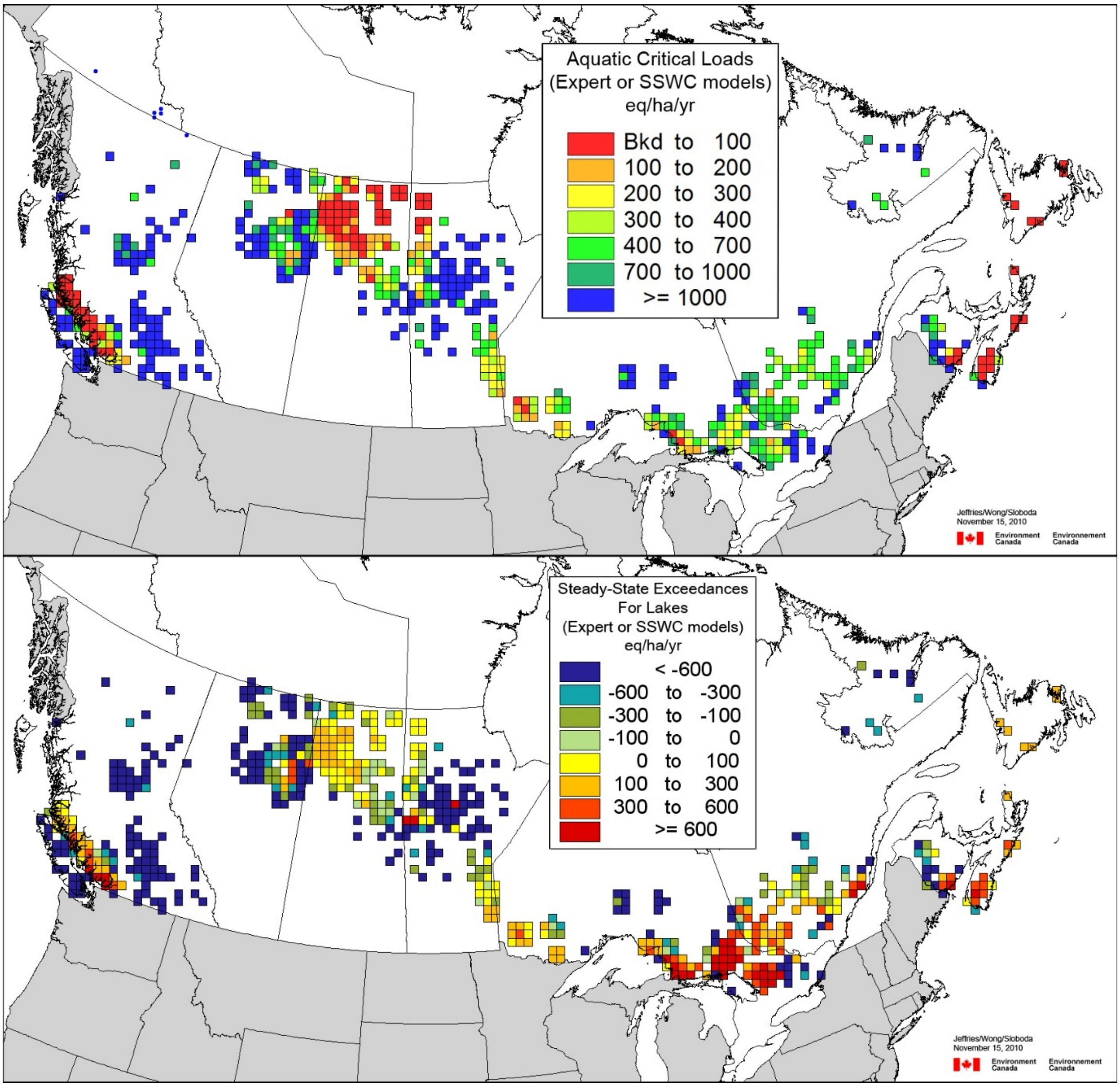
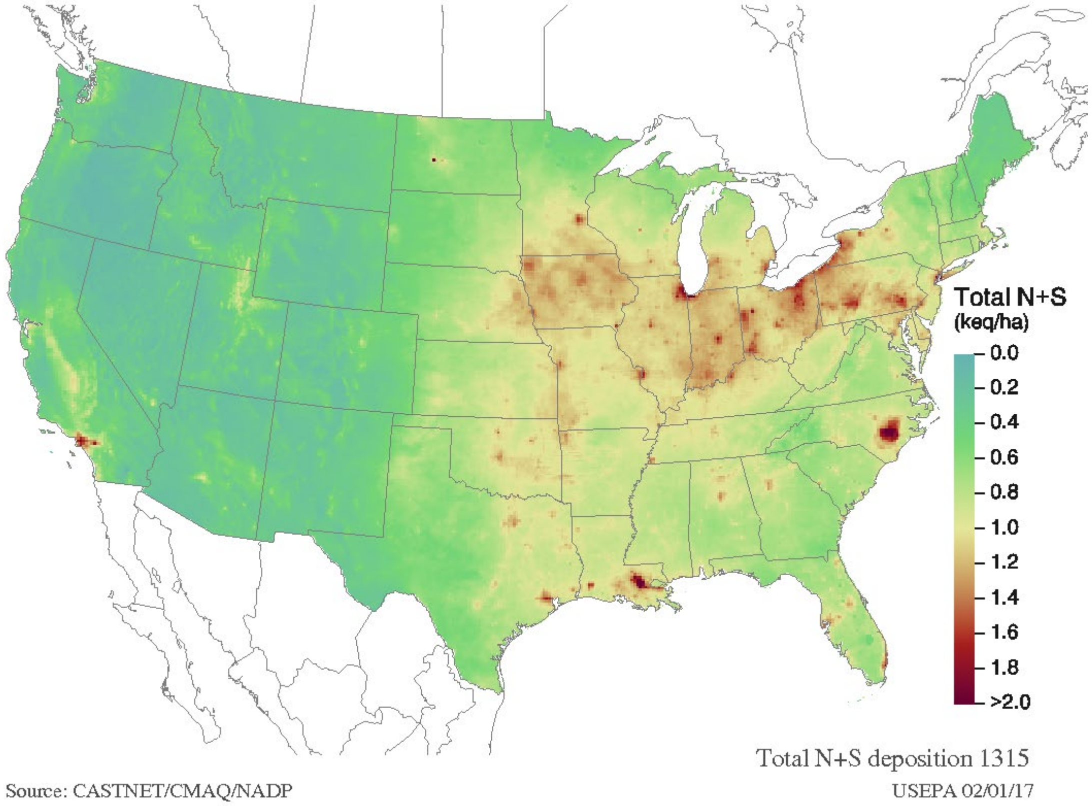


Figure 5 Critical loads for acidification of surface waters in Canada (top) and their exceedances using 2010 deposition.



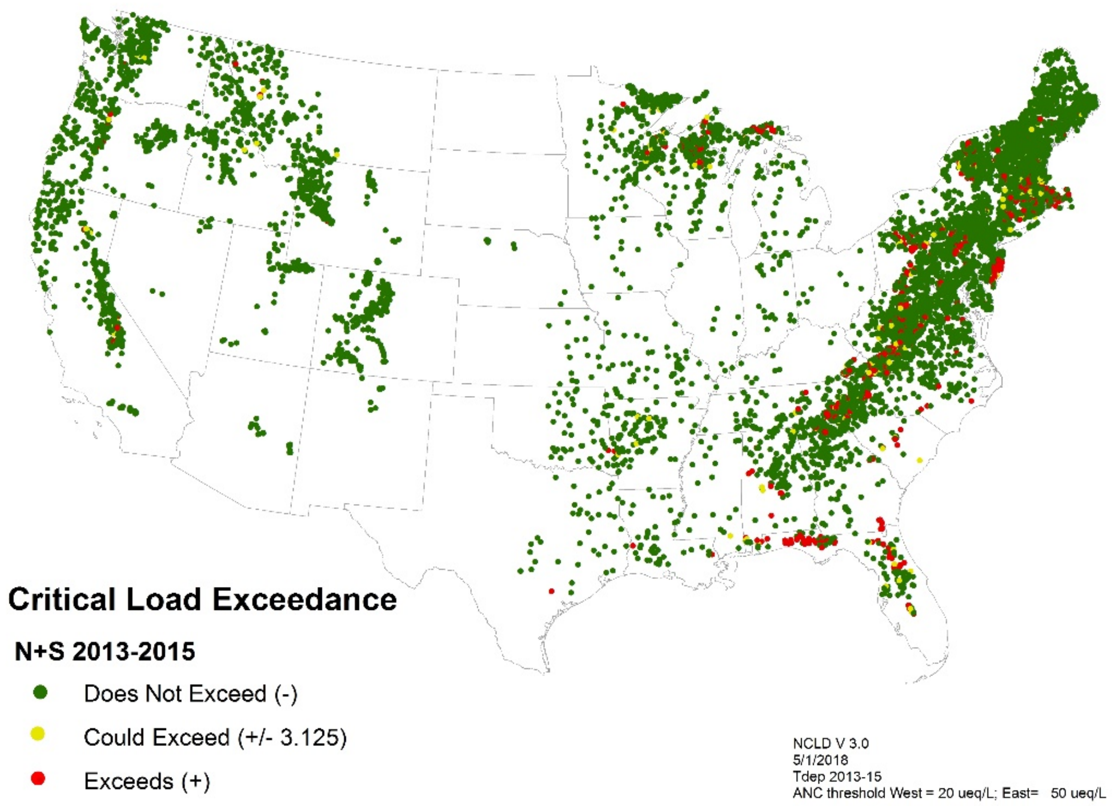


Figure 6 Top: Combined nitrogen and sulphur deposition in the US, average of 2013-2015. Bottom: Exceedance of critical loads for sulphur deposition in the US (cf. Figure 59) for 2013-2015. Source: Lynch et al. (2017).

3.2 National data on current extent of acidification

Data on current acidification status were submitted by 12 countries (Table 2). All submitted data were monitoring data. The spatial coverage of the submitted data varies considerably from between countries (Figure 7, Figure 8). In general, the data represent acid-sensitive sites or regions only and cannot be extrapolated to the country as a whole. A few countries collect national or regional data through extensive surveys and can state with some confidence the proportion of acidified lakes and/or streams within their acid-sensitive regions (Sweden, the US, Canada, Norway, Ireland); some countries monitor fewer sites in parts of their territory (Finland and Germany); while others focus on particularly sensitive or affected regions (the Czech Republic, Switzerland, Italy, Poland). The United Kingdom (UK) survey was based only on sites where critical loads had previously been found to be exceeded and were still expected to be exceeded by 2020, while the data from the Netherlands represent a particularly sensitive habitat type. Care must be taken in comparing the summary results for such different data sets.

Table 2 Overview of data submitted by the countries

Country	Lakes	Rivers/ Streams	Time period	Type of data
Canada	1581		2010-2016	Acid-sensitive lakes, regional surveys and long-term monitoring
Czech Republic	6	14	2010-2015	Streams in small forested catchments in acid-sensitive regions, Bohemian forest glacial lakes (acid sensitive)
Finland	18		2014-2016	Remote headwater forest lakes of the most acidified and acid-sensitive areas. Scattered around the country, covering gradients in deposition, climate and landscape
Germany		19	2012-2016	Acid-sensitive stream sites scattered around the country, selected to assess the regional degree of acidification of surface waters
Ireland	92		2010-2017	Acid-sensitive lakes (WFD surveillance monitoring and monitoring of small, upland headwater lakes)
Italy	30	1	2013-2017	Acid-sensitive sites in the alpine and subalpine areas of north-western Italy (Lake Maggiore watershed)
Netherlands	11		2013-2014	Small, acid-sensitive moorland pools in the eastern and southern parts of the country
Norway	178	52	2014-2017	Lakes in acid-sensitive areas, scattered around the country, but with a bias towards southern Norway. Six intensively monitored acid-sensitive streams or outlets of small lakes. Reference rivers unaffected by point sources scattered around the country
Poland	11	10	2011-2016	Sites in acid-sensitive regions, covering a variability in types of water bodies, location and land use
Sweden	94	52	2010-2016	Lakes and streams from national monitoring with catchment area < 1000 km^2 and which are not located in areas with calcareous bedrock or soil
Switzerland	52		2015-2017	Acid-sensitive lakes in the Central Alps
United Kingdom	119	41	2010	Acid-sensitive sites where deposition estimates suggested critical loads would continue to be exceeded by 2020
United States	74 (231)	60 (320)	2013-2016 (1991-1994)	Representative surveys of four acid-sensitive regions (combined to North-east lakes and Mid-Atlantic streams). The non-sensitive sites (in parentheses) within the regions are not monitored regularly.

The largest proportion of sites with low ANC (< 20 µeq/l) is found in the Netherlands, the UK, Norway, the Czech Republic, Switzerland and Finland (Table 3). The highest percentage is observed for the Netherlands and the UK, which are also the countries where the bias in the site selection towards potentially acidified sites probably is largest. For the UK, Norway and Finland, the sites are spread across the countries to a larger degree than for the remaining three countries. In the UK a majority of the low ANC sites occur in north England and Scotland, in Norway in the south and south-west and in Finland in the central/northern part of the country – thus broadly corresponding to the most sensitive areas (Figure 1).

Using the ANC_{oaa} criterion changes the picture somewhat[18]. The proportion of sites below the applied critical limit (< 8 µeq/l) is higher for the UK, the Czech Republic and to some extent the Netherlands, and somewhat lower for Norway and Finland. For Switzerland the proportion of sites below the critical limit is far lower, due to the very low concentrations of DOC in these lakes. This is also reflected in the low proportion of sites with pH < 5.6 in Switzerland. In Ireland the proportion of sites below the critical limit nearly doubles when using ANC_{oaa}, but remains relatively low. In Germany it stays the same, at an intermediate level, while in Canada, Italy, Poland, Sweden and the US the proportion of sites below both the ANC and the ANC_{oaa} critical limit is low. In Canada the relatively small proportion represents a large number of sites, though. These are mainly found in eastern Ontario/western Quebec (the Canadian Shield region) and in central, coastal British Columbia.

Table 3 Summary results per country, percentage of sites below selected critical limits for ANC, ANC_{oaa} and pH, and above 6 mg/l of DOC.

	ANC		ANC_{oaa}		pH		DOC	
Country	**Total**	**% < 20 µeq/l**	**Total**	**% < 8 µeq/l**	**Total**	**% < 5.6**	**Total**	**% > 6 mg/l**
Canada	1581	6	1581	8	1581	14	1581	43
Czech Republic	20	30	20	40	20	50	20	50
Finland	18	22	18	17	18	22	18	22
Germany	19	16	19	16	19	32	15	33
Ireland	92	7	92	13	92	27	92	49
Italy	31	3	31	0	31	0	31	0
Netherlands	11	45	6	50	11	82	7	86
Norway	230	33	230	28	230	35	230	19
Poland	12	8			21	5	4	0
Sweden	146	1	146	5	146	16	146	75
Switzerland	52	25	52	10	52	2	52	0
United Kingdom	157	39	108	63	118	53	118	64
United States lakes	8677[a]	0.1[b]	8677[a]	1[b]	8677[a]	7[b]	8677[a]	35[b]
United States streams	185617 km[c]	3[d]	185617 km[c]	3[d]	185617 km[c]	7[d]	185617 km[c]	5[d]

[a] Estimated population size, based on probability sample of 305 lakes
[b] The percentage results from a weighted analysis and represents the entire lake population within the region
[c] Estimated population size, based on probability sample of 380 streams
[d] The percentage results from a weighted analysis and represents the entire stream length within the region

[18] Not available for Poland, and for somewhat fewer sites in the Netherlands and the UK

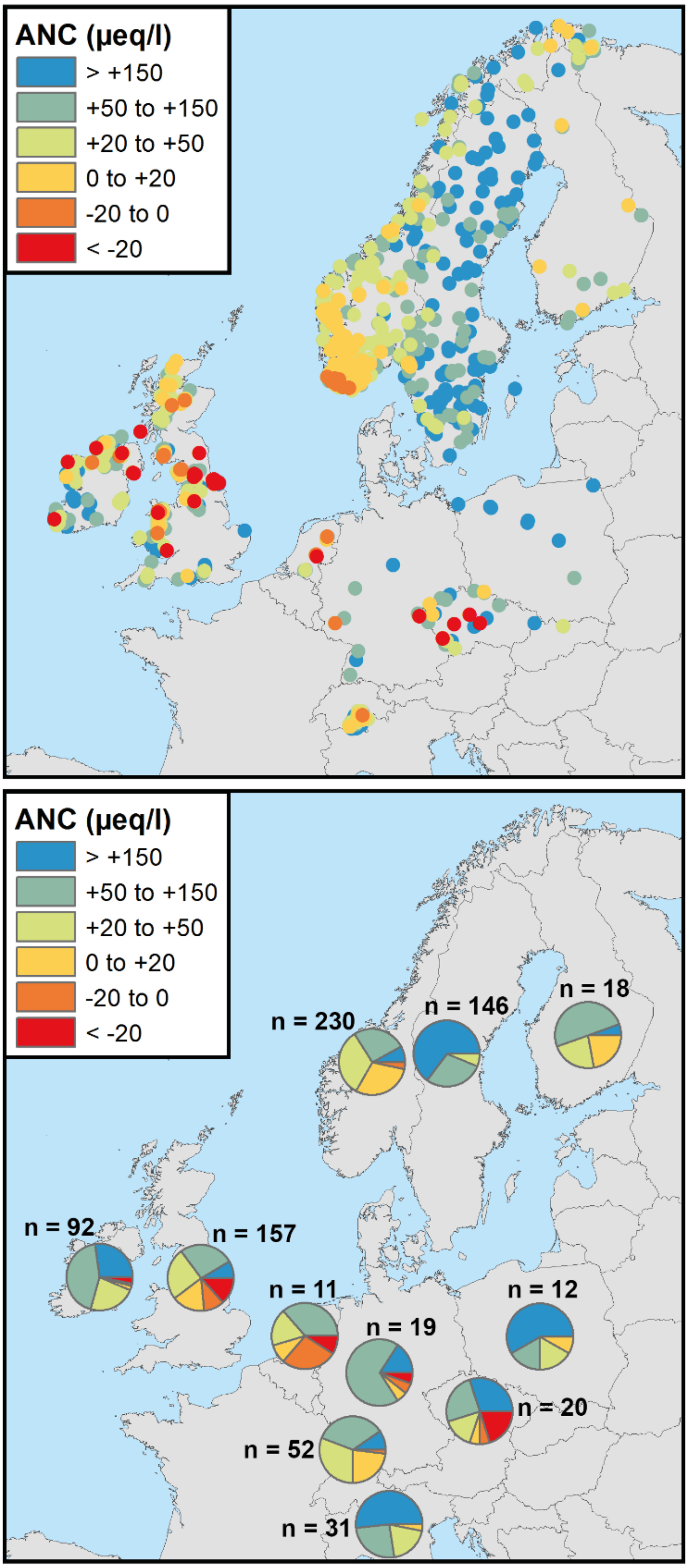


Figure 7 Average ANC (µeq/l) per site (top) and summary of ANC data (bottom) for European sites. The data represent the acid-sensitive parts of the countries, and the strategy for selection of sites varies between countries (cf. Table 2).

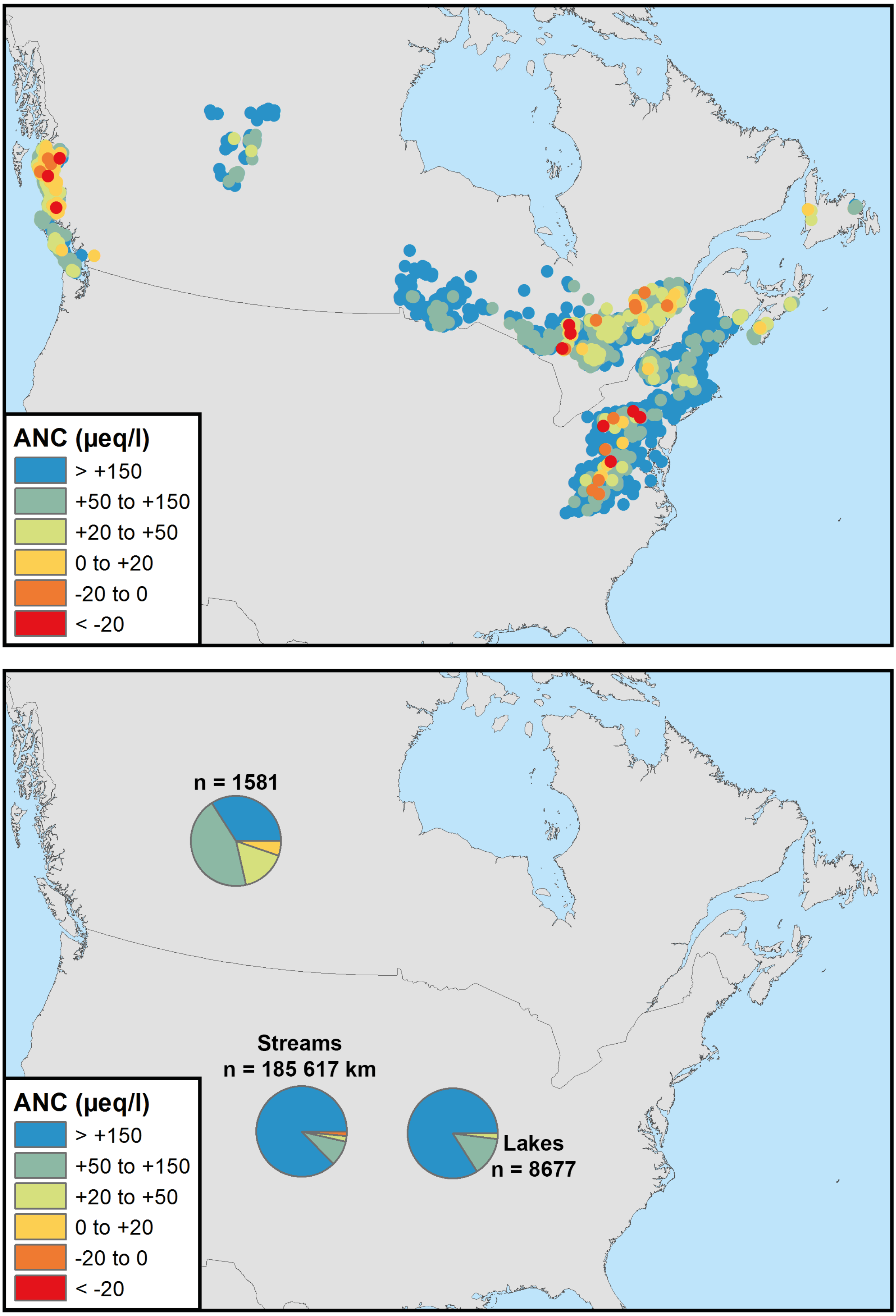


Figure 8 Average ANC (µeq/l) per site (top) and summary of ANC data (bottom) for North American sites. The data represent the acid-sensitive parts of the countries, and the strategy for selection of sites varies between countries (cf. Table 2). The distributions for the data from the United States (bottom) result from weighted analyses and represent the entire lake population/stream length within the regions, not only the monitored sites (top).

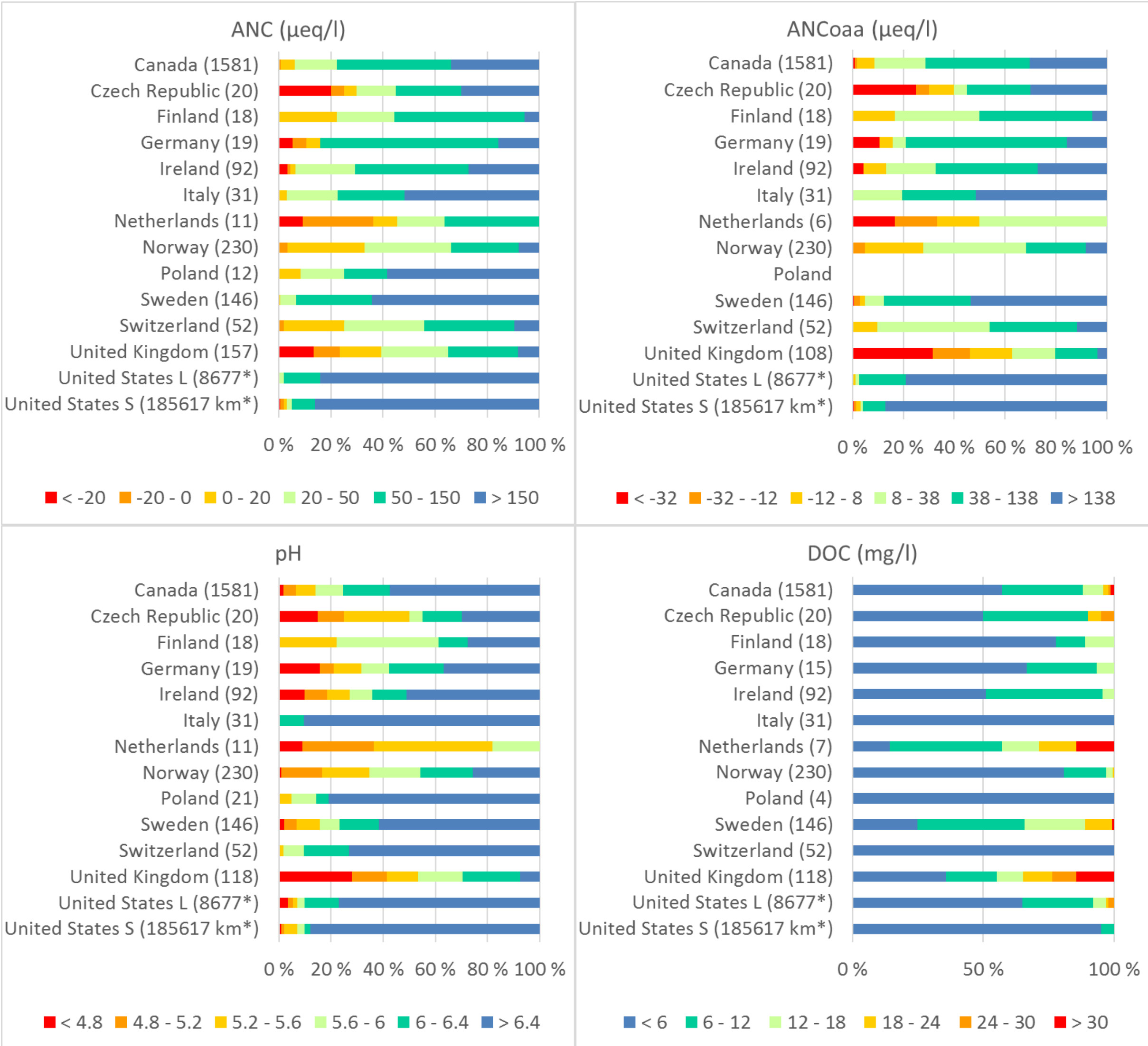


Figure 9 Distribution of sites among different ANC, ANC_{oaa}, pH and DOC classes per country. The numbers in parentheses are the total number of sites per country, which vary slightly between the parameters. The distributions for the data from the United States (L = lakes, S = streams) result from weighted analyses and represent the entire lake population/stream length within the regions, and the numbers in parentheses (*) are the estimated population sizes (cf. Table 3). ANC_{oaa} was not available for Poland.

Many countries have a large proportion of sites in the category ANC 20-50 µeq/l or ANC_{oaa} 8-38 µeq/l (Figure 9). This includes countries such as Italy, Ireland, Poland and Canada, where the proportion of sites with ANC < 20 µeq/l was low. Such water bodies are at risk of being acidified, and potentially vulnerable to episodic acidification events.

A majority of the countries have pH < 5.6 at more than 20% of the sites, but this partly reflects a high proportion of sites with medium to high DOC. Figure 10 shows how the pH can be quite low even if the ANC is acceptable, due to high DOC concentration. These are waters that are naturally acid. For sites with DOC < 6 mg/l, sites with pH below 5.6 generally have low ANC (< 50 µeq/l).

All in all, despite variable levels of acidification, all countries include acidified sites.

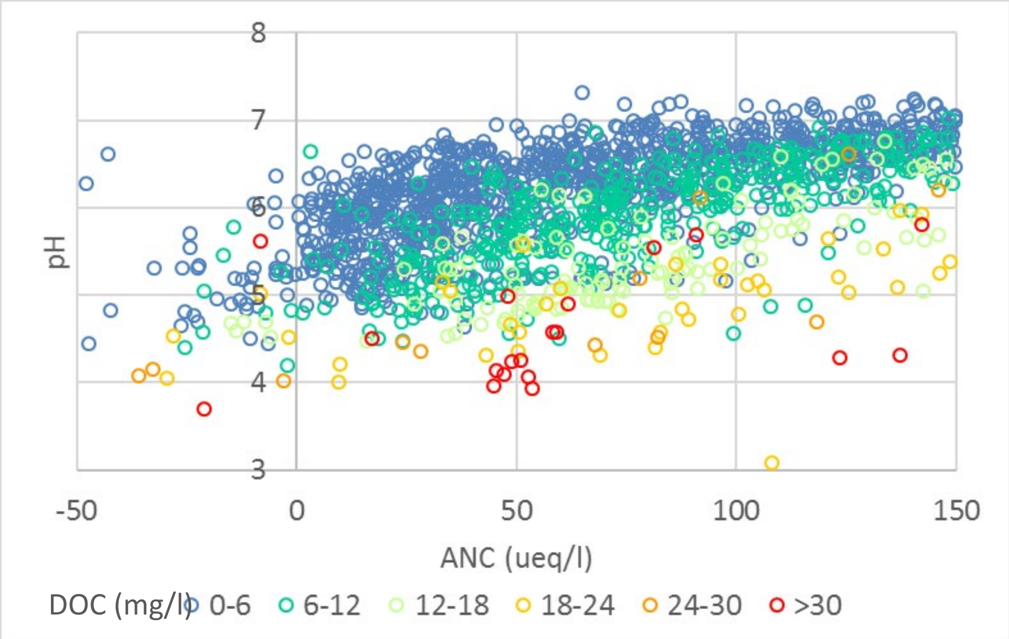


Figure 10 pH versus ANC for submitted sites grouped by DOC concentration (mg/l).

3.3 Acidification status as reported under the WFD

A number of countries do not classify acidification status under the WFD for any of their lake and/or river water bodies, i.e. acidification status is reported only as “Not applicable“, “Monitored but not used” or “Unknown” (Figure 11). Of the countries that do classify acidification status a high proportion (>25%) of unclassified water bodies is reported for Belgian, Bulgarian, Finnish, French and Portuguese rivers, Czech and UK lakes, and Estonian, Spanish and Swedish lakes and rivers, making the interpretation of the results more uncertain.

Lakes tended overall to show the highest percentage of water bodies in less than good acidification status (Table 4). Six countries had less than good status in more than 20% of their water bodies, and in Belgium, the Netherlands and Portugal more than 50% of the lake water bodies were in less than good status. For rivers, Poland was the only country with less than good status in more than 20% of its water bodies. The percentage with less than good acidification status is higher when calculated for the classified water bodies only, but in some cases the proportion of classified water bodies is very low. 8% of the water bodies with less than good acidification status still had good or high ecological status.

Atmospheric deposition is frequently, but far from always, reported as a significant pressure for water bodies with less than good acidification status. Germany, Finland, Sweden, the UK and Luxembourg report acid deposition as a significant pressure for all/most of these lake and/or river water bodies. A high proportion is also reported with atmospheric deposition for Czech rivers and lakes, Belgian rivers and Dutch lakes. Of the countries not covered by the country reports, only Luxembourg, Belgium and Hungary report atmospheric deposition as a significant pressure.

Only a small proportion of the water bodies with less than good acidification status is reported as being subject to other pressures that can potentially cause acidification. Overall, many countries have a high proportion of water bodies with less than good acidification status where this status cannot be explained by any of the relevant pressures. The main exceptions to this are Sweden, rivers in Germany and Finland, and some other countries where the total number of water bodies with less than good acidification status is low. There is also a number of water bodies with less than good acidification status in Poland, and also Finland, with no significant pressure.

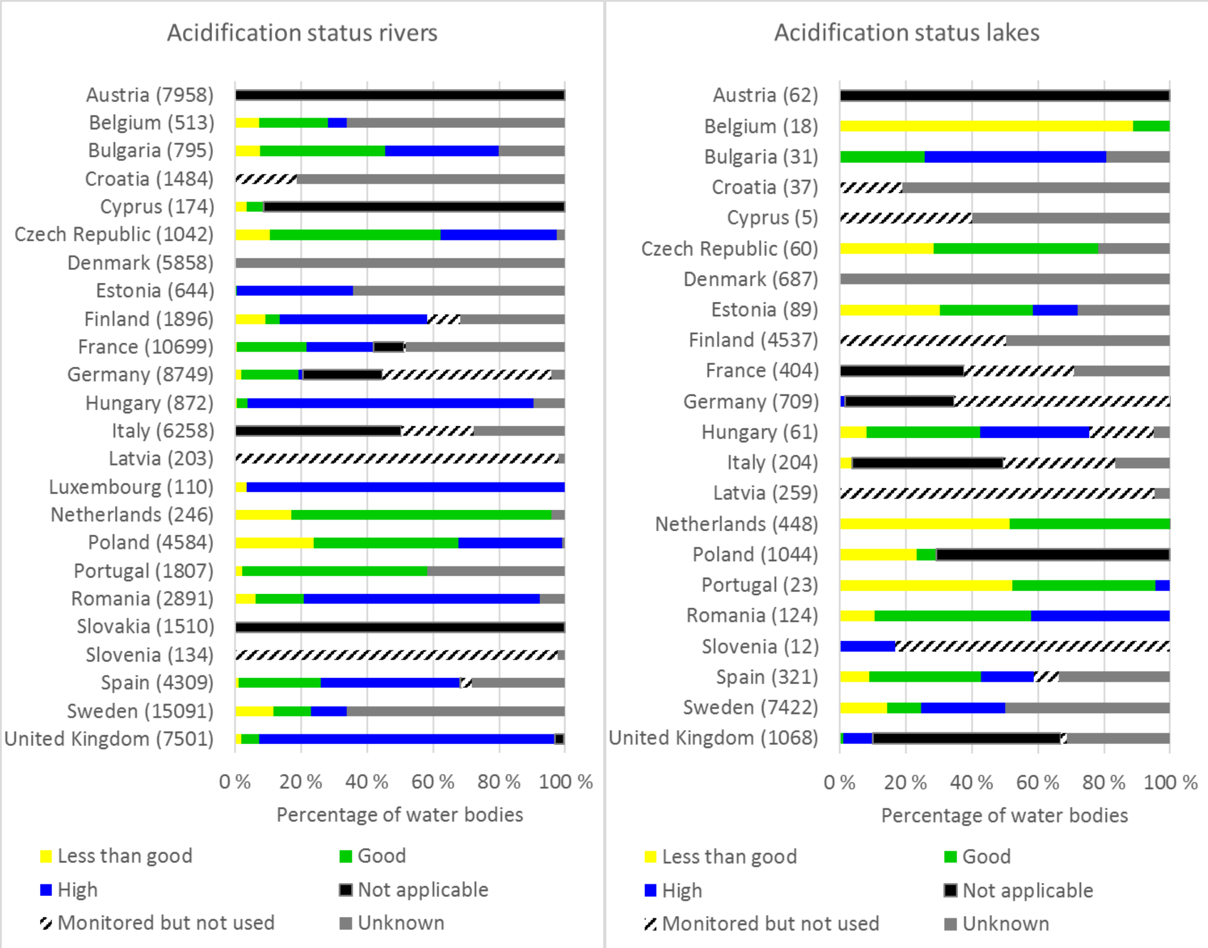


Figure 11 Acidification status of river (left) and lake (right) water bodies which are classified with respect to ecological status, as reported under the WFD. Several countries did not classify any water bodies with respect to acidification status (Austria, Croatia, Denmark, Italy, Latvia, Slovakia and Slovenia for rivers; Austria, Croatia, Cyprus, Denmark, Finland, France and Latvia for lakes). Luxembourg and Slovakia do not have lake water bodies as defined in the WFD. Greece, Ireland, Lithuania and Norway had not yet reported at the time of data extraction. The total number of water bodies is given on the vertical axis.

Belgium, Hungary, Sweden and the UK report the impact acidification for most/all their lake and/or river water bodies that are in less than good acidification status, but eight countries report this impact for no or very few of these water bodies. And again, there are several Polish and Finnish water bodies with less than good acidification status, but no significant impacts.

Overall, for the countries having water bodies with less than good acidification status, only eight report both the pressure atmospheric deposition and the impact acidification for at least some of these water bodies, and this pressure and impact is reported for most of the water bodies in less than good acidification status in Sweden (and UK lakes) only. Of the countries not covered by the country reports, this combination of acidification status, pressure and impact occurs in Belgium only (for 30% of the water bodies in less than good acidification status).

Table 4 Number and percentage of water bodies with less than good acidification status, in total and with different combinations of impacts and pressures. Percentages are calculated relative to all water bodies (numbers in parentheses in Figure 11) or only relative to those classified with respect to acidification status. Other relevant pressures are all pressure types other than atmospheric deposition that could possibly cause acidification (pressures associated with industry, contaminated sites, mining or historical pollution).

Country	< good	% < good (of all)	% < good (of classified)	With < good ecological status	With impact acidification	With pressure atmospheric deposition	With impact acidification and pressure atmospheric deposition	% with impact acidification and pressure atmospheric deposition (of all)	% with acidification and atmospheric deposition (of classified)	With other relevant pressures and not atmospheric deposition	With no significant pressures	With no significant impact
Rivers												
Belgium	37	7	21	37	33	16	13	3	8	7		
Bulgaria	60	8	9	59	2			0	0	36	1	1
Cyprus	6	3	40	2				0	0		2	
Czech Republic	109	10	11	109	43	29	12	1	1	5		
Estonia	1	0	0	1				0	0			
Finland	176	9	16	95	71	136	53	3	5	1	12	12
France	52	0	1	52	14			0	0	4		
Germany	168	2	9	167	28	164	24	0	1	1		
Hungary	4	0	1	4	4	1	1	0	0			
Luxembourg	4	4	4	4		4		0	0			
Netherlands	42	17	18	42		9		0	0	1		
Poland	1087	24	24	1073	19	7	6	0	0	1	225	225
Portugal	37	2	4	37				0	0	7		
Romania	179	6	7	179	4			0	0	5		
Spain	48	1	2	48	30			0	0	17		
Sweden	1744	12	34	1603	1652	1744	1652	11	32			
United Kingdom	141	2	2	141	31	51	14	0	0	3		
Lakes												
Belgium	16	89	89	16	16	3	3	17	17			
Czech Republic	17	28	36	17	7	5	1	2	2			
Estonia	27	30	42	17				0	0	1		
Hungary	5	8	11	4	5			0	0			
Italy	7	3	88	7				0	0	1		
Netherlands	231	52	52	231		91		0	0	7		
Poland	243	23	80	161				0	0	8	82	82
Portugal	12	52	52	12				0	0			
Romania	13	10	10	13				0	0			
Spain	29	9	15	29				0	0			
Sweden	1059	14	28	923	974	1059	974	13	26			
United Kingdom	3	0	3	3	3	3	3	0	3			

4 Country reports

4.1 Canada

Julian Aherne[1], Suzanne Couture[2], Andrew Paterson[3], Jocelyne Heneberry[3], Dean Jeffries[2]
[1] Trent University
[2] Environment and Climate Change Canada
[3] Ontario Ministry of the Environment and Climate Change

4.1.1 Acid sensitivity

Large areas of Canada are underlain by noncarbonate-bearing and weathering-resistant bedrock, overlain by shallow coarse-textured soils (Figure 12). Although acid sensitive regions span the width and breadth of the country, historically, scientific and political activity on 'acid rain' was focused primarily on eastern Canada, owing to the spatial coincidence of acid sensitive bedrock and elevated levels of acidic deposition associated with emissions from coal-fired power plants in the industrialised Ohio River Valley (Aherne and Jeffries, 2015). More recently there has been a focus on the impacts of sulphur and nitrogen deposition in western Canada (Aherne and Shaw 2010), owing to increasing emissions from the oil and gas sector.

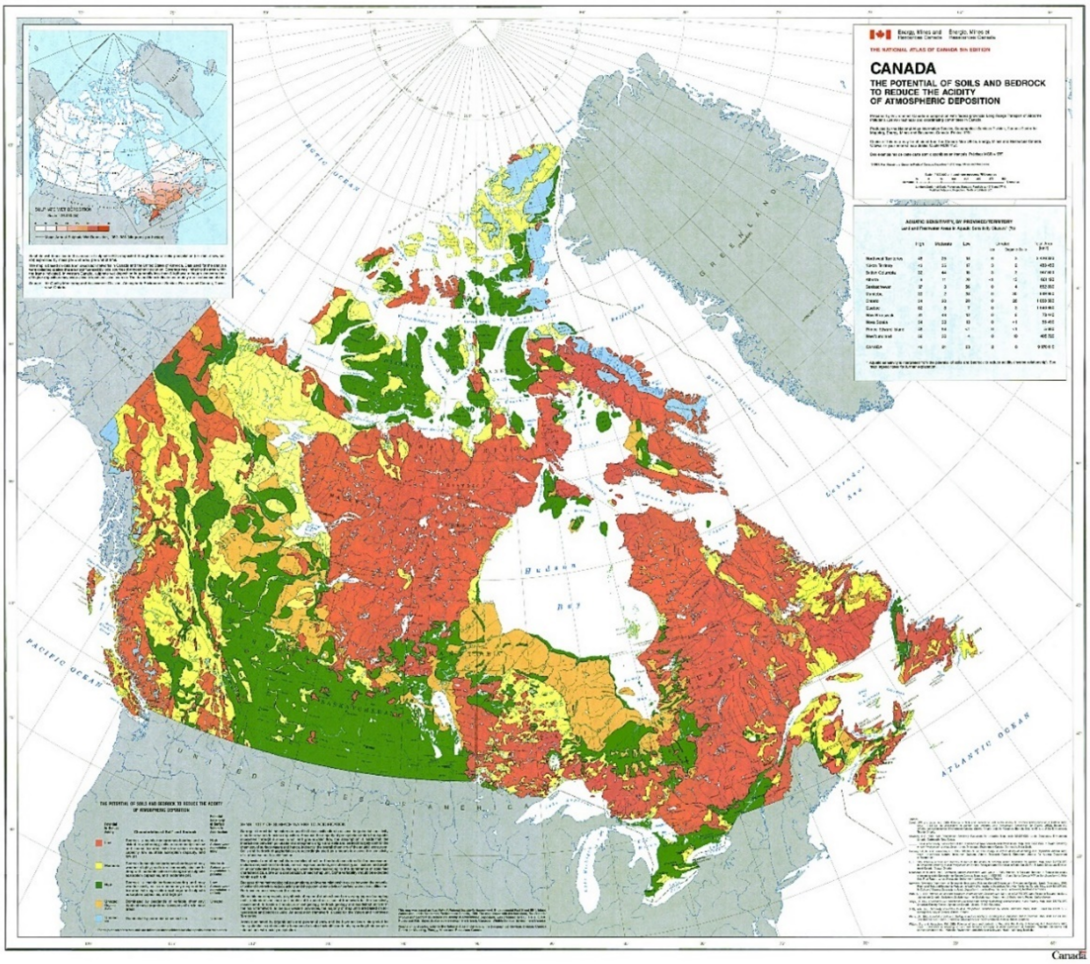


Figure 12 The potential of soils and bedrock to reduce the acidity of atmospheric deposition. For a more complete description see Natural Resources Canada (2001). The most sensitive regions (reddish-brown colour) are associated with noncarbonate-bearing and weathering-resistant bedrock, and shallow coarse-textured soils with low cation exchange capacity.

4.1.2 Monitoring and assessment approach

Hydrochemical monitoring of acid sensitive lakes is carried out federally by Environment and Climate Change Canada, and typically also by Ministries of the Environment on a provincial level, e.g., the Ontario Ministry of the Environment and Climate Change have maintained a provincial network of long-term monitoring lakes since the 1970s. The monitoring of acid sensitive surface waters during the 1980s and 1990s, focused primarily on eastern Canada (Aherne and Jeffries, 2015). These regional surveys and the long-term monitoring at a network of sites have underpinned several national 'acidic deposition science assessments', the most recent assessment was 2004 (Environment Canada, 2005). A sub-set of the long-term lakes (n=18) have been included in the ICP Waters programme since 1980.

The impacts of acidic deposition on surface waters in eastern Canada were a significant driver underpinning bilateral agreements on emission reductions (US-Canada 1983). However, more recently, monitoring of acid sensitive waters has been reduced in eastern Canada, owing to changing governmental priorities, lost institutional capacity, and the perception that 'acid rain' has been solved. In contrast, there has been a growth in monitoring in western Canada (Aherne and Shaw 2010), specifically focusing on large-scale surveys in British Columbia, and the development of long-term networks in Alberta and Saskatchewan, owing to concerns associated with emissions from the oil and gas sector. The current assessment collated hydrochemical data from regional surveys, and well established long-term monitoring networks (in eastern Canada). Previous assessments have included a wider regional coverage of lakes (Aherne et al., 2011); however, the current assessment was limited to 2010–2016.

4.1.3 Acidification status

During the period 2010–2016, average lake pH ranged from 3.93 to 7.50, with an overall average pH of 5.75 across the study lakes (n = 1581). Lakes with lower pH were primarily located on the north-western coast, and in eastern Canada (Figure 13). The average DOC was 6.8 mg/l and ranged from 0.1 to 46.8 mg/l. Approximately, 9–12% of the study lakes are considered to be highly acid sensitive and 'acid impacted' based on ANC or organic acid adjusted ANC.

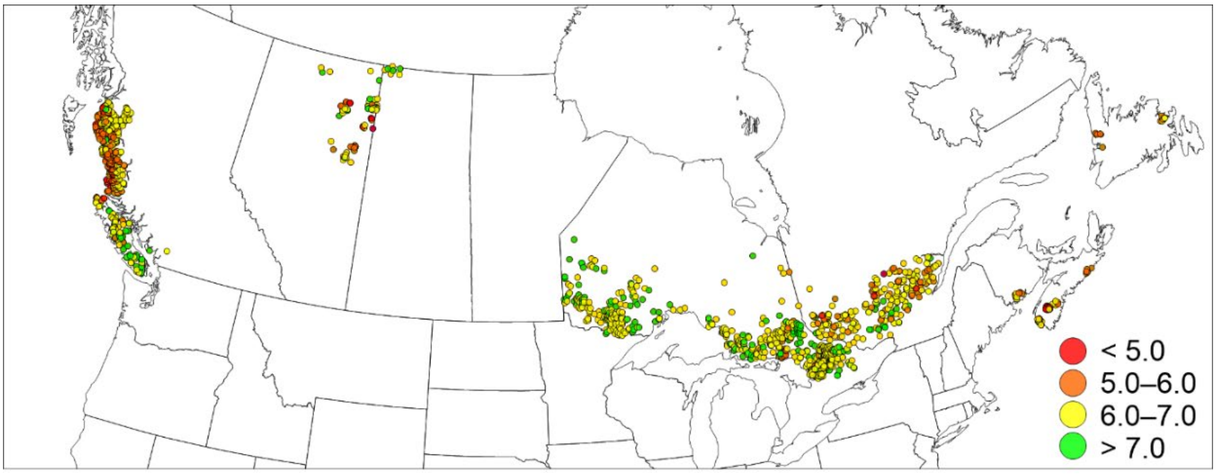


Figure 13 Average pH in lakes (n = 1581) located in 'acid sensitive' regions, during the period 2010–2016. Filled circles indicate the location of the assessment lake, with pH displayed as four coloured classes; red and orange indicate the lower pH categories.

During the past three decades there has been an improvement in the acid status of many lakes, owing to the reduction in acidic (sulphur) deposition (Environment Canada, 2005). In Nova Scotia, analysis of long-term monitoring lakes (n=16), indicated that average Gran alkalinity in lakes (n=16) increased by 1.5 mg/l $CaCO_3$ during the period 2000–2016 (Figure 14).

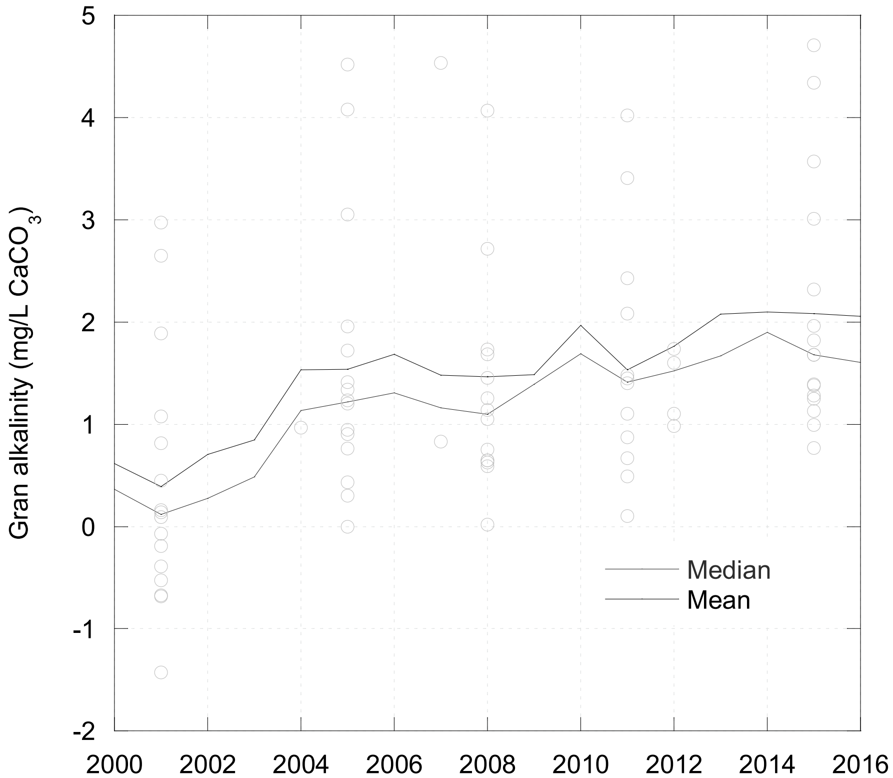


Figure 14 Time-series of Gran alkalinity (mg/l CaCo3) during the period 2000–2016 for long-term monitoring lakes in Nova Scotia, Canada (n = 16). The annual median (lower line) and mean (upper line) concentration are also shown.

Recent efforts at regionalisation have used regression kriging to upscale observations from survey lakes (n < 900) to regional coverage (n > 90,000) in northern Saskatchewan (Cathcart et al., 2016). The approach used a suite of catchment predictor variables to model hydrochemistry across > 90,000 lakes in northern Saskatchewan (Figure 15). The predicated hydrochemistry was used to determine ciritical loads for the > 90,000 lakes (catchments); exceedance was estimated at 12% of the study lakes under modelled 2010 sulphur and nitrogen deposition (Cathcart et al., 2016). The approach presents a promising method for national assessments.

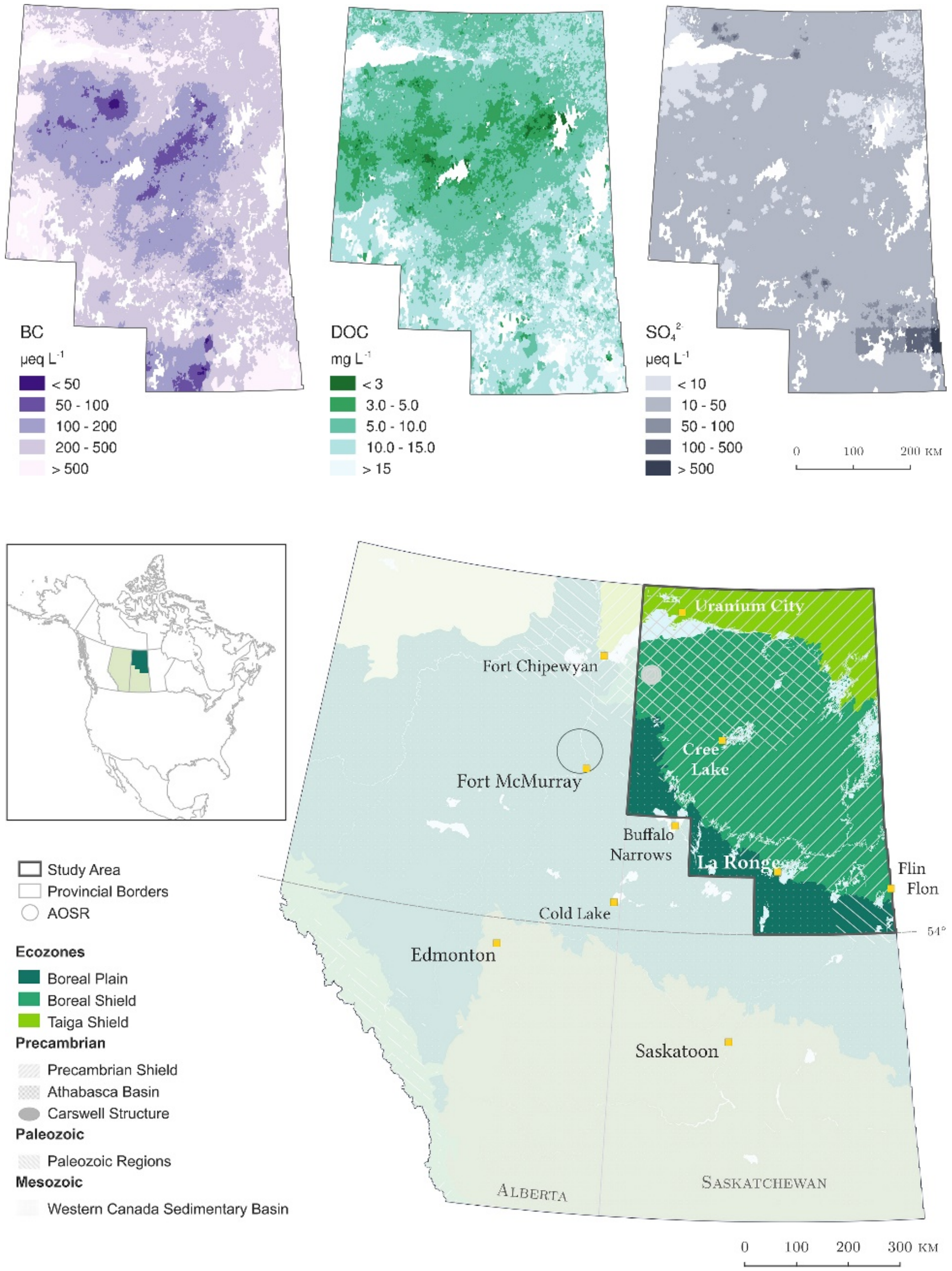


Figure 15 Upper panel: Predicted base cation (BC), sulphate (SO_4^{2-}) and dissolved organic carbon (DOC) in >90,000 lakes in northern Saskatchewan (Cathcart et al., 2016), produced via regression kriging from lake observations of DOC (845 lakes), BC (818 lakes) and SO_4^{2-} (846 lakes). Lower panel: Saskatchewan and Alberta with the study area in northern Saskatchewan outlined. Major geologic features (Precambrian Shield, Athabasca Basin, Western Canada Sedimentary Basin) are illustrated in crosshatching and ecozones (Boreal Plain, Boreal Shield, Taiga Shield) are shaded. An encircled 50 km radius north of Fort McMurray represents the Athabasca Oil Sands Region (AOSR).

Acknowledgement: This study was supported by Environment and Climate Change Canada through agreement GCXE15Z268.

4.1.4 References

Aherne, J. and Shaw, D.P. 2010. Impacts of sulphur and nitrogen deposition in western Canada. J. Limnol., 69:1, 1-3.

Aherne, J., Wolniewicz, M., Clair, T.A., Dennis, I. and Jeffries, D. 2011. Canada—National Focal Centre Report. In (editors) Posch, M., Slootweg, J. and Hettelingh J-P., Modelling Critical Thresholds and Temporal Changes of Geochemistry and Vegetation Diversity. Report no. 80359003. Coordination Centre for Effects (CCE), The Netherlands.

Aherne, J. and Jeffries, D. 2015. Critical load assessments and dynamic model applications for lakes in North America. In, W. de Vries, J.-P. Hettelingh and M. Posch (eds), Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems, Chapter 19. Springer.

Environment Canada, 2005. Canadian acid deposition science assessment 2004. Environment Canada. Meteorological Service of Canada, 479p

Natural Resources Canada, 1991. The Potential of Soils and Bedrock to Reduce the Acidity of Atmospheric Deposition. Atlas of Canada, 5th Edition. URL open.canada.ca/data/en/dataset/7412aa15-5905-5964-9e2a-79dd45316e5e

US-Canada, 1983. Memorandum of intent on transboundary air pollution. Report of the Impact Assessment Working Group I, Section 3 Aquatic effects.

4.2 Czech Republic

Jakub Hruška[1], Vladimír Majer[1], Jaroslav Vrba[2], Filip Oulehle[1], Tomáš Chuman[1], Pavel Krám[1]
[1] Czech Geological Survey
[2] Faculty of Science, University of South Bohemia

4.2.1 Introduction

Czech Republic is well known as a country that historically suffered from very high atmospheric deposition of sulphur and nitrogen. Deposition peaked in the 1980´s and has declined significantly to the present day. Between 1979 and 2012 the average sulphur deposition in throughfall declined by 82%, nitrate by 38 % and NH_4 by 46% (Oulehle et al. 2016). Despite the very high load of acidifying compounds, surface waters were affected only in specific regions, mostly located at high altitudes and on acid-sensitive slowly-weathering bedrock (e.g. Hruška and Krám 2003). The majority of the country was not affected significantly as the thick soils developed on highly weatherable bedrock dominate the territory of the country and have buffered atmospheric acidity. Thus only ca. 2% of surface waters was classified as acidified in the 1980´s (Veselý et al. 1998), mostly in the mountainous northern part of the country in close vicinity to lignite-burning power plants on the border between Poland, Germany and the Czech Republic, formerly known as the “Black Triangle” region.

4.2.2 Regions susceptible to acidification

An indirect estimate of the extent of acidification was done by GIS analysis combining several layers of parameters important for acidification. We used a linear combination of deposition data (sulphur and nitrogen deposition), a geological map where bedrock types were classified according their geochemical reactivity (Chuman et al. 2013), a forest ecosystem classification, precipitation amount and mean annual temperature. All layers were classified on the scale 1-4 where a lower number was associated with resistance and a higher with susceptibility to acidification. The total score was divided into 4 classes (Figure 16) with respect to acidification sensitivity of soil and surface water. As forest type and structure play an important role, only forested areas were taken into account.

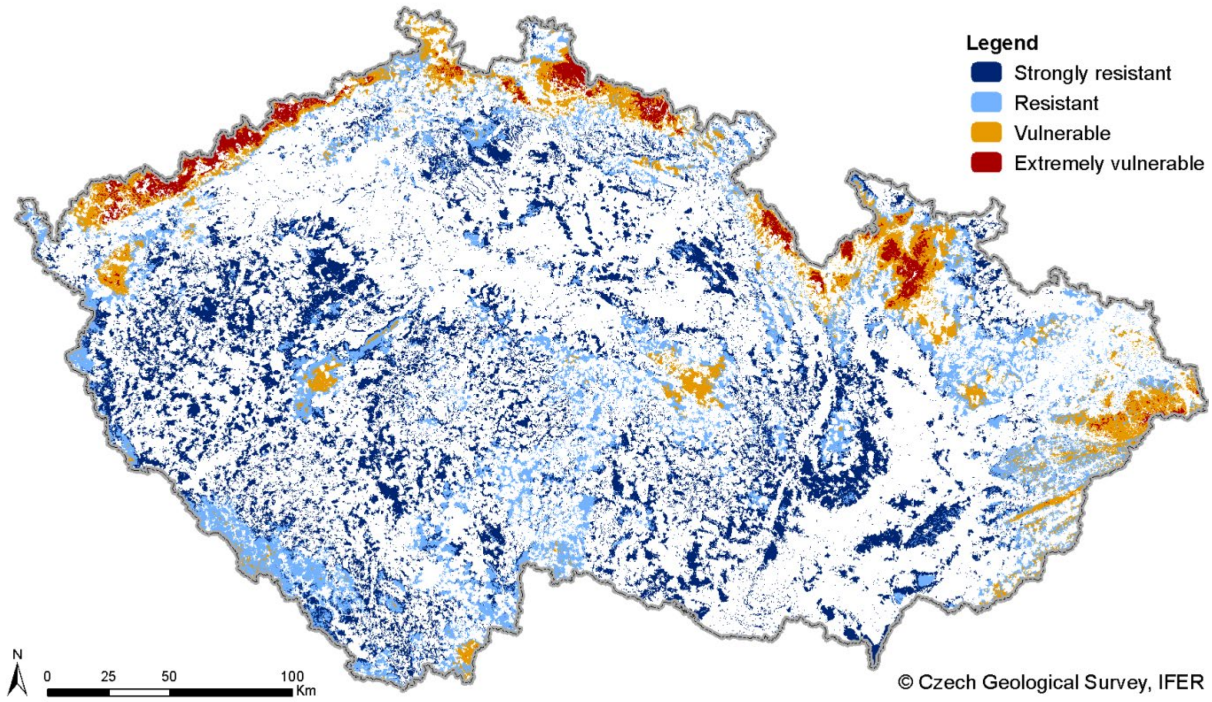


Figure 16 Areas susceptible to acidification (only forested landscape) within the Czech Republic.

4.2.3 Regions with acidified streams

The areas sensitive to acidification were last assessed in 2007–2010. In total 5,764 individual streams were sampled during summer–autumn base flow conditions (Majer et al. 2012). Only a small proportion of the streams were acidic at base flow runoff (2% had ANC<20 µeq/l). Low ANC (<100 µeq/l; Figure 17) and low pH (<6.0; Figure 18) were observed mostly at higher altitudes in the Krkonoše Mts. and Jizera Mts. in the northern part of the country. Both are underlined by gneiss and granite and receive a high amount of precipitation (over 1500 mm on the peaks) as well as acidic deposition from the nearby coal-fired power plants in the Czech Republic and Poland. Another acidified region is located on the ridge of the Krušné Mts. (Ore Mts.) and the Slavkov Forest in the western part of the country. This region is underlined by slowly weathering granite making the region naturally very sensitive to acidification. It is also a higher altitude area receiving around 1000 -1200 mm annual precipitation. Acidic deposition was historically just moderate. Parts of the Ore Mts. (mostly eastern part), Krušné hory Mts. and Jizera Mts. were extensively limed between the 1970´s and present.

The third acidified region is located in the Bohemian Forest (Šumava in Czech) in the south-west. This region was locally glaciated and there are five glacial lakes, all of which have been heavily acidified for several decades. The combination of high altitude (1008–1087 m.a.s.l.), sensitive bedrock (gneiss, quartzite, granite), very thin, poorly developed soil and moderate acidic deposition resulted in the long lasting severe acidification (a more detailed case study follows). Acidification is limited only to the lakes themselves and their close vicinity and/or to the highest elevations of the Bohemian Forest (altitudes higher than ca. 1000 m.a.s.l.). Streams below this elevation were not acidified.

A small, but significantly acidified region is located in the Brdy Mts., south-west from Prague. The Brdy received moderate acidic deposition, but the area is underlain by very nutrient poor and slow weathering quartzite bedrock and fully covered by spruce monoculture accelerating dry sulphur deposition.

There are small acidic areas (low pH, but mostly not low ANC) outside the previously mentioned regions. These are isolated high DOC peat bogs (usually few km^2), and streams draining the bogs usually have very high DOC concentrations (typically 20-50 mg/l DOC). Thus, pH is low, but ANC is highly positive. The largest such region is the Třeboň Basin in south Bohemia. A significant proportion of DOC derived acidity was also identified on the top of Czech-Moravian Highland (750–836 m.a.s.l.) in the middle of the Czech Republic, but this region is only slightly affected by acidic deposition.

The extent of acidified surface water bodies is at present <1% of the country territory. The area is divided into many small regions situated mostly on the ridges of mountains and highlands except in the east of the Czech Republic. While limited in area, they often present regions with very high conservation value due to the location of environmentally protected areas (e.g. National parks at Krkonoše and Bohemian Forest).

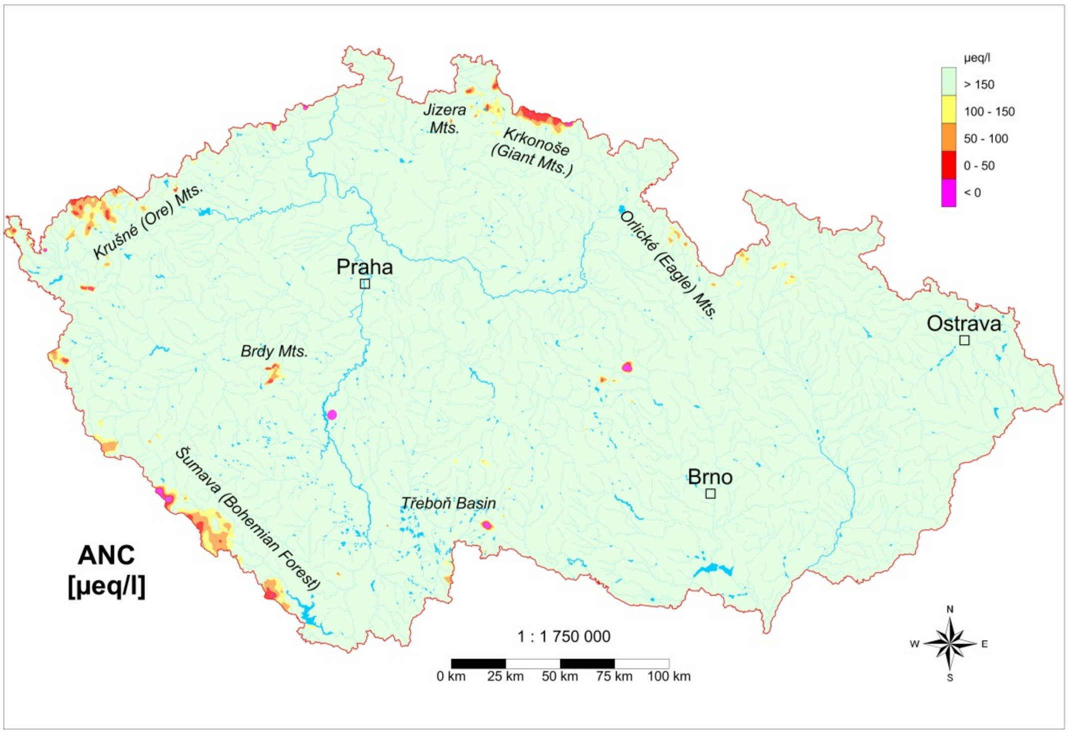


Figure 17 Stream water ANC during base flow conditions (ANC <0 µeq/l covers 60 km^2, ANC = 0–50 µeq/l at 179 km^2, ANC = 50–100 µeq/l at 515 km^2, and ANC = 100–150 µeq/l at 734 km^2).

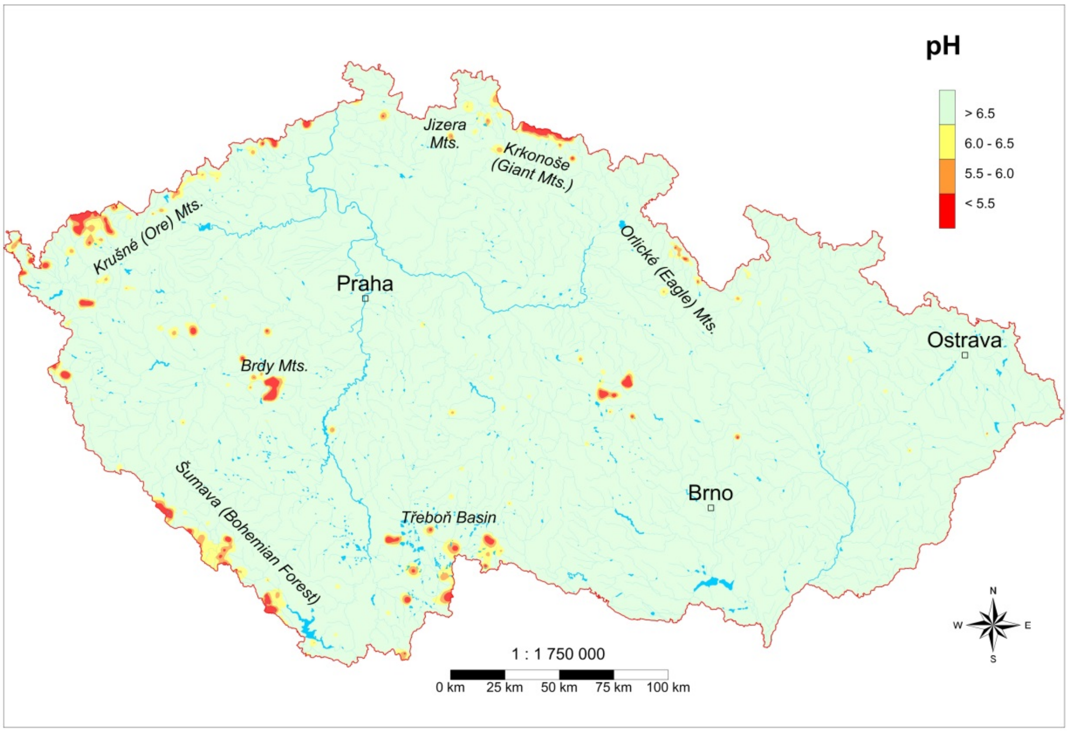


Figure 18 Stream water pH during base flow conditions, pH <5.5 covers 380 km^2, pH = 5.5–6.0 covers 529 km^2, and pH = 6.0–6.5 covers 1400 km^2.

4.2.4 Case study of Černé Lake

Lakes in the Bohemian Forest (SW Czech Republic) were acidified by acid rain continuously since the first half of the 20th century. The largest lake Černé was the most resistant and hosted fish (brown trout and later brook trout) and several species of benthos and zooplankton until the 1970´s, when fish population as well as zooplankton was completely lost and benthos diversity was significantly reduced (Figure 19). pH was reduced to 4.5 (pH >6.0 had been measured until the 1960´s). After three decades, sulphur deposition was reduced by 90% and inorganic nitrogen (TIN) by 50%. This resulted in pH increases to present values around 5.0, and decline of Al concentrations. Benthic species recovered significantly, and one of the originally six zooplankton species returned as well. Despite significant reductions of atmospheric deposition, fish have not yet returned. According to model predictions it will take at least three more decades for more complete biological recovery.

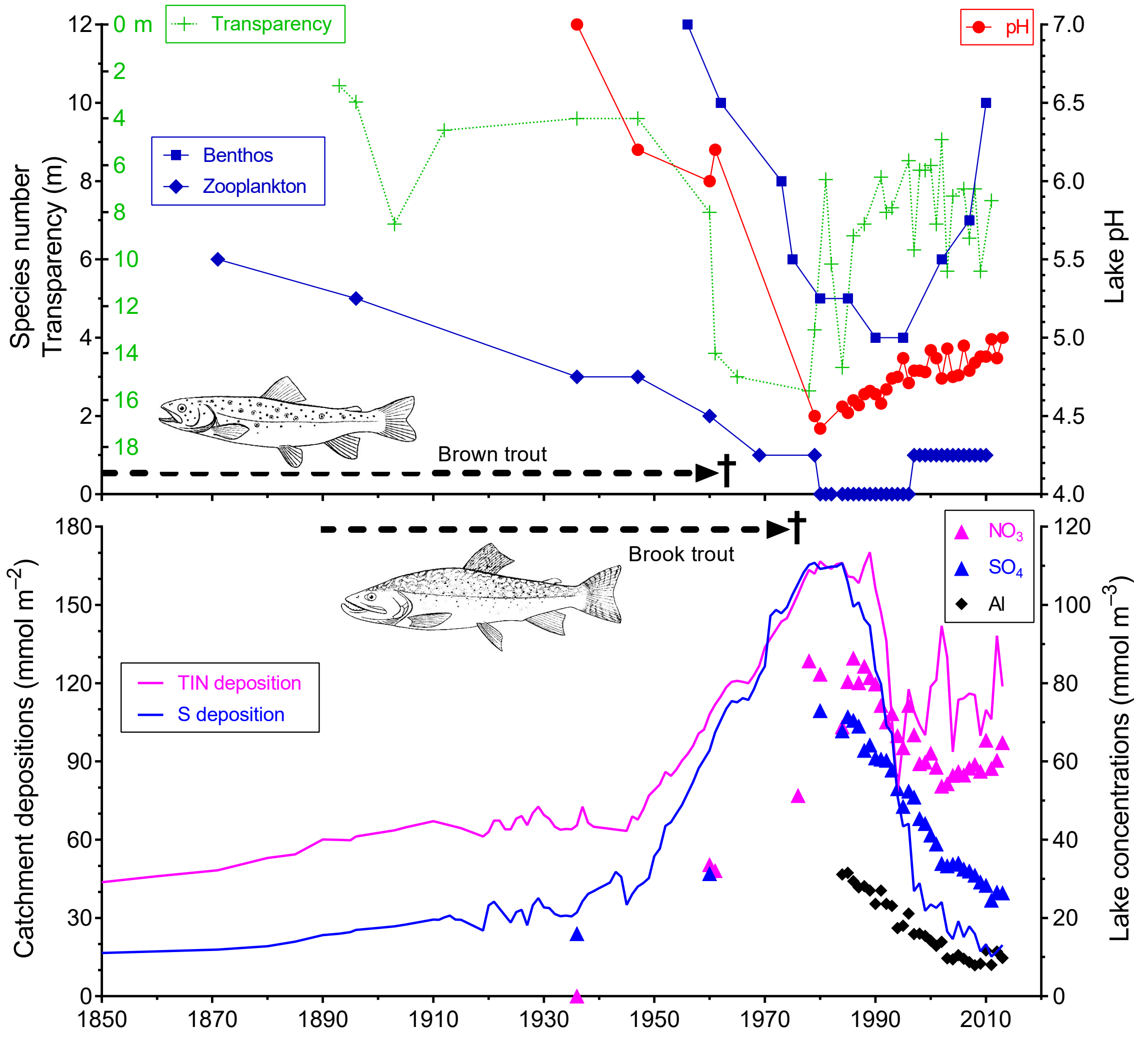


Figure 19 Long-term changes of biota, lake chemistry and atmospheric deposition. Černé Lake, the Bohemian Forest, Czech Republic (Vrba et al., 2016).

4.2.5 Acidification monitoring

There are two long-term acidification monitoring programmes (Figure 20). Both are part of the Czech LTER network. Data from these programmes were submitted for the overall analysis in this report. The first programme is a monitoring network of small forested catchments – GEOMON (Oulehle et al. 2017), which consists of 14 catchments monitored regularly on monthly basis since 1994, but many catchments were monitored longer, typically from the mid 1980´s, or more frequently. The longest record of surface water chemistry is available since 1978 for the Jezeří catchment (JEZ, Figure 20). The monitoring is operated on a monthly basis including bulk and throughfall deposition, runoff chemistry, and hydrological measurements. From time to time also soil chemistry and structure, forest and ground vegetation inventories, and chemistry are measured. Benthic species are monitored at selected catchments (mostly ICP waters catchments, UHL and LYS) bi-annually. This monitoring is operated by the Czech Geological Survey and Global Change Research Institute of the Czech Academy of Sciences (CAS). There is no systematic governmental support for this monitoring. It is maintained by *ad hoc* projects and institutional support.

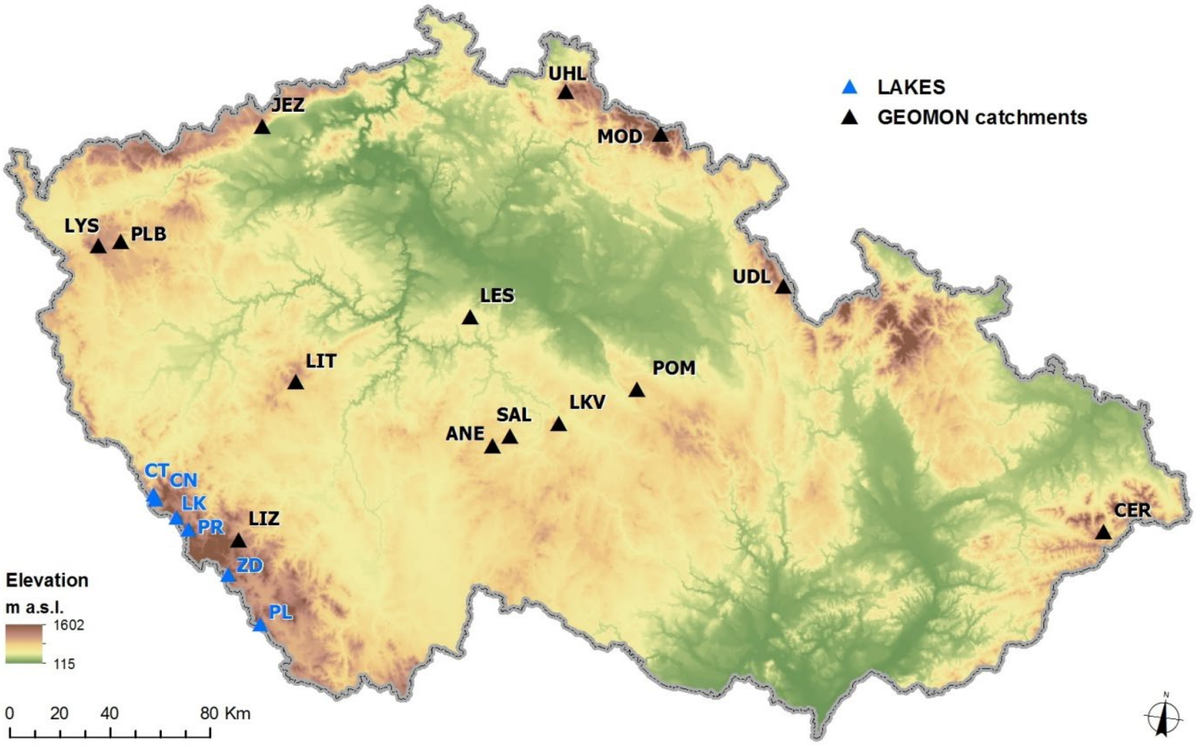


Figure 20 Long-term sites focused on acidification monitoring. The GEOMON forested catchment network was established in 1994, regular monitoring of Bohemian Forest lakes started in 1984.

The second programme of long-term monitoring is focused on the Bohemian Forest glacial lakes. The Czech Geological Survey supports water chemistry analyses of all lakes twice per year (summer and autumn) since 1984 and provides these data to ICP Waters (Oulehle et al. 2012). At some lakes (Plešné – PL, Černé – CT) intensive limnological and biogeochemical studies have been running for the last two decades. These studies were conducted by the Institute of Hydrobiology, Biology Centre CAS (e.g. Kopáček et al. 2017, Vrba et al. 2016). As for GEOMON, *ad hoc* projects are the only source for monitoring support.

Acknowledgements: This work was supported by the Czech Ministry of Environment and Ministry of Education, Youth and Sports of the Czech Republic - MEYS (projects LM2015075, EF16_013/0001782).

4.2.6 References

Hruška, J., Krám, P. (2003) Modeling long-term changes in streamwater and soil chemistry in catchments with contrasting vulnerability to acidification (Lysina and Pluhův Bor, Czech Republic). Hydrology and Earth System Sciences 7, 525-539.

Chuman, T., Gürtlerová, P., Hruška, J., Adamová, M. (2013) Geochemical reactivity of rocks of the Czech Republic, Journal of Maps, DOI:10.1080/17445647.2013.867418

Kopáček, J., Fluksová, H., Hejzlar, J. Kaňa, J., Porcal, P., Turek, J. (2017) Changes in surface water chemistry caused by natural forest dieback in an unmanaged mountain catchment. Science of the Total Environment 584–585, 971–981.

Majer, V., Hruška, J., Zoulková, V., Holečková P., Myška, O. (2012) Atlas chemismu povrchových vod České republiky Stav v letech 1984-1996 a 2007-2009 (Atlas of surface water chemistry in the Czech Republic. Situation 1984-1996 and 2007-2009). Czech Geological Survey, Praha, 104 p. ISBN 978-80-7075-780-2.

Oulehle, F., Chuman, T., Majer, V., Hruška, J. (2013) Chemical recovery of acidified Bohemian lakes between 1984 and 2012: The role of acid deposition and bark beetle induced forest disturbance. Biogeochemistry DOI 10.1007/s10533-013-9865-x.

Oulehle, F., Kopáček, J., Chuman T, Černohous V., Hůnová, I., Hruška, J. et al. (2016) Predicting sulphur and nitrogen deposition using a simple statistical method. Atmospheric Environment 140, 456-468.

Oulehle, F., Chuman, T., Hruška, J., Krám, P. et al. (2017) Recovery from acidification alters concentrations and fluxes of solutes from Czech catchments. Biogeochemistry DOI 10.1007/s10533-017-0298-9.

Veselý, J., Majer., V. (1998) Hydrogeochemical mapping of Czech freshwaters. Bulletin of the Czech Geological Survey 73, 3, 183-190.

Vrba, J., Bojková, J., Chvojka, P., Fott, J., Kopáček, J. et al. (2016) Constraints on the biological recovery of the Bohemian Forest lakes from acid stress. Freshwater Biology 61, 376–395.

4.3 Finland

Jussi Vuorenmaa, Martin Forsius, Jaakko Mannio
Finnish Environment Institute

4.3.1 Acid sensitivity in Finland

The bedrock in Finland is granitic, covered with a relatively shallow till layer and occasionally with glaciofluvial sand and gravel deposits. Thin soils and acidic geochemical characteristics of the bedrock together with cold and humid climate have resulted in low cation exchange capacity and low ionic strength of the lake water. Inherently acid-sensitive catchments with low buffering capacity are characteristic throughout Finland, but most commonly in south, western-central and eastern Finland and in southern and north-eastern parts of North Finland (Figure 21, Kämäri et al. 1991).

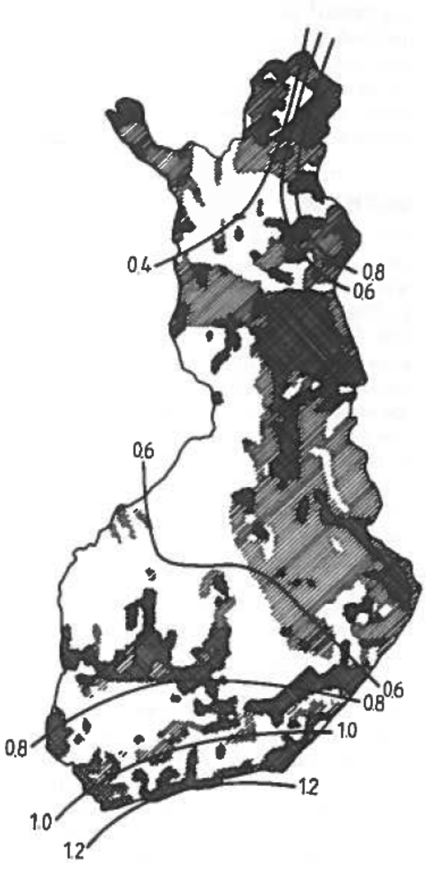


Figure 21 Map showing the regional relative sensitivity of surface water to acidic deposition in Finland, based on the bedrock type and soil properties, runoff and terrestrial relief. A darker shading indicates an increased sensitivity to acidic deposition, and the isolines depict the estimated total sulphur deposition (g S m^{-2} yr^{-1}) in Finland in 1987 (Kämäri 1990; Kämäri et al. 1991). The present-day sulphur deposition is in north Finland < 0.1 and south Finland < 0.3 g S m^{-2} yr^{-1}.

4.3.2 Monitoring of lake acidification

The regular monitoring of acidification of lakes in Finland started in 1987/1990 with ca. 160 lakes, but is presently being carried out in only 18 lakes. At most of the lakes, the monitoring started in 1987 in connection with the national Finnish lake acidification survey, conducted as a part of the Finnish Acidification Research Programme (HAPRO) by subjectively choosing lakes from the survey lake population. The objective of the selection was to cover the whole country, with emphasis on the remote headwater lakes of the most acidified and acid-sensitive areas. Since 1990, the lakes have been monitored regularly using a seasonal sampling strategy (samples in winter, spring, summer and autumn) with 4–7 samples per year. Some lakes were monitored once a year during the autumn circulation in 1990–1999, and a seasonal sampling strategy has been applied in these lakes since 2000. Out of these 18 lakes, 10 lakes belong to the UNECE ICP Waters network and three lakes belong to the UNECE ICP Integrated Monitoring network. Over 40 variables are analysed from the water samples including key acidification parameters and heavy metals.

The monitored lakes are small (median area=0.3 km^2), headwater, forest lakes, and are distributed throughout the country thus reflecting gradients in deposition, climate and landscape. The monitored headwater lakes were chosen to form an "early warning" system, because they have been shown to be good indicators of air pollution impacts (Mannio & Vuorenmaa 1995, Mannio 2001). They are more sensitive to acidification than the lake population as a whole, with low base cation concentrations, low alkalinity and pH and, in some lakes, elevated labile aluminium concentrations. The lakes have no upstream lakes, and the catchments have low catchment-to-lake area ratio. Most of the catchments (60%) are located in protected areas, and the land cover is mainly forested (median proportion of forest 91% of the terrestrial catchments excluding lakes in upland heaths (fell lakes)). The non-protected catchments have not been exposed to direct human impacts, except some minor forestry practices.

4.3.3 Acidification status in the 1980s and 1990s

The first statistically based survey of lake acidification in Finland took place in 1987 in connection with HAPRO (1985–1990) when 987 lakes (lake area ≥0.01 km^2) were randomly selected (Forsius et al. 1990a, b). Based on this survey, about 18% of the lakes had little or no bicarbonate buffering capacity (Gran alkalinity < 10 µeq/l) (Kämäri et al. 1991), and a total of 4900 lakes had Gran alkalinity ≤ 0 µeq/l, representing 12% of the lakes. A survey of lakes throughout northern Europe consisting of 5700 lakes ≥ 0.04 km^2 from six countries (873 lakes from Finland) was conducted in 1995 (Henriksen et al. 1998). Based on this survey, the number of acidic (Gran alkalinity ≤ 0 µeq/l) lakes ≥ 0.04 km^2 in Finland was estimated to be 1000 (Mannio et al. 2000). The number of acid-sensitive lakes (alkalinity < 20 µeq/l) was estimated to be 2700 lakes (9% of Finnish lakes ≥ 0.04 km^2) (Henriksen et al. 1998). The number of Finnish lakes of 0.04 to 1 km^2 in area susceptible to acidification was estimated to be around 5100 (Mannio 2001; Forsius et al. 2003).

The 1987 lake survey demonstrated that acidic or poorly buffered lakes were to be found throughout the country, but most commonly in south, western-central and eastern Finland (Kämäri et al. 1991). An extensive additional inventory (n=914) of small lakes in north Finland during 1987–1991 demonstrated that a considerable number of acidified lakes were also to be found in Finnish Lapland. The proportions of acidic lakes (Gran alkalinity < 0 µeq/l) and lakes with poor buffering capacity (Gran alkalinity 0–50 µeq/l) in north Finland were 6% and 19%, respectively (Kähkönen 1996). The acidity of a large number of Finnish lakes is dominated by natural organic acidity, particularly in peatland-rich regions of western and central Finland and in parts of eastern Finland and south Lapland (Kortelainen & Mannio 1990, Kähkönen 1996). Over 45% of lakes with a Gran alkalinity ≤ 0 µeq/l were assumed to be naturally acidic due to high concentrations of humus matter (Forsius et al. 1990b). But in south Finland, where sulphur deposition both from Finland's own emissions and the load of transboundary air pollution has been highest, and occurrence of peatlands is lower, minerogenic acidity commonly exceeds the catchment-derived organic acidity (Kortelainen & Mannio 1990). However, in humic lakes receiving acidic deposition, the influence of strong mineral acids is superimposed on organic acid contributions to acidity, and deposition has probably further decreased the pH in these lakes (Kämäri et al. 1991). Sulphate acidified clear-water lakes are found scattered throughout the country. Even in remote north Finland lakes acidified from long-range transported sulphur are found, and many lakes in upland heaths in north-eastern Lapland are affected by sulphur deposition from neighbouring Cu-Ni smelters in Kola Peninsula (Kähkönen 1996).

The effects of acidic deposition on the lake biota at different trophic levels were studied in a survey of 140 sensitive lakes located mainly in the southern and central part of the country (Eloranta 1990, Heitto 1990, Huttunen & Turkia 1990, Kippo-Edlund & Heitto 1990, Meriläinen & Hynynen 1990, Sarvala & Halsinaho 1990). Results indicated that acidification of waters had affected species composition and biodiversity of different biological communities. Acidification had effects on the structure of benthic assemblages also in north Finland (Yakovlev 1999).

The fish status survey in 1985–1987 reported declines and even extinctions of sensitive fish species in small lakes due to acidification in south and central Finland (Rask & Tuunainen 1990). The number of lakes in south and central Finland in which recent acidification has affected the growth or population structure of fish populations was estimated to be between 2200 and 4400, and out of these, the number of lakes in which fish populations have disappeared due to acid deposition was approximately 1000–2000 (Rask et al. 1995). Almost 60% of the affected or lost populations were roach (*Rutilus rutilus*), and less than 15% was European perch (*Perca fluviatilis*), the most common species in small lakes in south and central Finland. The fish status survey of Finnish lakes in connection with the Northern Europe Lake Survey in 1995 suggested that the number of lakes in which roach had been lost was 470 and 520 had affected roach stocks, and 410 lakes had affected perch stocks (Tammi et al. 2003). Both fish surveys suggested that perch was not extinct in any lakes, although there were records of lost perch stocks outside the coverage of the lake size range of these studies; that is, lakes smaller than 0.01 km² (Rask & Tuunainen 1990).

In a fish status and water chemistry survey carried out at 103 sites in three areas in north-eastern Lapland in 1991–1993, acid-induced damage was found for local minnow (*Phoxinus phoxinus*) populations in acid-sensitive low-alkalinity lakes (< 50 µeq/l) in the Vätsäri area, which has been exposed to acidic deposition from industrial emissions on the Kola Penisula (Lappalainen et al. 1995).

4.3.4 Present acidification status

The regional lake surveys in 1987 and 1995 clearly demonstrated that thousands of small forest lakes in Finland are susceptible to acidification and a number of lakes were acidified – to different degrees – due to the atmospheric sulphate deposition. Regional lake surveys after 1995 have not been carried out in Finland, and therefore the present-day situation of the extent of regional lake acidification cannot be empirically presented. However, it is evident that the number of very acidic lakes has decreased in the course of proceeding recovery from acidification through 1990s and 2000s, which has taken place in Finland (Vuorenmaa & Forsius 2008; Rask et al. 2014) as well as in wide areas in Europe and North America (Stoddard et al. 1999; Garmo et al. 2014). The data of selected 42 acidified monitoring lakes (mean ANC < 20 µeq/l in 1990–1992) showed that the annual average of ANC increased between the periods 1990–1992 and 2001–2003 from < 0 µeq/l to > 0 µeq/l in 70% of the lakes, and in 31% of the lakes ANC increased to a level ≥ 20 µeq/l between these two periods (Vuorenmaa 2007). The comparison of ANC levels between the periods 1990–1992 and 2014–2016 suggests that ANC level is now > 0 µeq/l in all monitored lakes (Figure 22). The mean values of ANC and ANC_{oaa} increased from 45 µeq/l to 75 µeq/l and from 33 µeq/l to 57 µeq/l, respectively, between these two periods. These results give strong evidence that the extent of regional acidification has decreased in terms of critical ANC thresholds, but the buffering capacity of many lakes is still low and still sensitive to acidic episodes and any future increase in acid deposition. In conditions of decreasing minerogenic acidification, increased catchment-derived organic acid episodes have become proportionally more important in affecting recovery process of sensitive lakes in Finland (Vuorenmaa & Forsius 2008; Rask et al. 2014). The regional-scale recovery of the lakes was also successfully predicted using MAGIC modelling (Helliwell et al. 2014).

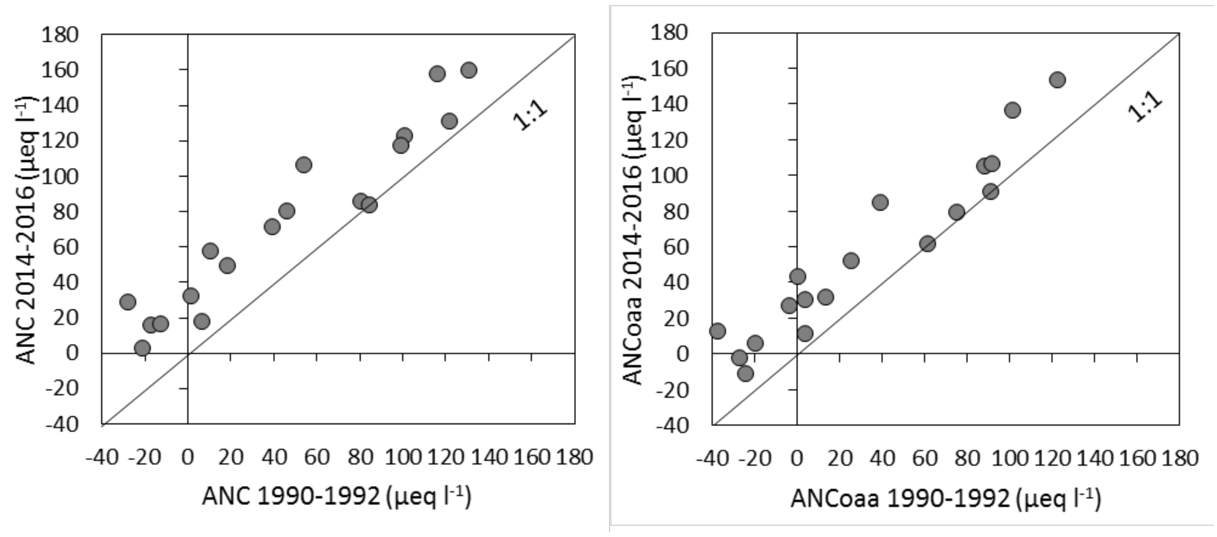


Figure 22 Mean annual ANC (calculated, μeq/l) (left) and ANC_{oaa} (calculated, μeq/l) (right) for the period 1990–1992 vs. the period 2014–2016 in 18 acidification monitoring lakes in Finland. ANC_{oaa} refers to strong organic acid adjusted concentration in which permanent anionic charge from strong organic acids is included (ANC_{oaa}=ANC - (3.4 x TOC)).

4.3.5 References

Eloranta P. 1990. Periphytic diatoms in the Acidification Project lakes. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 985–994.

Forsius M., Kämäri J., Kortelainen P., Mannio J., Verta M. & Kinnunen K. 1990a. Statistical lake survey in Finland: Regional estimates of lake acidification. In: Kauppi P., Anttila P. & Kenttämies K. (eds.). *Acidification in Finland*. Springer, Berlin, pp. 751–781.

Forsius M., Malin V., Mäkinen I., Mannio J., Kämäri J., Kortelainen P. & Verta M. 1990b. Finnish lake acidification survey: Survey design and random selection of lakes. *Environmetrics* 1: 79–88.

Forsius, M., Vuorenmaa, J., Mannio, J. & Syri, S. 2003. Recovery from acidification of Finnish lakes: regional patterns and relation to emission reduction policy. *The Science of the Total Environment* 310: 121–132.

Heitto L. 1990. Macrophytes in Finnish forest lakes and possible effects of airborne acidification. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland,* Springer, Berlin, pp. 963–972.

Helliwell, R.C., Wright, R.F., Jackson-Blake, L.A., Ferrier, R.C., Aherne, J., Evans, C.D., Forsius, M., Hruska, J., Jenkins, A., Kram, P., Kopacek,, J., Majer, V., Moldan, F., Posch, M., Potts, J.M., Rogora, M., and Schöpp, W. 2014. Assessing recovery from acidification of European surface waters in the year 2010: Evaluation of projections made with the MAGIC model in 1995. *Environmental Science & Technology* 48 (22), 13280-13288.

Henriksen A., Skjelkvåle B., Mannio J., Wilander A., Harriman R., Curtis C., Jensen J.P., Fjeld E. & Moiseenko T. 1998. Northern European Lake Survey, 1995. Finland, Norway, Sweden, Denmark, Russian Kola, Russian Karelia, Scotland and Wales. *Ambio* 27: 80–91.

Huttunen P. & Turkia J. 1990. Surface diatom assemblages and lake acidity. In: Kauppi P., Anttila P. & Kenttämies K. (eds.). *Acidification in Finland*. Springer, Berlin, pp. 995–1008.

Kippo-Edlund P. & Heitto A. 1990. Phytoplankton and acidification in small forest lakes in Finland. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 973–983.

Kortelainen P. & Mannio J. 1990. Organic acidity in Finnish lakes. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 849–863.

Kähkönen A.-M. 1996. Soil geochemistry in relation to water chemistry and sensitivity to acid deposition in Finnish Lapland. *Water Air Soil Pollut*. 87: 311–327.

Kämäri, J. 1986. Sensitivity of surface waters to acidic deposition in Finland. *Aqua Fennica* 16: 211–219.

Kämäri J., Forsius M., Kortelainen P., Mannio J. & Verta M. 1991. Finnish lake survey: present status of acidification. *Ambio* 20: 23–27.

Lappalainen A., Mähönen O., Erkinaro J., Rask M. & Niemelä, E. 1995. Acid deposition from the Russian Kola Peninsula: are sensitive fish populations in northeastern Finnish Lapland affected? *Water Air Soil Pollut*. 85: 439–444.

Mannio J. & Vuorenmaa J. 1995. Regional monitoring of lake acidification in Finland. *Water Air Soil Pollut*. 85: 571–576.

Mannio J., Räike A. & Vuorenmaa J. 2000. Finnish Lake Survey 1995: Regional characteristics of lake chemistry. *Verh. Internat. Verein. Limnol*. 27: 362–367.

Mannio J. 2001. Responses of headwater lakes to air pollution changes in Finland. *Monographs of the Boreal Environment Research*. Finnish Environment Institute, Helsinki.

Meriläinen J.J. & Hynynen J. 1990. Benthic invertebrates in relation to acidity in Finnish forest lakes. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 1029–1049.

Sarvala J. & Halsinaho S. 1990. Crustacean zooplankton of Finnish forest lakes in relation to acidity and other environmental factors. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland,* Springer, Berlin, pp. 1009–1027.

Rask M. & Tuunainen P. 1990. Acid-induced changes in fish populations of small Finnish lakes. In: Kauppi P., Anttila P. & Kenttämies K. (eds.), *Acidification in Finland*, Springer, Berlin, pp. 911–927.

Rask M., Mannio J., Forsius M., Posch M. & Vuorinen P.J. 1995. How many fish populations in Finland are affected by acid precipitation? *Environ. Biol. Fish.* 42: 51–63.

Rask M., Vuorenmaa J., Nyberg K., Tammi J., Mannio J., Olin M., Kortelainen P., Raitaniemi J. & Vesala S. 2014. Recovery of acidified lakes in Finland and subsequent responses of perch and roach populations. *Boreal Env. Res*. 19: 222–234.

Tammi J., Appelberg M., Beier U., Hesthagen T., Lappalainen A. & Rask M. 2003. Fish status of Nordic lakes: effects of acidification, eutrophication and stocking activity on present fish species composition. *Ambio* 32: 98–105.

Vuorenmaa, J. 2007. Recovery responses of acidified Finnish lakes under declining acid deposition. Yhteenveto: Pienentyneen laskeuman aiheuttamat toipumisprosessit Suomen happamoituvissa järvissä. Helsinki, Finnish Environment Institute. *Monographs of the Boreal Environment Research*, 30.

Vuorenmaa, J. and Forsius, M. 2008. Recovery of acidified Finnish lakes: Trends, patterns and dependence of catchment characteristics. Hydrol. Earth Syst. Sci. 12: 465–478.

Yakovlev V. 1999. Acidity of small lakes in Finnish Lapland - based on aquatic macroinvertebrate studies in 1993–1995. *The Finnish Environment* 234, Lapland Regional Environment Centre, Rovaniemi, Finland.

4.4 Germany

Jens Arle
German Environment Agency

4.4.1 Introduction

In 1983 the Convention on Long-range Transboundary Air Pollution (CLRTAP) entered into force to control air pollutant emissions in Europe and North America aiming to improve the environmental status of natural ecosystems. Over the past 30 years, Germany has contributed physical, chemical and biological data of surface waters to the International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters) in order to contribute to the documentation of the effects of the implemented Protocols under CLRTAP. Starting with 36 German monitoring sites (32 stream stations and 4 lake or reservoir stations) the monitoring programme was designed to representatively assess the regional degree of acidification of surface waters and of changes in acidification of streams and lakes caused by emission control measures and technological improvements.

The aim of the following report is to provide updated information on general trends of major physical, chemical and biological variables at the most consistently monitored stations of the official German stream monitoring network under the CLRTAP including also the most recent data collected between 2010 and 2016. An additional aim was to describe ranges of data available for different variables and data gaps in the German dataset.

4.4.2 Methods

Monitoring of physical and chemical factors followed the standardisations of sampling and analytical methodologies addressed in the ICP Waters Programme Manual (ICP Waters Programme Centre 2010). Only a small subset of chemical variables was analysed for the present report: pH, chloride (Cl^-), sulphate (SO_4^{2-}) and water temperature.

The sampling frequency for the monitoring sites varied from a single annual sample (most biological samples), to monthly sampling or more (some physical and chemical variables at some sites) and the frequency of observations for some monitoring sites differed between years.
The biological monitoring programme included regular sampling of macroinvertebrates using common kick net approaches. Biological sampling started at all German monitoring sites in the 1980`s or 1990`s (eastern parts of Germany). At some sites the monitoring activity was reduced, or the sites were declared “inactive” during the past 10 years. For the present analyses only 20 sites with biological, physical and chemical data for at least 24 years were used.

Site were selected if:

- data were available from biological as well as physical and chemical monitoring
- data were available for at least 20 years
- the most recent data were post 2012 for physical and chemical data and 2009 for biological data

Two biological metrics were calculated to assess the impact of acidification on macroinvertebrate assemblages: the number of taxa and the number of Ephemeroptera, Plecoptera and Trichoptera taxa, so-called EPT-taxa in each sample.

Statistical analyses were performed on raw, untransformed, disaggregated data for physical and chemical variables. Temporal trends in the biological, physical and chemical data were assessed using

Spearman rank order correlation. A value of $P<0.05$ was chosen as the determinate of statistical significance.

4.4.3 Results

Results of the trend analyses and additional descriptive information of the data sets for individual sites are summarised in Table 5.

At all sites a significant decrease in SO_4^{2-} concentrations was observed. pH value increased significantly at all sites. Chloride decreased at 14 sites, increased at 1 site and remained without a trend at two sites. At three sites chloride was not measured sufficiently continuous to allow a trend analysis. Water temperature increased significantly at 11 sites and remained without a significant trend at 8 sites (at one site no trend analysis was done).

A considerable increase of taxa number was observed at 19 sites, and only at one site was the positive trend not statistically significant. EPT - taxa richness increased at 14 sites, and at four sites no statistically significant trend was indicated although all Spearman coefficients were positive for these sites (for two sites no analysis was done). A total number of 298 EPT – taxa were found in the dataset and the following 10 taxa were the most regular occurring taxonomic groups in the dataset: *Leuctra sp., Protonemura sp., Nemoura sp., Leuctra nigra, Plecocnemia conspersa, Diura bicaudata, Amphinemura sp., Isoperla sp., Rhyacophila sp.* and *Nemurella pictetii*.

4.4.4 Discussion

During the last 30 years the emissions of many air pollutants were significantly reduced in Europe and North America (e.g. Garmo et al. 2014, Monteith et al. 2014). The observed trends for major physical and chemical variables at ICP Waters monitoring sites in German streams described in this report confirm the pattern of general improvement of water quality due to reduced airborne acidifying deposition. At about 50% of the monitoring sites a small, but significant increase in water temperature was observed in parallel to the strong reductions in sulphate, chloride and other ions. There was a pronounced increase in benthic invertebrate diversity at all sites. At most sites sensitive taxonomic groups of Ephemeroptera, Plecoptera and Trichoptera contributed significantly to the overall increase in taxa richness. The positive biological trends confirm earlier findings (e.g. Murphy et al. 2014, Velle et al. 2016). Most of the 20 German ICP sites are now in a good or very good ecological status according to the benthic invertebrate assessment system of the Water framework directive (Asterics / Perlodes, data not shown). For many taxa that have recolonized the formerly acidified stream sites it is unclear whether they have survived locally in refuges or in adjacent (in the past often organic polluted) downstream sites or in neighbouring catchments, which were less impacted by acidification due to higher buffer capacity. A reappearance of sensitive taxa is evident in the datasets of most German ICP water sites, but there is still sparse knowledge about the source populations for this recolonization. The very pronounced increase in diversity at most German ICP waters sites indicates that the most important and limiting pressure (airborne acidifying deposition) has been addressed and reduced in these stream systems. Many of the formerly acidified and species-poor stream networks have now a high species richness. After more than 30 years these streams may act for considerable number of EPT-Taxa again as source population networks.

The continuity of the monitoring varied at German ICP waters sites during the last 30 years, with some sites showing a very continuous and high frequency monitoring for many biological, physical and chemical variables. For some other monitoring sites only interrupted or terminated time series are to be found in the dataset. Some physical and chemical variables were measured more continuously than others at some sites, and some variables were measured at some sites but not at others.

The long term datasets collected under the CLRTAP are expected to be useful as baseline for future environmental changes, and they allow a very detailed picture of benthic invertebrate assemblages and their response to environmental changes. The invertebrate community datasets represent some of the oldest, most detailed and most continuous historical datasets of benthic invertebrates available in Germany. We hope and expect the continuation of the datasets will be secured in the next years by the new National Emissions Ceilings (NEC) Directive (2016/2284/EU) of the European Union, which requires European Member States to follow the methodologies agreed upon by the UNECE CLRTAP. Possibly the requirements of the NEC Directive might bring about a "reactivation" of the monitoring activities at the sites that were formerly declared “inactive”. This would be an essential step to make use of the data from these monitoring sites for future trend analyses.

Acknowledgement: I thank the German data contributors for their support of the ICP Waters monitoring and reporting during the last years.

4.4.5 References

Garmo, Ø. A., Skjelkvåle, B. L., de Wit, H. A., Colombo, L., Curtis, C., Fölster, J., Hoffmann, A., Hruška, J., Høgåsen, T., Jeffries, D.S., Keller, W.B., Krám, P., Majer, V., Monteith, D.T., Paterson, A.M., Logora, M., Rzychon, D., Steingruber, S., Stoddard, J.L., Vuorenmaa, J., Worsztynowicz A. & Keller, W. B. (2014). Trends in surface water chemistry in acidified areas in Europe and North America from 1990 to 2008. *Water, Air, & Soil Pollution*, *225*(3), 1880.

ICP Waters Programme Centre (2010). ICP Waters Programme Manual 2010. *ICP Waters report 105/2010*. 91 pp.

Monteith, D. T., Evans, C. D., Henrys, P. A., Simpson, G. L., & Malcolm, I. A. (2014). Trends in the hydrochemistry of acid-sensitive surface waters in the UK 1988–2008. *Ecological Indicators*, *37*, 287-303.

Murphy, J. F., Winterbottom, J. H., Orton, S., Simpson, G. L., Shilland, E. M., & Hildrew, A. G. (2014). Evidence of recovery from acidification in the macroinvertebrate assemblages of UK fresh waters: a 20-year time series. *Ecological Indicators*, *37*, 330-340.

Velle, G., Mahlum, S., Monteith, D.T., de Wit, H., Arle, J., Eriksson, L., Fjellheim, A., Frolova, M., Fölster, J., Grudule, N., Halvorsen, G.A., Hildrew, A., Hruška, J., Indriksone, I., Kamasová, L., Kopáček, J., Krám, P., Orton, S., Senoo, T., Shilland, E.M., Stuchlik, E., Telford, R.J., Ungermanová, L., Wiklund, M.-L. & Wright, R.F (2016). Biodiversity of macro-invertebrates in acid-sensitive waters: trends and relations to water chemistry and climate. *ICP Waters report 127/2016*. 33 pp.

Table 5 Results of the trend analyses and additional descriptive information on the long term data sets for German ICP sites. Time series trend description: ↑ - increasing, ↓ - decreasing, ≈ - no change), R = Spearman Correlation Coefficient and P-value; Max (year) = Maximum values in the dataset and the year(s) in which the maximum was/were measured; Min (year) = Minimum values in the dataset and the year(s) in which the maximum was/were measured; Sample size (n) = Number of measurements, time range (year – year) = the last year and the first year of the monitoring time series available. n.a. = not analysed (sample size not sufficient or limit of detection not appropriate)

Site No.	ICP Water Site name	pH - value	SO_4^{2-} - Sulphate (mg/l)	Cl^- - Chloride (mg/l)	Water temperature (°C)	Taxon number	EPT-Taxa-Number
DE01	Schwarzwald, Dürreychbach	↑ **R=0.758; P<0.0001** **7.9 (2011) –** **3.9 (1988)** **n=337; 2016 -1987**	↓ **R=-0.422; P<0.0001** **10.3 (1994) –** **1.8 (1995)** **n=331; 2016 -1987**	≈ R=-0.08; P>0.05 11.9 (1999) – 0.6 (2014) n=330; 2016 -1987	↑ **R=0.170; P=0.0017** **11.7 (2014) –** **0.3 (2010)** **n=338; 2016 -1987**	≈ R=0.324; P=0.0751 32 (2008) – 9 (2002) n=31; 2013 - 1987	≈ R=0.321; P=0.0774 20 (2008) – 6 (1988, 2000, 2002) n=31; 2013 - 1987
DE02	Fichtelgebirge, Eger	↑ **R=0.239; P<0.0001** **7.7 (2004) – 3.6 (2002)** **n=422; 2016 -1982**	↓ **R=-0.227; P<0.0001** **20.0 (1983) – 2.2 (1985)** **n=400; 2016 -1983**	↑ **R=0.567; P<0.0001** **16.2 (1991) – 1.9 (1987)** **n=408; 2016 -1982**	↑ **R=0.168; P=0.0019** **12.5 (1992) – 0.0 (1987)** **n=337; 2016 -1982**	↑ **R=0.617; P<0.0001** **38 (2016) – 8 (1990)** **n=31; 2016 -1989**	↑ **R=0.495; P<0.005** **19(2016) – 5 (1991)** **n=31; 2016 -1989**
DE03	Rothaargebirge, Elberndorfer Bach	↑ R=**0.223; P<0.0001** **8.4 (1997) –** **4.6 (1999)** **n=474; 2013 -1986**	↓ **R=-0.727; P<0.0001** **20.0 (1988, 1989, 1992)** **– 2,7 (1993)** **n=403; 2013 -1988**	n.a.	↑ **R=0.231; P<0.0001** **19.1 (1997) –** **0.1 (1986, 1994, 2010)** **n=484; 2013 -1986**	↑ **R=0.801; P<0.0001** **61 (2013) –** **6 (1988, 1989)** **n=48; 2016 -1988**	↑ **R=0.511; P<0.0001** **37 (2004) –** **4 (1988)** **n=48; 2016 -1988**
DE05	Schwarzwald, Goldersbach	↑ **R=0.341; P<0.0001** **7.6 (2003) –** **5.4 (2001)** **n=323; 2016 -1986**	↓ **R=-0.761; P<0.0001** **9.3 (1986) –** **1.0 (2014)** **n=320; 2016 -1986**	≈ R=0.03; P>0.05 24.8 (1999) – 0.25 (2015, 2016) n=323; 2016 -1986	≈ R=-0.01; P>0.05 16.8 (2003) – 0.1 (1991, 1997) n=277; 2016 -1986	↑ **R=0.698; P<0.0001** **50 (2014) –** **6 (1988)** **n=46; 2014 -1986**	↑ **R=0.705; P<0.0001** **35 (2014) –** **3 (1988)** **n=46; 2014 -1986**
DE06	Hunsrück, Gräfenbach	↑ **R=0.694; P<0.0001** **5.9 (2011) –** **3.6 (2001)** **n=279; 2016 -1982**	↓ **R=-0.899; P<0.0001** **51.0 (1987) –** **10.0 (2016)** **n=277; 2016 -1982**	n.a.	≈ R=0.10; P>0.05 16.1 (2011) – 0.00 (1988, 1993,1994, 2000) n=279; 2016 -1982	↑ **R=0.505; P=0.044** **28 (2015) –** **1 (1990)** **n=16; 2015 -1982**	≈ R=0.337; P>0.05 19 (2015 - 0 (1990) n=16; 2015 -1982
DE07	Erzgebirge, Große Pyra	↑ **R=0.595; P<0.0001** **6.6 (2012) –** **3.8 (2003)** **n=239; 2016 -1982**	↓ **R=-0.931; P<0.0001** **50.5 (1982) –** **6.0 (2016)** **n=240; 2016 -1982**	↓ **R=-0.498; P<0.0001** **12.9 (1982) –** **0.6 (2016)** **n=225; 2016 -1982**	≈ R=0.08; P>0.05 14.9 (2012) – 0.1 (1998) n=185; 2012 -1982	↑ **R=0.681; P<0.0001** **33 (2007, 2015) –** **5 (1992, 1994)** **n=66; 2016 -1992**	↑ **R=0.501; P<0.0001** **19 (2007) –** **3 (2004)** **n=66; 2016 -1992**
DE08	Bayerischer Wald, Grosse Ohe	↑ **R=0.328; P<0.0001** **7.7 (2013) –** **3.8 (1986)** **n=1770; 2016 -1979**	↓ **R=-0.566; P<0.0001** **47.6 (1989) –** **0.25 (1987)** **n=1447; 2016 -1979**	↓ **R=-0.408; P<0.0001** **26.0 (1981) –** **0.25 (1983-1990)** **n=1701; 2016 -1979**	↑ **R=0.079; P=0.017** **20.1 (2013) – 0.2 (1980, 1983, 1986 1994, 1997, 2006)** **n=899; 1979-2016**	↑ **R=0.833; P<0.0001** **55 (2016) –** **19 (1983)** **n=26; 2015 -1983**	↑ **R=0.476; P=0.0143** **34 (1997, 2004) –** **13 (1983)** **n=26; 2015 -1983**

Site No.	ICP Water Site name	pH - value	SO_4^{2-} - Sulphate (mg/l)	Cl^- - Chloride (mg/l)	Water temperature (°C)	Taxon number	EPT-Taxa-Number
DE10	Bayerischer Wald, Hinterer Schachtenbach	↑ **R=0.389; P<0.0001** **7.6 (2013) –** **4.2 (1987, 2000)** **n=403; 2016 -1979**	↓ **R=-0.713; P<0.0001** **13.0 (1987) –** **0.8 (1986, 1991)** **n=361; 2012 -1983**	↓ **R=-0.538; P<0.0001** **5.9 (1991) –** **0.3 (2009)** **n=351; 2014 -1984**	≈ R=0.05; P>0.05 18.1 (1983) – 0.0 (1994) n=377; 2016 -1979	↑ **R=0.722; P<0.0001** **52 (2011) –** **10 (1983)** **n=24; 2011 -1983**	↑ **R=0.732; P<0.0001** **30 (2003, 2011) –** **9 (1983)** **n=24; 2011 -1983**
DE11	Schwarzwald, Kleine Kinzig Huettenhardt	↑ **R=0.502; P<0.0001** **7.9 (2011) –** **5.2 (2002)** **n=160; 2016 -2001**	↓ **R=-0.818; P<0.0001** **16.0 (2001, 2002) –** **1.00 (2014)** **n=159; 2016 -2001**	↓ **R=-0.496; P<0.0001** **40.0 (2004) –** **1.9 (2015)** **n=157; 2016 -2001**	≈ R=0.049; P>0.05 15.2 (2002) – 0.1 (2002) n=159; 2016 -2001	↑ **R=0.866; P<0.0001** **59 (2014) –** **10 (2001)** **n=45; 2014 -1985**	↑ **R=0.876; P<0.0001** **45 (2014) –** **7 (2001)** **n=45; 2014 -1985**
DE12	Harz, Lange Bramke	↑ **R=0.182; P<0.0001** **7.4 (1976) –** **4.3 (1981)** **n=1635; 2016 -1970**	↓ **R=-0.331; P<0.0001** **17.98 (1976) –** **2.5 (2016)** **n=1382; 2016 -1970**	↓ **R=-0.405; P<0.0001** **4.9 (1991) –** **1.4 (2004)** **n=1483; 2016 -1970**	n.a.	↑ **R=0.791; P<0.0001** **25 (2009) –** **2 (1988)** **n=37; 2009 -1986**	↑ **R=0.790; P<0.0001** **16 (2008) –** **2 (1986, 1988)** **n=37; 2009 -1986**
DE18	Fichtelgebirge, Röslau	↑ **R=0.431; P<0.0001** **8.7 (1988) –** **3.7 (2002)** **n=412; 2016 -1982**	↓ **R=-0.857; P<0.0001** **29.0 (1983) –** **5.9 (2012, 2013)** **n=400; 2016 -1982**	↓ **R=-0.654; P<0.0001** **5.0 (1982) –** **0.6 (1985)** **n=410; 2016 -1982**	↑ **R=0.146; P=0.004** **14.3 (2015) –** **-0.1 (2006)** **n=388; 2016 -1982**	↑ **R=0.664; P<0.001** **19 (2016) –** **4 (1991)** **n=31; 2016 -1989**	↑ **R=0.665; P<0.001** **14 (1993) –** **2 (1991)** **n=31; 2016 -1989**
DE21	Erzgebirge, Rote Pockau	↑ **R=0.557; P<0.0001** **8.3 (1994) –** **4.1 (1993, 1995)** **n=229; 2016 -1979**	↓ **R=-0.814; P<0.0001** **51.0 (1994) –** **13.0 (2014)** **n=226; 2016 -1992**	↓ **R=-0.445; P<0.0001** **40.0 (1997) –** **1.9 (2007)** **n=226; 2016 -1979**	↑ **R=0.162; P=0.017** **15.3 (2015) –** **0.1 (1994, 2003)** **n=215; 2016 -1979**	↑ **R=0.790; P<0.0001** **44 (2009) –** **3 (1992)** **n=64; 2016 -1992**	↑ **R=0.666; P<0.0001** **23(2009) –** **3 (1992)** **n=64; 2016 -1992**
DE23	Bayerischer Wald, Seebach	↑ **R=0.267; P<0.0001** **7.3 (1983, 2013) –** **4.1 (1983)** **n=407; 2014 -1983**	↓ **R=-0.801; P<0.0001** **14.7 (1987) –** **0.8 (1986)** **n=388; 2014 -1983**	↓ **R=-0.483; P<0.0001** **5.0 (1984, 1988) –** **0.3 (2009, 2014)** **n=358; 2014 -1983**	≈ R=0.06; P>0.05 15.8 (1985) – 0.0 (1994) n=403; 2014 -1983	↑ **R=0.644; P=0.0016** **53 (2011) –** **11 (1983)** **n=21; 2011 -1983**	↑ **R=0.692; P<0.0001** **30 (2006, 2008) –** **9 (1983)** **n=21; 2011 -1983**
DE26	Hunsrück, Traunbach 1	↑ **R=0.599; P<0.0001** **7.3 (2003) –** **3.4 (1995)** **n=365; 2016 -1982**	↓ **R=-0.517; P<0.0001** **38.7 (1998) –** **1.30 (2003, 2008)** **n=357; 2016 -1982**	↓ **R=-0.256; P<0.0001** **38.0 (1985) –** **3.0 (2003)** **n=349; 2016 -1982**	↑ **R=0.144; P=0.006** **17.3 (2015, 2016) –** **0.2 (1999)** **n=356; 2016 -1982**	↑ **R=0.821; P<0.0001** **24 (2015) –** **1 (1985)** **n=19; 2015 -1983**	≈ R=0.364; P>0.05 7 (1999, 2015) – 1 (1985, 2008) n=19; 2015 -1983
DE27	Bayerischer Wald, Vorderer Schachtenbach	↑ **R=0.213; P<0.0001** **7.6 (2013) –** **4.3 (2002)** **n=405; 2014 -1983**	↓ **R=-0.766; P<0.0001** **10.0 (1983) –** **0.5 (1991)** **n=387; 2014 -1983**	↓ **R=-0.496; P<0.0001** **5.8 (1998) –** **0.3 (1984, 2009, 2014)** **n=351; 2014 -1983**	≈ R=0.07; P>0.05 15.2 (2012) – 0.0 (1994) n=403; 2014 -1983	↑ **R=0.820; p<0.0001** **62 (2011) –** **14 (1983)** **n=24; 2011 -1983**	n.a.

Site No.	ICP Water Site name	pH - value	SO_4^{2-} - Sulphate (mg/l)	Cl^- - Chloride (mg/l)	Water temperature (°C)	Taxon number	EPT-Taxa-Number
DE29	Oberpfälzer Wald, Waldnaab 8	↑ **R=0.503; P<0.0001** **7.3 (1995) – 3.8 (1989)** **n=97; 2016 -1986**	↓ **R=-0.753; P<0.0001** **37.5 (1986) – 6.1 (2015)** **n=85; 2016 -1986**	↓ **R=-0.774; P<0.0001** **4.2 (1986) – 1.4 (2015)** **n=88; 2016 -1986**	↑ **R=0.226; P=0.028** **14.8 (2014) – 0.1 (2016)** **n=95; 2016 -1986**	↑ **R=0.543; P<0.0001** **39 (2008) – 5 (1984)** **n=54; 2016 -1984**	n.a.
DE30	Erzgebirge, Wilde Weisseritz	↑ **R=0.482; P<0.0001** **8.1 (2010) –** **3.8 (1975)** **n=613; 2016 -1967**	↓ **R=-0.505; P<0.0001** **135.0 (1989) –** **7.00 (1968)** **n=314; 2016 -1967**	↓ **R=-0.640; P<0.0001** **30.0 (1978) –** **1.5 (2010)** **n=590; 2016 -1967**	≈ R=0.03; P>0.05 19.4 (2007) – 0.0 (2010) n=550; 2016 -1967	↑ **R=0.860; P<0.0001** **51 (2015) –** **3 (1993, 1996)** **n=74; 2016 -1992**	↑ **R=0.871; P<0.0001** **28 (2012, 2014) –** **2 (1996)** **n=74; 2016 -1992**
DE31	Erzgebirge, Wolfsbach	↑ **R=0.154; P=0.037** **7.8 (2006) – 6.2 (1994)** **n=185; 2012 -1992**	↓ **R=-0.722; P<0.0001** **150.0 (2003)–14.0 (2012)** **n=185; 2012 -1992**	↓ **R=-0.214; P=0.004** **35.0 (1992) – 4.0 (2003)** **n=182; 2012 -1992**	↑ **R=0.236; P=0.002** **20.3 (2012) – 0.4 (2004)** **n=168; 2012 -1992**	↑ **R=0.439; P=0.0138** **40 (1997) – 4 (1992)** **n=31; 2012 -1992**	≈ R=0.338; P>0.05 23 (1997) – 2 (1992) n=31; 2012 -1992
DE32	Rothaargebirge, Zinse	↑ **R=0.149; P=0.0013** **8.2 (2013) –** **4.2 (1999)** **n=469; 2013 -1986**	↓ **R=-0.419; P<0.0001** **19.0 (1988, 1994) –** **2.0 (1993)** **n=442; 2013 -1986**	n.a.	↑ **R=0.254; P<0.0001** **16.3 (1993) –** **-0.1 (2013)** **n=482; 2013 -1986**	↑ **R=0.750; P<0.0001** **63 (2013) – 3 (1989, 1992, 1993, 1994)** **n=50; 2016 -1988**	↑ **R=0.526; P<0.0001** **34 (2013) – 3 (1989, 1990,1992 1993, 1994)** **n=50; 2016 -1988**
DE33	Fichtelgebirge, Zinnbach	↑ **R=0.572; P<0.0001** **4.8 (1983, 2004, 2014) –** **3.3 (1989, 1993)** **n=378; 2016 -1983**	↓ **R=-0.861; P<0.0001** **43.0 (1983) –** **5.0 (1983)** **n=366; 2016 -1983**	↓ **R=-0.823; P<0.0001** **8.1 (1986) –** **0.7 (2014)** **n=371; 2016 -1983**	↑ **R=0.183; P<0.0001** **15.4 (2015) –** **-0.1 (2006)** **n=350; 2016 -1983**	↑ **R=0.662; P<0.0001** **14 (2012) –** **2 (1991)** **n=30; 2016 -1989**	↑ **R=0.549; P=0.0018** **8 (1998, 2008) –** **1 (1991)** **n=30; 2016 -1989**

4.5 Ireland

Julian Aherne[1], Wayne Trodd[2], Deirdre Tierney[2], Gary Free[2], John McEntagart[2]
[1] Trent University
[2] Environmental Protection Agency

4.5.1 Acid sensitivity

Large areas of Ireland are considered sensitive to acidification due to a combination of poor mineral soils, typically podsols or peaty podsols, and organic soils overlying slowly weathering base poor geologies such as granite, quartzite, schists and gneiss (Aherne and Farrell, 2000; Aherne et al. 2002; Figure 23). These extensive areas with acid sensitive water bodies lie along the western seaboard and on the middle east coast in Ireland. It has long been recognized that the surface waters in these areas typically have low alkalinity and consequently a poor buffering capacity (Bowman, 1986 and 1991). The prevailing wind is predominantly westerly and the associated precipitation is unlikely to be anthropogenically acidified; however, precipitation associated with easterly air masses has been shown to be acidic (Bowman and McGettigan, 1994); as such, the eastern seaboard typically receives higher levels of acidic precipitation (Aherne and Farrell, 2002).

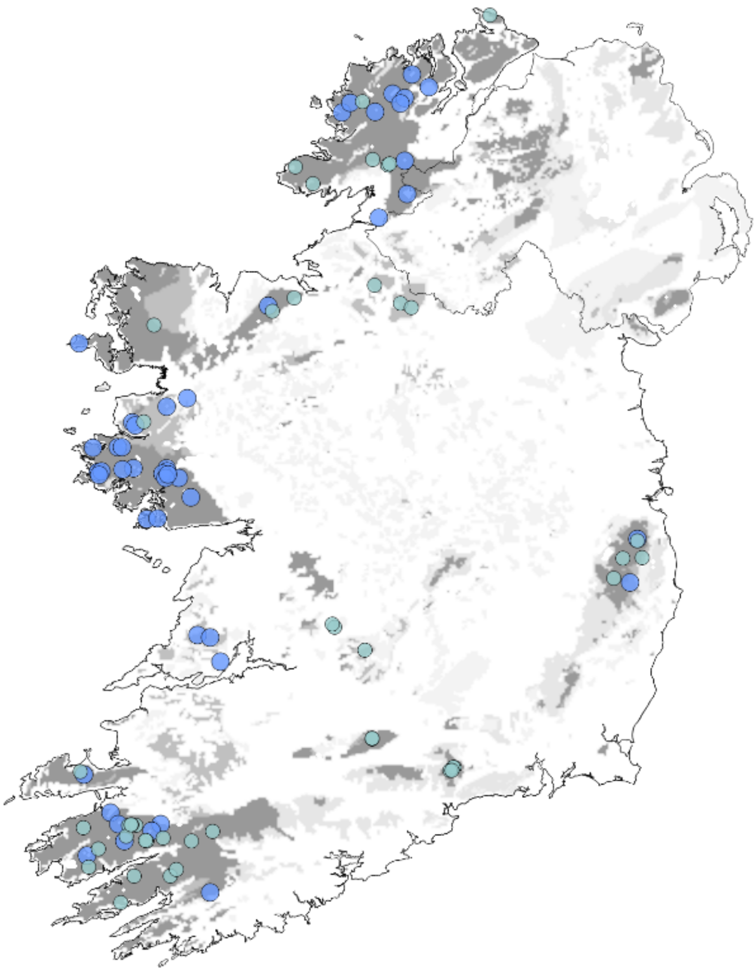


Figure 23 Relative sensitivity of surface waters to acidic deposition (darkening shade implies increasing sensitivity to the acidic deposition). The location of the survey lakes (n=92) is also shown; larger blue-filled circles indicate the location of 'acid sensitive' lakes under the Water Framework Directive, and the smaller blue-green-filled circles indicated the upland survey lake (see Aherne et al., 2002).

4.5.2 Monitoring and assessment approach

The monitoring of acid sensitive lakes is carried out by the Irish EPA[19] primarily under the Water Framework Directive. The 49 'acid' lakes are sampled for hydrochemistry six times a year, with a sub-set (n = 20) monitored up to 12 times a year (once every four years) under the WFD surveillance monitoring programme. In addition, macrophytes, phytoplankton and invertebrates are regularly monitored. The lakes were selected to be representative of well-established acid-sensitive regions (Figure 23), with many of the lakes occasionally monitored since the 1980s (Bowman, 1986 and 1991). However, accurate data archiving has only been carried out since the implementation of the WFD. Three of these lakes (Glendalough, Lough Veagh and Lough Maumwee) have been included in the ICP Waters programme since the 1980s; expanded to fourteen lakes in 2014 (with data from 2004 onwards).

The assessment of acid-sensitive lakes is also carried out under the Survey of Upland Acid Systems (SUAS), which is a decadal survey focused on the hydrochemistry of small upland headwater lakes in acid-sensitive regions (Aherne et al., 2002; Burton et al., 2012). These lakes have been surveyed since 1997, albeit with a reduction in sample population from 200 in 1997 to 50 in 2017, the latter surveys have focus on the higher elevation lakes. As such, compared with the WFD lakes, the SUAS lakes are smaller (average lake area: 1 km^2 [WFD] compared with 0.4 km^2 [SUAS]), located at higher elevations, and have lower concentrations of dissolved organic carbon [DOC] and acid neutralizing capacity adjusted for organic acids (ANC_{oaa}: see Figure 24). However, sampling frequency varied greatly between the two datasets; during the period 2010–2017, the WFD lakes were sampled 33 times per lake (on average, with a max of 19), compared with twice for the SUAS lakes (max of 14).

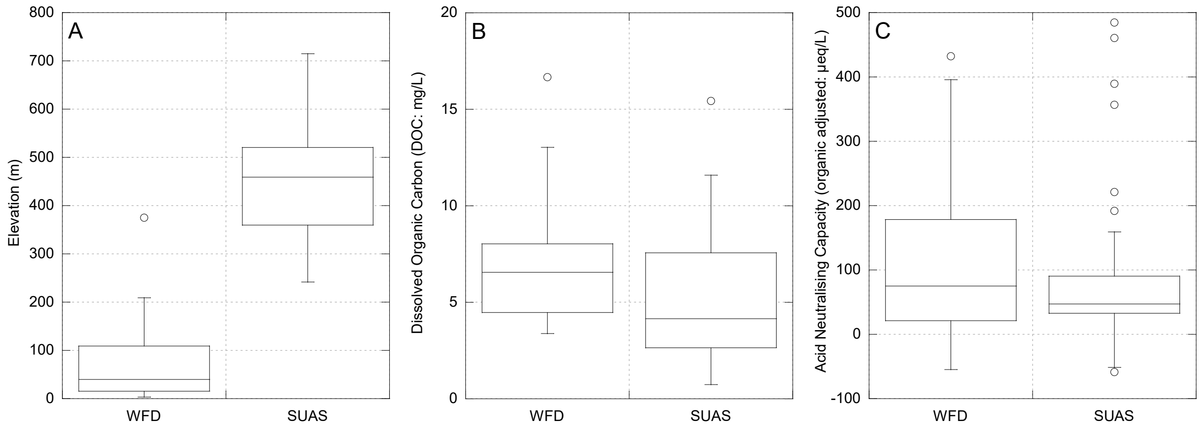


Figure 24 Box plot of elevation (m), dissolved organic carbon (DOC: mg/l) and organic acid adjusted Acid Neutralising Capacity (ANC_{oaa}: µeq/l) in the Water Framework Directive (WFD) lakes (n=47) and Survey of Upland Acid Systems (SUAS) lakes (n=45).

4.5.3 Acidification status

During the period 2010–2017, average lake pH ranged from 4.48 to 7.58, with an overall average pH of 5.4 across the study lakes (n = 92). Lakes with lower pH were primarily located on the middle east coast and the north-western seaboard (Figure 25). In general, these lakes tended to have higher concentrations of DOC (Figure 25), associated with the coverage of organic soils. The average DOC was 6.0 mg/l and ranged from 0.7 to 16.7 mg/l. Nonetheless, 10–15% of the study lakes (n=92) are considered to be highly acid sensitive and still 'acid impacted' based on ANC (Figure 25) or organic acid adjusted ANC. It is important to note that this is not representative of the national-scale, as monitoring and assessment was focused on acid sensitive regions but 10% 'acid impacted' is considered to be a reasonable estimate for acid sensitive regions.

[19] www.epa.ie

During the past two decades there has been an improvement in the acid status of many lakes, owing to the reduction in acidic (sulphur and nitrogen) deposition. Upland lakes (SUAS: n= 30) surveyed during 1997, 2007 and 2017 have shown a statistically significant decrease in sulphate concentrations and increase in alkalinity (Figure 26). Given Ireland's location on the western periphery of Europe, surface waters are strongly influenced by sea salts, Atlantic storms and changes in DOC (owing to the higher coverage of organic soils), i.e., the hydrochemical signal to noise ratio is often low. Nonetheless, the decadal surveys of upland lakes suggest that average sulphate decreased by > 1.5mg/l and alkalinity increased by 1.0 mg/l $CaCO_3$ during the past two decades.

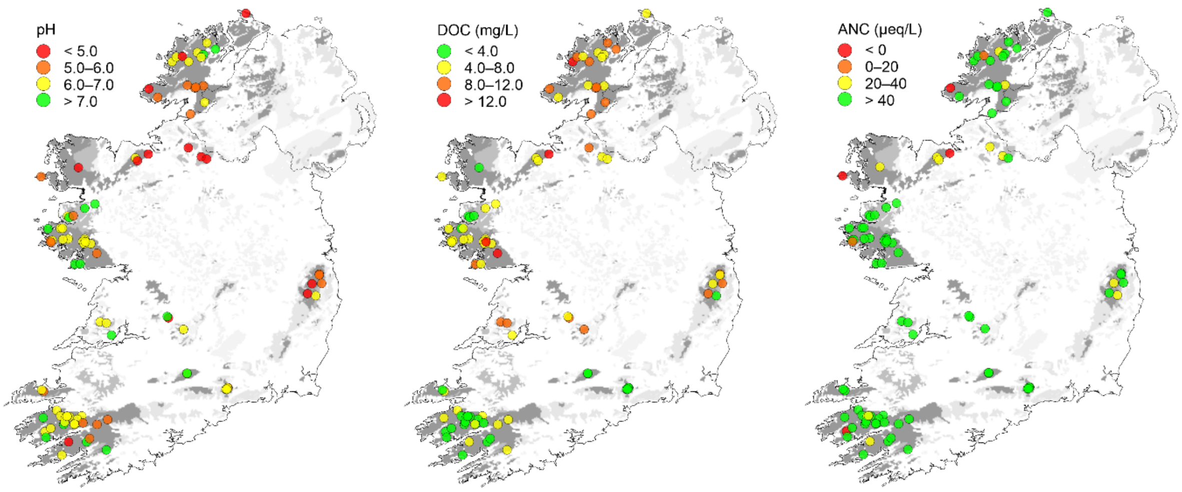


Figure 25 Average hydrochemistry for lakes (n = 92) located in 'acid sensitive' regions during the period 2010–2017 (note: darkening shade implies increasing sensitivity to the acidic deposition). Lake chemistry is displayed in four classes for pH, dissolved organic carbon (DOC: mg/l) and Acid Neutralising Capacity (ANC: µeq/l).

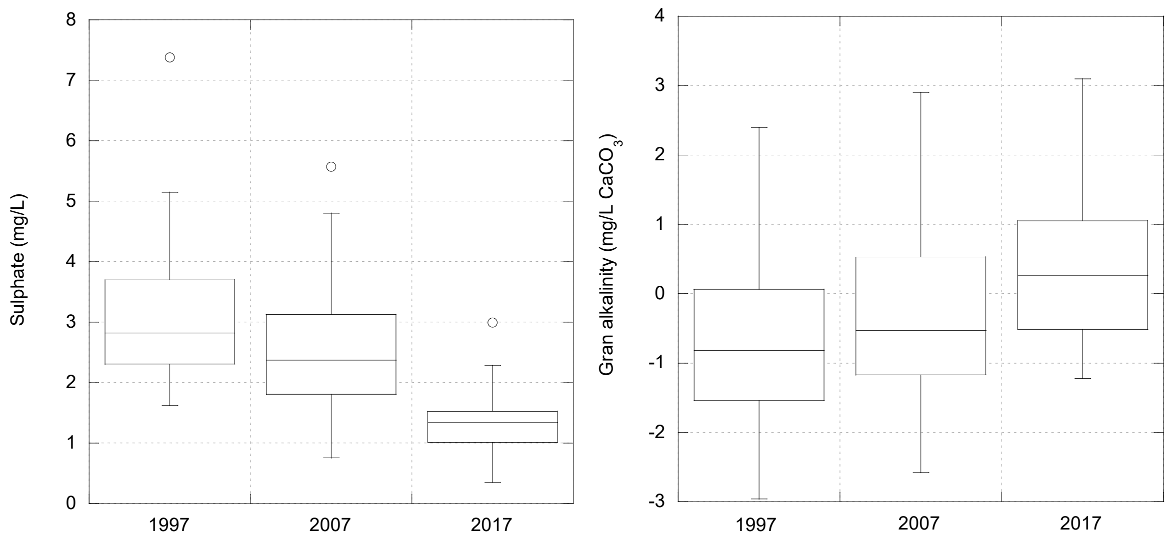


Figure 26 Box plot of concentrations of sulphate (mg/l) and Gran alkalinity (mg/l CaCo3) during 1997, 2007 and 2017 in upland acid sensitive lakes (n = 30).

Acknowledgement: This study was supported by the Irish EPA through project 2016-CCRP-MS.43.

4.5.4 References

Aherne, J. and Farrell, E.P, 2000. Sensitivity of Irish lakes to acidification. Verh. Internat. Verein. Limnol. 27: 2467–2470.

Aherne, J. and Farrell, E.P. 2002. Deposition of sulphur, nitrogen and acidity in precipitation over Ireland: chemistry, spatial variation and long-term trends. Atmospheric Environment 36: 1379–1389.

Aherne J., Kelly-Quinn, M. and Farrell, E.P. 2002. A survey of lakes in the Republic of Ireland: hydrochemical characteristics and acid sensitivity. Ambio 31: 452–459.

Bowman, J. 1986. Precipitation Characteristics and the Chemistry and Biology of Poorly Buffered Irish Lakes: A Western European Baseline for 'Acid Rain' Impact Assessment. An Foras Forbartha, Dublin, Ireland, 132 pp.

Bowman, J. 1991. Acid Sensitive Surface Waters in Ireland. Environment Research Unit, Dublin, Ireland, 321 pp.

Bowman, J. and McGettigan, M. 1994. Atmospheric deposition in acid sensitive areas of Ireland: The influence of wind direction and a new coal burning electricity generation station on precipitation quality. Water Air Soil Pollut. 75, 159–175.

Burton, A. and Aherne, J. 2012. Changes in the chemistry of small Irish lakes. Ambio 41:2, 170–79.

4.6 Italy

Michela Rogora, Aldo Marchetto, Rosario Mosello, Gabriele Tartari
CNR Institute of Ecosystem Study, Verbania Pallanza

4.6.1 Introduction

The Institute of Ecosystem Study of the National Research Council of Italy (CNR ISE) in Verbania Pallanza has been the National Focal Centre for ICP Waters since 1995, under the direction and coordination of the Italian Ministry for the Environment, Land and Sea. The Italian network and the monitoring activities are described in detail in Mosello et al. (2001). Within the ICP Waters aims, the main pressures identified as important at the Italian sites in the network are acidification and nitrogen deposition (Rogora et al., 2012, 2013). Furthermore, especially in recent times, attention has been paid to the effects of climate change, in interaction with the other pressures.

Acidification of surface waters due to acid rain has been a problem in the 1970s and 1980s for some high-altitude lakes in the Central Alps, characterised by low alkalinity and limited buffering capacity due to the geological composition of the catchments (Marchetto et al., 1994). Further acid sensitive sites, both rivers and lakes, were identified in the area of the Lake Maggiore catchment in north-western Italy (Mosello et al., 2000). This area is subject to high deposition of atmospheric pollutants, due to its location north of the Po Plain, one of the most densely inhabited, industrialised and urbanised areas of Europe (Rogora et al., 2006).

In Italy, major effects of acid rain were detected at a limited number of sites in the Central Alps, showing significant pH decrease (below 5.6), complete loss of alkalinity and an increase in aluminium concentrations. Although only a few lakes were effectively acidified, most of the lakes in this part of the Alps proved to be sensitive to acidic inputs and potentially threatened, if deposition had remained at the same level (Marchetto et al., 1994). Palaeolimnological studies outlined that lake acidification paralleled the increasing input of long-range transported industrial pollutants, traced by spherical carbonaceous particles (Marchetto et al., 2004).

Most of the sensitive sites which underwent acidification in the 1980s, partially recovered from the mid-1990s in response to the decreasing deposition of acidifying compounds, and showed an increase of alkalinity and pH (Rogora et al., 2001; 2013). This recovery was mainly due to a sharp decrease in the deposition of acidity and sulphate, as an effect of decreasing emissions of SO_2; on the other hand, deposition of nitrogen, both as ammonium and nitrate, did not change significantly or decreased only slightly in the last few years (Rogora et al., 2016).

4.6.2 Study sites and acidification assessment approach

To update the assessment of the acidification status of water bodies in the area of Lake Maggiore watershed, we performed a new monitoring campaign in 2017. In addition to the lake and rivers sites formally included in the ICP Waters network, the CNR ISE has monitored the chemistry of about 30 high altitude lakes regularly since the early 1980s. The monitoring of these lakes and the analyses of results have been regularly performed in close cooperation with the Swiss colleagues in charge for the ICP Waters network in Switzerland (Rogora et al., 2013).

In total, 31 sites, mainly high-altitude lakes located in the Ossola valley, Central Alps, were sampled in 2017 and analysed for the main chemical variables at the CNR ISE water chemistry laboratory. Details on the analytical methods and the QA/QC in use at the laboratory can be found at the laboratory web site[20].

[20] http://www.idrolab.ise.cnr.it

We compared average pH and ANC values (calculated as $Ca^{++}+Mg^{++}+Na^{+}+K^{+}-SO_4^{=}-NO_3^{-}-Cl^{-}$) measured in the 1980s with those of the most recent survey (2017), to assess temporal changes in the acidification status. As an example of recovery from acidification at a sensitive site, we also assessed long-term trends (1984-2017) of selected chemical variables at the ICP Water site Lake Paione Superiore (IT03).

4.6.3 Results

The comparison of average pH and ANC values measured in the 1980s and in 2017 showed a recovery for the most acid-sensitive sites, located in the central-southern part of the area (Figure 27). These sites lie on bedrock mainly consisting of metamorphic rocks (gneisses, granites). Sites in the northern part of the area partly lie in catchments made up of more soluble, basic rocks (carbonate schists, limestone, dolomites): these sites are much less sensitive towards acidification and their pH and ANC values did not change over time.

Despite a general tendency towards higher pH and ANC, a few sites are still acidic or characterised by a high sensitivity towards acidification: for instance, 7 lakes still show ANC values below 50 µeq/l in 2017, and several sites have ANC between 50 and 200 µeq/l (Figure 27).

Long-term trends of selected chemical variables for the ICP Waters site Lake Paione Superiore (PAS; ICPW site IT03) are shown in Figure 28. The lake showed positive trends of pH and ANC since the mid-1990s, mainly as an effect of the sharp decrease of sulphate. pH values reached 6.5-6.6 in recent years and ANC is presently around 25 µeq/l (Figure 28). Some first signs of biological recovery were also detected at this site, such as change in diatom flora and appearance of sensitive species among benthic insects (Marchetto et al., 2004).

In contrast to sulphate, nitrate concentrations at the study sites did not change significantly, except for a slight decrease in the most recent period (Rogora et al., 2012). For instance, nitrate concentrations in Lake Paione Superiore are presently around 15 µeq/l, compared to 20-25 µeq/l in the 1980s-1990s (Figure 28).

A recent study performed on about 40 lakes in Italy (Lake Maggiore watershed) and Switzerland (Canton Ticino) showed that some lakes are still affected by acidification, especially during snowmelt, when alkalinity may be fully depleted (Rogora et al., 2013). The study also highlighted the prominent role of N deposition in this area: at present, nitrate is the dominant acidifying agent in the studied lakes, due to the high input of nitrogen compounds from atmospheric deposition.

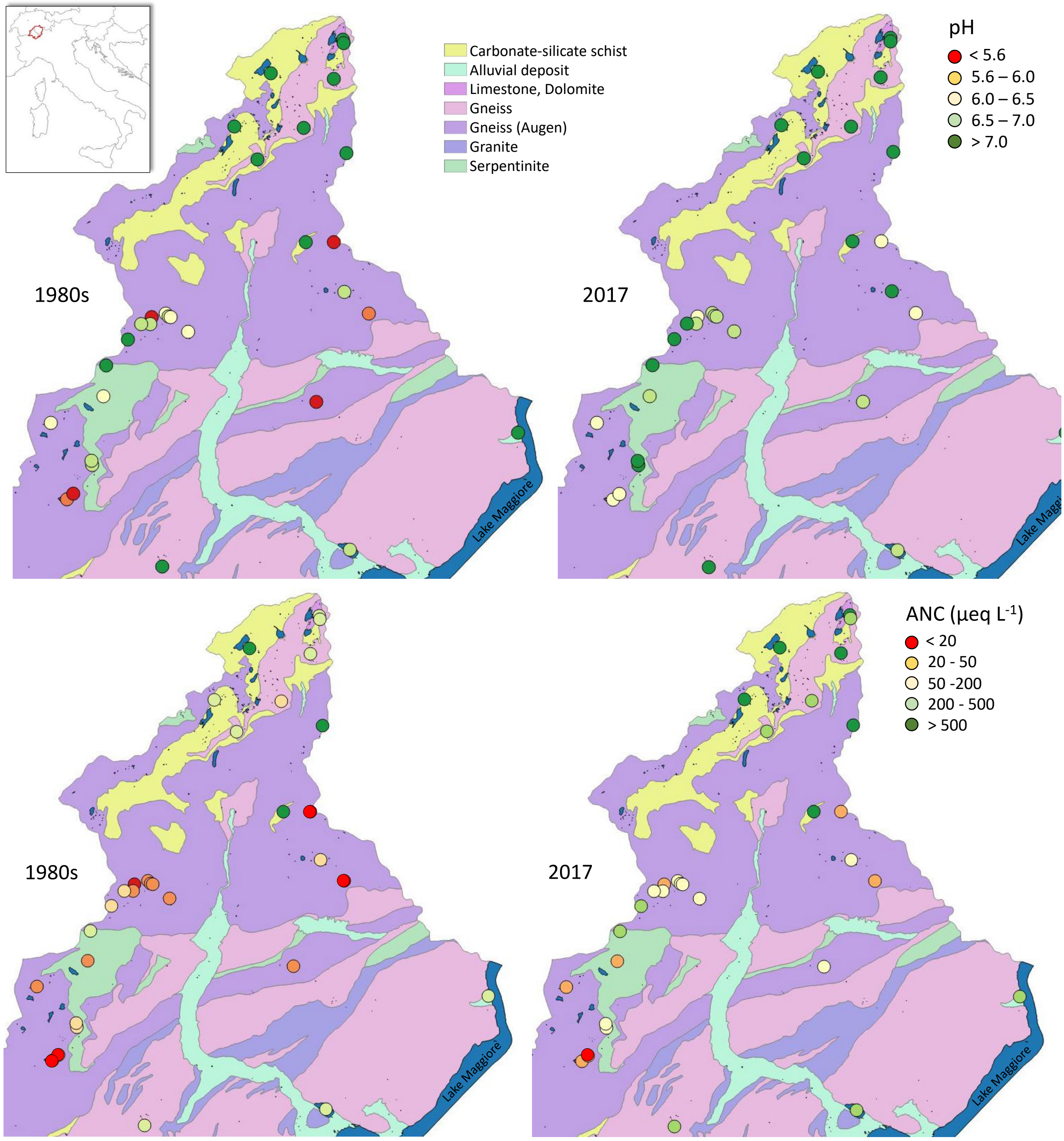


Figure 27 Comparison between the values of pH (above) and ANC (below) in the 1980s and in 2017 for 31 sites in the area of Lake Maggiore watershed, north-western Italy. The map also shows the main lithological features of the area.

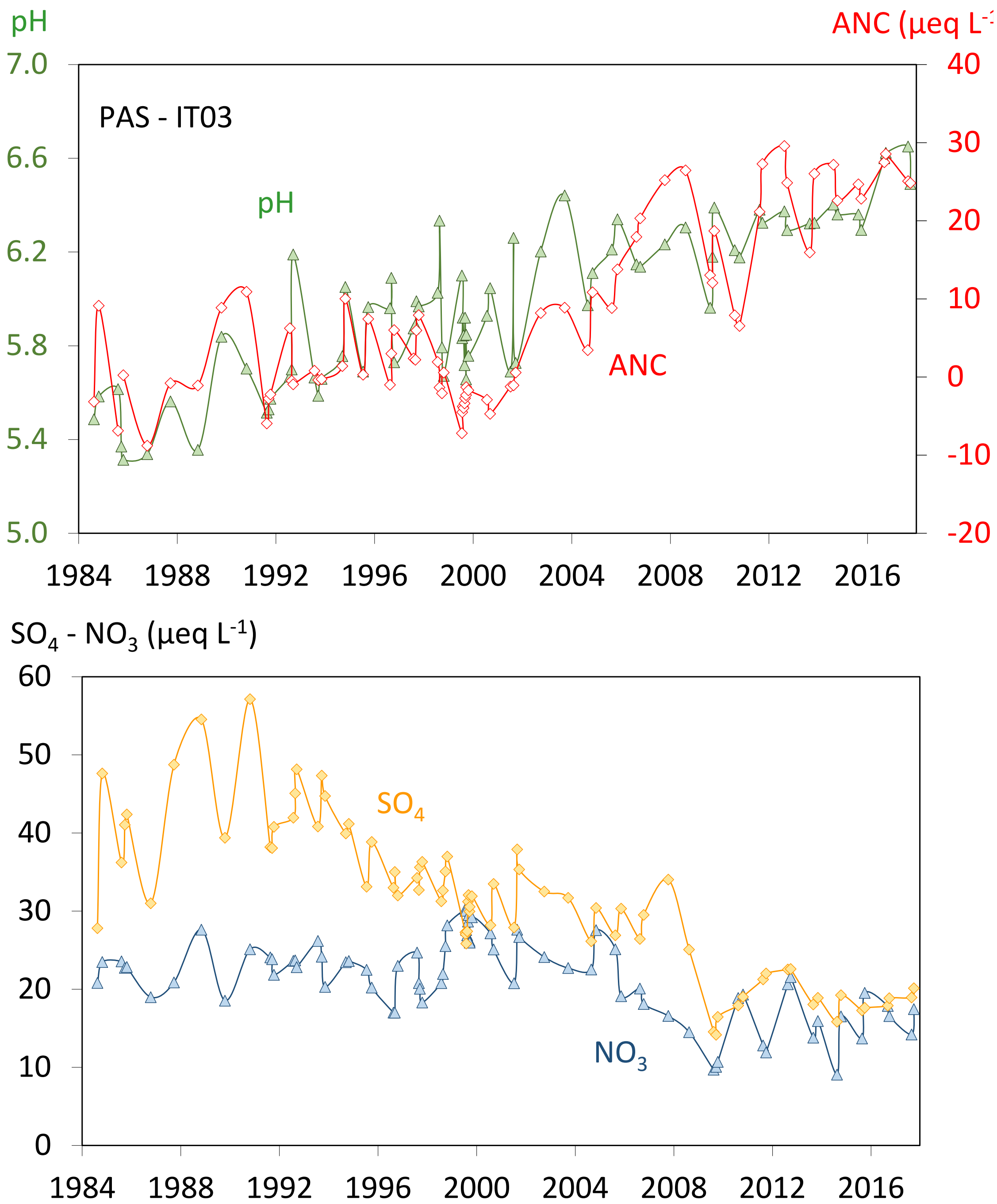


Figure 28 Long-term trends of pH, ANC (above), SO_4 and NO_3 (below) in the ICP Waters site Lake Paione Superiore (IT03).

4.6.4 Conclusion

The results of the acidification assessment in north-western Italy highlight the benefits of achieving emission reduction targets. Surface water response to decreasing acid deposition was widespread, even though some critical issues, such as the high amount of N deposition, still affect high altitude lakes and sensitive sites in general in the alpine and subalpine areas of Italy.

Furthermore, the analysis of long-term trends at the monitoring sites demonstrated that climatic factors interact with atmospheric deposition affecting the chemical changes in surface water. Some high-altitude lakes, for instance, showed an increasing trend of sulphate and base cations concentrations, despite the huge decrease in sulphate input from the atmosphere Rogora et al., 2016). Some climate-related effects have been suggested to explain these trends, including the decrease of snow cover duration and the degradation of permafrost due to climate warming (Rogora et al., 2013). The observed effects on lake chemistry are probably the result of several interplaying processes and drivers. Warmer air temperatures may enhance physical erosion of rocks and soils and the export of weathering products to lake water (Kopaček et al., 2016). Changes in precipitation may also regulate major element budgets and their mobilization from internal watershed sources (Mitchell et al., 2013). In addition, decrease of snow cover in space and time increases the exposure time of incoming precipitation with soluble rocks in lake watersheds, promoting higher solute transfer toward lakes (Rogora et al., 2013).

In the future, the recovery patterns, both from acidification and from N saturation, will be increasingly influenced by climatic drivers, also through indirect effects (hydrological changes, snow cover decrease, cryosphere thawing).

Acknowledgements: The National Research Council of Italy partly supported These studies, in the framework of a Convention with the Ministry for Environment, Land and Sea Protection of Italy for the management of the National Focal Centre of the ICP Waters. We would like to thank the staff of the chemical laboratory at CNR ISE and all the students and friends participating in sampling and field work.

4.6.5 References

Kopáček, J., Stuchlik, E., & Wright, R. F. 2005. Long-term trends and spatial variability in nitrate leaching from alpine catchment-lake ecosystems in the Tatra Mountains (Slovakia- Poland). Environmental pollution (Barking, Essex: 1987), 136(1), 89-101.

Marchetto, A., R. Mosello, R. Psenner, A. Barbieri, G. Bendetta, D. Tait, And G. A. Tartari. 1994. Evaluation of the level of acidification and the critical loads for Alpine lakes Ambio. 23: 150-154.

Marchetto, A., R. Mosello, M. Rogora, M. Manca, A. Boggero, G. Morabito, S. Musazzi, G.A. Tartari, A.M. Nocentini, A. Pugnetti, R. Bettinetti, P. Panzani, M. Armiraglio, P. Cammarano & A. Lami. 2004. The chemical and biological response of two remote mountain lakes in the Southern Central Alps (Italy) to twenty years of changing physical and chemical climate. J. Limnol. 63: 77-89.

Mitchell, M. J., Driscoll, C. T., McHale, P. J., Roy, K. M., & Dong, Z. 2013. Lake/watershed sulfur budgets and their response to decreases in atmospheric sulfur deposition: watershed and climate controls. Hydrol. Process., 27, 710–720.

Rogora, M., A. Marchetto & R. Mosello. 2001. Trends in the chemistry of atmospheric deposition and surface waters in the Lago Maggiore watershed. Hydrol. Earth System Sci., 5: 379-390.

Rogora, M., S. Arisci, A. Marchetto. 2012. The role of nitrogen deposition in the recent nitrate decline in lakes and rivers in Northern Italy. Science of the Total Environment 417-418C: 219-228.

Rogora M., L. Colombo, F. Lepori, A. Marchetto, S. Steingruber & O. Tornimbeni. 2013. Thirty years of chemical changes in alpine acid-sensitive lakes in the Alps. Water Air Soil Pollut. 224:1746.

Rogora M., Colombo L., Marchetto A., Mosello R., Steingruber S. 2016. Temporal and spatial patterns in the chemistry of wet deposition in Southern Alps. Atmos. Environ. 146: 44-54.

4.7 Latvia

Marina Čičendajeva, Marina Frolova
Latvian Environment, Geology and Meteorology Centre

4.7.1 Acid sensitivity

It is commonly known that the territory of Latvia is not acid-sensitive, because of predominantly calcareous soils with high initial concentrations of sulphate, calcium and magnesium. River and lake water bodies monitored under the WFD in Latvia show high pH values (there is a very low proportion of values below pH 7, and almost no observations of pH < 6), as well as high values of acid neutralising capacity (obtained from calculations). Nevertheless, monitoring of precipitation shows that there is a higher proportion of weakly acidic precipitation in the south-west and north-east regions of Latvia (Figure 29).

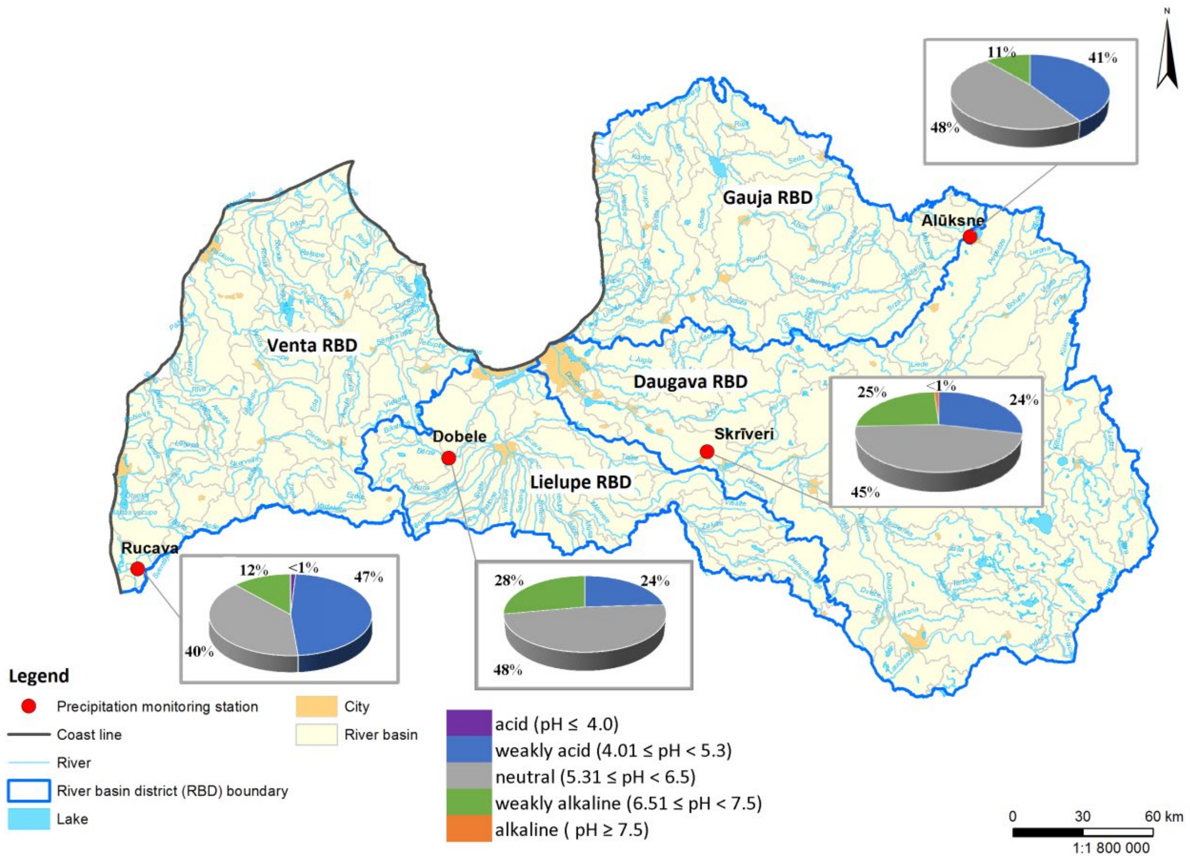


Figure 29 Distribution of precipitation samples with pH data for the period 2013-2016 to different acidity classes at Latvian precipitation stations

4.7.2 Monitoring and assessment approach

There is no specific monitoring designed to assess the level of acidification of surface waters, taking into account that generally the territory of Latvia is not acid-sensitive. Data obtained within the regular monitoring under the WFD were used to estimate if there are regional differences present in the characteristics of surface waters.

Data from 240 river and lake monitoring stations under the WFD were used, covering the time period from year 2013 to 2016. Stations are distributed more or less evenly across the country. Analysed

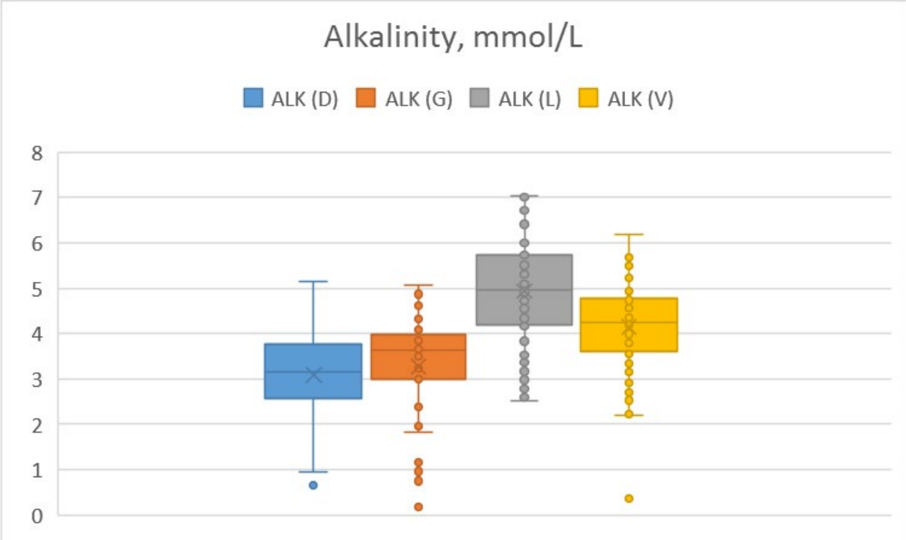


Figure 30 Box-whisker plots of alkalinity values in the river basin districts of Latvia. (D = Daugava RBD; G = Gauja RBD; L = Lielupe RBD; V = Venta RBD)

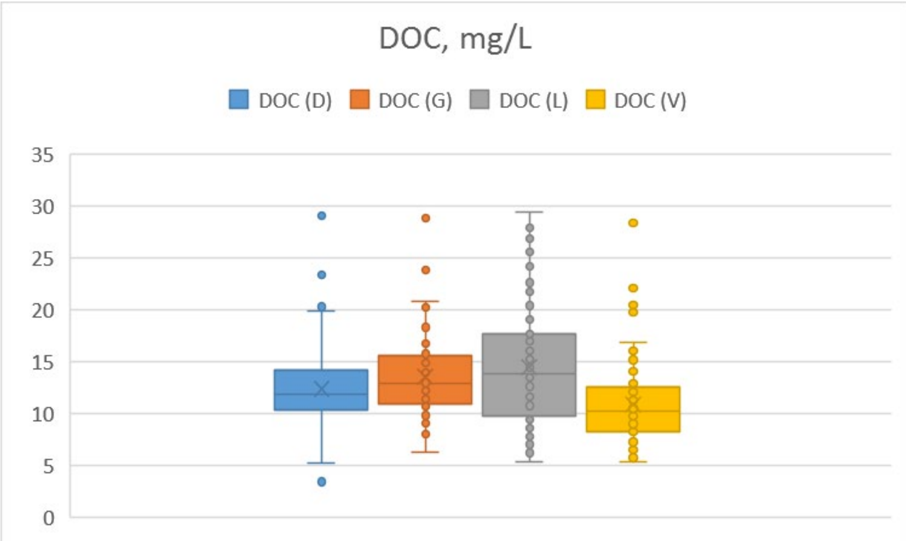


Figure 31 Box-whisker plots of DOC values in the river basin districts of Latvia

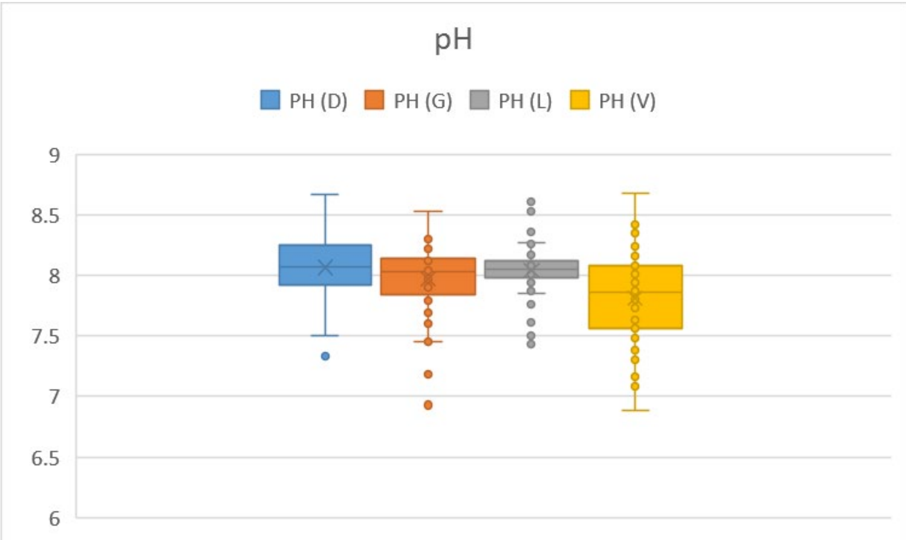


Figure 32 Box-whisker plots of pH values in the river basin districts of Latvia

data include pH, dissolved organic carbon (DOC) and alkalinity. For every monitoring station, *median values* of available data on pH, DOC, and alkalinity for the time period 2013 – 2016 were calculated. The available data amount varies from 4 seasonal samples (in ca. 80 stations with lowest monitoring intensity) up to 30 – 45 seasonal samples (in 19 intensive monitoring stations – mostly HELCOM and ICP-Waters stations).

4.7.3 Acidification status

There are some regional differences observed when comparing pH, DOC, and alkalinity values in 4 river basin districts in Latvia (Figure 30-Figure 32).

The Venta river basin district located in the western part of Latvia shows moderate to high alkalinity and low DOC, compared to the other RBDs. Nevertheless, observed pH values are in many cases lower than in the other RBDs, although numerically these pH values are above pH 7. Relatively low pH values (between pH 7 and 7.5) in the Venta RBD are observed in different water body types – not only in the water bodies with an impact of humic compounds in their catchment. Regular monitoring within the framework of the WFD will keep track of future changes.

Additionally, an international study based on the year 2000-2002 integrated monitoring data (Holmberg et al., 2013) has shown that ANC values for Latvian integrated monitoring sites exceed 2000 µeq/l and critical acidification loads are not exceeded. ANC was calculated as (Ca^{2+} + Mg^{2+} + Na^+ + K^+) – (SO_4^{2-} + Cl^- + NO_3^-). In the frame of this study, surface waters with ANC > 200 µeq/l were considered to be insensitive to acidification.

A study based on modelling of critical loads to Latvian forest ecosystems (Steinberga, 2011) has shown that overall critical loads for acidification have not been exceeded. Higher potential for acidification exists in the eastern part of Latvia.

4.7.4 References

Holmberg, M., Vuorenmaa, J., Posch, M., Forsius, M., Lundin, L., Kleemola, S., Augustaitis, A., Beudert, B., de Wit, H.A., Dirnböck, T., Evans, C.D., Frey, J., Grandin, U., Indriksone, I., Kram, P., Pompei, E., Schulte-Bisping, H., Srybny, A. and Vana, M., 2013. Relationship between critical load exceedances and empirical impact indicators at Integrated Monitoring sites across Europe. Ecological Indicators 24: 256-265.

Steinberga, I., 2011. Modelling and Mapping of Critical Loads of Acidification and Eutrophication on Forest Ecosystems in Latvia. Scientific Journal of Riga Technical University Vol. 7 (2011) 106-112.

4.8 Netherlands[21]

Herman van Dam[1], Adrienne Mertens[2]
[1] Consultancy for Water and Nature
[2] Diatomella

4.8.1 Acid sensitivity

In the originally poorly-buffered sandy regions in the eastern and southern part of the country there are several thousands of vulnerable, small, shallow and poorly buffered lakes (moorland pools), with a typical size between 0.5 and 5 ha and a depth between 1 and 3 m (Figure 33, left). They are close to industrial sources of air pollution and to areas with high emissions of ammonia from intensive cattle breeding (Figure 33, right). In the Natura 2000 European network they are classified as natural dystrophic lakes and ponds or oligotrophic waters of sandy plains, containing very few minerals.

Many of these soft water pools ('vennen') are included in nature reserves. After brief surveys it appeared that in some well-studied lakes a vast array of acid-sensitive species of algae and water plants had been replaced by a small number of acid-resistant species between 1920 and 1976 (Coesel *et al*. 1978, Van Dam & Kooyman-van Blokland 1978, Roelofs 1983).

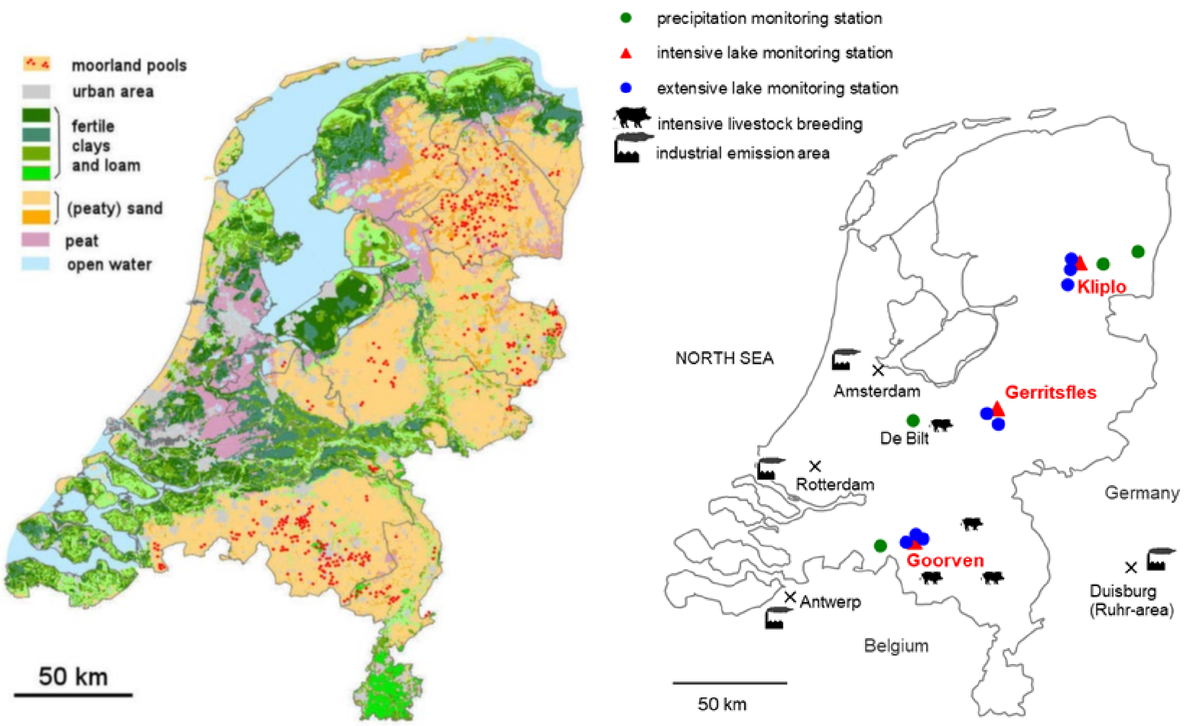


Figure 33 Left: Moorland pools are widely distributed on poorly-buffered sandy soils. Right: Location of precipitation and pool monitoring stations.

4.8.2 Monitoring and assessment

Regular sampling of eleven isolated pools throughout the country was started in 1978 (Van Dam & Mertens 2015). The pools are in protected areas and are exclusively fed by rainwater or some very local groundwater and have relatively minor disturbance from human activities, although Scots pine was planted or colonised spontaneously in the original open landscape of heathlands and sand dunes around the lakes in the last century (Figure 34, left).

[21] This country report is an abridged and updated version of the paper by Van Dam & Mertens (2013).

The monitoring focusses on chemistry (pH, major ions, nutrients) and diatoms (microscopic algae). Three pools are monitored intensively (at least four chemical and two diatom samples each year) and eight pools are monitored extensively (at least one chemical and one diatom sample in early autumn of every fourth year).

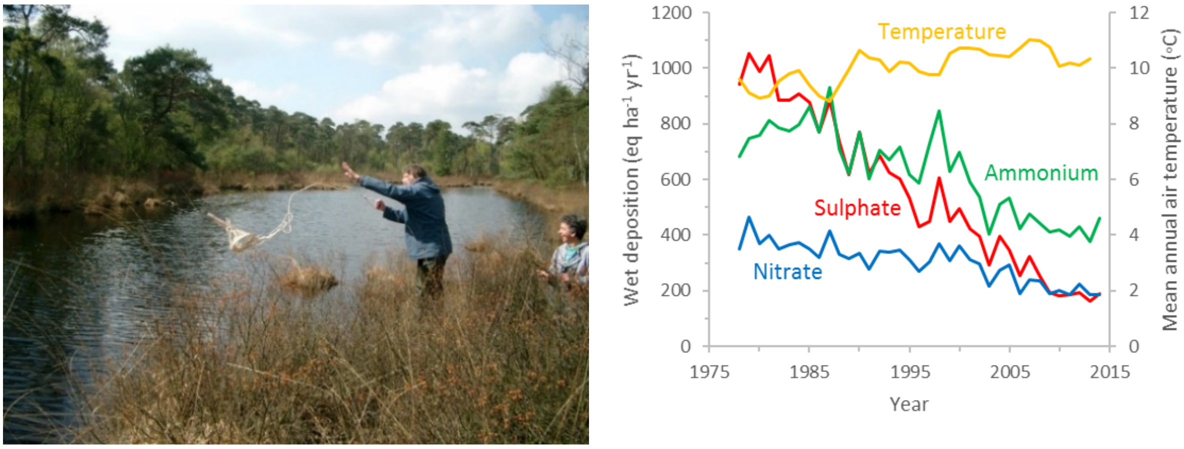


Figure 34 Left: Sampling diatoms in Goorven in 2004 (left; photo: Martijn Bellemakers). Right: Mean annual wet deposition at precipitation monitoring stations (data: www.lml.rivm.nl/gevalideerd) and three-year running means of temperature in De Bilt (data: www.KNMI.nl), see Figure 33 (right).

4.8.3 Atmospheric deposition and temperature

There are no exact data on total (potential) acid deposition on Dutch soft water pools. Wet deposition seems to be the best estimate, with an underestimation of probably 30 – 50% for sulphur compounds and 20 – 30% for nitrogen compounds (Arts *et al.* 2002). Over the period 1978 – 2014 the wet deposition of nitrate and ammonium has decreased significantly by about 50% and that of sulphate even by 82% ($p < 0.001$; Figure 34, right).

Current total nitrogen deposition on the lakes, based on these data, is about 650 eq/ha/yr, while the critical load for poorly-buffered lakes in The Netherlands is between 400 and 600 eq/ha/yr (Arts *et al.* 2002; Van Dobben *et al*. 2015). The current sulphur deposition is about 200 eq/ha/yr and well below the critical load of 800 eq/ha/yr (Arts *et al.* 2002).

The average air temperature in The Netherlands rose significantly by two degrees centigrade between 1978 and 2010 ($p < 0.001$; Figure 34, right). Consequently, the water temperature of the pools has increased by the same amount (Van Dam & Mertens 2008).

4.8.4 Chemical recovery

The median concentration of sulphate in water decreased from 230 mmol/m^3 in 1978 to 26 mmol/m^3 in 2014 (Figure 35). The concentrations in 1978 were excessively high, due to oxidation of accumulated airborne reduced sulphur in the sediments, which was oxidized when the bottom of most pools desiccated after the extremely dry year 1976 (Van Dam 1988). Gradually sulphate decreased and after other dry periods (e.g. 1996 – 1997) the increase was much smaller than after 1976. During periods of elevated sulphate concentrations after dry years sulphur eventually was lost from the lake system by discharge to the groundwater. The average decrease of sulphate (corrected for a change of chloride) in the shallow Dutch lakes between 1978 and 2002 amounts to 6.0 mmol/m^3/yr, which is considerably higher than in the deeper lakes of other European countries (Skjelkvåle *et al.* 2005). This is probably due to the much larger impact of sediment associated sulphate reduction in the shallow pools. Moreover, these primarily rain-fed pools receive only very minor amounts of sulphate stored in the catchment soils.

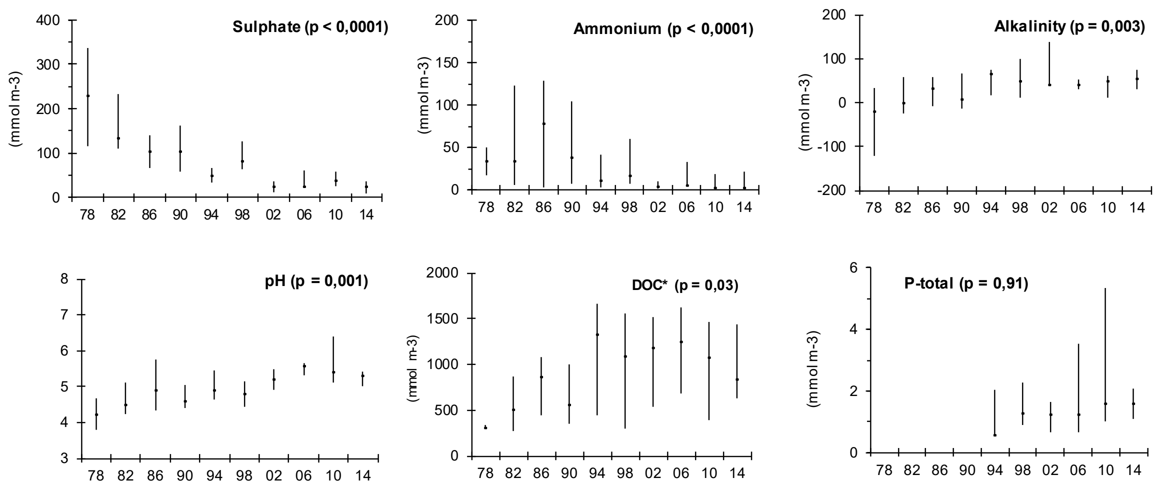


Figure 35 Changes in median values and 25th and 75th percentiles of selected chemical variables in the surface water of 11 isolated moorland pools between 1978 and 2014 (* = 8 pools only).

Ammonium peaked (as in the deposition) in 1986 to a median value of 80 mmol/m^3 and decreased later to values of 4 mmol/m^3 (Figure 35). The rate of the decrease between 1986 and 2002 (corrected for chloride changes) amounts to 2.4 mmol/m^3/yr. Nitrate concentrations were usually below the detection limit of 4 mmol/m^3. In other European lakes nitrogen is mainly present as nitrate, which has decreased in Central Europe by about 1 mmol/m^3/yr, while nearly no changes or even increases were found in the Alps, Scandinavia and the United Kingdom (Skjelkvåle *et al.* 2005).

The median pH increased from 4.3 in 1978 to 5.3 in 2010, along with the alkalinity. After correction for chloride changes the rate of the decrease of the proton concentration is 0.7 mmol/m^3/yr, while much lower rates were recorded from the rest of Europe (Skjelkvåle *et al.* 2005). The chemistry of the pools has changed considerably, much more than in other European countries. This is not only a consequence of the policy-mediated decrease of potential acid atmospheric deposition, but also due to the strong impact of sediment-related processes of sulphate reduction and denitrification, which are strongly enhanced by increased ambient temperatures, as have occurred over recent decades (Feijtel et al. 1989, Holmer & Storkholm 2001, Veraart et al. 2011).

4.8.5 Internal eutrophication by acidification and climatic warming

Due to the increased breakdown of organic material by bacterial processes phosphorus release from the sediments, which promotes eutrophication, has increased due to rising temperature and pH (Van Kleef *et al.* 2010). Maximal total phosphorus concentrations seem to have increased in some pools (Figure 35).

Internal eutrophication is particularly evident in Kliplo, a small, intensively-monitored pool that has not been acidified as seriously as the other investigated pools. Because the shores are comparatively steep and the sediments never run dry, a huge amount of organically-bound sulphur, derived from atmospheric deposition, has accumulated in the sediments over the years, due to bacterial sulphate reduction (Marnette & Stein 1993). Sulphides were bound to iron, and phosphorus, which was previously bound to iron, was released (Smolders *et al.*, Van Kleef *et al.* 2010).

Monthly measurements indicate that nutrients were released abruptly in 2008. This is not reflected by an increase of total phosphorus, but small green algae have proliferated since then in the plankton, as indicated by the high chlorophyll concentrations, the nearly permanent green colour of the water and high oxygen consumption by the algae in the dark (BOD) (Figure 36, left). The mean pH, which was between 4.6 and 6.1 between 1978 and 2005 increased to 7.1 – 7.2 in later years, with maximum values up to pH 8.7 in summer, due to elevated photosynthesis. Such high values have also recently been measured in some of the extensively sampled lakes. The changes may have been triggered by the high temperatures in 2006 – 2007, which were two of the warmest years since the onset of the measurements in 1706 (www.knmi.nl). Signs of internal eutrophication are also recorded in a set of 68 shallow soft water lakes from the south of The Netherlands (Van Kleef *et al*. 2010).

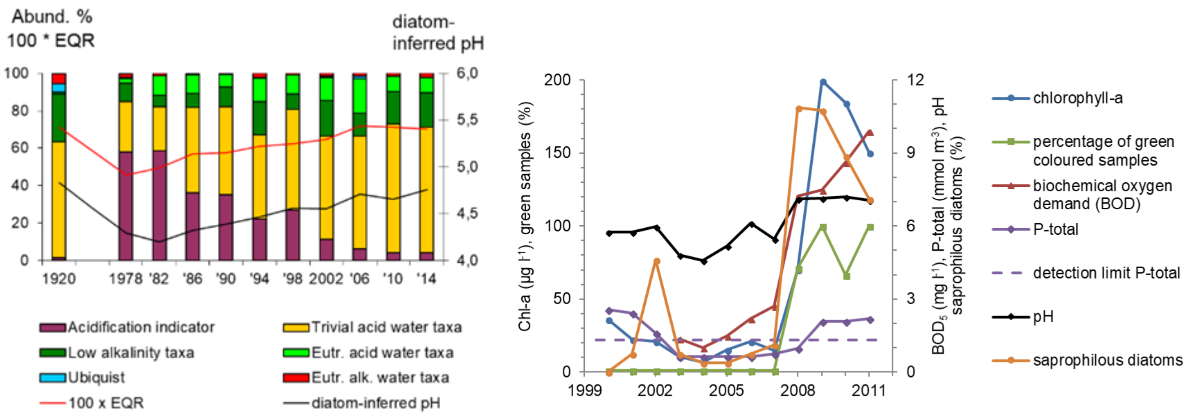


Figure 36 Left: Changes of annual monthly values of selected biological and chemical data from the pool Kliplo (data from Waterschap Drents-Overijsselse Delta). Right: Changes of mean percentage abundance of ecological groups of diatoms, diatom-inferred pH and Ecological Quality Ratio (EQR) in 11 isolated moorland pools. Eutr. = eutraphentic, alk. = alkaline.

4.8.6 Diatoms

For all pools diatom samples collected around 1920 – preserved in the collection of Naturalis (Leiden) – were studied as a reference. Both the historical and modern samples were collected in the same way (Figure 34, left) and consistent taxonomy was applied over all the years. In each sample 400 diatoms were identified and the relative contribution of each species was calculated, along with diatom-inferred pH-values (Ter Braak & Van Dam 1989) and ecological quality ratios (EQR), as required for the European Water Framework Directive (Van der Molen *et al.* 2012).

In total 145 diatom species and varieties were recorded in 141 samples. The species have been split in six ecological groups, listed in the legend of Figure 36 (right). In the samples from about 1920 the majority of the diatoms were common acid water species and also low alkalinity species were well represented. In the latter group often rare species from (moderately) nutrient poor habitats were present. In 1978 and 1982 most lakes were dominated by the diatom *Eunotia exigua,* a well-known indicator of acidified lakes and streams. From 1986 onwards the common acid water species gradually increased. The low alkalinity taxa were partly replaced by a group of species which are characteristic for acid, but more or less eutrophic water.

Thus, although diatom-inferred pH-values and the ecological quality ratios almost returned to historical values, the changed species composition of the diatom assemblages indicates partial recovery from acidification. The lakes have become less acid over the last few decades, but more eutrophic than in earlier times.

4.8.7 Conclusions

Due to national and international policy measures (LRTAP Convention) the atmospheric deposition of nitrogen and sulphur compounds to Dutch acid sensitive moorland pools over the last four decades has decreased by about 50 and 90%, respectively. Simultaneously the concentrations of sulphate and ammonium in the lakes decreased substantially, much faster than in deep lakes in other European countries, due to sediment-related processes of sulphate reduction and denitrification.

The increased decomposition of the sediment, also enhanced by climatic change, has caused internal eutrophication, particularly in the lakes with steep shores, where the sediments do not desiccate in dry years. The diatoms indicate the recovery from acidification since the 1980s, but the assemblages did not return to the reference state, due to internal eutrophication. Compounds of nitrogen and sulphur were stored in the sediments, as a legacy of excessive atmospheric deposition in the past.

Careful removal of accumulated organic matter might prevent further eutrophication. However, this is not a sustainable measure, as long as the atmospheric deposition remains above the critical load. The predicted reduction of the atmospheric nitrogen deposition by 15 – 20% until 2030 (Velders *et al* 2017), will not be sufficient for the sustainable preservation of soft water moorland pool ecosystems.

The regular monitoring of acid sensitive moorland pools in The Netherlands was state funded in the period 1978 and the results were regularly transferred to ICP Waters until 1995, when state funding was cut. From that year onwards, the measurements were funded year-to-year by regional water authorities, provincial governments, private foundations and the authors. As there is no long-term funding, the continuation of the measurements and transfer of the results to ICP Waters is very uncertain.

Acknowledgements: The authors are grateful to the proprietors of the study sites (Staatsbosbeheer, De Hoge Veluwe, Ministry of Defence, Natuurmonumenten, Brabants Landschap), the Province of Drenthe, the water authorities Drents-Overijsselse Delta, Vallei & Veluwe and De Dommel and the laboratories Aqualysis, Aquon and Waterproef for their cooperation.

4.8.8 References

Arts, G.H.P., H. van Dam, F.G. Wortelboer, P.W.M. van Beers & J.D.M. Belgers (2002): De toestand van het Nederlandse ven. Alterra, Wageningen / AquaSense, Amsterdam / RIVM, Bilthoven. 123p.

Coesel, P.F.M., R. Kwakkestein & A. Verschoor (1978): Oligotrophication and eutrophication tendencies in some Dutch moorland pools, as reflected in their desmid flora. Hydrobiologia 61: 21-31.

Feijtel, T.C., Y. Salingar, C.A. Hordijk, J.P.R.A. Sweerts, N. van Breemen & T.E. Cappenberg (1989): Sulfur cycling in a Dutch moorland pool under elevated atmospheric S-deposition. Water, Air, and Soil Pollution 44: 215-234.

Holmer, M., P. Storkholm (2001): Sulphate reduction and sulphur cycling in lake sediments: a review. Freshwater Biology 46: 431-451.

Marnette, E.C.L. & A. Stein (1993): Spatial variability of chemical compounds related to S-cycling in two moorland pools. Water Research 27: 1003-1012.

Roelofs, J.G.M.(1983): Impact of acidification and eutrophication on macrophyte communities in soft waters in The Netherlands. I. Field observations. Aquatic Botany 17: 139-155.

Skjelkvåle, B.L., J. Stoddard, D.S. Jeffries, K. Torseth, T. Hogåsen, J. Bowman, J., Mannio, D.T. Monteith, R. Mosello, M. Rogora, D. Rzychon, J. Vesely, J. Wieting, A. Wilander & A. Worsztynowicz (2005): Regional scale evidence for improvements in surface water chemistry 1990-2001. Environmental Pollution 137: 165-176.

Smolders, A.J.P., L.P.M. Lamers, E.C.H.E.T. Lucassen, G. van der Velde & J.G.M. Roelofs (2006): Internal eutrophication: how it works and what to do about it - a review. Chemistry and Ecology 22: 93-111

Ter Braak, C.J.F. & H. van Dam (1989): Inferring pH from diatoms: a comparison of old and new calibration methods. Hydrobiologia 178: 209-223.Van Dam & H. & A. Mertens (2013): Partial recovery of shallow acid-sensitive lakes from acidification. Environmental Scientist 22(2): 36-40.

Van Dam, H. (1988): Acidification of three moorland pools in The Netherlands by acid precipitation and extreme drought periods over seven decades. Freshwater Biology 20: 157-176.
Van Dam, H. & H. Kooyman-van Blokland (1978): Man-made changes in some Dutch moorland pools, as reflected by historical and recent data about diatoms and macrophytes. Internationale Revue der gesamten Hydrobiologie 63: 587-607.
Van Dam, H. & A. Mertens (2008): Vennen minder zuur maar warmer. H_2O 41 (12): 36-39.
Van Dam, H. & A. Mertens (2015): Monitoring herstel verzuring en klimaatverandering vennen 1978-2014: temperatuur, hydrologie, chemie, kiezelwieren [Monitoring recovery of acidification and effects of climate change in shallow soft water lakes in The Netherlands: Temperature, hydrology, chemistry, diatoms]. Rapport 1303. Herman van Dam, Adviseur Water en Natuur, Amsterdam. 123p.
Van der Molen, D.T., R. Pot, C.H.M. Evers & L. van Nieuwerburgh (Eds) (2012): Referenties en maatlatten voor natuurlijke watertypen voor de Kaderrichtlijn Water 2015-2021. STOWA, Amersfoort. 378p.
Van Dobben, H.F., A. van Hinsberg, D. Bal, J.P. Mol-Dijkstra, H.J.J. Wieggers, J. Kros & W. de Vries (2015): Derivation of critical loads of nitrogen for habitat types and their exceedances in The Netherlands. In: W. de Vries, J.-P. Hettelingh & M. Posch (Eds) Critical loads and dynamic risk assessments: Nitrogen, acidity and metals in terrestrial and aquatic ecosystems. Springer, Dordrecht. p. 547-572.
Van Kleef, H.H., E. Brouwer, R.S.E.W. Leuven, H. van Dam, A. de Vries-Brock, G. van der Velde & H. Esselink (2010): Effects of reduced nitrogen and sulphur deposition on the water chemistry of moorland pools. Environmental Pollution 158: 2679-2685.
Velders, G.J.M., J.M.M. Aben, G.P. Geilenkirchen, H.A. den Hollander, L.E. Nguyen, E. van der Swaluw, W.J. de Vries & R.J. Wichink Kruit (2017): Grootschalige concentratie- en depositiekaarten Nederland. Rapportage 2017. Rapport 2017-0117. RIVM, Bilthoven. 57p.
Veraart, A.J., J.J.M. de Klein & M. Scheffer (2011): Warming can boost denitrification disproportionately due to altered oxygen dynamics. PLoS ONE 6(3): e18508.

4.9 Norway

Øyvind Garmo[1], Espen Lund[1], Kari Austnes[1], Brit Lisa Skjelkvåle[2]
[1] Norwegian Institute for Water Research
[2] University of Oslo

4.9.1 Acid sensitivity

Much of southern Norway is characterized by resistant types of bedrock that weather slowly and are relatively poor neutralizers of acid (Figure 37). In large areas the bedrock is either bare or covered only by thin layers of unconsolidated glacial till and soil. This geology in combination with the steep terrain and high runoff found in the mountainous western parts, produces dilute surface waters prone to acidification. The forested south-eastern part of Norway is flatter and receives less precipitation than the western parts, producing surface waters with relatively high concentrations of natural organic acids that can give low pH even without high deposition of anthropogenic acid. Sedimentary and metamorphic bedrock, some of it calcareous, dominates in the middle parts of Norway and gives relatively low acid sensitivity, whereas the north-east is characterized by more resistant sandstone and gneiss.

The critical loads map (Figure 37) corresponds fairly well with the acid sensitivity predicted from bedrock, reflecting the high sensitivity found in southernmost Norway and in the western part of South Norway. However, the high sensitivity north and south-east of Oslo predicted by the geological map is not reflected in the critical loads map. This points to the uncertainty related to assessing acid sensitivity purely from the bedrock.

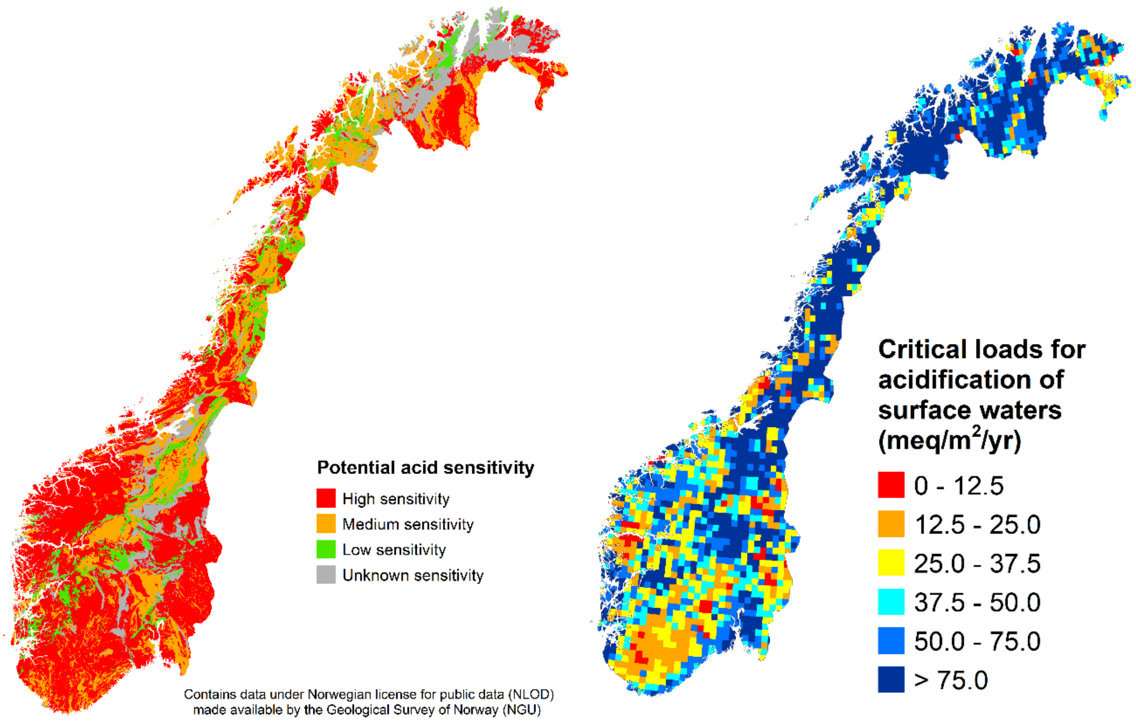


Figure 37 Left: Potential acid sensitivity derived from a map of bedrock types (Norwegian Geological Survey) and their expected relative weathering rates. Bedrock type is not the only determinand of acid sensitivity, but the map is an indication of where sensitive areas can be found. The sensitivity categories were assigned by geologists at the University of Oslo. Right: Critical loads for acidification derived from water chemistry and discharge, using the Steady-State Water Chemistry model (Henriksen and Posch, 2001).

4.9.2 Monitoring and assessment approach

The regular Norwegian monitoring for assessment of acidification now comprises 94 lakes that are sampled 1-4 times annually (Garmo and Skancke, 2017). Of these, 18 are subject to annual monitoring of biological indicators of acidification. The hydrochemistry of most of the 94 lakes have been monitored since 1986 when they were part of a national survey of 1000 acid-sensitive lakes (Lien et al., 1987). These 1000 lakes were headwater lakes, almost all with a lake area larger than 0.2 km^2. Their catchments were relatively undisturbed and situated on acid-sensitive bedrock. The 1000 lakes represented acid-sensitive waters and were not randomly selected and thus not meant to give the true distribution of Norwegian hydrochemistry at the time. A later survey based on a stratified random selection showed that a high fraction of the lakes in the areas that are marked as “high sensitivity” and “medium sensitivity” in Figure 37 were rather poor in base cations (Skjelkvåle et al., 1996). It can therefore be stated that the 94 lakes that remain in the monitoring programme, which are all located in these areas, give a good indication of the situation in the acid-sensitive regions. In the period 1995-2003 the 94 lakes were supplemented by an additional 100 lakes (selected according to similar criteria as in 1986), which were resampled in 2016. For the current exercise we have included data from this extended set of 200 lakes.

In addition, 6 calibrated catchments (of which 5 are ICP Waters sites), i.e. gauged streams or outlets of small lakes where water chemistry responds rapidly to changes in deposition, are sampled 26-52 times annually. These were also included. In recent years there have been few river sites in the regular programme although many are sampled frequently as part of the follow-up of river liming projects. We chose not to include these here. Instead we have included data from 47 reference rivers that are part of a programme that was started in 2017 to meet the requirements of the Water Framework Directive (WFD) for surveillance monitoring of rivers (Moe et al., 2018). These rivers are not necessarily acid sensitive, but they are distributed over a wide area and are expected to be unaffected by local point source pollution. Also, most of Norway is acid sensitive (cf. Figure 37). The lake and calibrated catchment data that were submitted for the overall analysis in this report are mean values for the period 2014-2016[22]. The river data represent arithmetic average of monthly samples in the period May-December 2017.

The official Norwegian criteria for defining a water body as acidified from unnatural causes (WFD classification) relies on a categorisation according to prehistoric calcium (alkalinity) and DOC (colour) levels. For each category a set of thresholds for the physicochemical parameters pH, ANC and labile/inorganic monomeric aluminium have been set that define the water body’s present state compared to its expected undisturbed reference condition. There are quite a few categories owing to the impact of base cations and humic acids on pH and ANC levels, especially in waters very poor in base cations. The submitted data are not sufficient to make a full assessment according to the mentioned criteria. Moreover, the sampling frequency of one sample per year during autumn turnover that applies to many of the lakes, is intended to document temporal trends over many years and not e.g. the seasonal variation in pH and labile aluminium that is important for understanding the effects on biota. The assessment of acidification status that is given in the next section is therefore based on ANC, which is not an intensity parameter like pH or labile aluminium. To estimate the size of the area that is not achieving “good status” because of acidification, we will rely on present day ANC, simulated with the MAGIC model, in the 1000 lakes that were sampled in 1995 (see above). The exact procedure for assigning the modelled chemistry of individual lakes to area of is described in detail in Austnes et al. (2016).

[22] First, an annual mean for each year was calculated as the arithmetic average. The mean value for the whole period was calculated as the arithmetic average of the annual means. For the approximately 100 lakes that were resampled in 2016 the value is based on just one sample.

4.9.3 Acidification status

The ANC, ANC_{oaa}, DOC and pH results from the Norwegian monitoring sites (Figure 38) reflect the natural conditions mentioned in the first section, as well as deposition of sulphur and nitrogen. Many of the sites have low ANC, ANC_{oaa} and pH, but there were only a few sites where the status according to the thresholds defined for ANC is now less than “good”, and they are almost exclusively found in the southernmost part of the country (Figure 39). If we had just considered the sites that were sampled at least seasonally and used the combined criteria for ANC, pH and labile aluminium, a somewhat larger fraction of sites would have been classified as less than good (not shown), but the spatial pattern would have been similar.

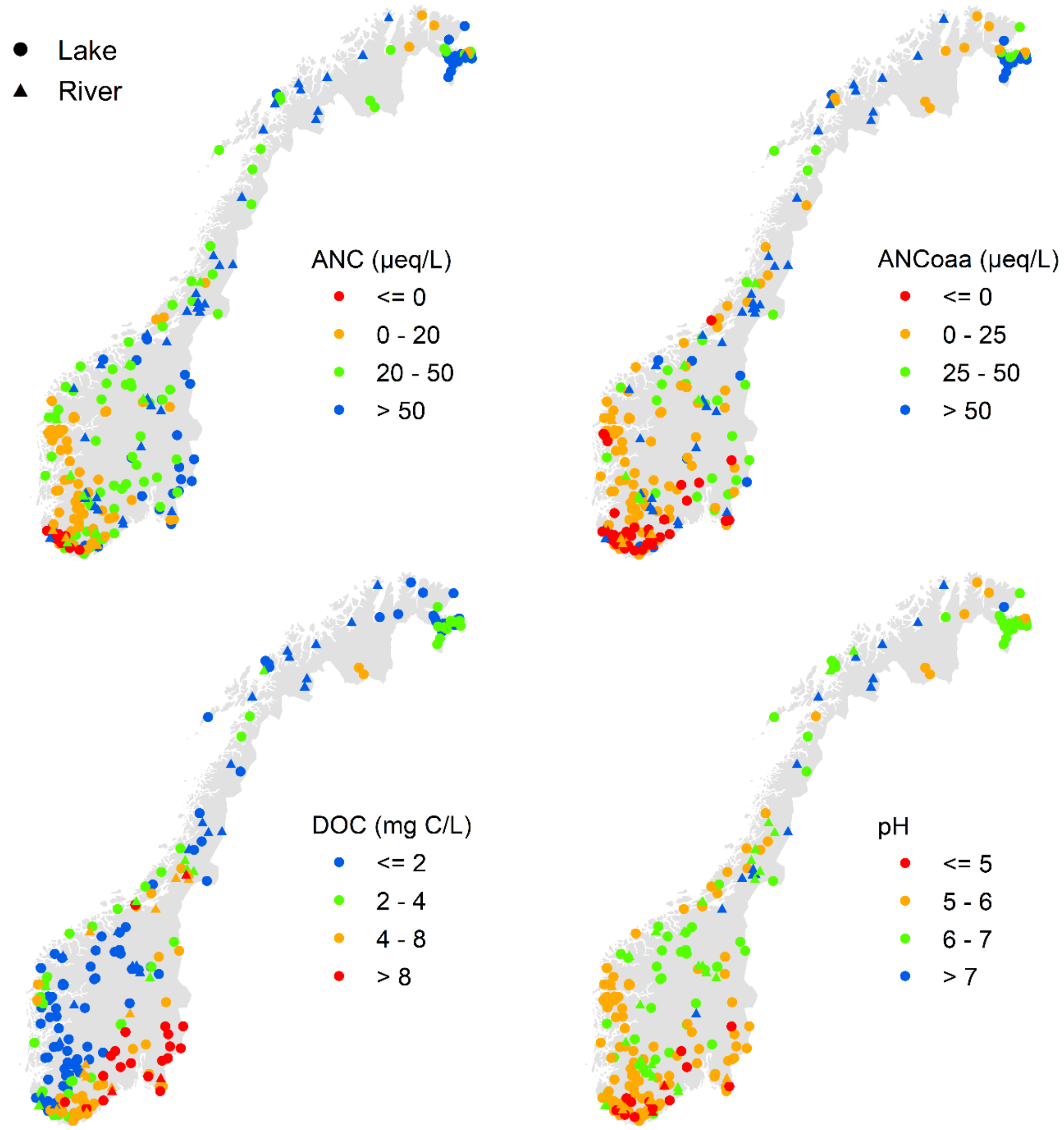


Figure 38 Mean values from selected Norwegian monitoring sites (see text) for the period 2014-2016.

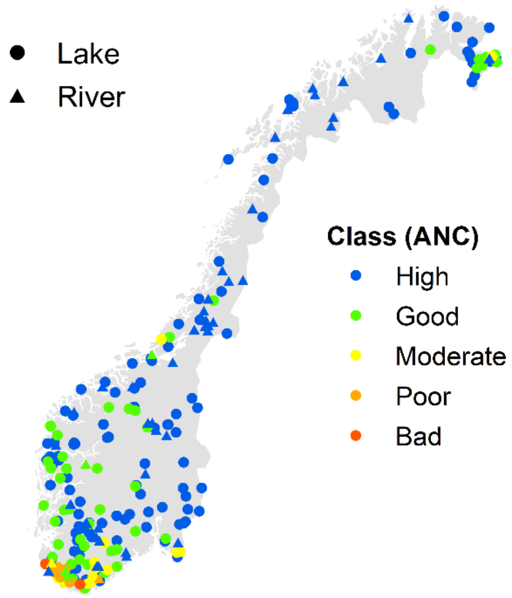


Figure 39 State of acidification according to Norwegian classification criteria and mean ANC values.

Dividing the country into 12*12 km^2 grid cells and using modelled results for lakes that were assigned to each cell, Austnes et al. (2016) found that lake waters in 7% of the country had not achieved "good" status or better by 2015 (Figure 40). The most likely scenario for future deposition indicates that this fraction will be reduced to 5% in 2033 (Figure 41).

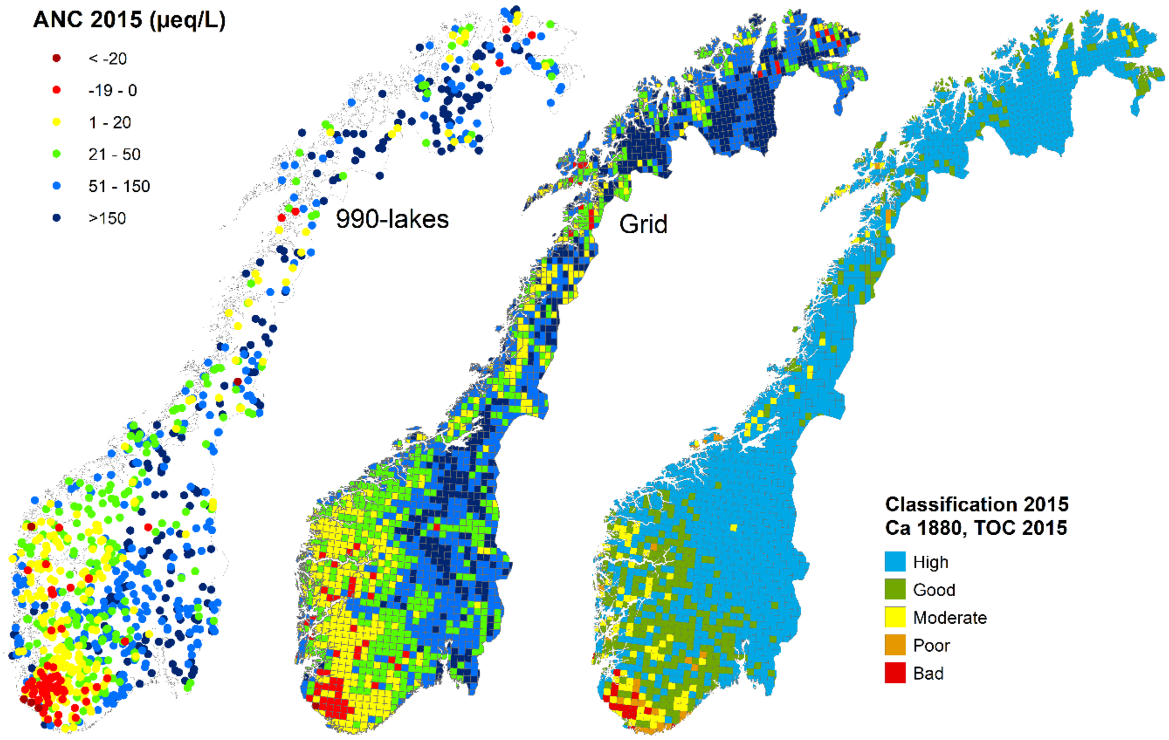


Figure 40 Acidification status in 2015 according to ANC thresholds as defined in the Norwegian implementation of the WFD (from Austnes et al., 2016).

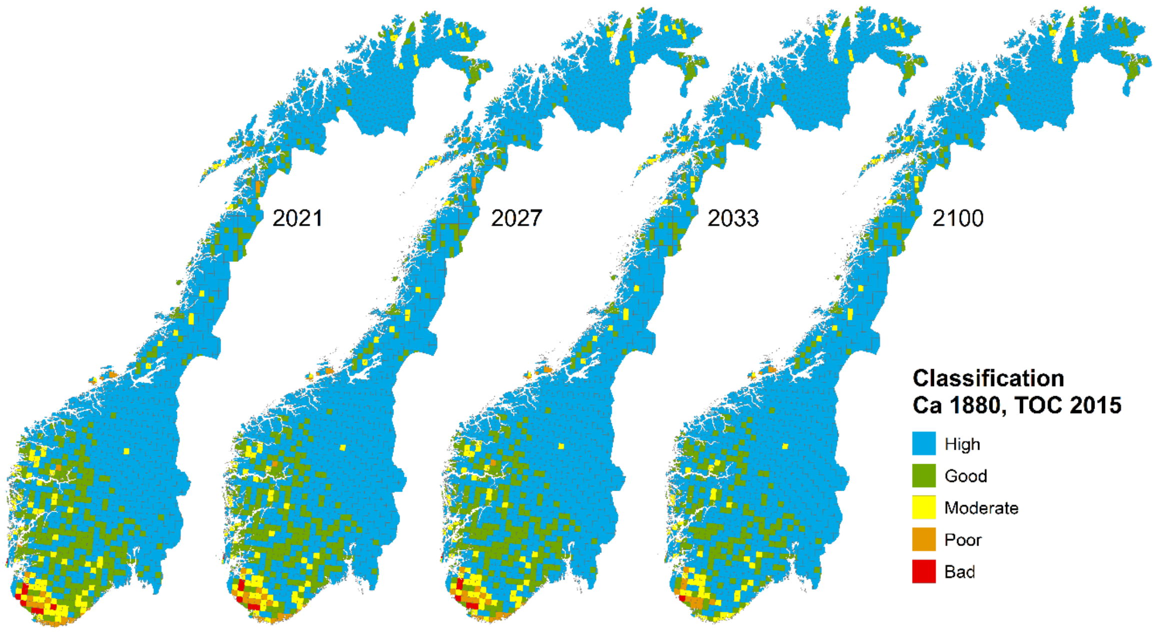


Figure 41 Future acidification status according to ANC thresholds defined in the Norwegian implementation of the WFD, and model results based on the most likely future deposition scenario (from Austnes et al., 2016).

Acknowledgement: The monitoring and modelling was funded by the Norwegian Environment Agency.

4.9.4 References

Austnes, K., Lund, E., Valinia, S. and Cosby, B.J. 2016. Model based classification of acidification status in lakes with no monitoring data. NIVA-report 7047-2016 (in Norwegian).

Garmo, Ø.A. and Skancke, L.B. 2017. Monitoring long-range transboundary air pollution. Water chemical effects 2017. Norwegian Environment Agency report M-836 (in Norwegian).

Henriksen, A. and Posch, M. 2001. Steady-state models for calculating critical loads of acidity for surface waters. Water, Air, and Soil Pollution Focus 1: 375–398.

Lien, L., Sevaldrud, T.S., Traaen, T. and Henriksen, A. 1987. 1000 lake survey 1986. Norwegian Pollution Control Authority report 282/87. SPFO-report TA-0624 (in Norwegian).

Moe, T.F., Thrane, J.E., Persson, J., Bækkelie, K.A., Myrvold, K.M., Olstad, K., Garmo, Ø.A., Grung, M. and de Wit, H. 2018. Surveillance monitoring of reference rivers in 2017. Norwegian Environment Agency report M-1002 (in Norwegian).

Skjelkvåle, B.L., Henriksen, A., Faafeng, B., Fjeld, E., Traaen, T., Lien, L., Lydersen, E. and Buan, A.K., 1996. Regional lake survey 1995. Norwegian Pollution Control Authority report 677/96. SPFO-report TA-1389 (in Norwegian).

4.10 Poland

Rafał Ulańczyk, Tomasz Pecka, Agnieszka Kolada, Krzysztof Skotak
Institute of Environmental Protection – National Research Institute

4.10.1 Acid sensitivity

In Poland, the acidification of surface waters caused by atmospheric deposition is not considered as a significant pressure at the national scale. Such status is reflected in both: the assessment of acid sensitivity and in the scope of state monitoring system (described in the next section). National scale assessments regarding the sensitivity to acid deposition in Poland were focused mainly on the Critical Load to soils calculated with the simple mass balance method (SMB) described in detail in the CLRTAP (2014) (Mill, 1995; Mill, 2007; Dróżdż et al., 2015). Due to the lack of national scale assessment of acid sensitivity of surface waters in Poland, a map of sensitive regions has been prepared, as a function of parent material of soil and based on the geological descriptors of surface water bodies according to the Water Framework Directive (WFD).

The first step in the sensitivity analysis was based mainly on the European Soil Database (ESDAC, 2006; Panagos et al., 2012), which includes geological data from national resources (Kondracki, 1978; Zawadzki et al., 1999). The 4-point scale of bedrock sensitivity was applied after McFee (1980), Shilts (1981) and Holowaychuk and Fessenden (1986), and it is as follow: 1) sensitive (less than 1% of the total area of Poland), 2) moderately sensitive (nearly 40% of the total area; dominating class in main lake districts located in northern Poland), 3) low sensitive (nearly 60% of the total area) and 4) insensitive (less than 2%). When assigning classes to particular regions of Poland, two sources of information in addition to the ESDAC database were of use: characteristic of soils and subsoils (Zawadzki, 1999; Brożek and Zwydak, 2003) and spatial distribution of soil types in Poland (Szostak, 1967; Lisicki, 1998; Gotkiewicz et al., 2004; Maruszczak, 2000; Kacprzak, 2003; Charzyński et al., 2007; Petelski, 2008; Wojciechowki, 2008; Sobolewski et al., 2014).

To some extent the typology of water bodies in accordance to the WFD reflects the acid sensitivity of bedrock, and therefore, in the second step of the acid sensitivity analysis the typology was used as an input. In case of the typology of lakes in Poland the geology was substituted by water quality parameters including concentration of calcium with 25 mg Ca/l as a threshold value (Hobot et al., 2014). In contrast to the majority of Polish lakes (greater than 50 ha) only 27 have the concentration of Ca less than 25 mg/l and they are characterized by relatively low alkalinity – less than 1.3 meq/l (Hobot et al., 2014; RBMP, 2016). For rivers (with catchment area greater than 10 km^2), the typology included a riverbed material which was divided into three groups according to the WFD's "system A" for characterisation of surface water body types, but it included also local characteristics (e.g. peat or alluvial soils) (Błachuta et al., 2005; Hobot et al., 2014).

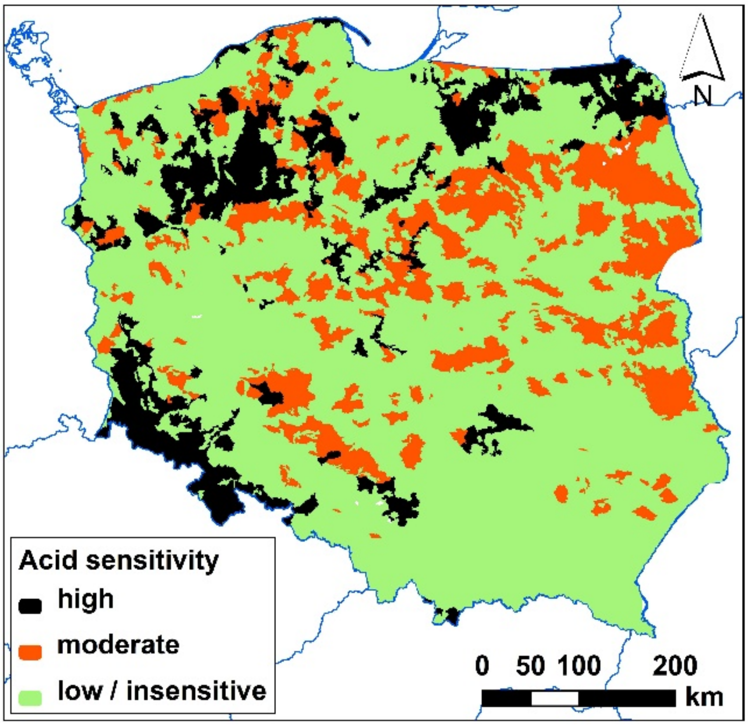


Figure 42 Final map of the acid sensitivity

4.10.2 The final map of acid sensitivity

Figure 42 is based on the combination of three datasets mentioned above: the bedrock and typologies of lakes and rivers in Poland. The final map includes three classes of acid sensitivity following Skjelkvåle and Wright (1990). High sensitivity is where the bedrock, or the body of surface water were assessed as highly sensitive. Additionally, this class includes the type "0" of surface water body if its catchment area was intersecting the high sensitive region. 12.5% of the country was classified as highly sensitive. Areas are considered as moderately sensitive if the surface water body and the bedrock of its catchment are both moderately sensitive. Moderately sensitive areas cover 17.3% of Poland. The other 70.2% of the area was classified as of low sensitivity or insensitive to the acidification.

4.10.3 Acidification monitoring in Poland

According to the "call for contribution", the water parameters of concern include acid neutralising capacity (ANC), ANC adjusted for organic acids (ANC_{oaa}) with pH and organic carbon as supporting parameters. Therefore, the description of data availability and the scope of monitoring in Poland refer to parameters used for the calculation of ANC, i.e. Ca^{2+}, Mg^{2+}, Na^{+}, K^{+}, NH_4^{+}, Cl^{-}, SO_4^{2-}, NO_3^{-}, alkalinity, pH, DOC and TOC.

The **state monitoring** of surface waters in Poland is based on a 6-year cycle according to the River Basin Management Plans (RBMP, 2016) and to the WFD. Monitoring campaigns are conducted in 6-year cycles (surveillance monitoring and monitoring of protected areas), twice if the water body is at risk of failure to achieve environmental objectives (operational monitoring) or every year if the body is a reference monitoring point or intensive monitoring point. In all cases the monitoring campaign is carried out during the full year (SEM, 2015). Since 2008, the scope of monitored parameters and the frequency of sampling is different for lakes and rivers, and for natural and artificial/modified bodies of water (Journal of Laws 2008, 2009, 2011, 2016). By 2008 the scope of monitored parameters (referring to the parameters of concern) was the same for all surface waters but varied in time (Journal of Laws 1962, 1970, 1975, 1987, 1991, 2004).

The State Environmental Monitoring also includes an air quality monitoring subsystem, which should (among others) help to monitor the changes in the acidification of the environment as a result of the deposition of pollutants (SEM, 2015). The subsystem includes **monitoring of chemical composition of precipitation** at 22 stations (e.g. SO_4, NO_x, Cl, Na, Ca, Mg, K, TN, pH) and the estimation of monthly deposition to soils (SEM, 2015). Additionally, as a part of the SEM, 11 **integrated monitoring stations** operate and include monitoring of deposition and most of them monitoring of surface waters with the scope consistent with the ICP Integrated Monitoring. According to the acid sensitivity analysis described in the previous section, 3 integrated monitoring sites are in the acid-sensitive area and 4 more are less than 1 km from such areas. The remaining four integrated monitoring sites are in insensitive regions but one of these points (Święty Krzyż) is worth mentioning because it was created in 1993 in order to assess impacts of high acid deposition.

In Poland there are 22 monitoring sites active in the **Long-Term Ecosystem Research in Europe** network (DEIMS, 2017). Ten of these sites include monitoring of surface water chemistry but at least half of these sites includes one of following types of water body / basin: large river, dammed reservoir, urban catchment area, basin with dominating organic matter and clay (DEIMS, 2017).
In addition to the State Environmental Monitoring System and the LTER, Poland has carried out research monitoring for the purpose of the ICP Waters Programme. This monitoring initially included two monitoring sites in the Tatra Mountains (since 1992) which were supplemented by two lakes in the Karkonosze Mountains in 2004. The location of sites was determined by the requirements of the ICP Waters Programme, e.g. no or limited sources of pollution other than atmosphere and additionally they are highly acid sensitive. Sampling frequency was twice a month and chemical analyses included among others: pH, alkalinity, Ca, Mg, Na, K, SO_4, Cl, NO_3, NH_4. At the end of 2012 the monitoring at all four sites was discontinued and in 2016 part of the reference monitoring points of the SEM (5 lakes: Jegocin, Długie Wigierskie, Krąpsko Długie, Głębokie and Łękuk Wielki) were proposed as new ICP Water monitoring sites. Selection of new sites was aimed to ensure low or no anthropogenic impacts other than atmospheric deposition, relatively long-term data availability (at least 10 years), and scope and frequency of analyses close to the required by the ICP Waters. Two of these lakes are in acid-sensitive areas and three are adjacent to sensitive areas. Unfortunately, reference monitoring points do not provide sufficient data for the calculation of ANC, and therefore, the assessment of impact of acid deposition is limited to pH and alkalinity.

4.10.4 Acidification monitoring data

The main form of water monitoring in Poland (surveillance and operational monitoring of surface water bodies) is only to some extent useful for the purpose of this report. Since the beginning of implementation of the WFD, the scope of physicochemical parameters used for the assessment of water quality was reduced (e.g. no pH in the assessment of lakes since 2008, not enough parameters to calculate the ANC) and, moreover, lakes smaller than 50 ha and rivers of catchment area smaller than 10 km^2 were excluded from the monitoring system. Therefore, for the purpose of this study, 21 monitoring points were selected from among integrated monitoring stations, reference monitoring points and river water bodies taking into account:

- Requirements of the acidifications status survey, i.e.: one value per parameter per site, data for current situation 2010-2016, data from acid-sensitive regions, parameters of concern: ANC, ANC_{oaa}, pH, DOC.
- Location of monitoring points and their catchment areas: all points except one are in or less than 1 km from sensitive regions (one point is not in the sensitive region in terms of the geology but it was intended to monitor the impact of high acid deposition).
- Scope of available data (parameters of concern monitored in the 2010-2016 period).
- Differentiation of types of water bodies, locations, land use of catchment areas.

4.10.5 Acidification status

Based on the ANC it can be stated, that the most acidified waters are small lakes in high mountains (6-100 µeq/l) (Tatra and Karkonosze). Relatively low ANC was also found at the Święty Krzyż integrated monitoring station, which is still under the significant impact of long-term acid deposition (124 µeq/l). In the northern part of Poland, where lake districts and the coast are partially classified as acid sensitive, only 1 of 7 analysed monitoring points had relatively low ANC (170 µeq/l) and the remaining points had the ANC ranging from 2662 to nearly 6000 µeq/l. This individual case is a small lake with very small, semi-natural catchment (17 ha).

The pH confirms conclusions based on the ANC. In the northern Poland the pH was low (yearly average 6.5) in the case of Czarne Lake in Strokowo integrated monitoring station only. Low pH was in the acidified basin of the Święty Krzyż integrated monitoring station, in the Tatra Mountains and in all 6 monitoring points in the Sudety Mountains. In case of the Sudety, it is worth mentioning, that lower monitoring points – rivers, had average pH 6.7-7.25 but yet below the target set for good ecological status of these types of rivers (Figure 43).

Highly sensitive areas cover 12.5% of Poland and the acidification occurs mainly in small lakes or streams and in heavily polluted ones (Święty Krzyż). That theory finds confirmation in the state monitoring of larger water bodies. Only 4 of 4586 river water bodies (catchment area greater than 10 km^2) had the pH below target set for good ecological status / potential in 2013 (RBMP, 2016). All these water bodies are in highly sensitive area of the Sudety Mountains, confirming that the south-western part of Poland (Sudety) is one of the most acid-sensitive and acidified regions.

Wolanin (2013) confirmed the importance of catchment area and geological conditions for the chemical characteristics of surface waters. The study included analysis of main 9 streams in the Tatra Mountains (8 samples per site in 2011). Only one stream had relatively low average pH (7.7, remaining ones 7.9-8.2) and ANC (485 µeq/l, where other streams had approx. 1000-3000). The mentioned stream was differentiated by fluvial and glacial sediments and crystalline rocks as bedrock, and by land cover with a relatively small share of forests (33%) and a large share of bare rock (34%) (Wolanin, 2013). The impact of catchment area on the acidification can be seen in case of one of the analysed streams (Sucha Woda) which does not show signs of acidification, even though there are acidified lakes (Zielony Staw and Długi Staw) in its source area.

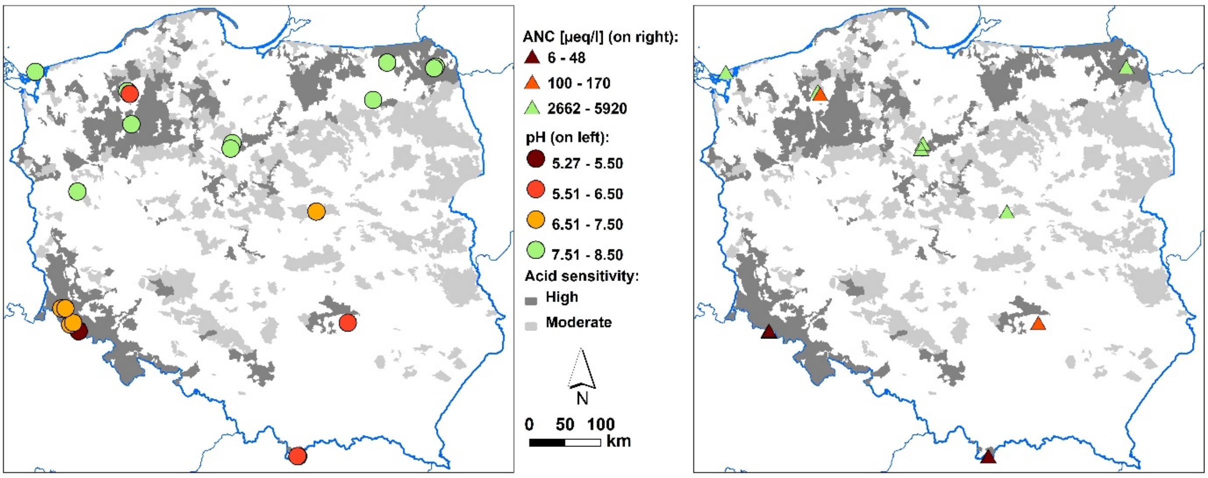


Figure 43 ANC and pH in the monitoring points used to derive submitted data

The high altitude part of Poland is considered as the most sensitive to acid deposition but there is a clear chemical recovery in response to the decreasing deposition of sulphur and nitrogen. In 4 mountain lakes used as ICP Waters monitoring sites, the ANC increased in 2004-2012 (Karkonosze) and 1992-2012 (Tatry), and the increase was statistically significant (Rzychoń, 2013). The acidification (natural and deposition-driven) was also a subject of biological studies with diatoms as indicators of changes in environmental parameters such as pH (Sienkiewicz, 2016). A long-term acidification history, based on the record of subfossil diatoms suggests that Mały Staw and Wielki Staw in the Karkonosze Mts. were naturally acidified, but the diatom-inferred pH values confirm that an increase in acidity began in the 1970s and affected Mały Staw more – a smaller and shallower lake with a larger catchment area. Changes in the phytoplankton communities toward an increase of diatom taxa preferring acidic conditions occurred and lasted until 2002, suggesting a significant delay in the biological recovery (Sienkiewicz, 2016).

4.10.6 References

Błachuta J., Czoch K., Kulesza K., Picińska-Fałtynowicz J., 2005. Typologia rzek i strumieni Polski i podział na jednolite części wód. IMGW oddział we Wrocławiu, IMGW Oddział w Krakowie. Proceedings: Wdrażanie Ramowej Dyrektywy Wodnej, Ocena stanu ekologicznego wód w Polsce, Poland, Łódź, 7-9.12.2005.

Brożek S., Zwydak M., 2003. Atlas gleb leśnych Polski, Centrum Informacyjne Lasów Państwowych, Warszawa.

Charzyński P., Bednarek R., Hulisz P., 2007. Pozycja systematyczna polskich gleb w międzynarodowej klasyfikacji WRB 2006, Roczniki Gleboznawcze, Tom LVIII, Nr 3/4, Warszawa.

CLRTAP, 2014. Guidance on mapping concentrations levels and deposition levels, Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. UNECE Convention on Long-range Transboundary Air Pollution

DEIMS, 2017. Dynamic Ecological Information Management System - Site and dataset registry (DEIMS-SDR). Available at: https://data.lter-europe.net/deims/ [accessed on 04-08-2017].

Dróżdż P., Dudzińska M., Hildebrand R., Jabłoński M., Kantorowicz W., Kluziński L., Kowalska A., Lech P., Małachowska J., Pierzgalski E., Piwnicki J., Stolarek A., Szczygieł R., Ślusarski S., Tyszka J., Wawrzoniak J., Wójcik J., Zajączkowski G., 2015. Stan uszkodzenia lasów w Polsce w 2014 roku na podstawie badań monitoringowych. Instytut Badawczy Leśnictwa, Zakład Zarządzania Zasobami Leśnymi. Raport opracowany w ramach VI etapu Umowy nr 20/2012/F z dnia 09.08.2012 r. pt. „Monitoring i ocena stanu zdrowotnego lasów w latach 2012 - 2014"

ESDAC, 2006. The European soil database. European Soil Data Centre esdac.jrc.ec.europa.eu, European Commission, Joint Research Centre

Gotkiewicz J., Piaścik H., Smołucha J., 2004. Ocen zasobów gleb mineralnych i hydrogenicznych pojezierza mazurskiego w aspekcie ich użytkowania i ochrony, Woda-Środowisko-Obszary Wiejskie, t.4, z.2a, IMUZ Falenty.

Hobot A., Banaszak K., Borzyszkowski J., Ciupak E., Dołęga M., Hubert K., Kolada A., Kołbut Ł., Kołodziejczyk A., Komosa M., Kraśniewski W., Krzymiński W., Kunert M., Kutyła S., Mutryn J., Pasak D., Pasztaleniec A., Skuza M.K., Soszka H., Stachura-Węgierek A., 2014. Aktualizacja wykazu JCWP i SCWP dla potrzeb kolejnej aktualizacji planów w latach 2015-2021 wraz z weryfikacją typów wód części wód – ETAP I – Metodyka

Holowaychuk N., Fessenden R.J., 1986. Soil sensitivity to acid deposition and the potential of soils and geology in Alberta to reduce the acidity of acidic inputs, Alberta Research Council,

ICP Waters Programme Centre, 2010. ICP Waters Programme Manual 2010. ICP Waters Report 105/2010. NIVA, November 2010

Journal of Laws, 1962. Regulation of the Prime Minister of 28 February 1962 concerning the allowed pollution of water and conditions to be met when introducing wastewater into water or ground (Journal of Laws of 1962, No 17, item 75)

Journal of Laws, 1970. Regulation of Ministers of 9 June 1970 concerning the allowed pollution of water and conditions to be met when introducing wastewater into water or ground (Journal of Laws of 1970, No 17, item 144)

Journal of Laws, 1975. Regulation of Ministers of 29 November 1975 concerning the Classification of water, conditions to be met by wastewater and fines for infringements of regulations (Journal of Laws of 1975, No 41, item 214)

Journal of Laws, 1987. Regulation of Ministers of 14 December 1987 concerning the Classification of water, conditions to be met by wastewater and fines for infringements of regulations (Journal of Laws of 1987, No 42, item 248)

Journal of Laws, 1991. Regulation of the Minister of Environment, Natural Resources and Forestry of 5 November 1991 concerning the Classification of water and conditions to be met when introducing wastewater into water or ground (Journal of Laws of 1991, No 116, item 503)

Journal of Laws, 2004. Regulation of the Minister of the Environment of 11 February 2004 concerning the Classification for the purpose of presenting the status of surface water and groundwater, and on the Form and Method of Monitoring, interpretation and presentation of its results (Journal of Laws of 2004, No 32, item 284)

Journal of Laws, 2008. Regulation of the Minister of the Environment of 20 August 2008 concerning the Classification of Surface Water Bodies (Journal of Laws of 2008, No 162, item 8654)

Journal of Laws, 2009. Regulation of the Minister of the Environment of 13 May 2009 on the Form and Method of Monitoring of Surface Water and Groundwater Bodies (Journal of Laws of 2009, No 81, item 685)

Journal of Laws, 2011. Regulation of the Minister of the Environment of 15 November 2011 on the Form and Method of Monitoring of Surface Water and Groundwater Bodies (Journal of Laws of 2011, No 258, item 1550)

Journal of Laws, 2016. Regulation of the Minister of the Environment of 19 August 2016 on the Form and Method of Monitoring of Surface Water and Groundwater Bodies (Journal of Laws of 2016, item 1178)

Kacprzak A., 2003. Pokrywy stokowe jako utwory macierzyste gleb Bieszczadów Zachodnich, Roczniki Gleboznawcze, Tom LIV, Nr 3, Warszawa.

Kondracki J., 1978. Geografia fizyczna Polski, PWN, Warszawa

Lisicki S., 1998. Osady interglacjału mazowieckiego w centralnej części Pojezierza Mazurskiego, Przegląd Geologiczny, vol. 46, nr 2.

Maruszczak H., 2000. Definicja i klasyfikacja lessów oraz utworów lessopodonych, Przegląd Geologiczny, vol. 48, nr 7.

McFee W.W., 1980. Sensitivity of Soil Regions to Acid Precipitation, Environmental Research Laboratory, US EPA, Cornvallis,

Mill W., 1995. Critical loads mapping in Poland: Lessons learned. Water, Air, and Soil Pollution, Volume 85, Issue 4, pp 2547–2552

Mill W., 2007. Update of critical load maps of acidity and eutrophication of selected terrestrial ecosystems of Poland (in Polish). Ochrona Środowiska i Zasobów Naturalnych nr 30, 2007

Panagos P., Van Liedekerke M., Jones A., Montanarella L., 2012. European Soil Data Centre: Response to European policy support and public data requirements. Land Use Policy, 29 (2), pp. 329-338. doi:10.1016/j.landusepol.2011.07.003

Petelski K., 2008. Ewolucja poglądów na budowę geologiczną i powstanie gardzieńskiej moreny czołowej, Landform Analysis, Vol. 7: 130–137.

Picińska-Fałtynowicz J., Błachuta J., Kotowicz J., Mazurek M., Rawa W., 2006. Wybór typów jednolitych części wód rzecznych i jeziornych do oceny stanu ekologicznego na podstawie fitobentosu wraz z rekomendacją metodyki poboru i analizy prób. Instytut Meteorologii i Gospodarki Wodnej, Oddział we Wrocławiu

RBMP, 2016. River Basin Management Plans, The Decree of the Council of Ministers of 18 October 2016, Journal of Laws, items 1818, 1911, 1914, 1915, 1917, 1919, 1929, 1959, 1967, available at: http://www.kzgw.gov.pl/index.php/pl/ramowa-dyrektywa-wodna-plany-gospodarowania-wodami (in Polish)

Rzychoń D., 2013. Wpływ opadów kwaśnych na wody powierzchniowe na przykładzie wybranych jezior w Tatrach i Karkonoszach. Otwarte seminaria 2013. Instytut Ekologii Terenów Uprzemysłowionych w Katowicach

Shilts W.W., 1981. Sensitivity of Bedrock to Acid Precipitation. Modification by Glacial Processes, Geological Survey of Canada

SEM, 2015. The State Environmental Monitoring Programme for the years 2016–2020. Chief Inspector of Environmental Protection.

Sienkiewicz E., 2016. Post-glacial acidification of two alpine lakes (Sudetes Mts., SW Poland), as inferred from diatom analyses. Acta Palaeobotanica 56(1): 65–77, 2016. DOI: 10.1515/acpa-2016-0002

Skjelkvåle B.L., Wright R.F., 1990. Overview of areas sensitive to acidification: Europe, Niva Report, Oslo.

Sobolewski W., Borowiak D., Borowiak M., Skowron R., 2014. Baza danych jezior Polski i jej wykorzystanie w badaniach limnologicznych, Uniwersytet Marii Curie-Skłodowskiej, Wydział Nauk o Ziemi i Gospodarki Przestrzennej, Lublin.

Szostak M., 1967. Pochodzenie Jeziora Śniardwy i jego zasoby wodne, Prace Geograficzne Nr 58, Państwowe Wydawnictwo Naukowe, Warszawa.

Wojciechowki A., 2008. Ewolucja jezior przybrzeżnych Niziny Gardzieńsko-Łebskiej na tle rozwoju środkowego wybrzeża Bałtyku w świetle badań malakologicznych, Landform Analysis, Vol. 7: 154–171.

Wolanin A., 2013. Physical and chemical properties of streamwater in the Tatra Mountains during April – November 2011 (in Polish). Prace Geograficzne, zeszyt 133. Instytut Geografii i Gospodarki Przestrzennej UJ. Kraków 2013, 49 – 60. doi: 10.4467/20833113PG.13.010.1100

Zawadzki S. (ed), 1999. Gleboznawstwo, PWRiL, Warszawa.

4.11 Spain

Manuel Toro Velasco
Centre for Hydrographic Studies (CEDEX)

4.11.1 Acid sensitivity

Maps of sensitive regions in Spain (Figure 44) were based on lithology information from the Spanish Geological Map (1:1.000.000)[23], weighted with estimated water contribution from each type of lithology in each catchment, and have been validated with data from water chemistry monitoring networks of reference stations and sites without significant pressures. In Spain, potential acid sensitive areas are located mainly in the western area, and in crystalline lithology mountainous areas in the NE (Pyrenees), small catchments in the C-NE (Iberian Range Mountains) and in the SE corner (Sierra Nevada and small littoral catchments). The rest of the country is covered with buffering lithology (e.g. calcareous, evaporite, mixed sedimentary).

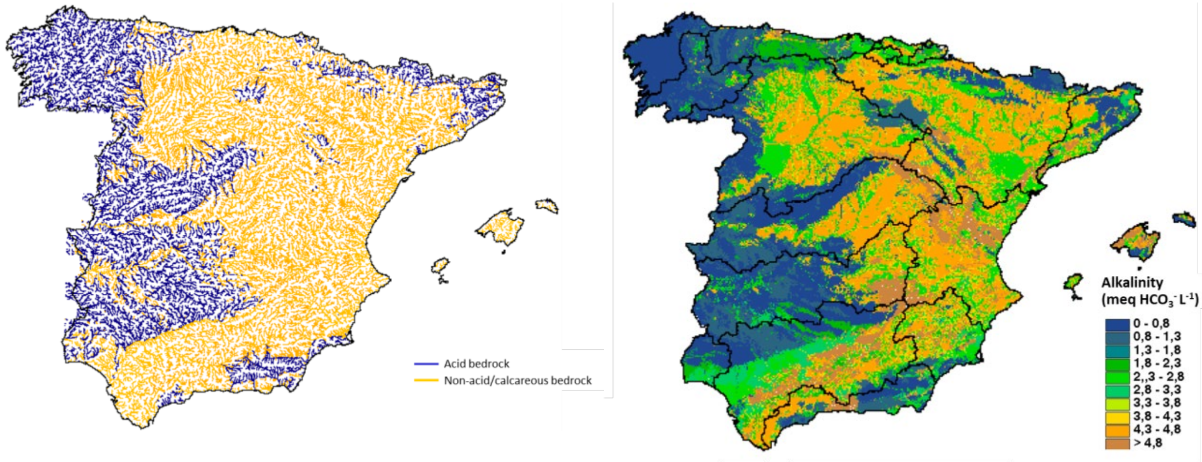


Figure 44 Potential acid sensitive areas in Spain based in lithology (left) and runoff alkalinity (right).[24]

4.11.2 Monitoring and assessment approach

There is not any specific water acidification monitoring network in Spain, since no significant effects on freshwater ecosystems at a regional scale have been observed or recorded in the past. Information on water chemistry in Spanish aquatic ecosystems is provided from several national monitoring networks, with yearly information in most rivers, lakes and reservoirs[25]. The main network is the Water Framework Directive surveillance programme, which includes about 2200 river monitoring stations, more than 400 reservoirs and about 170 lakes or wetlands. Figure 45 shows the location of 40 points selected from the national networks, being the only suitable places for monitoring acidification effects because they are not affected by any other human pressure, and have a variable frequency of measured data on pH and alkalinity from 1990. There are many areas, both in acid sensitive and non-sensitive regions, without any monitoring stations free of any human pressure or local sources of contaminants, or with a minimum frequency of monitoring. Reference sites in the Water Framework Directive monitoring network usually are sampled every 2-3 years,

[23] http://info.igme.es/cartografiadigital/datos/geologicos1M/Geologico1000_(1994)/jpgs/EditadoG1000_(1994).jpg
[24] http://nfp-es.eionet.europa.eu:8980/Public/irc/eionet-circle/phjornadas/library?l=/herramientas_implantacin/jornadasaquatool/cd_curso_aquatool/escenario001_1/caracterizacion/_EN_1.0_&a=i
[25] http://www.mapama.gob.es/es/agua/temas/estado-y-calidad-de-las-aguas/aguas-superficiales/programas-seguimiento/

which is not sufficient to get robust information on acidity trends, and many of them have minor alterations or pressures that could mask any possible response to atmospheric pollution, including acidification. At the same time, a small group of high mountain lakes, most of them located in acid-sensitive areas and free of any local pressure, have been monitored monthly since 20-30 years ago under scientific or natural protected areas monitoring programmes. Although they do not cover randomly the whole Spanish territory, these lakes provide a good long term approach to the state of the acidification in some on the main sensitive regions.

4.11.3 Acidification status

The mean values for pH and alkalinity (meq/l $CaCO_3$) of all recorded measurements in the selected 40 points from the national networks are shown in Figure 45. No significant trends or episodes of acidity have been observed during the registered monitoring periods (from last 3 to 20 years). The minimum value of pH recorded in any of these stations is higher than 5.8-6.0, even considering some high mountain lakes during the typical low pH snowmelt period.

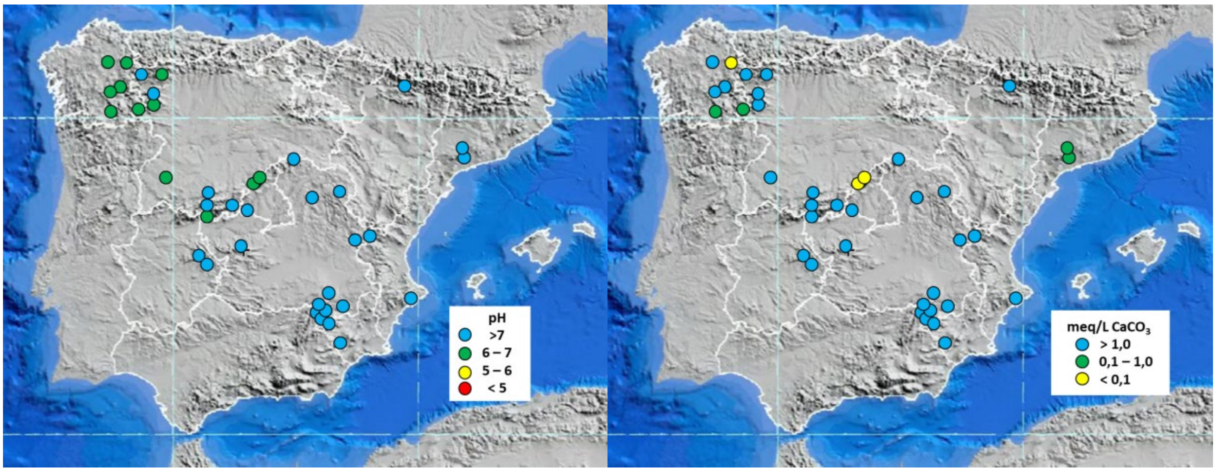


Figure 45 Mean values of pH and alkalinity in 40 rivers and lakes sites without any human pressure. Source: 2015-2017 data from National Monitoring Networks of Spanish General Directorate of Water.

A group of studied remote mountain lakes in the Iberian Peninsula in the 1990s did not show significant chemical and biological signs of water acidification in the recent past (Camarero et al, 1995). Most of these lakes would be considered sensitive to acidification according to categories established by Camarero and Catalan (1998) for Pyrenean lakes based on pH and alkalinity, with values of pH usually higher than 6.5 and alkalinity between 50 and 100 µeq/l HCO_3^-. A survey of 24 lakes conducted in 1995-97 in the Central Range (Toro et al., 2001) showed pH data with values between 5.8 to 7.2, with 70% of lakes higher than 6.5 and alkalinity between 30 and 167 µeq/l HCO_3^- (75% below 100 µeq/l). One of them, Lake Peñalara, is located 50 Km NW of Madrid city, and monthly data on pH and alkalinity/ANC since 1995 (Granados et al., 2006) show no significant trends of either parameter, with a typical seasonal oscillation from higher values during summer primary productivity, and minimum during snow/ice melting period at the end of the winter (Figure 46). Another remote lake in the Central Range, Lake Cimera, is located 140 Km SW of Madrid, and does also not show any acidity trend during last 20 years of yearly monitoring.

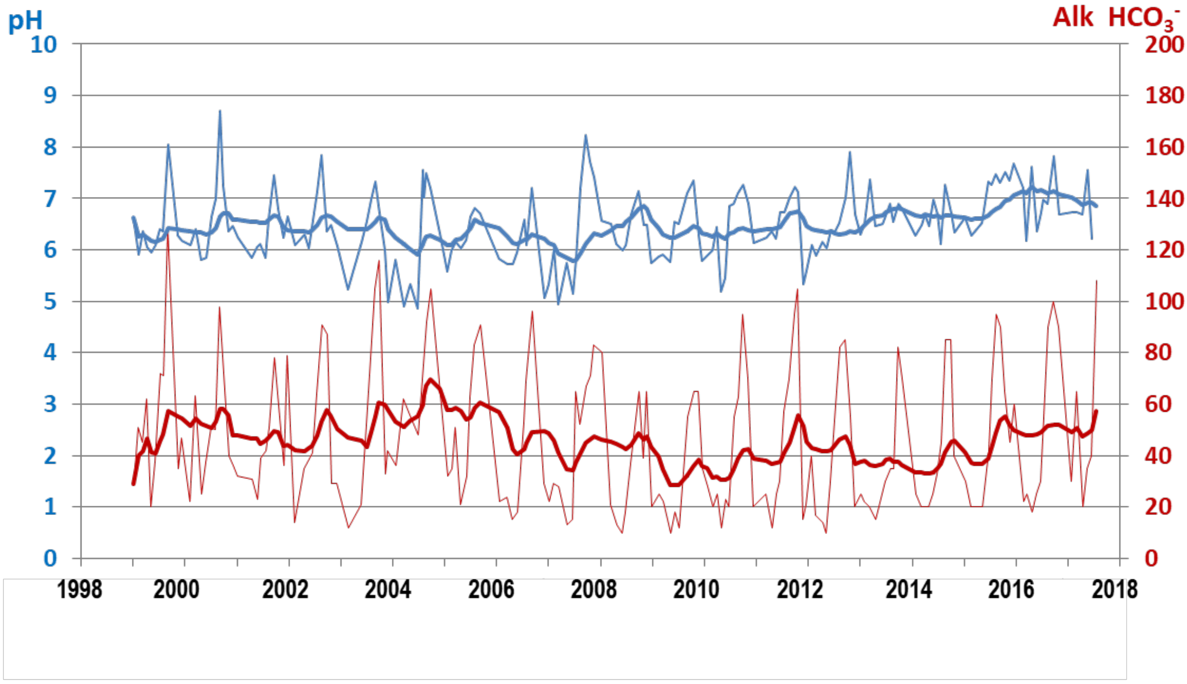


Figure 46 Long-term (20 years) trend of pH and alkalinity in surface water in Lake Peñalara (Central Spain).

In NW Spain, Lake Sanabria is the largest natural Spanish mountain lake, and it has been monthly monitored since 1986. Although there are some minor pressures in its basin (rural area with cattle grazing and tourism), it is an oligotrophic lake with a very low ionic concentration (9-17 µS/cm) and relatively low alkalinity (in the range from 0.44 to 0.62 meq/l $CaCO_3$ during last 5 years). The long-term trend of pH is positive for Lake Sanabria over the last 30 years, but there are no data on sulphate or nitrogen deposition during that period to relate this trend to recent changes in atmospheric pollution. Nevertheless, the relationship between time patterns of primary production in the lake (Chl *a* of phytoplankton) and pH values (Figure 47) indicates a possible biological factor that influences the pH trend due to the increase in the productivity of the lake in the last 30 years. More research on lake and environmental data is needed to clarify these hypotheses.

In the Pyrenees, where about 60% of the lakes are considered sensitive to acidification (Camarero, 2017), sulphate deposition and concentrations in lakes have decreased during the last four decades following a reduction in sulphur emissions (Avila and Rodá, 2002), and only a moderate acidification has been detected in some lakes, where nitrogen could be a more important agent of acidification (Camarero and Catalan, 1998). The recent increasing in the frequency and intensity of African dust (Rodríguez-Navarro et al., 2018) could explain the moderate or lack of signs of freshwater acidification in acid-sensitive regions in the Iberian Peninsula, due to its significant base cation load, which increases ANC in surface waters, minimizing acid deposition effects (Psenner, 1999; Morales-Baquero et al., 2006, 2013; Camarero, 2017).

Additionally, in Sierra Nevada lakes (Southern Spain), increased temperature and Saharan dust deposition containing phosphorus and calcium during the last few decades appear to be affecting biota and increasing productivity (Jiménez et al., 2018). Dust deposition with phosphorus could also be causing a possible shift from phosphorus to nitrogen limitation for phytoplankton in some Pyrenean lakes (Camarero and Catalan, 2012).

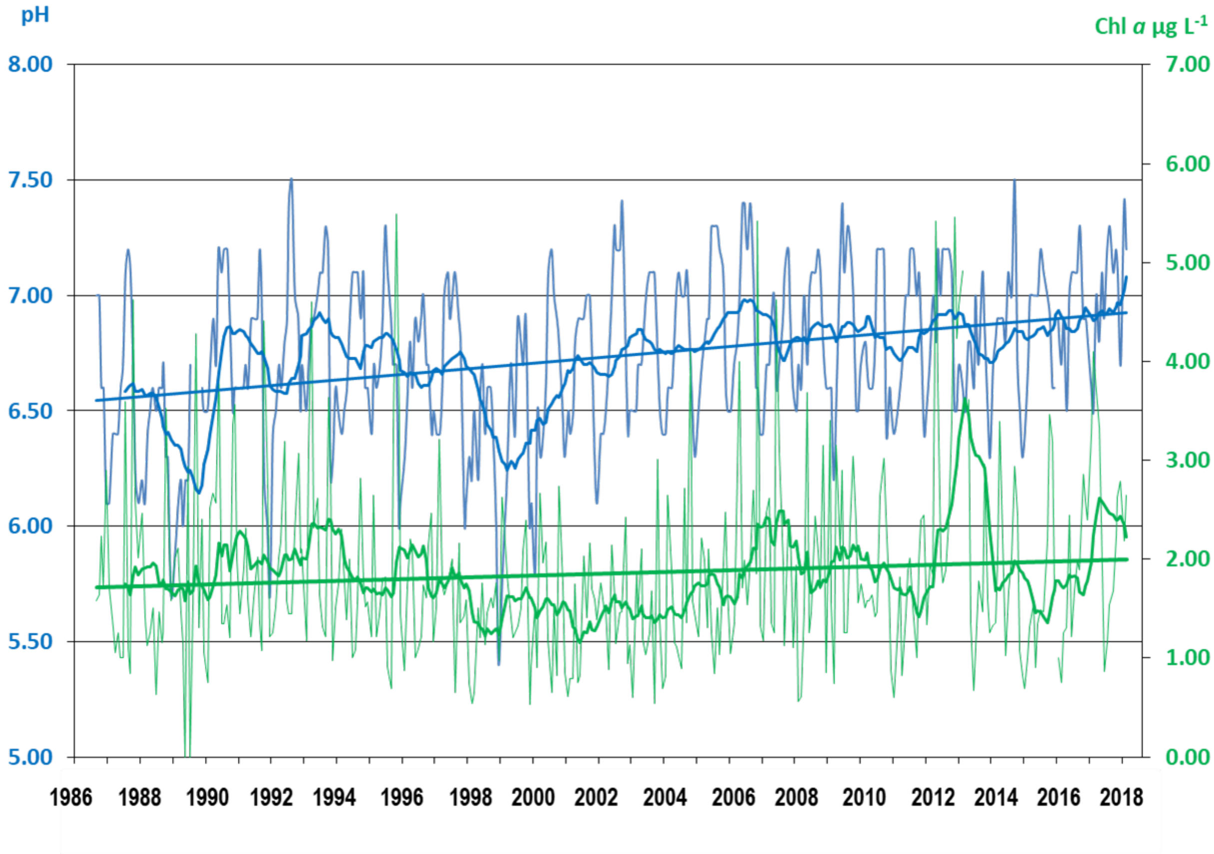


Figure 47 Long-term (30 years) trend of pH and chlorophyll in surface water in Lake Sanabria (NW of Spain). 12-month moving average and trend lines are shown for both parameters. Source: Limnological Monitoring Programme of Natural Park of Lake Sanabria[26].

Acknowledgements: We thank Alexandra Puig and Juan Alández from Spanish General Directorate of Water, and José Fernández del Pino from TRAGSA, for providing support and monitoring data from national networks. José Carlos Vega (Natural Park of Lake Sanabria) and Ignacio Granados (National Park of Guadarrama Mountains) have provided long-term data on lakes Sanabria and Peñalara, respectively.

[26] https://patrimonionatural.org/noticias/general/2013/11/14/los-datos-del-laboratorio-limnologico-del-lago-de-sanabria-certifican-la-buena-calidad-del-agua

4.11.4 References

Avila, A. and Rodá, F. 2002. Assessing decadal changes in rainwater alkalinity at a rural Mediterranean site in the Montseny Mountains (NE Spain). *Atmospheric Environment*, 36: 2881-2890.

Camarero, L., Catalan, J., Pla, S., Rieradevall, M., Jiménez, M., Prat, N., Rodríguez, A., Encina, L., Cruz-Pizarro, L., Sánchez Castillo, P., Carrillo, P., Toro, M., Grimalt, J., Berdie, L., Fernández, P. and Vilanova, R. 1995. Remote mountain lakes as indicators of diffuse acidic and organic pollution in the Iberian Peninsula (AL:PE 2 Studies). *Water, Air and Soil Pollution*, 85: 487-492.

Camarero, L. & Catalan, J. 1998. A simple model of regional acidification for high mountain lakes: application to the Pyrenean lakes (North-East Spain). *Water Research*, 32 (4): 1126-1136.

Camarero, L., and J. Catalán. 2012. Atmospheric phosphorus deposition may cause lakes to revert from phosphorus limitation back to nitrogen limitation. *Nature Communications*, 3, 1118. DOI:10.1038/ncomms2125.

Camarero, L. 2017. Atmospheric chemical loadings in the high mountain: current forcing and legacy pollution. In: J. Catalan et al. (eds.), *High mountain conservation in a changing world*, Advances in Global Change Research, 62. DOI 10.1007/978-3-319-55982-7_14. Springer, pp. 325-341.

Granados, I., Toro, M. and Rubio-Moreno, A. 2006. *Laguna Grande de Peñalara. 10 años de seguimiento limnológico*. Serie Técnica del Medio Natural. Edita Consejería de Medio Ambiente y Ordenación del Territorio. Comunidad de Madrid. 185 pp.

Jiménez, L., Rühland, K.M., Jeziorski, A., Smol, J.P. and Pérez-Martínez, C. 2018. Climate change and Saharan dust drive recent cladoceran and primary production changes in remote alpine lakes of Sierra Nevada, Spain. *Global Change Biology*, 24 (1): 139-158. DOI: 10.1111/gcb.13878.

Morales-Baquero, R., Pulido-Villena, E., Romera, O., Ortega-Retuerta, E., Conde-Porcuna, J.M., Pérez-Martínez and Reche, I. 2006. Significance of atmospheric deposition to freshwater ecosystems in the southern Iberian Peninsula. *Limnetica*, 25 (1-2): 171-180.

Morales-Baquero, R., Pulido-Villena, E. and Reche, I. 2013. Chemical signature of Saharan dust on dry and wet atmospheric deposition in the south-western Mediterranean region. *Tellus Series B*, 65, 8720, http://dx.doi.org/10.3402/tellusb.v65i0.18720.

Psenner, R. 1999. Living in a dusty world: airborne dust as a key factor for alpine lakes. *Water, Air and Soil Pollution*, 112 (3-4): 217-227. https://doi.org/10.1023/A:1005082832499

Rodriguez-Navarro, C., di Lorenzo, F. and Elert, K. 2018. Mineralogy and physicochemical features of Saharan dust wet deposited in the Iberian Peninsula during an extreme red rain event. *Atmos. Chem. Phys. Discuss.*, https://doi.org/10.5194/acp-2018-211.

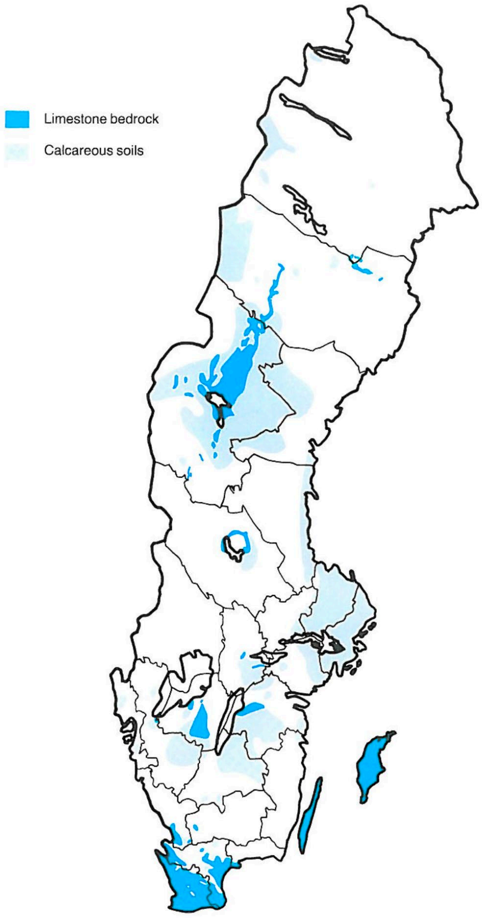
Toro, M. and Granados, I. (Eds.). 2001. *Las lagunas del Parque Regional de la Sierra de Gredos*. Monografías de la Red de Espacios Naturales de Castilla y León. Edita Junta de Castilla y León. Valladolid. 241 pp.

4.12 Sweden

Jens Fölster
Swedish University of Agricultural Sciences

4.12.1 Acid sensitivity

Most of Sweden is covered by till soils formed by slow-weathering silicates, mainly granites and gneisses. Exceptions to this are the large islands Öland and Gotland in the Baltic Sea, the southernmost county Skåne, the agricultural plains in south central Sweden, central northern Sweden where the soils are influenced by calcareous bedrock, and eastern central Sweden that is influenced by glacial deposits of limestone (Figure 48, left).



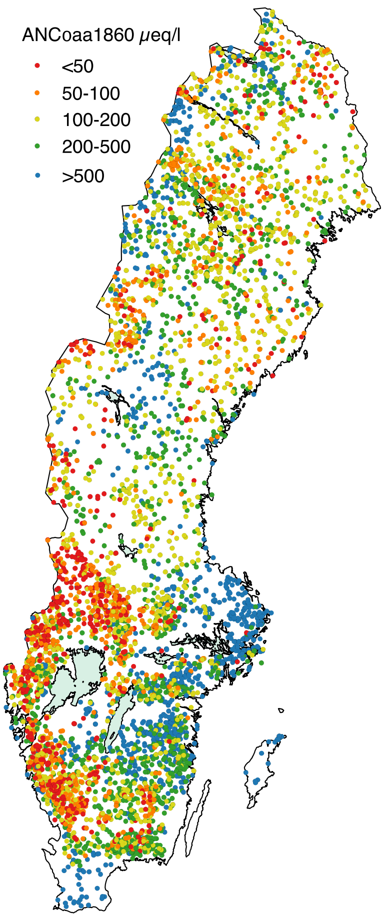


Figure 48 Left: Areas with lime stone bedrock (dark blue) and calcareous soils (light blue) in Sweden. Right: ANC_{oaa} in 1860 based on the MAGIC model in 5084 lakes sampled 2007-2012 after correction for anthropogenic liming. The density of dots reflects the higher spatial sampling frequency in southern Sweden.

Precipitation shows a strong gradient from west to east in Sweden. Higher precipitation leads to higher acid sensitivity under the same geological conditions due to dilution of weathering products. An additional factor controlling acid sensitivity is the concentration of organic carbon that contains natural organic acids. The concentration of TOC (total organic carbon) in Swedish lakes has a median of 8.9 mg/l 2007-2012 with 5 and 95 percentiles of 1.6 and 26 mg/l, respectively (Fölster et al., 2014a). The ANC_{oaa} (ANC adjusted for organic acids) at reference conditions was estimated from the MAGIC library for 5084 lakes, and shows the acid sensitivity of Swedish lakes (Figure 48, right). Acid-sensitive lakes, i.e. with a low ANC_{oaa}, are found in most of Sweden, except in the southernmost and central east regions. At smaller geographic scales, acid sensitivity also depends on catchment size, till depth, and coverage and location of mires in the catchment resulting in a local variability. This is for example found in the south-eastern parts of Sweden, where lakes with both high and low ANC_{oaa} are found close to each other.

4.12.2 Monitoring and assessment approach

The acidification status of Swedish surface waters is based on a national lake survey programme including c. 5000 lakes where 1/6 are sampled on a rotating basis once every six years during autumn circulation (Fölster et al., 2014a). The lakes are a stratified random selection from the national lakes register including all c. 100 000 lakes > 0.01 $km^{2,}$ with a bias towards larger lakes and lakes located in southern Sweden in relation to the real distribution. By destratifying the results, the true distribution of water chemistry in all Swedish lakes can be obtained. The programme started in 2007 and was preceded by lake surveys that occurred once every five years since 1972. The lake survey 1995 was a part of the larger Nordic lake survey (Henriksen et al., 1998).

A large number of the lakes has been sampled in several of the lake surveys allowing evaluation of changes over time, but for a more detailed monitoring of changes over time, there are national monitoring programmes including time series for lakes and streams. The lake programme comprises 108 lakes sampled for surface water chemistry four times a year. Phytoplankton and littoral and profundal invertebrates are sampled annually. A subset of the lakes is also sampled for fish every third year. Ten of the lakes are sampled more frequently for both chemistry and biology. Most trend lakes are acid sensitive and have been monitored since the mid-1980s. The national stream programme is comprised of 47 rivers sampled at the mouth and 67 smaller upland streams sampled monthly for water chemistry. 46 of the streams are sampled annually for phytobenthos and benthic invertebrates and 29 also for fish. The stream programme is more diverse than the lake programme and only a smaller number of streams are suitable for acidification assessments. For the common assessment in chapter 3.2 we assembled data from 94 lakes and 52 streams from these national monitoring programmes which have catchments less than 1000 km^2 and are located in regions with low occurrence of limestone bedrock or calcareous soils.

In addition to the national programmes, there are also regional monitoring programmes of which some have similar sampling schemes as the national stations. Further, there is a national programme for monitoring the effects of liming, which includes a smaller number of lakes and streams, both limed and not limed, that are monitored more intensively.

Sweden has an extensive programme of lake liming. 116 000 tons of lime are spread annually (2013), affecting around 8 000 lakes directly or indirectly (upstream liming). According to the Swedish national environmental objectives, liming is seen as an interim action to mitigate the harmful effects of acidification until natural recovery has occurred. Thus, acidification assessments are made on “non-limed” water chemistry calculated from the ratio of Ca/Mg from non-limed reference sites (Sjöstedt et al., 2013).

In Sweden, the criteria for acidification is a decrease in pH (>0.4 pH units) since 1860, as modelled by the MAGIC model (Fölster et al., 2007). Lakes or streams that have not been modelled using the MAGIC model are assessed using the most similar water body from a database of 2438 modelled lakes and 243 modelled streams called the MAGIC library (Moldan et al., 2013).

4.12.3 Acidification status

Ten percent of the lakes in Sweden are acidified, i.e. the pH value has decreased more than 0.4 units since preindustrial times. Most of the acidified lakes are found in west central Sweden and in southern Sweden, except for the southernmost county Skåne (Figure 49) (Fölster et al., 2014b). The assessment is based on simulations using the MAGIC model, which allows reconstruction of the number of acidified lakes since 1860. The reconstruction of pH in Swedish lakes shows that acidification started already during the first half of last century in the south-western part of the country (Figure 50). Acidification peaked in the 1980s in Sweden and was then followed by a recovery. In the northern and eastern parts of the country, where the degree of acidification was not as severe, a relatively large fraction of the acidified lakes has recovered. In the south-western region, however, almost half of the more heavily acidified lakes remain acidified, and a full recovery will not occur in the near future.

Although acidification still affects 10% of all lakes in Sweden, there has been a pronounced improvement in many lakes, especially in the south-western region, shown in the trend lakes (Figure 51). The rate of recovery has slowed, but there is still a pronounced decline in sulphate and increase in pH in lakes in south-western Sweden.

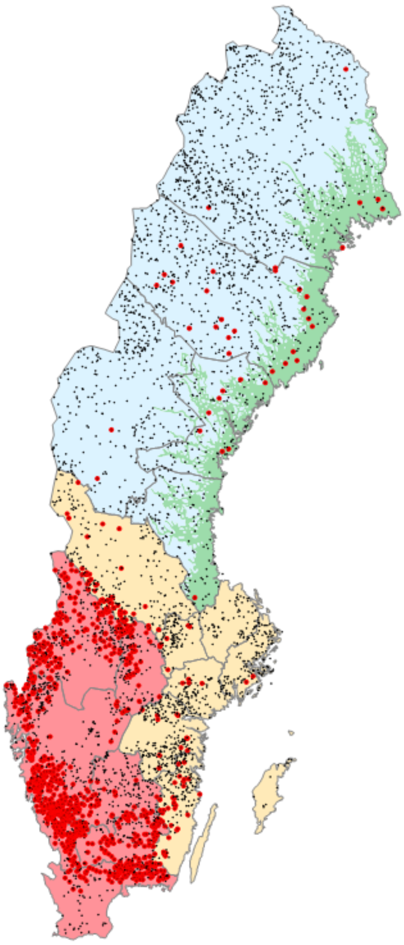


Figure 49 Acidification status of 5084 lakes selected by a random stratification of all 96 000 lakes > 1 ha in Sweden. The lakes were sampled during autumn circulation 2007-2012. Acidification is defined as a change in pH > 0.4 units since 1860 according to the MAGIC model (Moldan et al., 2013). Limed lakes were corrected for liming. Red dots denote acidified lakes and black dots non- acidified. The background colours represent four regions with different acidification pressure.

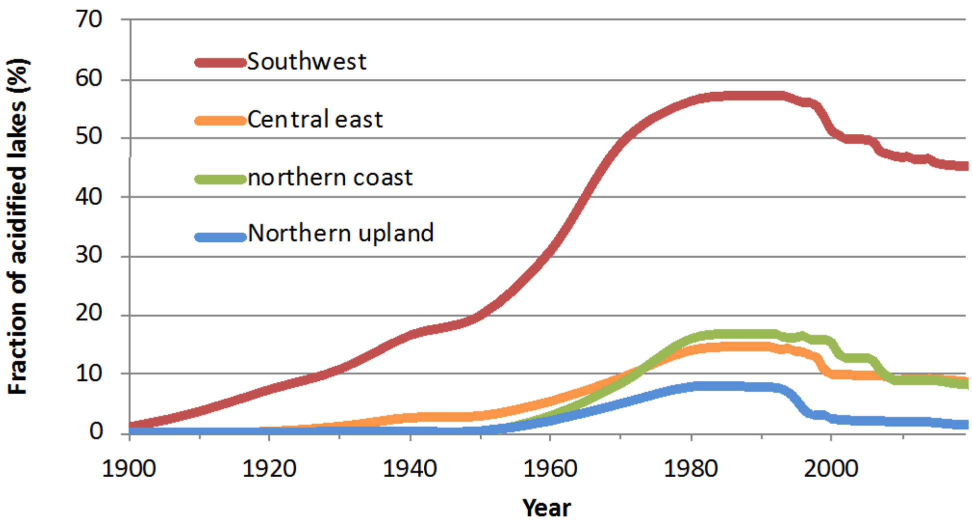


Figure 50 Development of acidification in lakes since 1860 in four regions in Sweden (see map in figure 3). The estimate is based on 5084 lakes selected by a random stratification of all 96 000 lakes > 1 ha in Sweden. The lakes were sampled during autumn circulation 2007-2012. Acidification is defined as a change in pH > 0.4 units since 1860 according to the MAGIC model (Moldan et al, 2013).

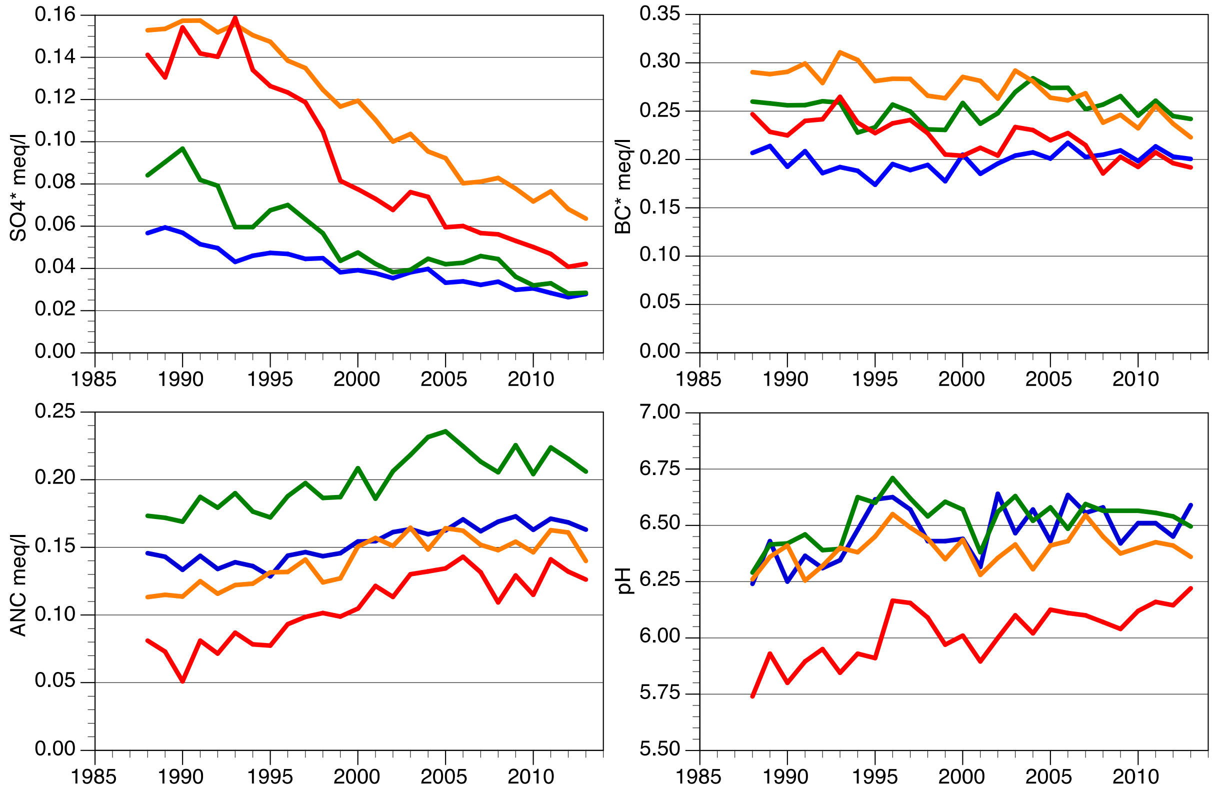


Figure 51 Development of water chemistry in time series lakes in four regions in Sweden (see map in Figure 49). Regional, annual medians of 95 national and regional trend lakes with ANC < 300 µeq/l. Orange = central east; red = south-west; green = northern coast; blue = northern upland.

Acknowledgements: The Swedish national monitoring data was financed by the Swedish Agency of Marine and Aquatic Management. The chemical analyses were all performed by the certified laboratory at the Swedish University of Agricultural Sciences.

4.12.4 References

Fölster, J., Andrén, C., Bishop, K., Buffam, I., Cory, N., Goedkoop, W., Holmgren, K., Johnson, R., Laudon, H. and Wilander, A. 2007. A Novel Environmental Quality Criterion for Acidification in Swedish Lakes – An Application of Studies on the Relationship Between Biota and Water Chemistry. Water Air and Soil Pollution: Focus 7(1): 331-338.

Fölster, J., Johnson, R.K., Futter, M.N. and Wilander, A. 2014a. The Swedish monitoring of surface waters: 50 Years of adaptive monitoring. Ambio 43: 3-18.

Fölster, J., S. Valinia, L. Sandin and Futter, M.N. 2014b. För var dag blir det bättre men bra lär det aldrig bli. Försurning i sjöar och vattendrag 2014. SLU, Vatten och miljö: Rapport 2014:20.

Henriksen, A., Skjelkvåle, B.L., Jaakko, M., Wilander, A., Ron, H., Curtis, C., Jensen, J. P., Erik, F. and Tatyana, M. 1998. Northern European Lake Survey, 1995: Finland, Norway, Sweden, Denmark, Russian Kola, Russian Karelia, Scotland and Wales. Ambio 27(2): 80-91.

Moldan, F., Cosby, B.J. and Wright, R.F. 2013. Modeling past and future acidification of Swedish lakes. Ambio 42(5): 577-586.

Sjöstedt, C., Andrén, C., Fölster, J. and Gustafsson, J.P. 2013. Modelling of pH and inorganic aluminium after termination of liming in 3000 Swedish lakes. Applied Geochemistry 35: 221-229.

4.13 Switzerland

Sandra Steingruber
Repubblica e Cantone Ticino, Sezione della protezione dell'aria dell'acqua e del suolo (Department of the territory of the Canton Ticino, Office for air, climate and renewable energy)

4.13.1 Acid sensitivity

Most freshwaters in Switzerland are protected from acidification by calcareous rock strata. Potentially acid-sensitive sites have small catchments and are situated on slow-weathering crystalline bedrock at higher altitudes; at lower altitudes the probability of catchments containing buffering minerals increases. According to earlier studies (Rihm 1994) most acid-sensitive lakes can be expected in the northern part of the Canton Ticino and in the neighbouring areas of other cantons. To identify the potentially acid-sensitive lakes in this region, lakes that satisfied the following criteria were selected:

- catchment situated prevalently on slow-weathering acidic rocks
- altitude over 1500 m.a.s.l.
- lake area greater than 0.5 ha
- not a reservoir

Figure 52 shows the occurrence of slow-weathering acidic rocks according to the Geological Map of Switzerland 1:500000 (lithological categories 50-53 and 58-63, i.e. gneiss, granite, quartzite, diorite; FOWG 2005) and the distribution of potentially acid-sensitive lakes in Switzerland according to the selection criteria described above (79 sites).

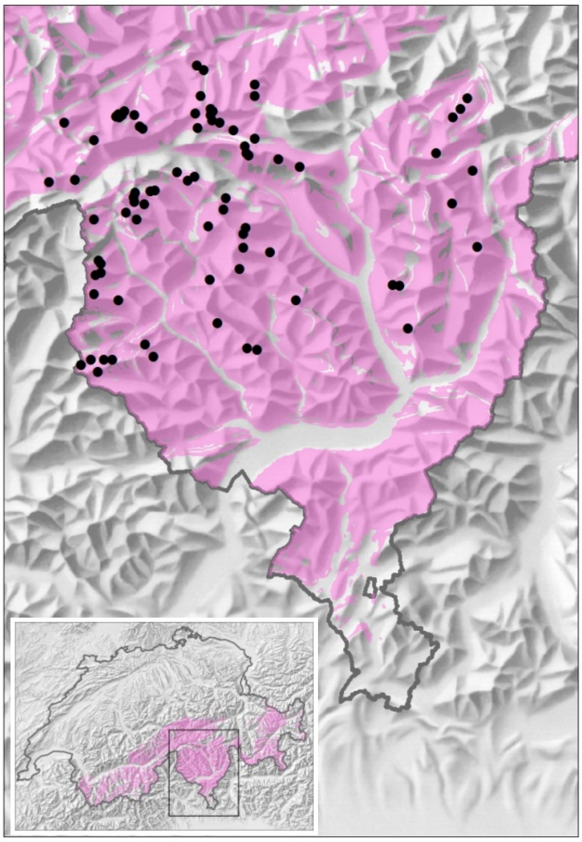


Figure 52 Potentially acid-sensitive areas (rose shading) and lakes (dots) in Switzerland (Relief map ©2011 swisstopo).

4.13.2 Study site and monitoring and assessment approach

Potentially acid-sensitive lakes in Switzerland are distributed over an area of about 2500 km^2. Besides having a base-poor lithology, this area is also characterized by high amounts of orographic precipitation (around 1300-3000 mm/yr, mean 1981-2010[27]) and of deposition of pollutants, due to its location at the foothills of the Alps, in proximity of the most industrialized part of Italy, the Po Valley, which includes cities such as Milan and Turin (Rogora et al. 2016, Steingruber 2015).

In order to monitor and assess acidification of freshwaters in Switzerland, water chemistry of 20 acid-sensitive high-altitude Alpine lakes have been monitored regularly since 2000 by the Canton of Ticino on behalf of the Federal Office for the Environment. From 2000 to 2005 lake surface water was sampled twice a year (once at beginning of summer, once in autumn). After 2006 lakes have been monitored three times a year (once at the beginning of summer, twice in autumn). Since 2000 four of these lakes belong to the ICP Waters monitoring network. Results including trend analysis are published in yearly reports[28].

Between 1980 and 2000 acid-sensitive Alpine lakes were monitored irregularly but in greater number (up to 50 lakes per year). The last large-scale survey was made in 1995. Since the aim of this report is to present an overview of the current situation that is as representative as possible, it was decided to increase the number of lakes sampled autumn 2017 from 20 to 52 and to include also acid-sensitive lakes that have been sampled irregularly in the past but, for financial reasons, not for the last 22 years. To limit the monitoring costs, only one autumn sampling was made. Unlike the selection of potentially acid -sensitive lakes shown in Figure 52, also some lakes smaller than 0.5 ha were monitored. With exception of lakes with other lakes in their catchment (63, 65, 224), for which the FAB-method has not been designed, critical loads of acidity (CLA(FAB)) were calculated for all monitored lakes (Posch et al. 2007, Achermann et al. 2015).

As regards national acidification criteria, only general requirements are prescribed for surface waters in the Swiss Waters Protection Ordinance (WPO). Annex 1 of the WPO prescribes that the communities of plants, animals and micro-organisms in surface waters and the surroundings influenced by them shall be close to the natural state and appropriate to the location as well as reproducing and regulating themselves and show a diversity and frequency of species that are specific for unpolluted or slightly polluted waters of the type in question. In addition, the water quality shall be such that naturally-occurring substances are not increased by human activities to concentrations that are above the range of natural concentrations. Since in Switzerland calculations of CLAs of Alpine lakes are based on a critical ANC value of 20 µeq/l (De Jong J.E. 1996, Posch et al. 2007, Rihm 1994), this critical ANC value is also used here in this report.

This contribution presents the actual acidification status of 52 acid-sensitive lakes in Switzerland (2015-2017) based on autumn samplings. To evaluate the impact of seasonal variations, autumn data for a subset of 20 lakes were compared with results from samplings after snowmelt at the beginning of July. Temporal changes were assessed comparing the present data with results from the past large-scale survey in 1995 (45 sites). For the 20 regularly analysed lakes trend analyses were also performed.

Furthermore, for lakes with existing CLA(FAB)s (Achermann et al. 2015) the percentage of lakes with exceeding CLA(FAB)s was compared with the percentage of lakes with ANC below the critical value of 20 µeq/l. The same comparison was made for CLAs calculated with the steady-state water chemistry model (CLA(SSWC)).

[27] http://www.meteoswiss.admin.ch/home/climate/past/climate-normals/norm-value-charts.html
[28] http://www4.ti.ch/dt/da/spaas/uacer/temi/aria/per-saperne-di-piu/rapporti-e-studi

4.13.3 Acidification status

At present (2015-2017) fully 25% of the analysed potentially acid-sensitive lakes (52) had autumn ANC values below 20 µeq/l and 10% had pH values below 6.0 (Figure 53, Figure 54). This was also the case when comparing autumn data for 20 lakes with data from after snowmelt sampling (25% for ANC and 15% for pH). However, mean ANC and pH values were lower after snowmelt (27 µeq/l and 6.4, respectively) than in autumn (374 µeq/l and 6.6, respectively), suggesting that during snowmelt the percentage of lakes with critical ANCs might be higher. In addition to seasonal variations, interannual variability of meteorological conditions also influences lake chemistry (Rogora et al. 2013). In particular, extreme episodes (e.g. heavy rainfall and snowfall) can temporarily decrease lake alkalinity by dilution with an overall acidification effect.

Compared to the past, the present acidification status has improved (Figure 53, Figure 54). During the large-scale survey in 1995, 40% and 29% of the 45 analysed lakes had autumn ANC and pH values below 20 µeq/l and 6.0, respectively. Trend analysis performed for 20 lakes and for the two periods 1980s-2016 and 2000-2016 showed that in the more recent period the number of lakes with significantly increasing total alkalinities (Gran Alkalinity) was lower (10) compared to the entire monitoring period (18), as well as the mean of the Kendall slopes (0.57±0.36 µeq/l/yr compared to 0.63±0.25 µeq/l/yr), indicating a slower in chemical recovery of ANC during the last years (Steingruber 2017).

Decreasing deposition of S is the main reason for the observed chemical recovery (Rogora et al. 2013). However, at present N accounts for about 80% of the acidifying deposition (Rogora et al. 2016), which means that for further recovery of lake chemistry, emissions of N must be significantly reduced.

Next to seasonality, meteorology and atmospheric depositions, climate change also indirectly influences lake chemistry. Analysis of meteorological data (MeteoSvizzera 2012) has shown for the Ticino region an average increase of air temperature during the last 50 years of 0.5°C per decade, with an increase of the temperature rise after 1990. Direct consequences have been decreasing snow falls and snow cover periods (MeteoSvizzera 2012), melting of glaciers (Bauder et al. 2017), permafrost and rock glaciers (PERMOS 2016). Lower snow pack and shorter snow cover periods cause less dilution during snowmelt and reduction of acidic spring episodes. At the same time shorter snow cover periods and thawing glaciers, permafrost and rock glaciers can increase the weatherable surface and together with increasing temperatures they can enhance physical erosion and the export of weathering products to lake water. In fact, an increase of the release of S and BC has been observed for lake catchments at higher altitudes and was related to the presence of thawing permafrost/rock glaciers (Steingruber 2017). Since for the studied lakes the increase in release of S and BC is almost of the same order of magnitude, lake alkalinity was not influenced by this phenomenon. Catchments with permafrost or rock glaciers releasing acidity were described in the Austrian Alps (Thies et al. 2013, Ilyashuk et al. 2014). In fact, crystalline bedrocks in the Alps may contain small amounts of pyrite that is probably preferentially oxidized in the presence of thawing permafrost/rock glaciers to produce sulphuric acid, which in turn can acidify lower situated surface waters, if not sufficiently neutralized through other weathering reactions (Ilyashuk et al. 2014, Williams et al. 2006, Thies et al. 2013). Increasing temperatures might also influence the N cycle and possibly N retention in the catchments. However, data on this are not available.

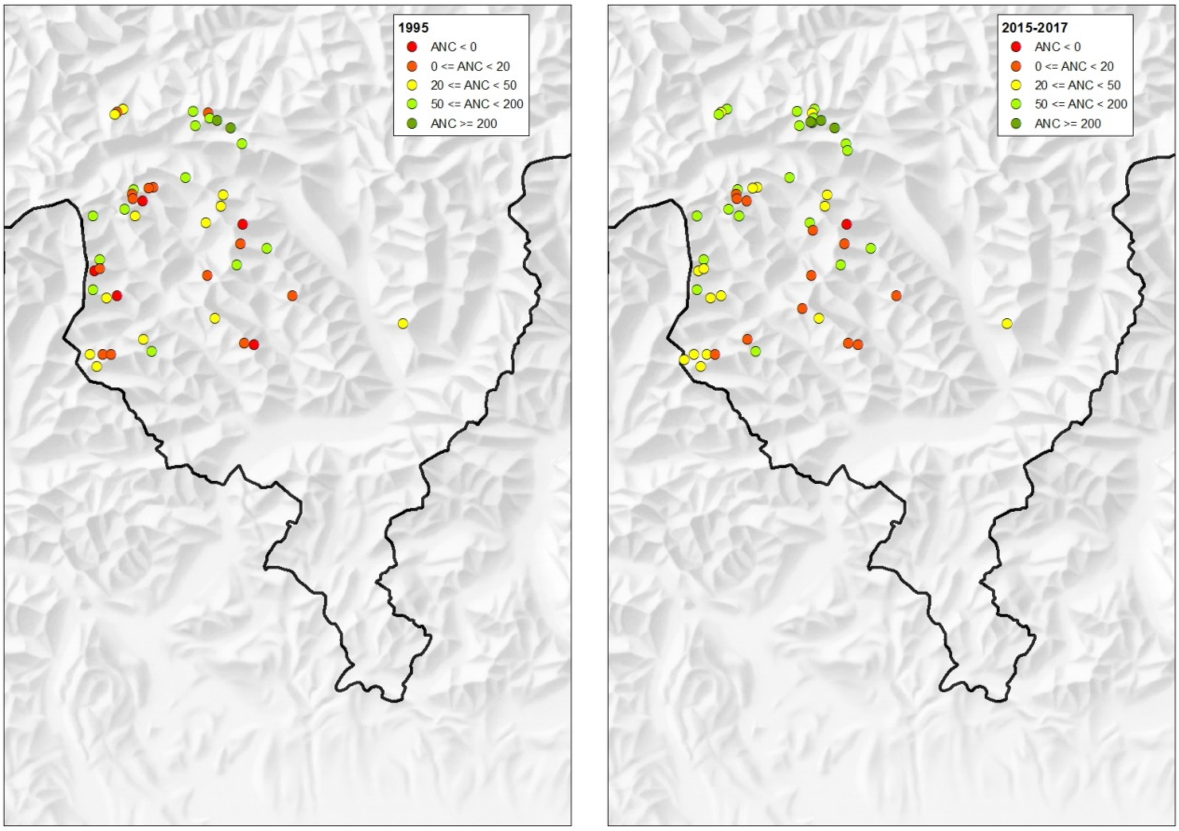


Figure 53 ANC (μeq/l) in potentially acid-sensitive lakes in Switzerland in 1995 and at present (2015-2017).

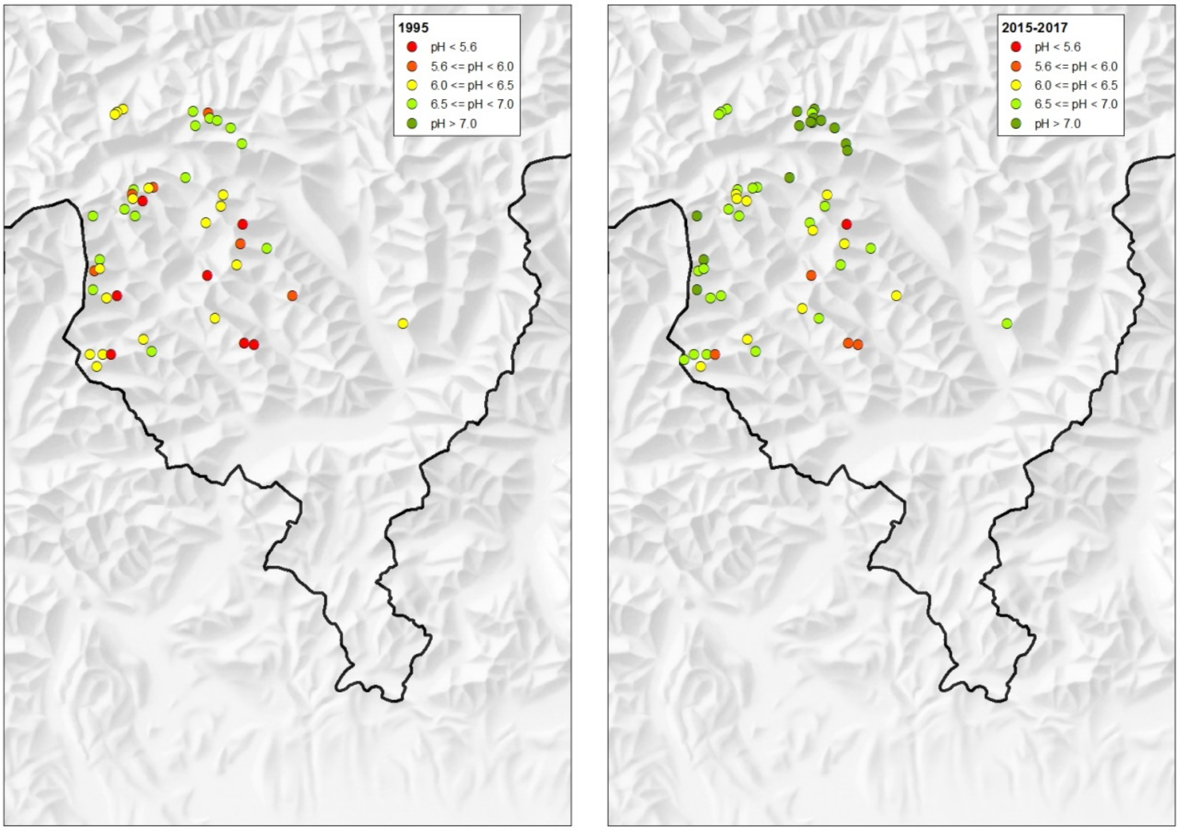


Figure 54 pH in potentially acid-sensitive lakes in Switzerland in 1995 and at present (2015-2017).

Modelled deposition of S and N exceeded CL(FAB)s in 87% of the monitored lakes during 1994-1997, while at present (2014-2016) about 75% are exceeded (see Steingruber 2017 for details on deposition estimates). The main reason for the discrepancy between exceedances of CL(FAB)s and the measured ANC values is an underestimation of the present weathering rates in the FAB model for most lakes (see comparison between modelled and measured BC fluxes in Posch et al. 2006). In fact, BC leaching was explicitly formulated in terms of its sources and sinks in the catchment and not derived from present-day water chemistry as it is done in in the SSWC model. Actually, not only output fluxes of BC were underestimated but for many lakes also output fluxes of S, because geologic S was not considered in the mass balance. Underestimation of both BC and S causes underestimation of CL(FAB) only when underestimation of BC is larger than underestimation of S, because then the errors do not cancel each other out. Similarly, the recently observed increase of BC and S release from lake catchments with thawing permafrost should not greatly influence the calculation of CLAs, because their rates of increase are almost the same (Steingruber 2017). On the other hand, for catchments with permafrost or rock glaciers releasing acidity as described elsewhere in the Alps (Thies et al. 2013, Ilyashuk et al. 2014) this new phenomenon must be considered in the CLA calculation. As regards comparison of measured ANC with exceedances of CLA(SSWC)s, results are more similar: exceedance occurred in 49% of the monitored lakes during 1994-1996 and in 15% of the lakes at present.

4.13.4 Conclusion

Because of the decrease of especially sulphur but also nitrogen deposition, the status of acid-sensitive Swiss lakes significantly improved: at present only 25% have autumn ANC values below the critical value of 20 µeq/l. However, this percentage might temporarily increase during snowmelt or heavy precipitation events. Similarly, although not the focus of these investigations, low ANC values may also occur in high altitude small watercourses with catchments on crystalline bedrock during the same periods. Climate change also influences lake water chemistry. In some lakes, especially if influenced by thawing permafrost or rock glaciers, concentrations of sulphate and base cations can increase as a consequence of enhanced weathering despite decreasing sulphate concentrations in rainwater. Finally, to continue chemical recovery of Alpine surface waters it is necessary to further decrease the emissions of acidifying pollutants, particularly of N, since it has become the dominating acidifying agent, but also because N deposition in the lake catchments presented here (9-14 kg N/ha/yr) far exceeds the critical loads for nutrients of 3-5 kg N/ha/yr suggested for alpine oligotrophic lakes (de Wit and Lindholm 2010).

Acknowledgement: This study was supported by the Swiss Federal Office for the Environment through the ICP Waters Programme.

4.13.5 References

Achermann B., Meier R., Rihm B., Kurz D., Braun S., Kohli L. and Roth T. 2015. National Focal Centre Report - Switzerland. In: Slootweg J., Posch M., Hettelingh J.-P. (eds.), Modelling and mapping the impacts of atmospheric nitrogen and sulphur: CCE Status report 2015, Coordination Centre for Effets (CCE), Bilthoven, The Netherlands, 184 pp.

Bauder A., Fischer M., Funk M., Gabbi J., Hoelzle M., Huss M., Keppenberger G. and Steinegger U. 2017. The Swiss Glaciers 2013/2014 and 2014/2015. Glaciological Report (Glacier) No 135/136. Cryospheric Commission of the Swiss Academy of Sciences, Zürich, Switzerland, 138 pp.

De Jong J.E. 1996. The methodology for the calculation of critical loads of acidity for high altitude lakes – application on lakes in Canton Ticino (Switzerland). Technical Report no. 44, Istituto Italiano di Idrobiologia, Pallanza, Italy.

de Wit H.A. and Lindholm M. 2010. Nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters – a review. NIVA-report 6007-2010. ICP Waters report 101/2010. Norwegian Institute for Waters Research (NIVA), Oslo, Norway, 39 pp.

FOWG. 2005. The 1:500000 geological map of Switzerland. Federal Office for Water and Geology (FOWG), Berne, Switzerland.

Ilayshuk B.P., Ilayshuk E.A., Psenner R., Tessadri R and Koinig K.A., 2014. Rock glacier out!ows may adversely affect lakes: lessons from the past and present of two neighboring water bodies in a crystalline-rock watershed. Environ. Sci. and Technol. 48: 6192-6200.

Meteo Svizzera 2012. Rapporto sul clima–Cantone Ticino 2012. Meteo Svizzera, Locarno Monti, Switzerland, 63 pp.

PERMOS. 2016. Permafrost in Switzerland 2010/2011 to 2013/2014. Glaciological Report (Permafrost) No. 12-15. Cryospheric Commission of the Swiss Academy of Sciences, Zürich, Switzerland, 138 pp.

Posch M., Eggenberger U., Kurz D. and Rihm B. 2007. Critical loads of acidity for Alpine lakes. A weathering rate calculation model and the generalized First-order Acidity Balance (FAB) model applied to Alpine lake catchments. Environmental studies no. 0709. Federal Office for the Environment (FOEN), Berne, 69pp.

Rihm B. 1994. Critical loads of acidity for forest soils and alpine lakes - steady-state mass-balance method. Environmental Series Air no. 238. Federal Office of Environment, Forests and Landscape (FOEFL), Berne, Switzerland, 68pp.

Rogora M., Colombo L., Lepori F., Marchetto A., Steingruber S. and Tornimbeni O. 2013. Thirty Years of Chemical Changes in Alpine Acid-Sensitive Lakes in the Alps. Water Air Soil. Pollut. 224: 1746.

Rogora M., Colombo L., Marchetto A., Mosello R., Steingruber S. 2016. Temporal and spatial patterns in the chemistry of wet deposition in Southern Alps. Atmos. Environ. 146: 44-54.

Steingruber S. 2015. Deposition of acidifying and eutrophying pollutants in Southern Switzerland from 1988 to 2013. Boll. Soc. Tic. Sci. Nat. 103: 29-37.

Steingruber S. 2017. Results from the participation of Switzerland to the International Cooperative Programme on Assessment and Monitoring Effects of Air Pollution on Rivers and Lakes (ICP Waters). Annual report 2016. Dipartimento del territorio del Canton Ticino, Bellinzona, Switzerland, 66pp.

Steingruber S. 2017. Trends in input-output budgets of S and N in Swiss high-altitude lakes. In: De Wit, H., Garmo, Ø. (editors). 2017. Proceedings of the 33rd Task Force meeting of the ICP Waters Programme in Uppsala, May 9-11, 2017. ICP Waters report 131/2017. Norwegian Institute for Waters Research (NIVA), Oslo, Norway, 60 pp.

Thies H., Nickus U., Tolotti M., Tessadri R. and Krainer K. 2013. Evidence of rock glacier melt impacts on water chemistry and diatoms in high mountain streams. Cold Reg. Sci. and Technol. 96: 77-85.

Williams M.W., Knauf M., Caine N., Liu F. and Verplanck P.L. 2006. Geochemistry and source waters of rock glacier outflow, Colorado Front Range. Permafrost and Periglac. Process. 17: 13-33.

4.14 United Kingdom

Don Monteith[1], Chris Curtis[2], Chris Evans[3]
[1] Centre for Ecology & Hydrology, Lancaster Environment Centre
[2] University of the Witwatersrand, Johannesburg
[3] Centre for Ecology & Hydrology, Bangor

4.14.1 Acid sensitivity

The diverse geology of the UK results in large spatial gradients in acid sensitivity. The most sensitive waters drain catchments characterized by acid soils overlying poorly weathering bedrock, including granites, gneisses, sandstones, schists and shales (Figure 55). These are most common in upland regions to the north and west, in areas including Dartmoor (south-west England), mid-Wales, Snowdonia (north-west Wales), the North York Moors (north-east England), the English Lake District (north-west England), Galloway (south-west Scotland), the Trossachs (central Scotland) and the Cairngorms (north-east Scotland).

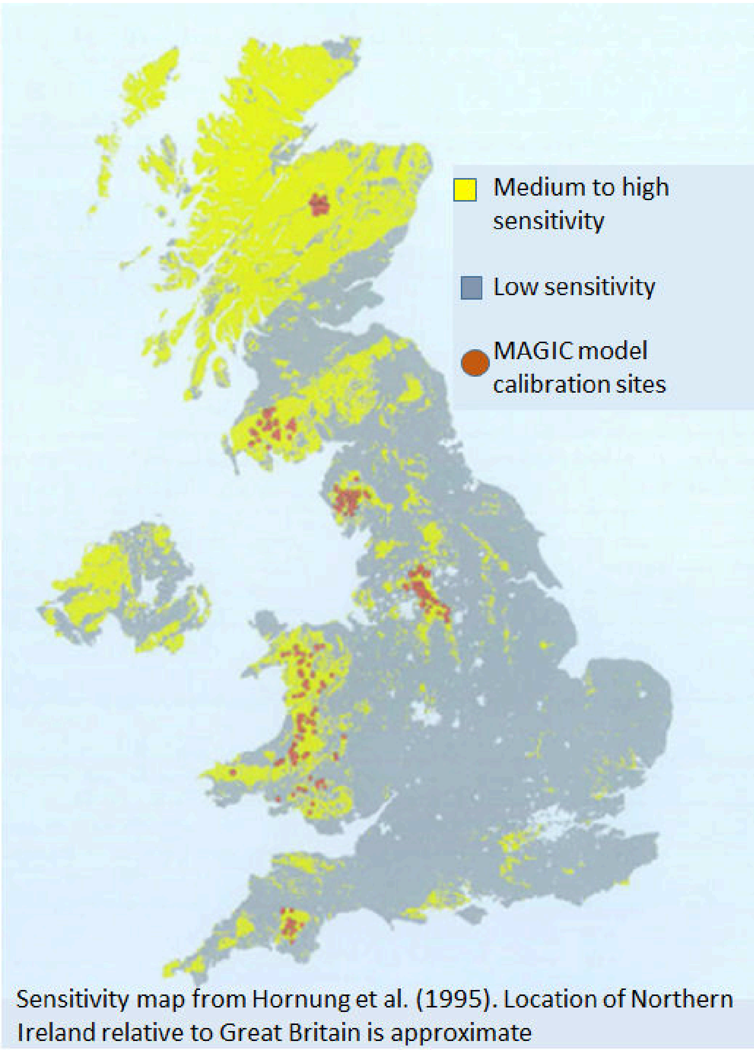


Figure 55 UK freshwater acidification sensitivity map (red dots = MAGIC modelling sites)

Vulnerability to acidification is dependent both on sensitivity and exposure to acid pollutants. Exposure in turn depends on proximity to pollutant sources, the amount of precipitation and the amount of interception provided by the vegetation canopy. Hence the acid deposition gradient also runs largely from south-east (high) to the north-west (low) of the UK, reflecting population densities and (historically) the location of major fossil fuel burning power stations and other industrial emission sources. Acid pollutant fluxes are also dependent on seeder-feeder processes, and are thus relatively higher at sites exposed to higher amounts of precipitation, and in catchments covered by coniferous forest plantations relative to most other vegetation types.

4.14.2 Monitoring and assessment approaches

Over the past three decades monitoring of surface water acidification has been conducted by the UK Upland Waters Monitoring Network (UWMN). The UWMN includes water chemistry sampling of the outflows from small lakes (quarterly) and low order streams (monthly), in addition to annual biological sampling (aquatic macrophytes, epilithic diatoms and macroinvertebrates), from many regions of the UK uplands (Battarbee et al. 2014). The UWMN is an expansion of the former UK Acid Waters Monitoring Network (AWMN), which was established to assess the efficacy of regional reductions in acid pollutants. AWMN sites were selected on the criteria of low geological acid-buffering capacity and the absence of marked local catchment interference or point source contamination that might otherwise complicate acidification/recovery signals. The main exception to the latter criterion was the selection of a subset of sites with significant conifer plantation forestry. The remaining sites were either subject to low-intensity grazing (mainly by sheep or deer) which maintains a grassland or heathland cover, or were located above the natural tree line. The last comprehensive assessment of trends in the chemical and biological status of UWMN sites was reported in a special issue of Ecological Indicators in 2014 (Battarbee et al., 2014).

UK data submitted for the current regional analysis are derived from a survey of lakes and streams conducted in the autumn of 2010, as part of the UK Defra-funded Freshwater Umbrella project (AQ0803). The purpose of the survey was to assess the acidification status of a subset of 445 previously sampled acid-sensitive sites, where deposition estimates provided by the FRAME model suggested critical loads would continue to be exceeded by 2020. Sites comprised natural lakes and ponds, reservoirs and streams ranging in altitude from close to sea level to 1123 m. A total of 163 of these sites (lake outflows and streams) were randomly selected for sampling following stratification by region. Chemistry data for the autumn 2010 survey are available for 157 sites. Major ion chemistry was determined by ion chromatography. Acid Neutralising Capacity for each sample was determined by subtracting the equivalent sum of acid anion concentrations from that of base cation concentrations. In addition, First-Order Acidity Balance (FAB) model critical loads and exceedances for 2020 were calculated.

4.14.3 Acidification status

Acid Neutralising Capacity of the 157 sites in the 2010 survey varied widely (Figure 56). The most acidic sites tended to occur in the North York Moors (mean ANC = -38 µeq/l). Four small streams in this region exhibited ANC values of less than -100 µeq/l. This is consistent with a recent assessment of 51 surface waters in this region by Evans et al. (2014), who concluded that their current severely acidified condition and apparent retarded recovery is primarily due to the release of a legacy sulphur store from the peaty catchment soils and the exacerbating effects of plantation forestry. Overall, the ANC of 24% of the sites was negative, and that of a further 16% of sites fell between 0 – 20 µeq/l. The ANC of samples from 42% of sites was greater than 40 µeq/l. While there are indications from UWMN monitoring data that nitrate is making an increasingly large contribution to total acid anion concentrations as sulphate concentrations have fallen, nitrate comprised on average 20% of the equivalent sum of nitrate and non-marine sulphate concentration, with the highest average contribution occurring in the Northern Irish sites (circa 30%) (Figure 56).

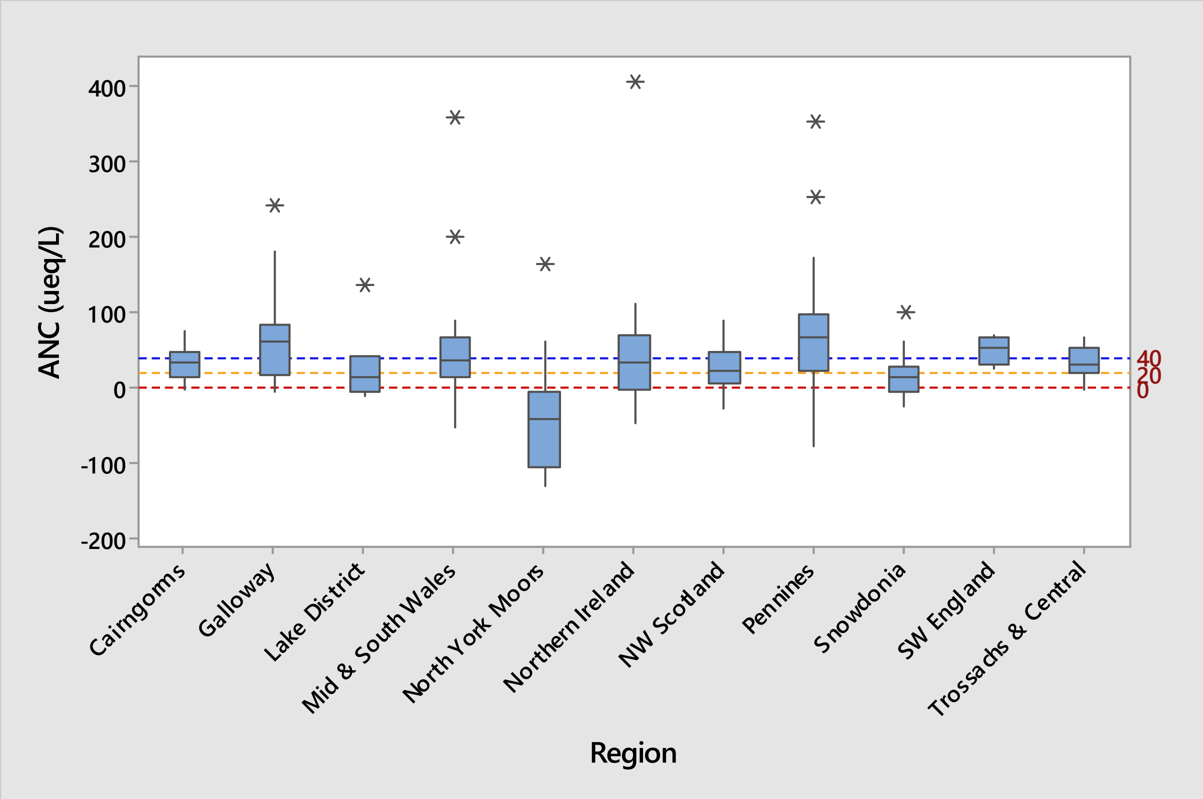
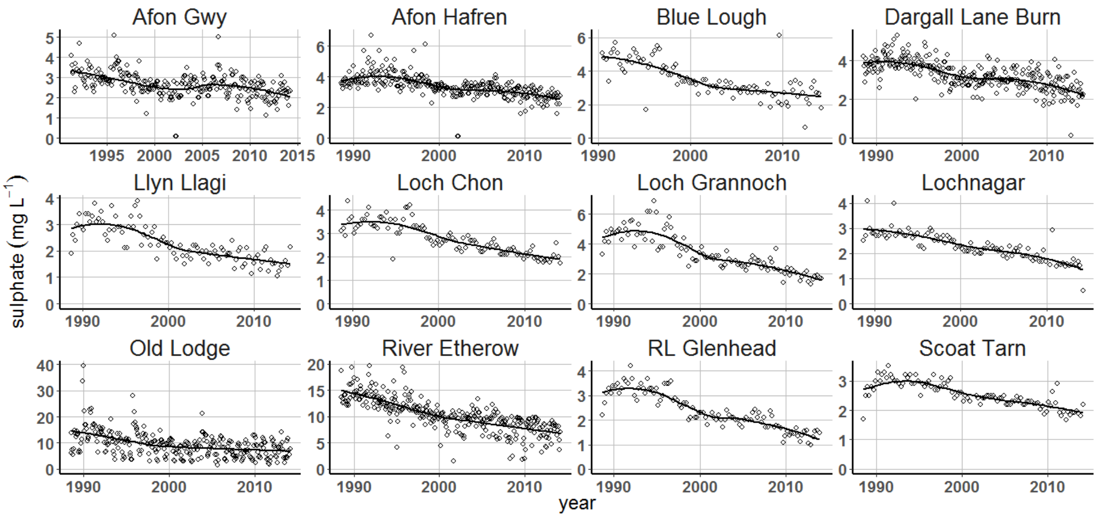


Figure 56 Boxplots illustrating the distribution of ANC values for the 157 sites included in the Autumn 2010 survey. Red, orange and blue lines indicate thresholds of 0, 20 and 40 μeq/l, respectively. See section 4.14.1 for region locations.

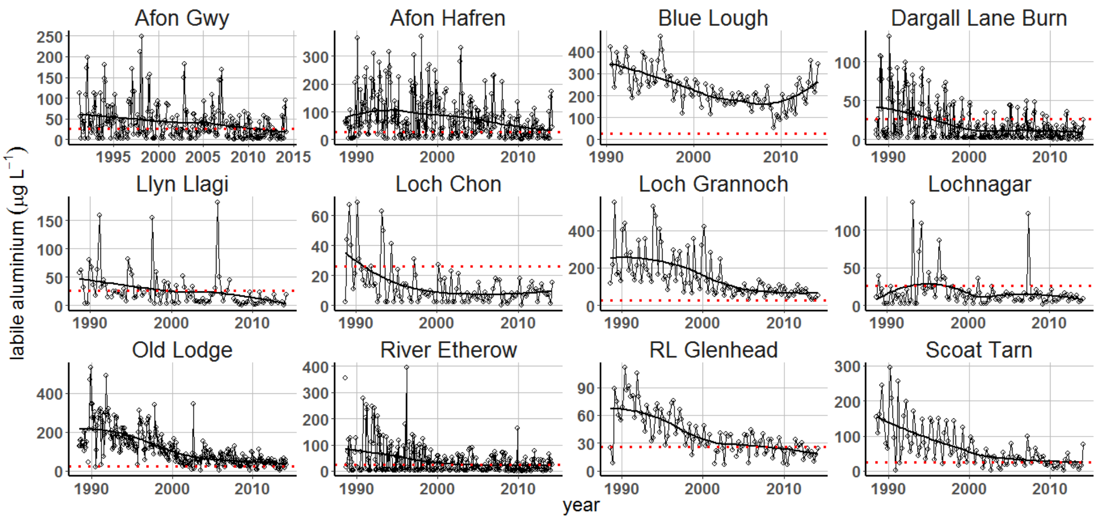
FAB modelling of these data together with additional measurements collected in the spring of 2011 indicated that 28% of sites (previously found to have been exceeded) no longer show exceedance based on 2020 deposition, but the majority of sites continue to be exceeded. This modelling also showed that nitrate contributed a larger proportion of the acidity flux indicated by exceedance, from a minimum of around 50% in some sites in NW Scotland, to a median of around 80% in the Lake District and Northern Ireland. However, a more conservative estimate of nitrate leaching (rather than the worst-case as indicated by FAB) would result in fewer exceeded sites in most regions, and a reduced magnitude of exceedance in all sites (Curtis et al., 2005).

Monitoring of UK Upland Water Monitoring sites has demonstrated progressive reductions in sulphate concentration, and concomitant increases in ANC and pH, and reductions in labile aluminium concentrations (Figure 57). An approximate doubling of dissolved organic carbon (DOC) concentrations in these waters, indicating increased solubility of soil organic matter, is also thought to be an integral part of this chemical recovery process (Monteith et al., 2014). Sulphate concentrations declined most rapidly during the second half of the 1990s, corresponding to a period of substantial reductions in sulphur emissions from UK power plants. Analysis of the first 20 years of data showed that while chemical recovery was occurring in both moorland and forested sites at a similar rate, the latter remain in a more acidified state (Monteith et al., 2014). Figure 57 illustrates that ANC in a number of the most acidic sites on the network now regularly exceeds 20 μeq/l, the UK critical limit for freshwater acidity, although values still fall considerably below this threshold at times of high rainfall and sea salt deposition events. The continued occurrence of severe acid episodes at several sites may at least partly explain why evidence for biological recovery (e.g. in macroinvertebrates and epilithic diatom communities) remains more patchy than the improvement in average chemical conditions might suggest (Monteith et al., 2005; Murphy et al., 2014).

a)

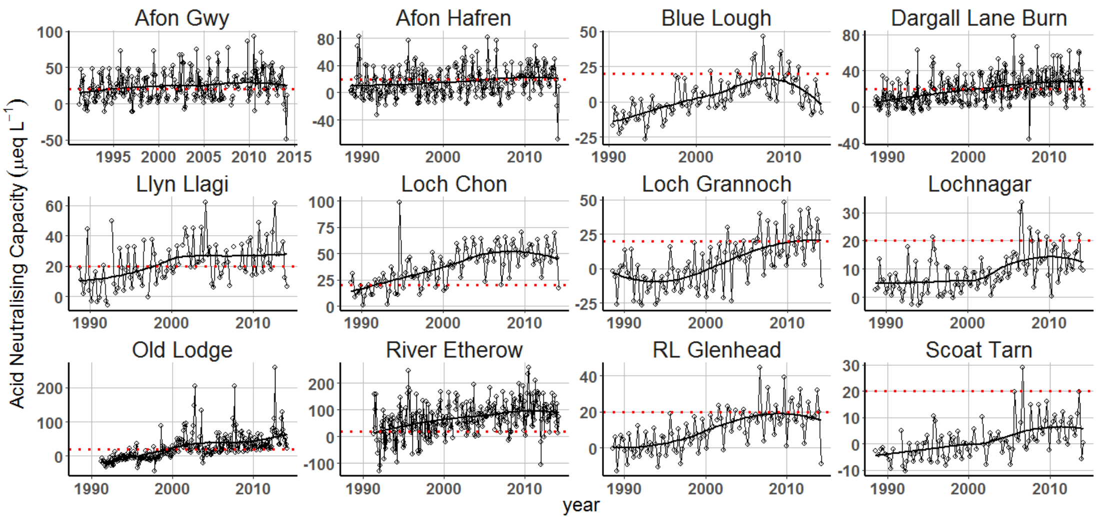


b)



Red line = 26 µg/l = 80% probability of trout in 2/3 or fishing reaches (Malcolm et al., 2014).

c)



Red line = 20 µeq/l

Figure 57 Trends in a) sulphate concentration, b) labile aluminium concentration and c) ANC, at a selection of UK UWMN sites between 1988-2015, including loess smoother (span = 0.75).

4.14.4 References

Battarbee, Richard W.; Shilland, Ewan M.; Kernan, Martin; Monteith, Donald T.; Curtis, Chris J.. 2014 Recovery of acidified surface waters from acidification in the United Kingdom after twenty years of chemical and biological monitoring (1988–2008) [in special issue: Threats to upland waters] Ecological Indicators, 37 B. 267-273.

Curtis, C.J., Evans, C., Helliwell, R.C., Monteith, D.T., 2005. Nitrate leaching as a confounding factor in chemical recovery from acidification in UK upland waters. Environmental Pollution 137, 73–82.

Evans, Chris D.; Chadwick, Tom; Norris, David; Rowe, Edwin C.; Heaton, Tim H.E.; Brown, Philip; Battarbee, Richard W.. 2014 Persistent surface water acidification in an organic soil-dominated upland region subject to high atmospheric deposition: the North York Moors, UK. Ecological Indicators, 37 (B), 304-316.

Hornung, M., Bull, K.R., Cresser, M., Ullyett, J., Hall, J.R., Langan, S., Loveland, P.J., Wilson, M.J., 1995. The sensitivity of surface waters of Great Britain to acidification predicted from catchment characteristics. Environmental Pollution. 87(2):207-14.

Malcolm, I.A., Bacon, P.J., Middlemas, S.J., Fryer, R.J., Shilland, E.M., Collen, P., 2014. Relationships between hydrochemistry and the presence of juvenile brown trout (Salmo trutta) in headwater streams recovering from acidification. Ecological Indicators 37, 351–364.

Monteith, D.T., Hildrew, A.G., Flower, R.J., Raven, P.J., Beaumont, W.R.B., Collen, P., Kreiser, A.M., Shilland, E.M., Winterbottom, J.H., 2005. Biological responses to the chemical recovery of acidified fresh waters in the UK. Environmental Pollution 137, 83–101.

Monteith, D.T.; Evans, C.D.; Henrys, P.A.; Simpson, G.L.; Malcolm, I.A.. 2014 Trends in the hydrochemistry of acid-sensitive surface waters in the UK 1988–2008. Ecological Indicators, 37 B. 287-303.

Murphy, J.F., Winterbottom, J.H., Orton, S., Simpson, G.L., Shilland, E.M., Hildrew, A.G., 2014. Evidence of recovery from acidification in the macroinvertebrate assemblages of UK fresh waters: a 20-year time series. Ecological Indicators 37 B, 330–340.

4.15 United States

John L. Stoddard
U.S. Environmental Protection Agency

4.15.1 Acid Sensitivity

Much of the land area of the United States is characterized by deep, well-buffered soils, and as a result is not sensitive to acidic deposition. Exhaustive research conducted during the National Acid Precipitation Assessment Program (NAPAP; NAPAP (1991)) has identified several regions where surface waters are at risk for acidification, primarily in the Northeastern, Mid-Atlantic and Western regions of the country. These areas are characterized by crystalline bedrock, thin soils and forested (or alpine) land cover (Eilers and Selle, 1991).

Areas especially sensitive to acidification, and where historical acidification has been documented (Figure 58), include forested and mountainous areas of the Northeast—the Adirondack mountains (Driscoll et al., 1991), New England (Kahl et al., 1991), the Northern Appalachian Plateau (Stoddard and Murdoch, 1991) - and southeastern streams in the Valley and Ridge Province and Blue Ridge Province (Cosby et al., 1991). Many lakes in mountainous areas of the West are extremely sensitive, but receive relatively low levels of acidic deposition (Turk and Spahr, 1989; Melack and Stoddard, 1991). Some high elevation western lakes, particularly in the Rocky Mountains, can become acidic during snowmelt, but very little chronic acidification has been observed in the West (Stoddard, 1995; Campbell et al., 2004).

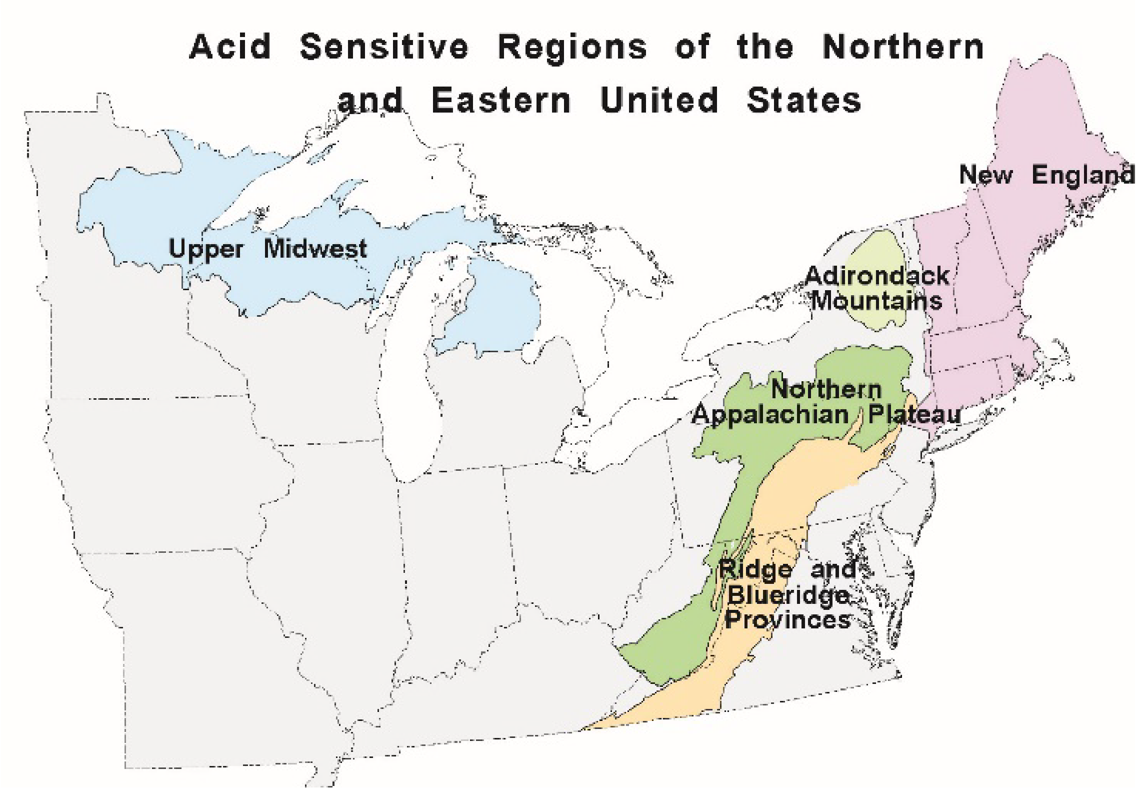


Figure 58 Monitoring for acidification is conducted annually in four regions of the eastern U.S. (Adirondacks, New England, Northern Appalachians, and Blueridge and Ridges). Monitoring is no longer conducted in the Upper Midwest.

Critical loads for surface water acidification (Figure 59) generally follow this same geographic pattern, as do monitoring efforts to assess changes in acidity of surface water in the U.S., with both site-specific and regional monitoring conducted primarily in the Northeast and Mid-Atlantic.

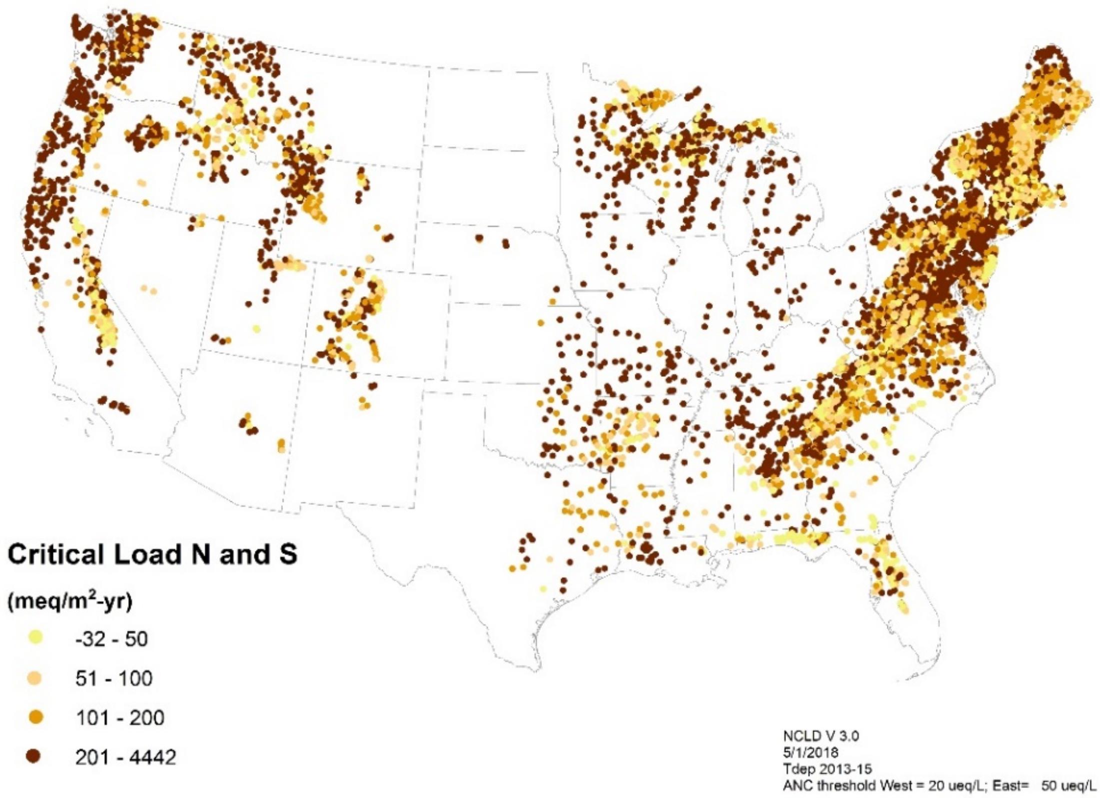


Figure 59 Map of critical loads of acidity for surface waters (Lynch et al., 2017).

4.15.2 Monitoring and assessment approach

An overall assessment of the extent of acidification of surface waters in the U.S. can be gleaned from national surveys of lakes and streams conducted every 5 years as part of an effort to quantify the ecological status of aquatic resources[29]. These are probability surveys, so that the results can be statistically summarized as describing the population of lakes and/or streams in the 48 conterminous states (excluding Hawaii and Alaska)[30]. Approximately 1000 lakes and 2000 streams and rivers are sampled in each round. The most recent lake survey and assessment was conducted in 2012 (U.S. Environmental Protection Agency, 2016). The most recent river and stream survey was conducted in 2013-2014 (U.S. Environmental Protection Agency, 2009).

Because acidification is known to be localized within a small number of regions, the U.S. also conducts probability surveys at higher site densities and higher frequency (annual) in four areas: (1) lakes in the Adirondack mountains; (2) lakes in New England; (3) streams in the Northern Appalachian Plateau; and (4) streams in the Blueridge and Valley and Ridge Provinces (Figure 58). High density complete surveys were conducted in each region in the early- to mid-1990s (1991-94 for lakes; 1993-94 for streams). The most acid-sensitive (defined as gran alkalinity < 100 µeq/l) lakes and streams found in each survey have been sampled annually in the intervening years. For the purposes of this report, we assume that the non-sensitive (and un-monitored) lakes and streams in each region

[29] https://www.epa.gov/national-aquatic-resource-surveys

[30] In a probability survey each site is assigned a weight corresponding to the probability of the site being included in the survey, and these weights are included in the analysis of the data

have not changed their acid/base status since the original surveys were conducted. The population estimates reported in chapter 3.2, and in this chapter, were created by combining the original survey data with the mean values observed at the monitored acid-sensitive sites over the 3 most recent years of sampling (2013-15 for the streams and 2014-2016 for the lakes).
In both the national and regional surveys, sites are sampled once during a summer index period, for a complete suite of chemical variables. The probability nature of the monitoring design makes it possible to present the results as population estimates (e.g., an estimate of the total number or percentage of lakes in the nation or region that are acidic). Because samples are collected in the summer, the results represent chronic acidification.

4.15.3 Acidification status

Nationally, surface water acidification is rare, occurring in less than 1% of lakes, and less than 1% of stream length in the conterminous U.S. (Table 6). Regionally, acidification is more common, particularly in the Adirondack (6% acidic), and Northern Appalachian Mountains (5% acidic).

It is also possible, and important, to place these numbers in a historical context. Figure 60 presents the time sequence for the percentage of acidic systems in each region, beginning with the results of the complete statistical survey completed in each region in the early to mid-1990s (U.S. Environmental Protection Agency, 2000; Whittier et al., 2002). Each panel in Figure 60 shows, at the far left, the percentage of regional systems that were acidic at the time of the last complete survey (1991-94 for lakes; 1993-94 for streams). The larger plots in each panel illustrate how this number has changed annually, up to the time of the most recent available data.

Table 6 Summary of the national and regional datasets used to assess acidification of surface waters in the U.S., with estimates of the number and proportion of acidic lakes/streams in each, according to the most recent available data.

Survey/Resource	Year(s)	Number Acidic	% Acidic
National Lake Survey	2012	310	0.3%
National Stream Survey	2013-2014	5,215*	0.2%
Adirondack Lakes	2014-2016	110	6%
New England Lakes	2013-2016	200	3%
No. Appalachian Streams	2012-2015	1,909*	5%
Blueridge/Ridges Streams	2012-2015	790*	2%

* Total acidic stream length (km)

In three of the acid-sensitive regions, the proportion of lakes or streams that are acidic has been roughly halved (from 13% to 6% in Adirondack lakes; from ca. 6% to 3% in New England lakes; and from ca. 12% to less than 5% for Northern Appalachian streams). These data suggest little or no recovery (measured as a decline in the percent of acidic streams) in the Blueridge/Ridges provinces. In all cases, there is considerable year-to-year variability in these estimates, suggesting that many lakes and streams are near the 0 ANC threshold we use to define acidification, and fluctuate from slightly acidic to non-acidic annually, depending on climatic factors.

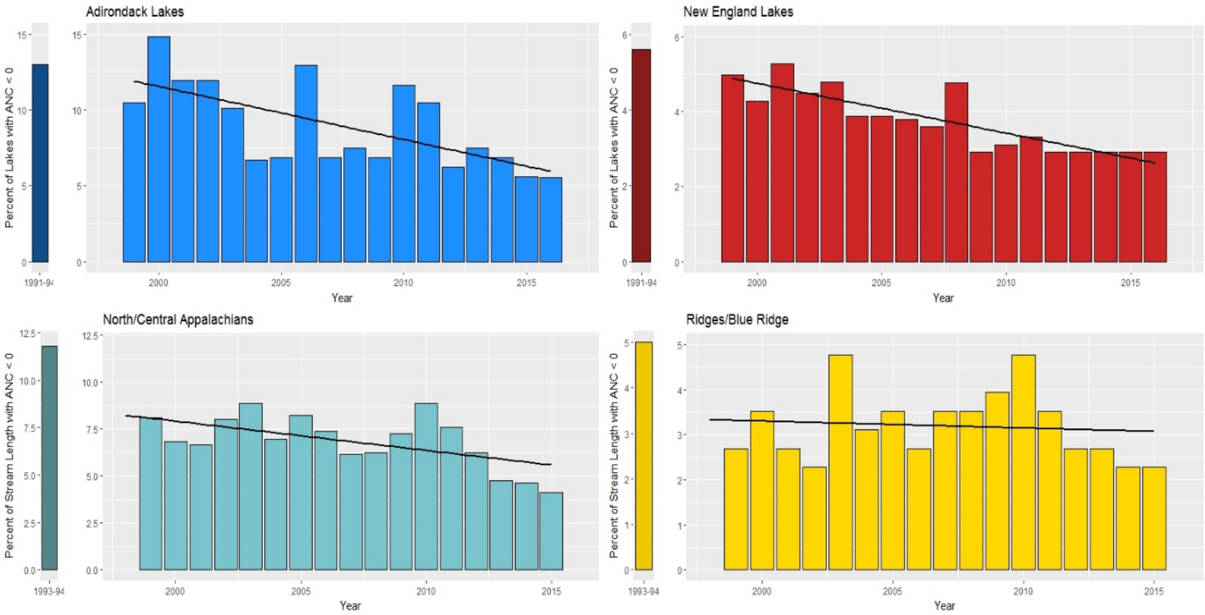


Figure 60 Percent of acidic lakes (top panels) and stream length (bottom panels) over time. Trend lines were generated by linear regression.

4.15.4 References

Campbell, D. H., E. Muths, J. T. Turk, and P. S. Corn. 2004. Sensitivity to acidification of subalpine ponds and lakes in north-western Colorado. Hydrological Processes 18:2817-2834.

Cosby, B. J., P. F. Ryan, R. Webb, G. M. Hornberger, and J. N. Galloway. 1991. Mountains of West Virginia. Pages 297-318 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

Driscoll, C. T., R. M. Newton, C. P. Gubala, J. P. Baker, and S. W. Christensen. 1991. Adirondack Mountains. Pages 133-202 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

Eilers, J. M., and A. R. Selle. 1991. Geographic Overview of the Regional Case Study Areas. Pages 107-125 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

Kahl, J. S., S. A. Norton, C. A. Cronan, I. J. Fernandez, L. C. Bacon, and T. A. Haines. 1991. Maine. Pages 203-235 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

Lynch, J.A., Phelan, J., Pardo, L.H., McDonnell, T.C. and Clark, C.M. 2017. Detailed Documentation of the National Critical Load Database (NCLD) for U.S. Critical Loads of Sulfur and Nitrogen, version 3.0. National Atmospheric Deposition Program, Illinois State Water Survey, Champaign, IL.)

Melack, J. M., and J. L. Stoddard. 1991. Sierra Nevada. Pages 503-530 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

NAPAP 1991. The experience and legacy of NAPAP. The Oversight Review Board of the National Acid Precipitation Assessment Program.

Stoddard, J. L. 1995. Episodic acidification during snowmelt of high elevation lakes in the Sierra Nevada Mountains of California. Water Air and Soil Pollution 85:353-358.

Stoddard, J. L., and P. S. Murdoch. 1991. Catskill Mountains. Pages 237-271 in D. F. Charles, editor. Acidic Deposition and Aquatic Ecosystems: Regional Case Studies. Springer-Verlag, New York, NY.

Turk, J. T., and N. E. Spahr. 1989. Chemistry of Rocky Mountain lakes. Pages 181-208 in D. C. Adriano and M. Havas, editors. Acid Precipitation. Volume 1. Case Studies. Springer-Verlag, New York.

U.S. Environmental Protection Agency. 2000. Mid-Atlantic Highlands Streams Assessment. EPA/903/R-00/015, U.S. Environmental Protection Agency, Region 3, Philadelphia, PA.

U.S. Environmental Protection Agency. 2009. National Rivers and Streams Assessment Field Operations Manual. U.S. Enviromental Protection Agency, Washington, DC.

U.S. Environmental Protection Agency. 2016. National Lakes Assessment 2012: A Collaborative Survey of the Nation's Lakes. U.S. Environmental Protection Agency, Washington, DC.

Whittier, T. R., S. G. Paulsen, D. P. Larsen, S. A. Peterson, A. T. Herlihy, and P. R. Kaufmann. 2002. Indicators of ecological stress and their extent in the population of northeastern lakes: A regional-scale assessment. BioScience 52:235-247.

5 Discussion

5.1 Current extent of acidification

5.1.1 Countries covered by the country reports

The country reports and data submitted by the NFCs give an overview of the current acidification status in a range of European countries, as well as the US and Canada. Many of the country reports also show and give examples of the acidification history leading up to the current situation. It is evident that the systems are generally in recovery: The chemistry at Canadian (Figure 14), Czech (Figure 19), Dutch (Figure 35), Finnish (Figure 22), Italian (Figure 27), Irish (Figure 26), Swiss (Figure 53) and UK (Figure 57) sites has clearly improved; The percentage acidified sites has declined in both Sweden (Figure 50) and the US (Figure 60); All the German sites showed a significant decline in sulphate and increase in pH (Table 5). Despite the observed chemical recovery, surface water acidification still occurs in most of the countries, although the extent and severity vary strongly.

It was not possible, based on the submitted data, to analyse the extent of surface water acidification in terms of total area where acidification occurs or the total number of acidified lakes or rivers/streams, but such analyses are available in some of the country reports. In the following, the different sources of information, including the WFD data, are summarised and compared.

In Europe the largest areas with acidification are found in **Norway**, **Sweden** and **the UK**. In Norway acidified waters are found in the southern and south-western parts of the country (Figure 38). Modelling shows that lakes in 7% of the country area are acidified, and in some areas they are strongly acidified (Figure 40). This percentage is not entirely comparable with the percentage of sites shown in Table 3, since it refers to the country area, but it is clearly lower. This is because the selected sites are acid-sensitive and have a bias towards acidified sites (except the rivers), and that lakes and rivers in part of the country have naturally low ANC and are thus not acidified according to the national WFD criteria (cf. section 4.9.2). In Sweden 10% of the lakes are acidified, according to the definition used in the country report, which is stricter than the ANC < 20 µeq/l criterion (cf. section 4.12.2). Most of the acidified lakes are located in southern Sweden (Figure 49). The WFD data indicate a slightly higher percentage of acidified lakes, as well as acidified rivers. However, the proportion of water bodies with unknown acidification status is high, so it is difficult to say how representative these data are. According to the NFC the WFD classified water bodies may be somewhat biased towards acidified water bodies. The 10% estimate, on the other hand, is based on a random selection of all Swedish lakes. The UK sites are selected among sites with predicted critical loads exceedance in 2020, so it is to be expected that the proportion of acidified sites is high. The distribution of the acidified sites (Figure 7) indicates that acidification in the UK is a regional issue, focussed particularly on upland sites in northern England (e.g. the North York Moors), but also north Wales and southern Scotland. The proportion of acidified UK water bodies according to the WFD reporting is considerably lower, reflecting a site selection that covers the whole country and includes larger, lower lying, and less sensitive catchments. However, about 1/3 of the lake water bodies have unknown acidification status.

The proportion of acidified sites in **Finland** and **Germany** is uncertain given the low number of submitted sites. The Finnish WFD data cover only river water bodies. Again the proportion of acidified water bodies is uncertain given that about 1/3 have unknown status, but the numbers are slightly lower than those for the submitted data for lakes, probably because they also represent less acid-sensitive areas. The distribution of the acidified lakes and the existence of sensitive areas throughout the country (Figure 21) indicate that acidification is present in many parts of the country, although it may not be as severe. This is also confirmed by the exceedance of critical loads (Figure 1).

For Germany, 2% of the river water bodies are reported as having less than good acidification status. Nearly all have atmospheric deposition as a significant pressure, while less than 20% of them have the impact acidification. According to the NFC, one cannot assume that the classified water bodies are representative of the acid-sensitive areas (which would give 9% acidified river water bodies in these areas), despite the low proportion in unknown status. The classification of acidification status is also inconsistent. However, the WFD results indicate that acidification still occurs and it can be associated with atmospheric deposition, but it affects only a small proportion of the water bodies. As for Finland, acidification occurs in different parts of the country, but it is less extensive while slightly more severe in Germany.

According to the country report, the extent of acidified surface water bodies in the **Czech Republic** is less than 1% of the country area, or 2% of the monitored streams. However, this is based on base flow stream data. The WFD data indicate acidification to a larger extent, although if considering only the water bodies with both atmospheric deposition pressure and acidification impact the proportion is similar to that of the base flow data. According to the NFC, several of the water bodies with less than good acidification status are not considered to be acidified, and the WFD classification criteria are unclear. The country report shows that acidification is limited to selected parts of the country only, and here the monitoring data suggest a high proportion (30-40%) of acidified sites. Many of the sites are also strongly acidified (ANC < -20 µeq/l). These include some streams at higher altitude in the middle part of the country, as well as glacial lakes in the Bohemian forest in the south-west. **Switzerland** has only a relatively small region with sensitive lakes in the south, but 10-25% of the lakes can be considered as acidified, depending on the criteria. In **Ireland**, about 10% of the lakes in the acid-sensitive areas are considered impacted by acid deposition. The sites are found in the north-west and south-west. Some lakes are severely acidified. In **the Netherlands** acid sensitivity is mainly associated with small moorland lakes and ponds, and around half of those monitored are acidified. Although some small headwater streams are susceptible to acidification (van Dam and Mertens, 1995), the proportion of water bodies in less than good acidification status according to the WFD seems unlikely. None of these water bodies are reported as having the impact acidification, so it is unclear why they are assigned to this status class.

Poland and **Italy** have each only one water body with ANC less than 20 µeq/l, but there are also water bodies at risk (ANC 20-50 µeq/l, especially in Italy). Italy does report seven lakes as having less than good acidification status under the WFD, but according to the NFC this could be an erroneous reporting or acidification can be ascribed to other causes than atmospheric deposition. Only small water bodies, mainly high altitude lakes, not covered by the WFD, are acidified or at risk in Italy. Similarly, Poland reports 1330 water bodies as acidified under the WFD, but hardly any with the impact acidification or the pressure atmospheric deposition. According to the NFC a negligible number of rivers fail the pH criterion, and for lakes pH is not even used in the assessment of ecological status. The basis for the reported acidification status is not known, and the results should not be considered relevant. The high number of water bodies in less than good acidification status with no significant pressure and/or impact also raises questions about this reporting.

The country report from **Spain** showed that although acid-sensitive surface waters are found in part of the country, acidification effects have been marginal, owing in part to the buffering of acid deposition by dust from North Africa. Only in some Pyrenean lakes a moderate acidification has been detected. The WFD data indicate that there is a small proportion of water bodies in less than good acidification status. However, neither have atmospheric deposition as a pressure, and few have the impact acidification. According to the NFC the acidification status of these water bodies is related to land-use pressures and not to atmospheric deposition. Surface waters in **Latvia** are generally not acid-sensitive, and acidification is not an issue. This is confirmed by the WFD data.

In the **US** and **Canada**, acidification occurs in some, large regions. From the national surveys in the US only 1% of lakes, and less than 1% of the stream length is acidic. In the four sensitive regions in the eastern part of the country, 3-6% of lakes or stream length is acidic, given the national criteria and statistically representing all lakes and streams in these regions. In Canada the percentage of acid impacted lakes in the sensitive regions is 9-12% according to the country report. Using common criteria (Table 3), the percentage of acidified sites is more similar, but still somewhat higher in Canada than in the US. The acidified lakes are mainly found in the Canadian Shield region and on the north-western coast.

In summary, despite reduced deposition levels and recovery, surface water acidification still occurs in most of the countries covered by the country reports (Table 7). In Norway, Sweden, the UK, Canada and the US it occurs in large regions. In Finland and Germany acidification is more scattered, while in the Netherlands, the Czech Republic, Ireland, Italy, Switzerland and Poland it is limited to a few smaller areas. The severity of acidification is not necessarily related to the spatial extent of the problem; local hot spots may occur even where acidification is not a major, regional issue, e.g. in the Czech Republic and Ireland. However, based on both extent and severity, surface waters in Italy and Poland and, to a lesser degree, Finland, Germany, Switzerland, Canada and the US seem closer to chemical recovery than waters in the other countries which submitted data. In Spain acidification is a limited issue, and in Latvia it is unlikely to occur at all.

5.1.2 Countries not covered by the country reports

The country reports give a good overview of surface water acidification in several countries. For North America, the national programmes of Canada and the United States have thoroughly evaluated the extent of surface water acidification, and it appears unlikely that major areas with acidified surface waters have been overlooked (see country reports). For Europe there may be areas with acidified surface waters outside those covered by the country reports. The analysis of bedrock and deposition in chapter 3.1.1 suggests some regions where acidification may occur. However, this can only be confirmed by the collection and analysis of water samples.

Several large European projects conducted in the 1980s and 1990s focused on mountain lakes, and included studies of acidification and recovery from deposition of long-range transported air pollutants. These projects entailed sampling and collating water chemistry data from small lakes, many of which lie in acid-sensitive terrain. The results have been analysed with respect to critical loads, exceedance, and recovery from acidification. Several of the sites are not included in the ICP Waters monitoring network. The first of these projects were AL:PE and AL:PE2 (Alpine Lakes: Paleolimnology and Ecology; Mosello et al., 1995). This was followed by MOLAR (Mountain Lake Research; The MOLAR Water Chemistry Group, 1999) and then EMERGE (European Mountain lake Ecosystems: Regionalisation, diaGnostic & socio-economic Evaluation; Curtis et al., 2005; Camarero et al., 2009). Finally RECOVER:2010 (Predicting Recovery In Acidified Freshwaters By The Year 2010, And Beyond) looked at many of these same mountain lakes and evaluated the recovery expected given full implementation of the Gothenburg protocol (Ferrier et al., 2001). A broad-scale study of the extent of surface water acidification in the UN-ECE region was conducted in the late 1980s (Merilehto et al., 1988) through e.g. literature review and questionnaires.

Belgium and Luxembourg: Small, acid-sensitive lakes and ponds, similar to the ones in the Netherlands (cf. chapter 4.7.4) are found in the northern part of Belgium (the Campine region) (Vangenechten et al., 1981), and evidence of anthropogenic acidification was found in the 1980s (Merilehto et al., 1988). Although rates of acid deposition are somewhat lower than in the Netherlands, nitrogen deposition especially is still high, so it is likely that some of these lakes are acidified also on the Belgian side of the border. Acid-sensitive streams are found in the south-eastern part of the country, but only natural acidification was reported in the 1980s (Merilehto et al., 1988).

No recent studies of surface water acidification in Belgium were found. In the WFD reporting, 3 and 17% of the river and lake water bodies, respectively, exhibited the combination of less than good acidification status, the impact acidification and the pressure atmospheric deposition. Only 2/3 of the river water bodies were classified, making these numbers more uncertain. However, the WFD reporting indicates that surface water acidification caused by air pollution is likely to be an issue in Belgium. No literature on surface water acidification in Luxembourg was found, but it may occur for landscape types similar to the acid-sensitive areas in Belgium. 4% of the river water bodies have less than good acidification status according to the WFD reporting. All are reported to be exposed to the pressure atmospheric deposition, but none have acidification as impact.

France: Potentially sensitive areas (red areas in Figure 2) have been studied in the past. In some areas buffering is enhanced due to e.g. the presence of clay in the sandstone, the presence of regoliths, or deep soils, while in other areas impermeable layers prevent vertical drainage and reduces the buffering. Surface water acidification has mainly been observed in the Vosges mountains (north-eastern France), a massif on silicate bedrock just west of the Rhine River valley, opposite the Black Forest, Germany, on the east of the Rhine River. The region receives acid deposition. Massabuau et al. (1987) reported acidified and fishless streams in the southern part of the massif, and Kreiser et al. (1995) reported on the state of its acidified lakes. In 1992, an exhaustive survey of 800 streams carried out in a joint study on critical loads in this area identified 7.5% of the streams as acidic (Party et al., 1995). A study using data up until 2005 (Angeli et al., 2009) showed a slow increase in stream pH on granite bedrock, but no recent recovery in pH on sandstone. Acidified streams have also been observed in the Ardennes (Fevrier et al., 1999). No recent surveys have been conducted in these areas. A very low proportion of the river water bodies are reported with less than good acidification status under the WFD, and the acidification is never due to atmospheric deposition, according to the pressure reporting.

Slovakia: Widespread and severe acidification in the small alpine lakes of the Tatra Mountains (also on the Polish side, cf. section 4.10.5) has been well-documented (Kopáček and Stuchlik, 1994) and (Rzychon and Worsztynowicz, 1995). Several of these lakes have earlier been included in ICP Waters assessments (see e.g. Lükewille et al., 1997). Recovery from acidification has also been documented, but many lakes are still acidified (Kopáček et al., 1998; Kopáček et al., 2017).

Austria: Acidification of mountain lakes has been observed (Psenner, 1989), and Austria reported data to ICP Waters until 2015. However, acidification due to atmospheric deposition is no longer considered an issue. This also corresponds with the WFD reporting. Monitoring of selected sites is still ongoing, demonstrating a climate impact on pH (mainly increasing, e.g. Sommaruga-Wögrath et al. (1997)) and other chemical variables (increasing alkalinity and solutes, see Rogora et al., 2018).

Russia: Acidified lakes were observed in surveys in European Russia (2003-2008) and Western Siberia (2011-2013) (Moiseenko et al., 2017). The percentage of acidified lakes in the survey were 2.0-6.9% and 5.0-17.2% (depending on the definition) in European Russia and Western Siberia, respectively.

Armenia: Acidified rivers are observed in parts of the country (e.g. the Debed basin) according to the NFC, but relevant data are limited.

Western Balkans: According to Figure 3, surface water acidification may also occur in other regions, such as northern Croatia, parts of Bosnia and Herzegovina and Serbia and western Albania, but no recent studies on acidification in these regions have been found. According to Merilehto et al. (1988), no acidification effects were observed for the lakes and rivers in the former Yugoslavia that were investigated. Small lakes at high altitude could be susceptible to acidification, but no systematic data from these were available.

Bulgaria and Romania: Bulgaria and Romania were not identified as particularly vulnerable from Figure 3, due to lower deposition levels. However, mountain lakes in these areas were included in the EMERGE project (Curtis et al., 2005). In Bulgaria, nine lakes in the Rila Mountains were studied. They were found to be acid sensitive, but the critical load was only exceeded for one of them in 2000. Six sites in the Retezat Mountains of Romania were studied and found to be acid-sensitive, and at five of them the critical loads were exceeded in the year 2000. Moreover, in a study of 40 lakes and ponds in the same area in Romania in 2004-2006, 20% of the water bodies had pH < 5 (Cogalniceanu et al., 2012). No more recent studies have been found. Bulgaria reports 60 river water bodies with less than good acidification status under the WFD, but none where atmospheric deposition is identified as a significant pressure and only two with the impact acidification, so it is questionable whether this relates to acidification causing biological damage and that it is due to air pollution. For Romania 179 river water bodies and 13 lake water bodies are reported as acidified under the WFD, but neither with atmospheric deposition as pressure and only four of the river water bodies with the impact acidification, so again the interpretation of these results is difficult.

Finally, no significant areas of **Portugal, Hungary, Estonia, Cyprus** were considered to be at risk from surface water acidification according to the analysis in section 3.1.1. While some water bodies (and for lakes in Estonia and Portugal a significant proportion) are identified as being of less than good acidification status according to the WFD reporting, none are reported with the pressure type atmospheric deposition (except one water body in Hungary), and only the Hungarian water bodies are attributed the impact acidification, so it is likely that this reporting does not reflect acidification effects on surface water ecosystems due to air pollution. Hungarian surface waters generally have high buffering capacity (Merilehto et al., 1988). Results from the AL:PE 2 project in the 1990s showed acid-sensitive waters in Portugal, but little indication of anthropogenic acidification, due to low deposition levels (Camarero et al., 1995). This is in line with Figure 3. No literature has been found for Cyprus or Estonia.

In summary, acidified surface waters are likely to occur also outside the countries covered by the country reports (Table 8). Acidification is observed in Slovakia, Russia and Armenia, and is very likely to occur in Belgium and potentially also in Luxembourg. Deposition-induced acidification has previously been observed in France, Bulgaria and Romania, but present conditions are not known. In the Western Balkans there are indications that surface water acidification may occur, but there are no monitoring studies to confirm this. There may also be small acid-sensitive areas with sufficient deposition to cause acidification that cannot be identified from the low resolution European maps (Figure 3).

5.1.3 Overview of information and results

Table 7 Overview of information and results for countries covered by the country reports

Country	Submitted data	Type of data	Critical loads	Exceedance of critical loads	Potentially acid-sensitive regions according to maps	Relevance of acidification related WFD data	Acidification extent and severity	NFC ICP Waters
Canada	Yes	Regional	Yes	Yes	n.a.	n.a.	Occurs mainly in two large regions (Canadian shield region and the north-western coast), moderate severity	Currently not
Czech Republic	Yes	Particularly sensitive areas	No	n.a.	Yes	Few relevant data, but these confirm other national data	Partly severe, but limited to certain areas	Yes
Finland	Yes	Scattered, regional	Yes	Yes	n.a.	Somewhat ambiguous and relatively large proportion of unknown for rivers, no information on lakes	Scattered across the central/northern parts of the country, less severe	Yes
Germany	Yes	Scattered, regional	No	n.a.	Yes	Few classified, but relevant data	Scattered (east and west in mid/southern Germany), sometimes severe, mainly not acidified	Yes
Ireland	Yes	Regional	Yes	Yes	n.a.	Not reported yet	Limited to the north-west and south-west, some local hotspots	Yes
Italy	Yes	Particularly sensitive area	No	n.a.	Yes	No relevant data	Occurs for small, mountain lakes, but not severe, closer to chemical recovery	Yes
Latvia	Not relevant		No	n.a.	No	Not assessed	Not an issue	Yes

Country	Submitted data	Type of data	Critical loads	Exceedance of critical loads	Potentially acid-sensitive regions according to maps	Relevance of acidification related WFD data	Acidification extent and severity	NFC ICP Waters
Netherlands	Yes	Particularly sensitive area/habitat type	Yes	Yes	n.a.	Ambiguous data, probably not relevant	Occurs for small pools on sandy soils (east), can be severe	Currently not
Norway	Yes	Regional	Yes	Yes	n.a.	Not reported yet	Regional issue (south/south-west), and frequently severe	Yes
Poland	Yes	Particularly sensitive areas	No	n.a.	Yes	Ambiguous data, not relevant	Low occurrence and severity, closer to chemical recovery	Yes
Spain	Not asked		No	n.a.	Yes	Ambiguous data, not relevant	Limited extent and severity	Currently not
Sweden	Yes	Regional, representative	Yes	Yes	n.a.	Large proportion of unknown, but relevant data	Regional (south), more extensive and severe according to national criteria/WFD than the submitted data	Yes
Switzerland	Yes	Particularly sensitive area	Yes	Yes	n.a.	n.a.	Limited to the south, moderate severity	Yes
United Kingdom	Yes	Regional, exceeded sites	Yes	Yes	n.a.	Ambiguous for rivers, few classified for lakes	Regional (north/west), frequently severe, but data biased towards exceeded sites	Yes
United States	Yes	Regional, representative	Yes	Yes	n.a.	n.a.	Occurs in some large regions (east), can be severe	Yes

Table 8 Overview of information and results for countries not covered by the country reports with potentially acidified surface waters or WFD data

Country	Potentially acid-sensitive regions according to maps	Relevance of acidification related WFD data	Older literature	Recent literature	Acidification extent and severity	NFC ICP Waters
Albania	Yes	n.a.	No	No	Could occur, but mainly unknown	Currently not
Armenia	Yes	n.a.	No	No	Observed according to NFC, but mainly unknown	Yes
Austria	Yes	Not classified	Yes	Not with deposition focus	No longer considered an issue	Yes
Belgium	Yes	Partly relevant data, large proportion unknown for rivers	Yes	No	Likely, in particular for lakes in the north. Extent and severity unknown	Currently not
Bosnia and Herzegovina	Yes	n.a.	Yes, limited	No	Could occur in the mountains, but mainly unknown	Currently not
Bulgaria	No	Ambiguous data, mainly not relevant	Yes	No	Previously observed in the Rila Mountains, current status unknown	Currently not
Croatia	Yes	Not classified	Yes, limited	No	Could occur in the mountains, but mainly unknown	Yes
Cyprus	No	Ambiguous data, not relevant	No		Very likely not an issue	Currently not
Estonia	No	Ambiguous data, not relevant	No		Very likely not an issue	Yes
France	Yes	Ambiguous data, not relevant	Yes	No	Previously observed in the Vosges mountains and the Ardennes, current status unknown	Currently not
Hungary	No	Ambiguous data, mainly not relevant	Yes	No	Most likely not an issue	Currently not

Country	Potentially acid-sensitive regions according to maps	Relevance of acidification related WFD data	Older literature	Recent literature	Acidification extent and severity	NFC ICP Waters
Luxembourg	Yes	Only partly relevant and few water bodies	No	No	May occur, but mainly unknown	Currently not
Portugal	No	Ambiguous data, not relevant	Yes	No	Most likely not an issue	Currently not
Romania	No	Ambiguous data, mainly not relevant	Yes	No	Previously observed in the Retezat Mountains, current status unknown	Currently not
Russia	Yes	n.a.	Yes, some	Yes, some	Occurs, information on extent and severity is limited	Yes
Serbia	Yes	n.a.	Yes, limited	No	Could occur in the mountains, but mainly unknown	Currently not
Slovakia	Yes	Not classified	Yes	Yes	Occurs for small lakes in the Tatra Mountains, recovery documented, but still acidified	Currently not

5.2 Do we have sufficient information about the extent of surface water acidification in Europe and North America?

The information summarised in 5.1 provides an overview of large and small regions where surface water acidification occurs, the levels of the observed acidification, and in some cases estimates of the actual extent in terms of percentage of water bodies or total land area that is affected. However, the analysis also revealed several shortcomings:

Limited reporting of critical loads for surface waters: Exceedance of critical loads provides a good indication of the likelihood of surface water acidification. However, only seven European countries submit critical loads for surface waters to the CCE. As critical loads are used to quantify the need for emission reductions, the countries do not necessarily need to submit critical loads for areas that are never likely to be exceeded, i.e. where the sensitivity is low. However, the overview of acidification status shows that critical loads are still likely to be exceeded also in areas outside these seven countries. There may be several reasons why critical loads for these areas are not submitted. In some cases, the areas may be so small and scattered that there is limited value to include them in such European overviews. Resources for monitoring and modelling may also be an issue, especially where acidification affects only a small proportion of the surface waters within a country. In any case it should be kept in mind that the overall exceedance maps issued do not reflect potential surface water acidification across all Europe. In the context of emission reductions there is also an issue if the critical loads used in negotiations do not fully protect surface waters.

Low/reduced monitoring: Several of the monitoring programmes described are small, have been reduced, have limited funding and/or are irregular. This may be a consequence of reduced acid deposition and recovery, i.e. the problem is actually smaller, and resources are directed towards more widespread and acute issues. However, acidification monitoring may also suffer from reduced attention and an impression of acid deposition as a solved problem. In any case, low level of monitoring affects the representativity and makes it difficult to upscale and compare the results. A proper evaluation of extent requires large-scale surveys. These are only reported to take place at regular intervals in Sweden and the US, although less extensive surveys as in Ireland and once-off surveys as conducted in the Czech Republic, Canada and Norway, and at a smaller scale in Switzerland and Italy are also highly valuable. Monitoring of surface water acidification has different requirements than monitoring for other purposes (parameters, sites) and are not necessarily fully addressed in non-targeted monitoring programmes. Lower frequency of monitoring may also prevent observation of seasonal or episodic acidification events (Rogora et al., 2013). This may be particularly crucial for sites undergoing recovery, where the water quality at other times may be satisfactory.

Regions with sparse/no recent information: The analysis showed that there are parts of Europe and neighbouring countries where surface water acidification occurs, is likely to occur or may occur, but where information is non-existent, sparse or not easily available. Again, there is a need for increased monitoring, and section 5.1.2 identifies the regions which should be the primary focus of this monitoring.

Shortcomings of the WFD reporting: One major drawback of the WFD in the context of surface water acidification is that it mainly focusses on the larger lakes and catchments. The countries can choose to define also smaller water bodies as WFD water bodies. Actually, 26% of the reported lake water bodies are less than 0.5 km^2 (numbers for catchment area are not available). Still, small headwater systems are usually considered only as part of larger river water bodies. At the same time the frequency of lakes tends to increase exponentially with declining lake size, so that lakes that fall below the WFD cut-off of 0.5 km^2 are particularly numerous (see for example Cael and Seekell, 2016)). Acid sensitivity is frequently higher in these systems, due to shallower soils, lower weathering rates and steeper slopes, while higher levels of precipitation in the uplands can accentuate acid deposition fluxes. In contrast, the larger catchments that dominate the WFD reporting tend to occupy lower altitudes, including former sea floor regions, and be closer to agricultural and urban areas that are likely to provide higher levels of buffering. The WFD will consequently never give the complete picture of surface water acidification and may even ignore substantial numbers of water bodies that are most at risk.

This set aside, the WFD assessment and reporting has the great advantage that it is intended to cover all water bodies. Even if all water bodies are not necessarily monitored, a representative picture of the status of all water bodies should be obtained through grouping, modelling and expert judgement. Unfortunately, when it comes to individual quality elements as acidification status, countries frequently do not report the status class. If acidification status is reported as "Not applicable" this is perfectly acceptable, as it means that acidification is not relevant in assessing ecological status (because the water body is not sensitive, or it is sensitive but acid deposition has been and is low). The same may be the case if they report "Monitored but not used", although this may also indicate that there is no assessment system in place. However, when the status is reported as "Unknown" it may be understood as "not relevant", but also as "relevant, but not known". If a high proportion of the water bodies are in this category, then the representativity of the data is highly reduced.

There are also ambiguities and uncertainties in the interpretation of the WFD acidification status results, related to the cause of the acidification, as well as whether "less than good acidification status" really implies damage to the ecosystem due to acidification. The pressures and impacts information should clarify these issues. Even though this information is also ambiguous, in that the pressure "atmospheric deposition" does not necessarily imply acid deposition, and that the impact "acidification" does not necessarily arise from air pollution, a water body assigned the combination of less than good acidification status, atmospheric deposition as a significant pressure and acidification as a significant impact, is likely to be acidified due to air pollution, and to an extent that the ecosystem is damaged. However, our analysis of the WFD data showed that this combination of reporting was rare. Many water bodies with less than good acidification status did not have any pressures attributed to them that would cause acidification and/or the acidification status was not reflected in the impact acidification.

The pressure reporting under the WFD is quite detailed, and the level of reporting indicates that not all countries have considered all pressure types. Likewise, not all countries may have considered all impact types. So there may be cases where the water bodies are acidified due to air pollution, giving significant impacts, but where this is not evident from the pressures and impacts reporting. On the other hand, there were also water bodies in less than good acidification status reported as having no significant pressures or no relevant pressures and/or as not having the impact acidification. There were also cases where water bodies with less than good acidification status still had good or high ecological status. This raises questions about the acidification status classification. The lack of intercalibration means that the assessment systems probably vary largely between countries. The concept of water body type specific assessment embedded in the WFD could give more precise assessments than those used to date in some countries. In other countries "less than good acidification status" may have been set even if the acidification is primarily natural and/or is not associated with biological damage. It is evident from the feedback from the NFCs that there are several cases where the basis for setting the acidification status to less than good is unclear and where acidification from air pollution causing significant damage to the ecosystem does not seem likely. The lack of a common approach to classification makes it difficult to interpret and compare the results from the different countries.

In summary, the WFD data are not fully representative of the type of surface water bodies that are frequently found to be vulnerable to acidification. Along with the ambiguity and uncertainty associated with the reported data, this means that the information reported under the WFD is at present of limited value in assessing the current extent of acidification of surface waters in Europe.

Monitoring requirements of the new **NEC Directive** can serve to meet some of these shortcomings. According to Article 9, the Member States are required to monitor "negative impacts of air pollution upon ecosystems based on a network of monitoring sites that is representative of their freshwater (…) ecosystem types". This legal obligation is a powerful means to halt and reverse the current reduction in surface water acidification monitoring. The requirement that the monitoring should be representative is essential, ensuring that the monitoring covers different surface water types and that the number of monitoring sites is sufficient to upscale the results to all areas at risk.

The design of the monitoring programmes under the NEC Directive is left to the Member States, but it will, and should, partly build on existing monitoring. Annex V provides a list of optional indicators, which in the case of freshwaters include both biological and chemical indicators. A specific guidance document[31] has been produced to assist the countries in setting up their monitoring programmes. It includes principles that are vital in monitoring of air pollution effects, i.e. that the sites should be sensitive and that the impacts of air pollution can be distinguished from other pressures. It stresses that the number and density of sites should be sufficient to allow analysis of spatial gradients, the importance of covering environmental gradients, and that reference sites should be included. The specific recommendations build largely on the methods and principles outlined in the ICP Waters manual (ICP Waters Programme Centre, 2010). The ultimate objective of the monitoring as stated in the guidance document is "to improve information on the impacts of air pollution, including the extent of any impacts and the recovery time when the impacts are reduced, and to contribute to review of critical loads and levels".

5.3 The future of acidified surface waters

During the past 40 years sulphur deposition has declined dramatically in nearly all the areas with acidified surface waters in Europe and North America (Garmo et al., 2014). Nitrogen deposition has also declined, *albeit* to a lesser extent (de Wit et al., 2015). Consequently, many formerly acidified waters have been recovering chemically, exhibiting rising levels of pH and ANC and lower concentrations of inorganic aluminium. Biological recovery has also begun with the return and increase in abundance of some acid-sensitive species reported in some systems (e.g. Murphy et al., 2014). Nevertheless, as shown in this report, many surface waters remain acidified in many areas of both North America and Europe. In some cases, they are continuing to respond to the recent declines in acid deposition with time lags resulting from processes such as the delayed release of sulphate stored in the catchment soils (Robison et al., 2013; Evans et al., 2014). Full chemical recovery of acidified waters may require replenishment of the base cation stores in soils through chemical weathering of soil minerals, a notoriously slow process (Kirchner and Lydersen, 1995). Whereas the depletion of soil base cations due to acid deposition and leaching of strong acid anions such as sulphate can occur within only a few decades, the replenishment is governed by weathering rates and may require many decades or even centuries. This asymmetry has been illustrated by application of process-oriented models such as MAGIC (Model for Acidification of Groundwater In Catchments) (Cosby et al., 1985) to acidified surface waters in, for example, Norway (Larssen et al., 2010), Sweden (Moldan et al., 2013), Czech Republic (Hruška et al., 2002; Hruška et al., 2014) and the Appalachian Mountains of the US (Sullivan et al., 2011) (see also Jenkins et al. (2002) and Figure 41).

[31] http://ec.europa.eu/transparency/regexpert/index.cfm?do=groupDetail.groupDetailDoc&id=35724&no=3

Other factors may influence the chemical recovery of acidified surface water in response to reduced levels of acid deposition (Skjelkvåle et al., 2006). Climate change and forestry practices are most prominent. Both have been shown to influence surface water chemistry and aquatic biology. Increased soil temperatures associated with climate change may increase rates of mineralisation of soil organic nitrogen, promoting the release of nitrate to surface waters (Wright, 1998). Increased removal of biomass from forests can accelerate the depletion of base cations in the soil, and impair recovery or even cause re-acidification of surface waters (Kirchner and Lydersen, 1995; Helliwell et al., 2001; Akselsson et al., 2007). Modelling studies conducted in Finland and Sweden indicate that both climate change and forestry practices pose risks for future acidification of acid-sensitive surface waters (Aherne et al., 2011; Moldan et al., 2017).

Biological recovery is predicated on adequate chemical recovery (Raddum et al., 2004). A stable target water chemistry above the critical limits of acid-sensitive species is necessary. Then hindrances and delays include dispersal of the species from a source population, survival and internal dispersal in the lake/river as well as time needed to reach natural fluctuations (Yan et al., 2003). Temporarily more acidic conditions may occur due to e.g. snowmelt, high rainfall, droughts and sea salt events (Lepori et al., 2003; Wright, 2008). Such acid episodes can cause set-backs of recovery of sensitive organisms (Raddum and Fjellheim, 2003). In general, biological recovery is a complex process, depending on environmental factors, resource stoichiometry and biotic interactions (Monteith et al., 2005; Vrba et al., 2016). The delayed biological recovery compared to chemical recovery is nicely illustrated in the Czech case study Lake Černé (Figure 19). Here sulphate concentrations are nearly back to pre-industrial levels and aluminium concentrations have declined. However, the initial decline in nitrate concentration has levelled off, reflecting the similar pattern for nitrogen deposition, the recovery of water pH and benthos has been modest, and the fish have yet to re-establish.

Several examples in this report show that chemical recovery is ongoing. But there are also examples of recovery slowing down and levelling off. Figure 60 shows how the proportion of acidic streams has basically remained unchanged over the past 20 years in the Blueridge/Ridges region in the US. In Swedish lakes ANC is levelling off and there are not yet any signs of increasing base cation concentrations (Figure 51). This may reflect the very slow base cation replenishment via weathering, but, given that critical loads are still exceeded in these areas, it is also likely to be an indication that the deposition remains too high. The slow decline in nitrogen deposition also causes reason for concern, especially in sparsely vegetated mountain watersheds, where nitrogen is more likely to cause acidification effects due to lower nitrogen retention capacity (Lepori and Keck, 2012).

Full chemical and biological recovery requires reductions in acid deposition such that critical loads for acidification of surface waters are no longer exceeded. The reduction in sulphur and nitrogen deposition needed to reach non-exceedance can be calculated based on the critical loads and deposition data. Figure 61 illustrates the reduction in sulphur and nitrogen deposition needed to reach non-exceedance in the European countries which have submitted critical loads for surface water acidification (Figure 1: left). The crosses represent grid cells with exceedance given current (2015) deposition (Figure 1:right/Figure 62: left). The figure shows that sulphur deposition constitutes the larger part of the exceedance (most of the grid cells are above the 1:1 line), while reduction in both components are needed in most cases (where the crosses are not on the vertical axis). For a majority of the grid cells relatively small reductions are needed (for 75% of the grid cells a maximum of 17 eq/ha/yr for sulphur and 6 eq/ha/yr for nitrogen). Still, Figure 62 shows that given current legislation, there will still be exceedance of critical loads in 2030. Both the size and extent of the exceedance is lower, but the continued exceedance implies that the current emission reduction targets may not be sufficiently ambitious for protection of surface waters from acidification. It should also be kept in mind that the critical loads represent the deposition that is sustainable in the long-term perspective. In the current situation deposition below the critical load would allow faster replenishment of soil base cations and thus faster recovery in areas with previous exceedance of critical loads (Kirchner and Lydersen, 1995). All in all, further emission reductions would be required to enable and increase rates of recovery.

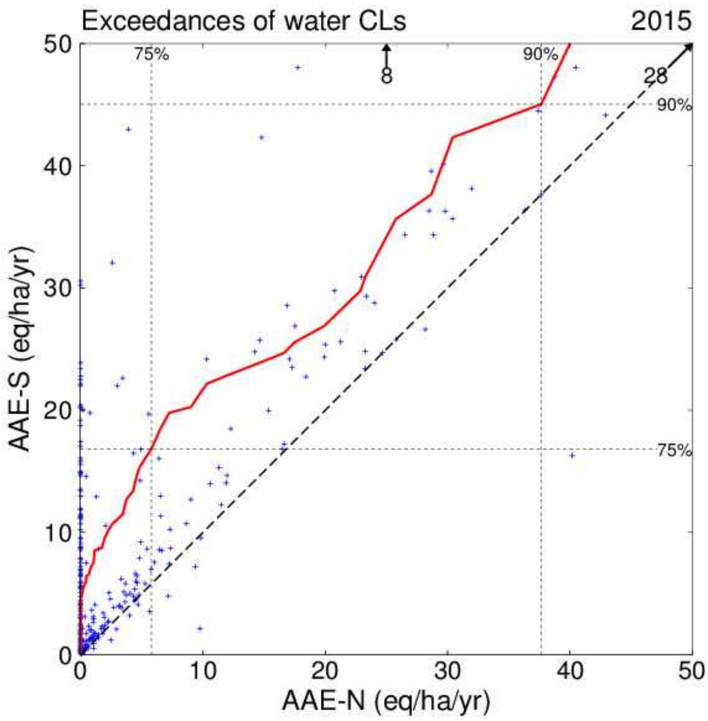


Figure 61 Average accumulated exceedance (AAE, i.e. area weighted average exceedance for the different reported areas per grid cell) of critical loads for surface waters by sulphur (S) and nitrogen (N), assuming the shortest route from exceedance to non-exceedance. The crosses represent the grid cells with exceedance in Figure 1: right/Figure 62: left (except 8 grid cells with S exceedance > 50 eq/ha/yr and 28 grid cells where both S and N exceedance are > 50 eq/ha/yr). The red line indicates where the percentiles of exceedances are equal for S and N.

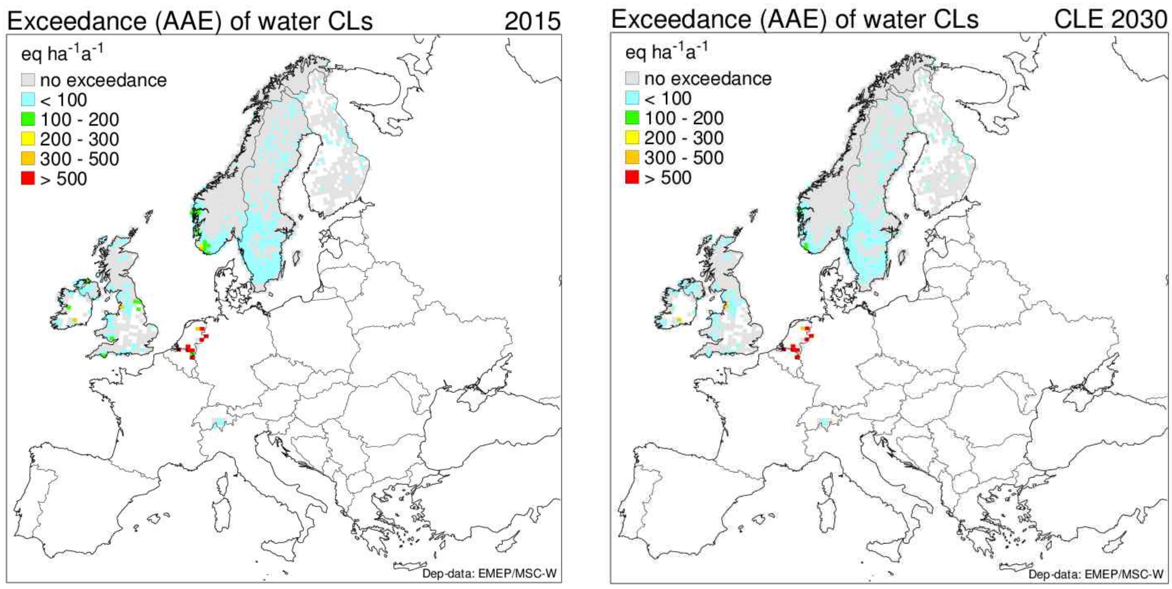


Figure 62 Exceedance of critical loads for acidification of surface waters for some European countries, given the deposition in 2015 (left; Same as Figure 1) and the deposition in 2030 given current legislation (right). The exceeded area constitutes 5.7 and 3.4% of the total area with critical loads in 2015 and 2030, respectively. AAE = average accumulated exceedance, i.e. area weighted average exceedance for the different reported areas per grid cell.

6 Conclusions and future perspectives

The current status of surface water acidification related to air pollution in Europe and North America has been assessed using country reports, monitoring data, critical loads and exceedance data, acid sensitivity and deposition maps, and data reported under the European Commission's Water Framework Directive (WFD). Acidification is still observed in many countries, despite widespread evidence of a gradual recovery of acid-sensitive waters in response to reduced sulphur and nitrogen deposition. However, the extent and severity of surface water acidification vary. In some countries acidification is an issue of regional concern, while elsewhere it is a smaller-scale issue. In some countries acidification remains severe in larger areas, and there is also evidence of acidification hot-spots in countries which are generally less affected. Countries that are close to chemical recovery will occasionally have sites at risk, vulnerable to short-term acidification events.

Maps of acid sensitivity and deposition suggest that surface water acidification is present in regions and countries for which no data or reports were delivered for the current assessment. Supporting evidence is in many cases available from previous studies, but recent monitoring data would be necessary for substantiation.

The submitted data show that existing national monitoring varies in the ability to assess the spatial extent of acidification and the recovery responses of acidified sites. In some countries there are extensive monitoring programmes or regular large-scale surveys, while in others the monitoring is limited. Monitoring under the WFD does not fully cover acid-sensitive waters, as these are often small headwater systems, which are not included in the scope of the WFD. The monitoring requirements under the European Union's National Emission Ceilings Directive, tailored to assess ecosystem impacts of air pollution, are expected to increase the representation of sensitive water bodies, and thereby address this shortcoming of the WFD monitoring, as well as reverse the recent decline in the number of monitoring sites observed in some countries.

The documented chemical recovery confirms that the mechanistic understanding of the effects of acid deposition on surface waters has been correct and that the resulting policy measures to reduce emissions of acidifying components to the atmosphere are having the intended effects. However, critical loads of acidity are still exceeded in many regions, indicating that the deposition is not yet at a sustainable level to protect surface waters from acidification. Moreover, chemical recovery can be slow, due, for example, to the release of sulphur stored in the soils and the slow rates of geological weathering that determine the potential rate of replenishment of base cations in catchment soils following many decades of base cation depletion. Other factors such as climate change and intensified forestry may further delay the recovery. Biological recovery can lag severely behind chemical recovery, due to the need for stable, tolerable conditions and hindrances such as species dispersal. Hence, further emission reductions will be required to enable significant ecological recovery of acidified surface waters in some areas and increase the rates of recovery across wider regions.

Based on the current assessment it is strongly recommended that the existing monitoring programmes in some countries are re-evaluated. Current stations may not be sufficiently representative, especially where monitoring is largely conducted in the context of the WFD. The frequency of monitoring may be too low, which is particularly crucial in ecosystems where episodic acidification events can delay biological recovery although average conditions may be satisfactory. Large-scale surveys are highly useful for estimating the spatial extent of acidification and the number of water bodies that are affected and should be considered where such surveys are not conducted regularly. A particular recommendation goes to the countries and areas identified as potentially having issues with surface water acidification, but where little or no documentation on the current status has been found. If such information is not available, investigative surveys should be conducted and, if needed, monitoring programmes should be established. In general, monitoring sites and parameters relevant to acidification are also highly relevant for detecting changes in water quality due to climate change, supporting the usefulness of keeping up and strengthening this type of monitoring.

Apart from the limited focus on headwater systems in the WFD, there are other shortcomings that should be addressed. Classification systems for acidification status are not consistent between countries, have too little documented relevance for biological status, and could thus benefit from intercalibration. Moreover, the reporting schemes allow for ambiguities, e.g. the lack of reporting of a certain pressure or impact may imply i) absence of the pressure or impact, ii) the pressure or impact is not considered, or iii) there is no information. Currently, the information reported under the WFD is of limited value in assessing the extent of acidification of surface waters in Europe.

This assessment documents that acidification of surface waters remains an environmental concern in many countries. Even by reaching the emission targets for acidifying compounds set for 2030, critical loads for surface waters will remain exceeded. This implies that the current emission reduction targets may not be sufficiently ambitious for protection of all surface waters from acidification. Critical load estimates provide a good basis for evaluating the current as well as future exceedances, implying that more countries could benefit from submitting critical loads for acidification of surface waters under the LRTAP Convention. Despite large and effective efforts across Europe and North America to reduce surface water acidification, air pollution still constitutes a threat to freshwater ecosystems.

7 Literature

Aherne, J., Posch, M., Forsius, M., Lehtonen, A. and Härkönen, K. 2011. Impacts of forest biomass removal on soil nutrient status under climate change: a catchment-based modelling study for Finland. Biogeochemistry 107: 471-488.

Akselsson, C., Westling, O., Sverdrup, H. and Gundersen, P. 2007. Nutrient and carbon budgets in forest soils as decision support in sustainable forest management. Forest Ecology and Management 238(1-3): 167-174.

Angeli, N., Dambrine, E., Boudot, J.P., Nedeltcheva, T., Guerold, F., Tixier, G., Probst, A., Party, J.P., Pollier, B. and Bourrie, G. 2009. Evaluation of streamwater composition changes in the Vosges Mountains (NE France): 1955-2005. Science of the Total Environment 407(14): 4378-4386.

Baker, J.P., Warrenhicks, W.J., Gallagher, J. and Christensen, S.W. 1993. Fish population losses from Adirondack lakes: The role of surface water acidity and acidification. Water Resources Research 29(4): 861-874.

Battarbee, R.W. and Charles, D.F. 1986. Diatom-based pH reconstruction studies of acid lakes in Europe and North America: A synthesis. Water Air and Soil Pollution 30(1-2): 347-354.

Battarbee, R.W., Simpson, G.L., Shilland, E.M., Flower, R.J., Kreiser, A., Yang, H. and Clarke, G. 2014. Recovery of UK lakes from acidification: An assessment using combined palaeoecological and contemporary diatom assemblage data. Ecological Indicators 37: 365-380.

Cael, B.B. and Seekell, D.A. 2016. The size-distribution of Earth's lakes. Scientific Reports 6: 29633.

Camarero, L., Rogora, M., Mosello, R., Anderson, N.J., Barbieri, A., Botev, I., Kernan, M., Kopáček, J., Korhola, A., Lotter, A.F., Muri, G., Postolache, C., Stuchlik, E., Thies, H. and Wright, R.F. 2009. Regionalisation of chemical variability in European mountain lakes. Freshwater Biology 54(12): 2452-2469.

Camarero, L., Catalan, J., Pla, S., Rieradevall, M., Jimenez, M., Prat, N., Rodriguez, A., Encina, L., CruzPizarro, L., Castillo, P.S., Carrillo, P., Toro, M., Grimalt, J., Berdie, L., Fernandez, P. and Vilanova, R. 1995. Remote mountain lakes as indicators of diffuse acidic and organic pollution in the Iberian peninsula (AL:PE 2 studies). Water Air and Soil Pollution 85(2): 487-492.

Clow, D.W., Mast, M.A. and Campbell, D.H. 1996. Controls on surface water chemistry in the upper Merced River basin, Yosemite National Park, California. Hydrological Processes 10(5): 727-746.

CLRTAP 2015a. Introduction, Chapter 1 of Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. UNECE Convention on Long-range Transboundary Air Pollution. Accessed on 14 April 2018 on Web at www.icpmapping.org.

CLRTAP 2015b. Exceedance calculations, Chapter VII of Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. UNECE Convention on Long-range Transboundary Air Pollution. Accessed on 11 December 2017 on Web at www.icpmapping.org.

CLRTAP 2017. Mapping critical loads for ecosystems, Chapter V of Manual on methodologies and criteria for modelling and mapping critical loads and levels and air pollution effects, risks and trends. UNECE Convention on Long-range Transboundary Air Pollution. Accessed on 11 December 2017 on Web at www.icpmapping.org.

Cogalniceanu, D., Bancila, R., Plaiasu, R., Samoila, C. and Hartel, T. 2012. Aquatic habitat use by amphibians with specific reference to Rana temporaria at high elevations (Retezat Mountains National Park, Romania). Annales De Limnologie-International Journal of Limnology 48(4): 355-362.

Cosby, B.J., Hornberger, G.M., Galloway, J.N. and Wright, R.F. 1985. Modeling the effects of acid deposistion: Assessment of a lumped parameter model of soil water and streamwater chemistry. Water Resources Research 21(1): 51-63.

Curtis, C.J., Botev, I., Camarero, L., Catalan, J., Cogalniceanu, D., Hughes, M., Kernan, M., Kopáček, J., Korhola, A., Psenner, R., Rogora, M., Stuchlik, E., Veronesi, M. and Wright, R.F. 2005. Acidification in European mountain lake districts: A regional assessment of critical load exceedance. Aquatic Sciences 67(3): 237-251.

de Wit, H.A., Hettelingh, J.-P. and Harmens, H. (eds.) 2015. Trends in ecosystem and health responses to long-range transported atmospheric pollutants. ICP Waters report 125/2015. NIVA report 6946-2015.

Escudero-Oñate, C. 2017. Intercomparison 1731: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca,Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. ICP Waters report 134/2017. NIVA report 7207-2017.

Evans, C.D., Chadwick, T., Norris, D., Rowe, E.C., Heaton, T.H.E., Brown, P. and Battarbee, R.W. 2014. Persistent surface water acidification in an organic soil-dominated upland region subject to high atmospheric deposition: the North York Moors, UK. Ecological Indicators 37 (B): 304-316.

Exley, C., Chappell, J.S. and Birchall, J.D. 1991. A mechanism for acute aluminum toxicity in fish. Journal of Theoretical Biology 151(3): 417-428.

Ferrier, R.C., Jenkins, A., Wright, R.F., Schopp, W. and Barth, H. 2001. Assessment of recovery of European surface waters from acidification 1970-2000: An introduction to the Special Issue. Hydrology and Earth System Sciences 5(3): 274-282.

Fevrier, C., Party, J.P. and Probst, A. 1999. Surface water acidification and critical loads for the French Ardennes Massif. Comptes Rendus De L Academie Des Sciences Serie Ii Fascicule a-Sciences De La Terre Et Des Planetes 328(1): 29-35.

Fjellheim, A. and Raddum, G.G. 2001. Acidification and liming of River Vikedal, western Norway. A 20 year study of responses in the benthic invertebrate fauna. Water Air and Soil Pollution 130(1-4): 1379-1384.

Garmo, Ø.A., De Wit, H.A. and Fjellheim, A. 2015. Chemical and biological recovery in acid-sensitive waters: trends and prognosis. ICP Waters report 119/2015. NIVA report 6847-2015.

Garmo, Ø.A., Skjelkvåle, B.L., de Wit, H.A., Colombo, L., Curtis, C., Fölster, J., Hoffmann, A., Hruška, J., Høgåsen, T., Jeffries, D.S., Keller, W.B., Krám, P., Majer, V., Monteith, D.T., Paterson, A.M., Rogora, M., Rzychon, D., Steingruber, S., Stoddard, J.L., Vuorenmaa, J. and Worsztynowicz, A. 2014. Trends in surface water chemistry in acidified areas in Europe and North America from 1990 to 2008. Water Air and Soil Pollution 225(3).

Gensemer, R.W. and Playle, R.C. 1999. The bioavailability and toxicity of aluminum in aquatic environments. Critical Reviews In Environmental Science And Technology 29(4): 315-450.

Gorham, E. 1961. Factors influencing supply of major ions to inland waters, with special reference to the atmosphere. Geological Society of America Bulletin 72: 795-840.

Hartmann, J. and Moosdorf, N. 2012. The new global lithological map database GLiM: a representation of rock properties at the Earth surface. Geochemistry, Geophysics, Geosystems 13: 12.

Helliwell, R.C., Ferrier, R.C., Johnston, L., Goodwin, J. and Doughty, R. 2001. Land use influences on acidification and recovery of freshwaters in Galloway, south-west Scotland. Hydrology and Earth System Sciences 5(3): 451-458.

Henriksen, A. and Posch, M. 2001. Steady-state models for calculating critical loads of acidity for surface waters. Water, Air, and Soil Pollution Focus 1: 375–398.

Herrmann, J., Degerman, E., Gerhardt, A., Johansson, C., Lingdell, P.E. and Muniz, I.P. 1993. Acid stress effects on stream biology. Ambio 22(5): 298-307.

Hesthagen, T. and Hansen, L.P. 1991. Estimates of the annual loss of Atlantic salmon, Salmo salar L., in Norway due to acidification. Aquaculture Research 22(1): 85-92.

Hesthagen, T., Fiske, P. and Skjelkvåle, B.L. 2008. Critical limits for acid neutralizing capacity of brown trout (Salmo trutta) in Norwegian lakes differing in organic carbon concentrations. Aquatic Ecology 42: 307-316.

Hettelingh, J.-P., Posch, M. and Slootweg, J. (eds.) 2017. European critical loads: database, biodiversity and ecosystems at risk. CCE Final Report 2017, Coordination Centre for Effects, RIVM Report 2017-0155, Bilthoven, Netherlands.

Howells, G.D., Brown, D.J.A. and Sadler, K. 1983. Effects of acidity, calcium, and aluminum on fish survival and productivity - a review. Journal of the Science of Food and Agriculture 34(6): 559-570.

Hruška, J., Moldan, F. and Krám, P. 2002. Recovery from acidification in central Europe - observed and predicted changes of soil and streamwater chemistry in the Lysina. catchment, Czech Republic. Environmental Pollution 120(2): 261-274.

Hruška, J., Krám, P., Moldan, F., Oulehle, F., Evans, C.D., Wright, R.F., Kopáček, J. and Cosby, B.J. 2014. Changes in soil dissolved organic carbon affect reconstructed history and projected future trends in surface water acidification. Water Air and Soil Pollution 225(7): 13.

ICP Waters Programme Centre 2010. ICP Waters Programme Manual 2010. ICP Waters report 105/2010. NIVA report 6074-2010.

Jenkins, A., Larssen, T., Moldan, F., Posch, M. and Wright, R.F. 2002. Dynamic modelling of surface waters: Impact of emission reduction - possibilities and limitations. ICP Waters report 70/2002. NIVA report 4598-2002.

Kirchner, J.W. and Lydersen, E. 1995. Base cation depletion and potential long-term acidification of Norwegian catchments. Environmental Science & Technology 29(8): 1953-1960.

Kopáček, J. and Stuchlik, E. 1994. Chemical characteristics of lakes in the high tatra mountains, Slovakia. Hydrobiologia 274(1-3): 49-56.

Kopáček, J., Hejzlar, J., Stuchlik, E., Fott, J. and Veselý, J. 1998. Reversibility of acidification of mountain lakes after reduction in nitrogen and sulphur emissions in Central Europe. Limnology and Oceanography 43(2): 357-361.

Kopáček, J., Kaňa, J., Bičárová, S., Fernandez, I.J., Hejzlar, J., Kahounová, M., Norton, S.A. and Stuchlik, E. 2017. Climate change increasing calcium and magnesium leaching from granitic alpine catchments. Environmental Science & Technology 51(1): 159-166.

Kreiser, A., Rose, N.L., Probst, A. and Massabuau, J.C. 1995. Relationships between lake-water acidification in the Vosges Mountains (France) and SO2 — NOx Emissions in western Europe. In: Landmann, G.M., Bonneau, M. (eds.) Forest decline and air pollution effects in the French mountains. Ecological studies. Springer Verlag: New York, pp. 363–369.

Lacoul, P., Freedman, B. and Clair, T. 2011. Effects of acidification on aquatic biota in Atlantic Canada. Environmental Reviews 19: 429-460.

Larssen, T., Cosby, B.J., Lund, E. and Wright, R.F. 2010. Modeling future acidification and fish populations in Norwegian surface waters. Environmental Science & Technology 44: 5345–5351.

Lepori, F. and Keck, F. 2012. Effects of atmospheric nitrogen deposition on remote freshwater ecosystems. Ambio 41(3): 235-246.

Lepori, F., Barbieri, A. and Ormerod, S.J. 2003. Causes of episodic acidification in Alpine streams. Freshwater Biology 48(1): 175-189.

Lien, L., Raddum, G.G., Fjellheim, A. and Henriksen, A. 1996. A critical limit for acid neutralizing capacity in Norwegian surface waters, based on new analyses of fish and invertebrate responses. Science of the Total Environment 177(1-3): 173-193.

Lydersen, E., Larssen, T. and Fjeld, E. 2004. The influence of total organic carbon (TOC) on the relationship between acid neutralizing capacity (ANC) and fish status in Norwegian lakes. Science of the Total Environment 326: 63-69.

Lükewille, A., Jeffries, D., Johannessen, M., Raddum, G., Stoddard, J. and Traaen, T. 1997. The nine year report: Acidification in Europe and North America. Long term developments (1980s and 1990s). NIVA report 3637-1997.

Lynch, J.A., Phelan, J., Pardo, L.H., McDonnell, T.C. and Clark, C.M. 2017. Detailed Documentation of the National Critical Load Database (NCLD) for U.S. Critical Loads of Sulfur and Nitrogen, version 3.0. National Atmospheric Deposition Program, Illinois State Water Survey, Champaign, IL.

Massabuau, J.C., Fritz, B. and Burtin, B. 1987. Acidification of fresh-waters (pH-less-than-or-equal-to-5) in the Vosges mountains (eastern France). Comptes Rendus De L Academie Des Sciences Serie Iii-Sciences De La Vie-Life Sciences 305(5): 121-124.

Merilehto, K., Kenttämies, K. and Kämäri, J. 1988. Surface water acidification in the ECE region. Nordic Council of Ministers. Miljørapport 1988:14.

Moiseenko, T.I., Gashkina, N.A., Dinu, M.I., Khoroshavin, V.Y. and Kremleva, T.A. 2017. Influence of natural and anthropogenic factors on water acidification in humid regions. Geochemistry International 55(1): 84-97.

Moldan, F., Cosby, B.J. and Wright, R.F. 2013. Modeling past and future acidification of Swedish lakes. Ambio 42(5): 577-586.

Moldan, F., Stadmark, J., Folster, J., Jutterstrom, S., Futter, M.N., Cosby, B.J. and Wright, R.F. 2017. Consequences of intensive forest harvesting on the recovery of Swedish lakes from acidification and on critical load exceedances. Science of the Total Environment 603: 562-569.

Monteith, D.T., Hildrew, A.G., Flower, R.J., Raven, P.J., Beaumont, W.R.B., Collen, P., Kreiser, A.M., Shilland, E.M. and Winterbottom, J.H. 2005. Biological responses to the chemical recovery of acidified fresh waters in the UK. Environmental Pollution 137(1): 83-101.

Mosello, R., Wathne, B.M., Lien, L. and Birks, H.J.B. 1995. AL:PE projects: Water chemistry and critical loads. Water Air and Soil Pollution 85(2): 493-498.

Murphy, J.F., Winterbottom, J.H., Orton, S., Simpson, G.L., Shilland, E.M. and Hildrew, A.G. 2014. Evidence of recovery from acidification in the macroinvertebrate assemblages of UK fresh waters: A 20-year time series. Ecological Indicators 37: 330-340.

Neville, C.M. 1985. Physiological response of juvenile rainbow trout, *Salmo Gairdneri*, to acid and aluminum - prediction of field responses from laboratory data. Canadian Journal of Fisheries and Aquatic Sciences 42(12): 2004-2019.

Nilsson, J. and Grennfelt, P. 1988. Critical loads for sulphur and nitrogen. Nordic council of Ministers. Report from a workshop held at Skokloster, Sweden, 19-24 March 1988. Miljørapport 15.

OECD 1977. The OECD Programme on Long Range Transport of Air Pollutants. Measurements and Findings. Organisation for Economic Co-operation and Development, Paris 1977. Report can be downloaded from http://www.nilu.no/projects/ccc/reports/paris_1977.pdf.

Party, J.P., Probst, A., Dambrine, E. and Thomas, A.L. 1995. Critical loads of acidity to surface waters in the Vosges massif (North-East of France). Water Air and Soil Pollution 85(4): 2407-2412.

Psenner, R. 1989. Chemistry of high mountain lakes in silicous catchments of the Central Eastern Alps. Aquatic Sciences 51(2): 108-128.

Raddum, G.G. and Fjellheim, A. 2003. Liming of River Audna, southern Norway. A large scale experiment of benthic invertebrate recovery. Ambio 32: 230-234.

Raddum, G.G., Erikson , L., Fott, J., Halvorsen, G.A., Heegaard, E., Kohout, L., Kifinger, B. and Schaumberg, J. 2004. Recovery from acidification of invertebrate fauna in ICP Water sites in Europe and North America. ICP Waters report 76/2004. NIVA report 4858-2004.

Reuss, J.O. and Johnson, D.W. 1986. Acid deposition and the acidification of soils and waters. Springer-Verlag, USA, p. 119.

Robison, A.L., Scanlon, T.M., Cosby, B.J., Webb, J.R. and Galloway, J.N. 2013. Roles of sulfate adsorption and base cation supply in controlling the chemical response of streams of western Virginia to reduced acid deposition. Biogeochemistry 116(1-3): 119-130.

Rogora, M., Colombo, L., Lepori, F., Marchetto, A., Steingruber, S. and Tornimbeni, O. 2013. Thirty years of chemical changes in alpine acid-sensitive lakes in the Alps. Water Air and Soil Pollution 224(10): 20.

Rogora, M., Frate, L., Carranza, M.L., Freppaz, M., Stanisci, A., Bertani, I., Bottarin, R., Brambilla, A., Canullo, R., Carbognani, M., Cerrato, C., Chelli, S., Cremonese, E., Cutini, M., Di Musciano, M., Erschbamer, B., Godone, D., Iocchi, M., Isabellon, M., Magnani, A., Mazzola, L., di Cella, U.M., Pauli, H., Petey, M., Petriccione, B., Porro, F., Psenner, R., Rossetti, G., Scotti, A., Sommaruga, R., Tappeiner, U., Theurillat, J.P., Tomaselli, M., Viglietti, D., Viterbi, R., Vittoz, P., Winkler, M. and Matteucci, G. 2018. Assessment of climate change effects on mountain ecosystems through a cross-site analysis in the Alps and Apennines. Science of the Total Environment 624: 1429-1442.

Rosseland, B.O. and Staurnes, M. 1994. Physiological mechanisms for toxic effects and resistance to acidic water: An ecophysiological and ecotoxicological approach. In: Steinberg, C.E.W., Wright, R.F. (eds.) Acidification of freshwater ecosystems: Implications for the future. John Wiley & Sons Ltd., pp. 227-246.

Rzychon, D. and Worsztynowicz, A. 1995. The role of nitrogen in acidification of Tatra mountains lakes. Water Air and Soil Pollution 85(2): 481-486.

Skjelkvåle, B.L. and Wright, R.F. 1990. Overview of areas sensitive to acidification in Europe. Norwegian Institute for Water Research.

Skjelkvåle, B.L. and Wright, R.F. 1998. Mountain lakes: sensitivity to acid deposition and global climate change. Ambio 27: 280-286.

Skjelkvåle, B.L., Forsius, M., Wright, R.F., De Wit, H., Raddum, G.G. and Sjøeng, A.M.S. 2006. Joint workshop on confounding factors in recovery from acid deposition in surface waters, 9-10 October 2006, Bergen, Norway; Summary and abstracts. ICP Waters report 86/2006. NIVA report 5310-2006.

Skjelkvåle, B.L., Stoddard, J.L., Jeffries, D.S., Tørseth, K., Høgåsen, T., Bowman, J., Mannio, J., Monteith, D.T., Mosello, R., Rogora, M., Rzychon, D., Vesely, J., Wieting, J., Wilander, A. and Worsztynowicz, A. 2005. Regional scale evidence for improvements in surface water chemistry 1990-2001. Environmental Pollution 137(1): 165-176.

Sommaruga-Wögrath, S., Koinig, K.A., Schmidt, R., Sommaruga, R., Tessadri, R. and Psenner, R. 1997. Temperature effects on the acidity of remote alpine lakes. Nature 387: 64-67.

Stoner, J.H., Gee, A.S. and Wade, K.R. 1984. The effects of acidification on the ecology of streams in the upper Tywi catchment in west Wales. Environmental Pollution Series a-Ecological and Biological 35(2): 125-157.

Sullivan, T.J., Cosby, B.J., Jackson, W.A., Snyder, K.U. and Herlihy, A.T. 2011. Acidification and prognosis for future recovery of acid-sensitive streams in the southern Blue Ridge province. Water Air and Soil Pollution 219(1-4): 11-26.

The MOLAR Water Chemistry Group 1999. The MOLAR Project: atmospheric deposition and lake water chemistry. J. Limnol. 58: 88-106.

van Dam, H. and Mertens, A. 1995. Long-term changes of diatoms and chemistry in headwater streams polluted by atmospheric deposition of sulphur and nitrogen compounds. Freshwater Biology 34(3): 579-600.

Vangenechten, J.H.D., Vanpuymbroeck, S., Vanderborght, O.L.J., Bosmans, F. and Deckers, H. 1981. Physico-chemistry of surface waters in the Campine region of Belgium, with special reference to acid moorland pools. Archiv Fur Hydrobiologie 90(4): 369-396.

Vrba, J., Bojková, J., Chvojka, P., Fott, J., Kopáček, J., Macek, M., Nedbalova, L., Papacek, M., Radkova, V., Sacherova, V., Soldan, T. and Sorf, M. 2016. Constraints on the biological recovery of the Bohemian Forest lakes from acid stress. Freshwater Biology 61(4): 376-395.

Watt, W.D., Scott, C.D., Zamora, P.J. and White, W.J. 2000. Acid toxicity levels in Nova Scotian rivers have not declined in synchrony with the decline in sulfate levels. Water Air and Soil Pollution 118(3-4): 203-229.

Wright, L.P., Zhang, L.M., Cheng, I., Aherne, J. and Wentworth, G.R. 2018. Impacts and Effects Indicators of Atmospheric Deposition of Major Pollutants to Various Ecosystems - A Review. Aerosol and Air Quality Research 18(8): 1953-1992.

Wright, R.F. 1983. Acidification of freshwaters in Europe. Water Quality Bulletin 8: 137-142.

Wright, R.F. 1998. Effect of increased CO_2 and temperature on runoff chemistry at a forested catchment in southern Norway (CLIMEX project). Ecosystems 1: 216-225.

Wright, R.F. 2008. The decreasing importance of acidification episodes with recovery from acidification: an analysis of the 30-year record from Birkenes, Norway. Hydrology and Earth System Sciences 12(2): 353-362.

Wright, R.F. and Henriksen, A. 1978. Chemistry of small Norwegian lakes with special reference to acid precipitation. Limnology and Oceanography 23: 487-498.

Yan, N.D., Leung, B., Keller, W., Arnott, S.E., Gunn, J.M. and Raddum, G.G. 2003. Developing conceptual frameworks for the recovery of aquatic biota from acidification. Ambio 32(3): 165-169.

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/

Escudero-Oñate, C. 2017. Intercomparison 1731: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. **ICP Waters report 134/2017**.

Halvorsen, G.A., Johannessen, A. and Landås, T.S. 2017. Biological intercalibration: Invertebrates 2017. **ICP Waters report 133/2017**.

Braaten, H.F.V., Åkerblom, S., de Wit, H.A., Skotte, G., Rask, M., Vuorenmaa, J., Kahilainen, K.K., Malinen, T., Rognerud, S., Lydersen, E., Amundsen, P.A., Kashulin, N., Kashulina, T., Terentyev, P., Christensen, G., Jackson-Blake, L., Lund, E. and Rosseland, B.O. 2017. Spatial and temporal trends of mercury in freshwater fish in Fennoscandia (1965-2015). **ICP Waters report 132/2017**.

Garmo, Ø., de Wit, H. and Fölster, J. (eds.) 2017. Proceedings of the 33rd Task Force meeting of the ICP Waters Programme in Uppsala, May 9-11, 2017. **ICP Waters report 131/2017**.

Anker Halvorsen, G., Johannessen, A. and Landås, T.S. 2016. Biological intercalibration: Invertebrates 2016. **ICP Waters report 130/2016**.

Escudero-Oñate, C. 2016. Intercomparison 1630: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. **ICP Waters report 129/2016.**

De Wit, H. and Valinia, S. (eds.) 2016. Proceedings of the 32st Task Force meeting of the ICP Waters Programme in Asker, Oslo, May 24-26, 2016. **ICP Waters report 128/2016**.

Velle, G., Mahlum, S., Monteith, D.T., de Wit, H., Arle, J., Eriksson, L., Fjellheim, A., Frolova, M., Fölster, J., Grudule, N., Halvorsen, G.A., Hildrew, A., Hruška, J., Indriksone, I., Kamasová, L., Kopáček, J., Krám, P., Orton, S., Senoo, T., Shilland, E.M., Stuchlík, E., Telford, R.J., Ungermanová, L., Wiklund, M.-L. and Wright, R.F. 2016. Biodiversity of macro-invertebrates in acid-sensitive waters: trends and relations to water chemistry and climate. **ICP Waters report 127/2016**.

De Wit, H., Valinia, S. and Steingruber, S. 2015. Proceedings of the 31st Task Force meeting of the ICP Waters Programme in Monte Verità, Switzerland 6th –8th October, 2015. **ICP Waters report 126/2015**.

De Wit, H., Hettelingh, J.P. and Harmens, H. 2015. Trends in ecosystem and health responses to long-range transported atmospheric pollutants. **ICP Waters report 125/2015**.

Fjellheim, A., Johannessen, A. and Landås, T.S. 2015. Biological intercalibration: Invertebrates 1915. **ICP Waters report 124/2015**.

Escudero-Oñate, C. 2015 Intercomparison 1529: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. **ICP Waters report 123/2015**.

de Wit, H., Wathne, B. M. (eds.) 2015. Proceedings of the 30th Task Force meeting of the ICP Waters Programme in Grimstad, Norway 14th –16th October, 2014. **ICP Waters report 122/2015**.

Fjellheim, A., Johannessen, A. and Landås, T.S. 2014. Biological intercalibration: Invertebrates 1814. **ICP Waters Report 121/2014**.

Escuedero-Oñate. 2014. Intercom¬parison 1428: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. **ICP Waters Report 120/2014**.

De Wit, H. A., Garmo Ø. A. and Fjellheim A. 2014. Chemical and biological recovery in acid-sensitive waters: trends and prognosis. **ICP Waters Report 119/2014**.

Fjellheim, A., Johannessen, A. and Landås, T.S. 2013. Biological intercalibration: Invertebrates 1713. **ICP Waters Report 118/2014**.

de Wit, H., Bente M. Wathne, B. M. and Hruśka, J. (eds.) 2014. Proceedings of the 29th Task Force meeting of the ICP Waters Programme in Český Krumlov, Czech Republic 1st –3rd October, 2013. **ICP Waters report 117/2014**.

Escuedero-Oñate, C. 2013. Intercomparison 1327: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. **ICP Waters Report 116/2013**.

Holen, S., R.F. Wright and Seifert, I. 2013. - Effects of long-range transported air pollution (LTRAP) on freshwater ecosystem services. **ICP Waters Report 115/2013**.

Velle, G., Telford, R.J., Curtis, C., Eriksson, L., Fjellheim, A., Frolova, M., Fölster J., Grudule N., Halvorsen G.A., Hildrew A., Hoffmann A., Indriksone I., Kamasová L., Kopáček J., Orton S., Krám P., Monteith D.T., Senoo T., Shilland E.M., Stuchlík E., Wiklund M.L., de Wit, H. and Skjelkvaale B.L. 2013. Biodiversity in freshwaters. Temporal trends and response to water chemistry. **ICP Waters Report 114/2013**.

Fjellheim, A., Johannessen, A. and Landås, T.S. 2013. Biological intercalibration: Invertebrates 1612. **ICP Waters Report 113/2013**.

Skjelkvåle, B.L., Wathne, B.M., de Wit, H. and Rogora, M. (eds.) 2013. Proceedings of the 28th Task Force meeting of the ICP Waters Programme in Verbania Pallanza, Italy, October 8 – 10, 2012. **ICP Waters Report 112/2013**.

Dahl, I. 2012. Intercomparison 1226: pH, Conductivity, Alkalinity, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni and Zn. **ICP Waters report 111/2012**.

Skjelkvåle, B.L., Wathne B. M. and Moiseenko, T. (eds.) 2012. Proceedings of the 27th meeting of the ICP Waters Programme Task Force in Sochi, Russia, October 19 – 21, 2011. **ICP Waters report 110/2012**.

Fjellheim, A., Johannessen, A., Svanevik Landås, T. 2011. Biological intercalibration: Invertebrates 1511. NIVA-report SNO 6264-2011. **ICP Waters report 109/2011**.

Wright, R.F., Helliwell, R., Hruska, J,. Larssen, T., Rogora, M., Rzychoń, D., Skjelkvåle, B.L. and Worsztynowicz, A. 2011. Impacts of Air Pollution on Freshwater Acidification under Future Emission Reduction Scenarios; ICP Waters contribution to WGE report. NIVA-report SNO 6243-2011. **ICP Waters report 108/2011**.

Dahl, I and Hagebø, E. 2011. Intercomparison 1125: pH, Cond, HCO3, NO3-N, Cl, SO4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 6222-2011. **ICP Waters report 107/2011**.

Skjelkvåle B.L. and de Wit, H. (eds.) 2011. Trends in precipitation chemistry, surface water chemistry and aquatic biota in acidified areas in Europe and North America from 1990 to 2008. NIVA-report SNO 6218-2011 **ICP Waters report 106/2011**.

ICP Waters Programme Centre 2010. ICP Waters Programme manual. NIVA SNO 6074-2010. **ICP Waters report 105/2010**.

Skjelkvåle, B.L., Wathne B. M. and Vuorenmaa J. (eds.) 2010. Proceedings of the 26th meeting of the ICP Waters Programme Task Force in Helsinki, Finland, October 4 – 6, 2010. **ICP Waters report 104/2010**.

Fjellheim, A. 2010. Biological intercalibration: Invertebrates 1410. NIVA-report SNO 6087-2010, **ICP Waters report 103/2010**.

Hovind, H. 2010. Intercomparison 1024: pH, Cond, HCO_3, NO_3-N, Cl, SO_4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 6029-2010. **ICP Waters report 102/2010**.

De Wit, H.A. and Lindholm M. 2010. Nutrient enrichment effects of atmospheric N deposition on biology in oligotrophic surface waters – a review. NIVA-report SNO 6007 - 2010. **ICP Waters report 101/2010**.

Skjelkvåle, B.L., De Wit, H. and Jeffries, D. (eds.) 2010. Proceedings of presentations of national activities to the 25th meeting of the ICP Waters Programme Task Force in Burlington, Canada, October 19-21 2009. NIVA-report SNO 5995 - 2010. **ICP Waters report 100/2010**.

Fjellheim, A. 2009. Biological intercalibration: Invertebrates 1309. NIVA-report SNO 5883-2009, **ICP Waters report 99/2009**.

Hovind, H. 2009. Intercomparison 0923: pH, Cond, HCO_3, NO_3-N, Cl, SO_4, Ca, Mg, Na, K, TOC, Al, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5845-2009. **ICP Waters report 98/2009**.

Ranneklev, S.B., De Wit, H., Jenssen, M.T.S. and Skjelkvåle, B.L. 2009. An assessment of Hg in the freshwater aquatic environment related to long-range transported air pollution in Europe and North America. NIVA-report SNO 5844-2009. **ICP Waters report 97/2009**.

Skjelkvåle, B.L., Jenssen, M. T. S. and De Wit, H (eds.) 2009. Proceedings of the 24th meeting of the ICP Waters Programme Task Force in Budapest, Hungary, October 6 – 8, 2008. NIVA-report SNO 5770-2009. **ICP Waters report 96/2009**.

Fjellheim, A and Raddum, G.G. 2008. Biological intercalibration: Invertebrates 1208. NIVA-report SNO 5706-2008, **ICP Waters report 95/2008**.

Skjelkvåle, B.L., and De Wit, H. (eds.) 2008. ICP Waters 20 year with monitoring effects of long-range transboundary air pollution on surface waters in Europe and North-America. NIVA-report SNO 5684-2008. **ICP Waters report 94/2008**.

Hovind, H. 2008. Intercomparison 0822: pH, Cond, HCO_3, NO_3-N, Cl, SO_4, Ca, Mg, Na, K, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5660-2008. **ICP Waters report 93/2008**.

De Wit, H. Jenssen, M. T. S. and Skjelkvåle, B.L. (eds.) 2008. Proceedings of the 23rd meeting of the ICP Waters Programme Task Force in Nancy, France, October 8 – 10 , 2007. NIVA-report SNO 5567-2008. **ICP Waters report 92/2008**.

Fjellheim, A and Raddum, G.G. 2008. Biological intercalibration: Invertebrates 1107. NIVA-report SNO 5551–2008. **ICP Waters report 91/2008**.

Hovind, H. 2007. Intercomparison 0721: pH, Cond, HCO3, NO3-N, Cl, SO4, Ca, Mg, Na, K, Fe, Mn, Cd, Pb, Cu, Ni, and Zn. NIVA-report SNO 5486-2007. **ICP Waters report 90/2007**.

Wright, R.F., Posch, M., Cosby, B. J., Forsius, M., and Skjelkvåle, B. L. 2007. Review of the Gothenburg Protocol: Chemical and biological responses in surface waters and soils. NIVA-report SNO 5475-2007. **ICP Waters report 89/2007**.

Skjelkvåle, B.L., Forsius, M., Wright, R.F., de Wit, H., Raddum, G.G., and Sjøeng, A.S.M. 2006. Joint Workshop on Confounding Factors in Recovery from Acid Deposition in Surface Waters, 9-10 October 2006, Bergen, Norway; Summary and Abstracts. NIVA-report SNO 5310-2006. **ICP Waters report 88/2006**.

De Wit, H. and Skjelkvåle, B.L. (eds) 2007. Trends in surface water chemistry and biota; The importance of confounding factors. NIVA-report SNO 5385-2007. **ICP Waters report 87/2007**.

Hovind, H. 2006. Intercomparison 0620. pH, K25, HCO3, $NO_3 + NO_2$, Cl, SO_4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-report SNO 5285-2006. **ICP Waters report 86/2006**.

Raddum, G.G. and Fjellheim, A. 2006. Biological intercalibration 1006: Invertebrate fauna. NIVA-report SNO 5314-2006, **ICP Waters report 85/2006**.

De Wit, H. and Skjelkvåle, B.L. (eds.) 2006. Proceedings of the 21th meeting of the ICP Waters Programme Task Force in Tallinn, Estonia, October 17-19, 2005. NIVA-report SNO 5204-2006, **ICP Waters report 84/2006**.

Wright, R.F., Cosby, B.J., Høgåsen, T., Larssen, T. and Posch, M. 2005. Critical Loads, Target Load Functions and Dynamic Modelling for Surface Waters and ICP Waters Sites. NIVA-report SNO 5166-2005. **ICP Waters report 83/2006**.

Hovind, H. 2005. Intercomparison 0317. pH, K25, HCO3, NO3 + NO2, Cl, SO4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-report SNO 5068-2005. **ICP Waters report 82/2005**.

Raddum, G.G. 2005. Intercalibration 0307: Invertebrate fauna. NIVA-report SNO 5067-2005. **ICP Waters report 81/2005**.

De Wit, H. and Skjelkvåle, B.L (eds.) 2005. Proceedings of the 20th meeting of the ICP Waters Programme Task Force in Falun, Sweden, October 18-20, 2004. NIVA-report SNO 5018-2005. **ICP Waters report 80/2005**.

Fjeld, E., Le Gall, A.-C. and Skjelkvåle, B.L. 2005. An assessment of POPs related to long-range air pollution in the aquatic environment. NIVA-report SNO 5107-2005. **ICP Waters report 79/2005**.

Skjelkvåle et al. 2005. Regional scale evidence for improvements in surface water chemistry 1990-2001. Environmental Pollution, 137: 165-176

Hovind, H. 2004. Intercomparison 0418. pH, K25, HCO3, NO3 + NO2, Cl, SO4, Ca, Mg, Na, K, Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-report SNO 4875-2004. **ICP Waters report 78/2004**.

Raddum, G.G. 2004. Intercalibration: Invertebrate fauna 09/04. NIVA-report SNO 4863-2004. **ICP Waters report 77/2004**.

Skjelkvåle, B.L. (ed) 2004. Proceedings of the 19th meeting of the ICP Waters Programme Task Force in Lugano, Switzerland, October 18-20, 2003. NIVA-report SNO 4858-2004. **ICP Waters report 76/2004.**

Raddum, G.G, et al. 2004. Recovery from acidification of invertebrate fauna in ICP Water sites in Europe and North America. NIVA-report SNO 4864-2004. **ICP Waters report 75/2004.**

Hovind, H. 2003. Intercomparison 0317. pH, K25, HCO3, NO3 + NO2, Cl, SO4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-report SNO 4715-2003. **ICP Waters report 74/2003.**

Skjelkvåle, B.L. (ed) 2003. The 15-year report: Assessment and monitoring of surface waters in Europe and North America; acidification and recovery, dynamic modelling and heavy metals. NIVA-report SNO 4716-2003. **ICP Waters report 73/2003.**

Raddum.G.G. 2003. Intercalibration 0307: Invertebrate fauna. NIVA-report SNO-4659-2003. **ICP Waters report 72/2003**.

Skjelkvåle, B.L. (ed.) 2003. Proceedings of the 18th meeting of the ICP Waters Programme Task Force in Moscow, October 7-9, 2002. NIVA-report SNO 4658-2003. **ICP Waters report 71/2003**.

Wright, R.F and Lie, M.C. 2002.Workshop on models for Biological Recovery from Acidification in a Changing Climate. 9-11 september 2002 in Grimstad, Norway. Workshop report. NIVA-report 4589-2002.

Jenkins, A. Larssen, Th., Moldan, F., Posch, M. and Wrigth, R.F. 2002. Dynamic Modelling of Surface Waters: Impact of emission reduction - possibilities and limitations. NIVA-report SNO 4598-2002. **ICP Waters report 70/2002**.

Halvorsen, G.A, Heergaard, E. and Raddum, G.G. 2002. Tracing recovery from acidification - a multivariate approach. NIVA-report SNO 4564-2002. **ICP Waters report 69/2002**.

Hovind. H. 2002. Intercomparison 0216. pH, K_{25}, HCO_3, NO_3 + NO_2, Cl, SO_4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-Report SNO 4558-2002. **ICP Waters Report 68/2002**.

Skjelkvåle, B.L. and Ulstein, M. (eds) 2002. Proceedings from the Workshop on Heavy Metals (Pb, Cd and Hg) in Surface Waters; Monitoring and Biological Impact. March 18-20, 2002, Lillehammer, Norway. NIVA-report SNO-4563-2002. **ICP Waters report 67/2002**.

Raddum.G.G. 2002. Intercalibration 0206: Invertebrate fauna. NIVA-report SNO-4494-2002. **ICP Waters report 66/2002**.

Bull, K.R. Achermann, B., Bashkin, V., Chrast, R. Fenech, G., Forsius, M., Gregor H.-D., Guardans, R., Haussmann, T., Hayes, F., Hettelingh, J.-P., Johannessen, T., Kryzanowski, M., Kucera, V., Kvaeven, B., Lorenz, M., Lundin, L., Mills, G., Posch, M., Skjelkvåle, B.L. and Ulstein, M.J. 2001. Coordinated Effects Monitoring and Modelling for Developing and Supporting International Air Pollution Control Agreements. Water Air Soil Poll. 130:119-130.

Hovind, H. 2001. pH, K_{25}, HCO_3, NO_3 + NO_2, Cl, SO_4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-Report SNO 4416-2002. **ICP Waters report 64/2001**.

Lyulko, I. Berg, P. and Skjelkvåle, B.L. (eds.) 2001. National presentations from the 16th meeting of the ICP Waters Programme task Force in Riga, Latvia, October 18-20, 2000. NIVA-report SNO 4411-2001. **ICP Waters report 63/2001**.

Raddum.G.G. 2000. Intercalibration 0005: Invertebrate fauna. NIVA-report SNO4384-2001. **ICP Waters report 62/2001**.

Raddum, G.G. and Skjekvåle B.L. 2000. Critical Load of Acidifying Compounds to Invertebrates In Different Ecoregions of Europe. Water Air Soil Poll. 130:825-830.

Stoddard, J. Traaen, T and Skjelkvåle, B.L. 2001. Assessment of Nitrogen leaching at ICP-Waters sites (Europe and North America). Water Air Soil Poll. 130:825-830.

Skjelkvåle, B.L. Stoddard J.L. and Andersen, T. 2001. Trends in surface waters acidification in Europe and North America (1989-1998). Water Air Soil Poll.130:781-786.

Kvaeven, B. Ulstein, M.J., Skjelkvåle, B.L., Raddum, G.G. and Hovind. H. 2001. ICP Waters – An international programme for surface water monitoring. Water Air Soil Poll.130:775-780.

Wright, R.F. 2001. Note on: Effect of year-to-year variations in climate on trends in acidification. NIVA-report SNO 4328-2001. **ICP Waters report 57/2001**.

Hovind, H. 2000. Trends in intercomparisons 8701-9812: pH, K_{25}, NO_3 + NO_2, Cl, SO_4, Ca, Mg, Na, K and aluminium - reactive and nonlabile, TOC, COD-Mn. NIVA-Report SNO 4281-2000, **ICP Waters Report 56/2000**.

Hovind, H. 2000. Intercomparison 0014. pH, K_{25}, HCO_3, NO_3 + NO_2, Cl, SO_4, Ca, Mg, Na, K, total aluminium, aluminium - reactive and nonlabile, TOC, COD-Mn. Fe, Mn, Cd, Pb, Cu, Ni and Zn. NIVA-Report SNO 4281-2000. **ICP Waters Report 55/2000**.

Skjelkvåle, B.L., Olendrzynski, K., Stoddard, J., Traaen, T.S, Tarrason, L., Tørseth, K., Windjusveen, S. and Wright, R.F. 2001. Assessment of trends and leaching in Nitrogen at ICP Waters Sites (Europe And North America). NIVA-report SNO 4383-2001. **ICP Waters report 54/2001.**

Stoddard, J. L., Jeffries, D. S., Lükewille, A., Clair, T. A., Dillon, P. J., Driscoll, C. T., Forsius, M., Johannessen, M., Kahl, J. S., Kellogg, J. H., Kemp, A., Mannio, J., Monteith, D., Murdoch, P. S., Patrick, S., Rebsdorf, A., Skjelkvåle, B. L., Stainton, M. P., Traaen, T. S., van Dam, H., Webster, K. E., Wieting, J., and Wilander, A. 1999. Regional trends in aquatic recovery from acidification in North America and Europe 1980-95. Nature 401:575- 578.

Skjelkvåle, B. L., Andersen, T., Halvorsen, G. A., Raddum, G.G., Heegaard, E., Stoddard, J. L., and Wright, R. F. 2000. The 12-year report; Acidification of Surface Water in Europe and North America; Trends, biological recovery and heavy metals. NIVA-Report SNO 4208/2000. **ICP Waters report 52/2000**.

Reports before year 2000 can be listed on request.

NIVA: Norway's leading centre of competence in aquatic environments

NIVA provides government, business and the public with a basis for preferred water management through its contracted research, reports and development work. A characteristic of NIVA is its broad scope of professional disciplines and extensive contact network in Norway and abroad. Our solid professionalism, interdisciplinary working methods and holistic approach are key elements that make us an excellent advisor for government and society.

NIVA
Norwegian Institute for Water Research

Gaustadalléen 21 • NO-0349 Oslo, Norway
Telephone: +47 22 18 51 00 • Fax: 22 18 52 00
www.niva.no • post@niva.no