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# **Ionic composition in Norwegian lakes from 1986 to 2014. - Dominating processes and trends**

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**Ionic composition in Norwegian lakes from 1986 to 2014.  
- Dominating processes and trends.**

“Brand”

(Henrik Ibsen, 1866):

”Værre tider, værre syner gjennom fremtidsnatten lyner! Brittens kvalme stenkullsky sænker sort seg over landet smudser alt det friske grønne kvæler alle spirer skjønne stryger lavt med giftstoff blandet stjæler sol og dag fra egnen, dysser ned som askeregnen over oldtids dømte by”.

The slogan in the 70's was:  
“Think globally and act locally!”

## Summary

In the 70s anthropogenic emissions of sulphur dioxide (SO<sub>2</sub>) and nitrous oxides (NO<sub>x</sub>) in Europe and Russia lead to acidification and fish deaths in many lakes in the South and North of Norway. Therefore the Gothenburg protocol with the aim of reducing SO<sub>2</sub> and NO<sub>x</sub> emissions was developed and a monitoring program on the ionic composition in Norwegian lakes was initialized.

Different interactions between ions in the catchment area and in the lakes are known to play a crucial role in Lake Acidification. While some ions can become toxic (aluminium), increase the H<sup>+</sup> release into the lakes (sodium and chloride), reduce buffer capacities (calcium and magnesium), or act as acids (humus), other ions are important buffers themselves (bicarbonate) and may counteract acidification. Several processes, such as weathering, sea-salt episodes, vegetation changes, or eutrophication, can affect the ionic composition.

The current study evaluates whether climate change related processes can lead to lake acidification, in spite of the reduced acid rain during the last 29 years and change the ionic composition.

A dataset on the ionic composition of 77 Norwegian lakes from 1986 to 2014 is used to test for time trends in their ionic compositions and buffer capacities.

It appears that the lakes are still recovering in the initially most impacted areas in the South and North of Norway. The buffer capacity and pH is increasing and acid based weathering seems to decrease. In less impacted areas, however, weathering seems to increase, as indicated by increased base cation concentrations. My proposed explanations for this increase are climate change related factors, such as increased precipitation and temperature. Another potentially climate change related change is the increased Brownification (TOC concentrations). Besides the increasing pH, thawing permafrost in the north and afforestation are potential explanations. Storm related sea-salt episodes are not increasing, but can be expected for the future.

I conclude that the reduction of SO<sub>2</sub> and NO<sub>x</sub> emissions in Europe and Russia yielded in a recovery of most Norwegian lakes since 1986. However, climate change caused by the increase of CO<sub>2</sub> emissions worldwide leads to a new hazard for Norwegian lakes and should be the target for further efforts.

## Samandrag

På 70-tallet førte antropogene utslepp av svoveldioksid (SO<sub>2</sub>) og nitrogenoksid(NO<sub>x</sub>) i Europa og Russland til forsuring og død fisk i mange innsjøar i Sør- og Nord-Norge. Som en konsekvens av Gøteborgprotokollen med sikte på å redusere SO<sub>2</sub> og NO<sub>x</sub> vart det utvikla eit overvåkingsprogram for langtransportert forureining. Ei grein av dette forskingsprosjektet tok føre seg ione samansetjinga i norske innsjøar

Interaksjonar mellom i nedbørsfelt og innsjøar er kjent for å spele ei avgjerande rolle i forsuring av norske innsjøar. Medan nokre ion kan vere toksiske (e.g. Al), fører til auka forsuring utslepp av H<sup>+</sup> ved kationebyte og redusere buffer kapasiteten ved auka ionemengd. Nokre fungerer som syrer i seg sjølve, slik som Sulfat og humussyre, medan andre ion er viktige bufferar i seg sjølve, slik som e.g. bikarbonater og kan motverke forsuring. I tillegg vert ionekomposisjonen påverkar av klimatiske forhold og naturlige eller antropogene, sjøatepisodar og forvitring av geologi, endra vegetasjon og biologi, eller eutrofierings og fargeendringar i innsjøen.

Denne masteroppgåva vil sjå på endringar i ionesamansetjinga i 77 Norske innsjøar gjennom dei siste 29 åra; belyse prosessane bak forsuring og saltepisodar, og vise korleis nedgang i forureiningar i Europa og klima endringar har og kan kome til å påverke inn innsjøkvaliteten i framtida.

Det ser ut til at innsjøane framleis er i forbetningsfasa i dei områda som var mest påverka av sur nedbør, i sør og Nord aust i Norge. Bufferkapastiteten og pH aukar, og forvitring grunna forsuring ser ut til å minske. På den andre sida ser dei mindre påverka områda ut til å auke i forsuring, noko som er indikert ved auka basekation konsentrasjon. Eg foreslår at denne auken er grunna faktorar som inngår i klima endringar, e.g. endringar i nedbør og temperatur.

Auka fargetal i skandinaviske innsjøar, grunna påverknad frå ein auke i total organisk karbon (TOC). I tillegg til auke i alkalinitet og temperatur, kan tining av permafrost og auke av skoggrensa vere mulege forklaringar er ein anna faktor som kan relaterast til Klima endringar. Storm episodar med tilførsel av sjøvattn syner inga endring enno, men det kan ventast.

Eg konkluderer med at handteringa av SO<sub>2</sub> og NO<sub>x</sub> utslepp frå Europa og Russland har ført til ei betring i dei fleste norske innsjøar sidan 1968. Likevel hevdar eg at klimaendringar, blant anna ved auka CO<sub>2</sub>, utslepp verda over kan føre til ei ny trussel for Norske innsjøar og bør vere et mål for vidare handtering.

## List of abbreviations

ANC	Acid neutralisation capacity
AL-R	Reactive aluminium (Al.R)
ANCoaa	Acid neutralisation capacity, including TOC
BC	Base cations
HCO <sub>3</sub>	Bicarbonate (HCO <sub>3</sub> <sup>-</sup> ),
Ca	Calcium (Ca <sup>2+</sup> )
CEC	Cation exchange capacity
Cl	Chloride (Cl <sup>-</sup> )
CO <sub>2</sub>	Carbon dioxide (CO <sub>2</sub> )
CO <sub>3</sub>	Carbon acid (CO <sub>3</sub> <sup>-</sup> )
DN	Norwegian directorate of nature management Direktoratet for naturforvaltning Direktoratet for naturforvaltning
DOC	Dissolved organic carbon
Fe	Iron (Fe <sup>2+</sup> )
H <sup>+</sup>	Hydronium ion (H <sub>3</sub> O <sup>+</sup> )
IPCC	Intergovernmental panel of climate change Fn's klimapanel
K	Potassium (K <sup>+</sup> )
KLIF	Norwegian climate and pollution agency Klima- og forurensningsdirektoratet
LAL	Labile aluminium (Lal)
MD	Norwegian environment agency, (a merge of before DN and KLIF) Miljødirektoratet
Mg	Manganese (Mg <sup>2+</sup> )
Na	Sodium (Na <sup>+</sup> )
NGU	Norwegian geological survey Norges geologiske undersøkelse Norges geologiske undersøkelse
NH	Ammonium (NH <sub>4</sub> <sup>+</sup> )
NILU	Norwegian institute for air research Norsk institutt for langtransportert forurensning
NMBU	Norwegian University of Life Sciences Norges miljø- og biovitenskapelige universitet
NO	Nitrate (NO <sub>3</sub> <sup>-</sup> )
NO <sub>3</sub>	Nitrate (NO <sub>3</sub> <sup>-</sup> )
NOM	Natural organic matter
NVE	The Norwegian Water Resources and Energy Directorate Norges vassdrags- og energidirektorat
PO	Phosphate (PO <sub>4</sub> <sup>2-</sup> )
SFT	Statens forurensningstilsyn

	Norwegian Pollution Control Authority
SINTEF	The Foundation for Scientific and Industrial Research
	Stiftelsen for industriell og teknisk forskning
SO <sub>4</sub>	Sulphate (SO <sub>4</sub> <sup>2-</sup> )
TOC	Total organic carbon
TOT-N	Total nitrogen
TOT-P	Total phosphorus

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## 1 Introduction

In the early 70s severe fish deaths in Southern Norway have been observed. Later it became apparent that the release of toxic Al species after lake acidification is the main reason (Henriksen et al., 1989). In the 80s another period of sudden fish deaths was observed and the introduction of high amounts of sea-salt after storm events were found to be the cause (Hindar et al., 1995). Different interactions between ions in the catchment area and in the lakes are known to play a crucial role in Lake Acidification. While some ions can become toxic (aluminium), increase the  $H^+$  release into the lakes (sodium and chloride), reduce buffer capacities (calcium and magnesium), or act as acids themselves (humus), other ions are important buffers themselves (bicarbonate) and may counteract acidification (vanLoon and Duffy, 2011). Thus, the processes that change the ionic compositions in lakes and catchments are important factors, controlling the acidity of the lake. Anthropogenic emissions of sulphur dioxide and nitrous oxides were thought to be the main cause for Lake Acidification in the 70s (Henriksen et al., 1989). Later sea-salt episodes became important. Nowadays, the lakes seem to recover from the early anthropogenic impacts and the increase of humic substances and weathering appears to become more important (vanLoon and Duffy, 2011). Both processes are thought to increase with climate change. Before the role of these processes can be understood, different geographical, climatological, and chemical aspects have to be understood and will be explained in the following chapters.

### 1.1 Geography, Geology and Hydrology in Norway

The lakes of the current study are located in all regions of Norway (Figure 1). They range from lakes in coastal areas at 0 m.a.s.l., to mountain lakes at 1250 m.a.s.l (Figure 4). The lakes chosen for the dataset are located in mountain and forest areas (Skjelkvåle and Wright, 1998). Height above sea level as well as distance from the coast has an impact on weathering speed, as will be described in chapter 1.2.6. The geology (Chapter 1.1.2) plays a main role in ion composition and acidification processes, along with precipitation, snowmelt and air transported ions from industry in populated areas.

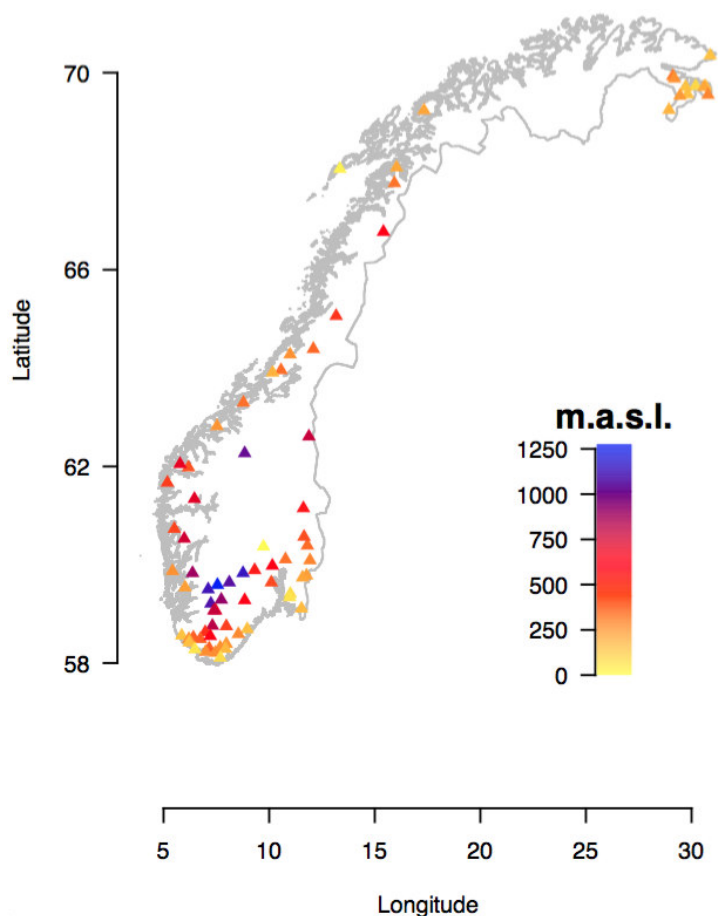


Figure 1: The lakes locations in Norway and their height above sea level.

### 1.1.1 Industry and Population

Anthropogenic environmental impacts in Norway can be caused by industry, agriculture or urbanization. The impact of urbanization is usually limited to a local scale (Booth, 1991). In Norway the highest population densities are found in the Oslo region (Southeast, Figure 2). Agriculture can have impacts on a larger scale via release of fertilizers to catchment areas (Smith et al., 1999). The highest agricultural land use is in the southeast (Figure 2). Besides the land use by agricultural fields, the forest type can have direct impacts on the environment and ionic compositions of the catchment areas. Conifer forests are known to release humic acids, which can enhance acidification, while hardwoods can have the opposite effect (Cannell, 1999). At the moment, most conifer forests are located in the south and most hardwood forests in the north (Figure 2). However, climate change may allow the agricultural use of trees in the north, which would have implications for the ionic composition (Sykes and Prentice, 1996; Cannell, 1999).

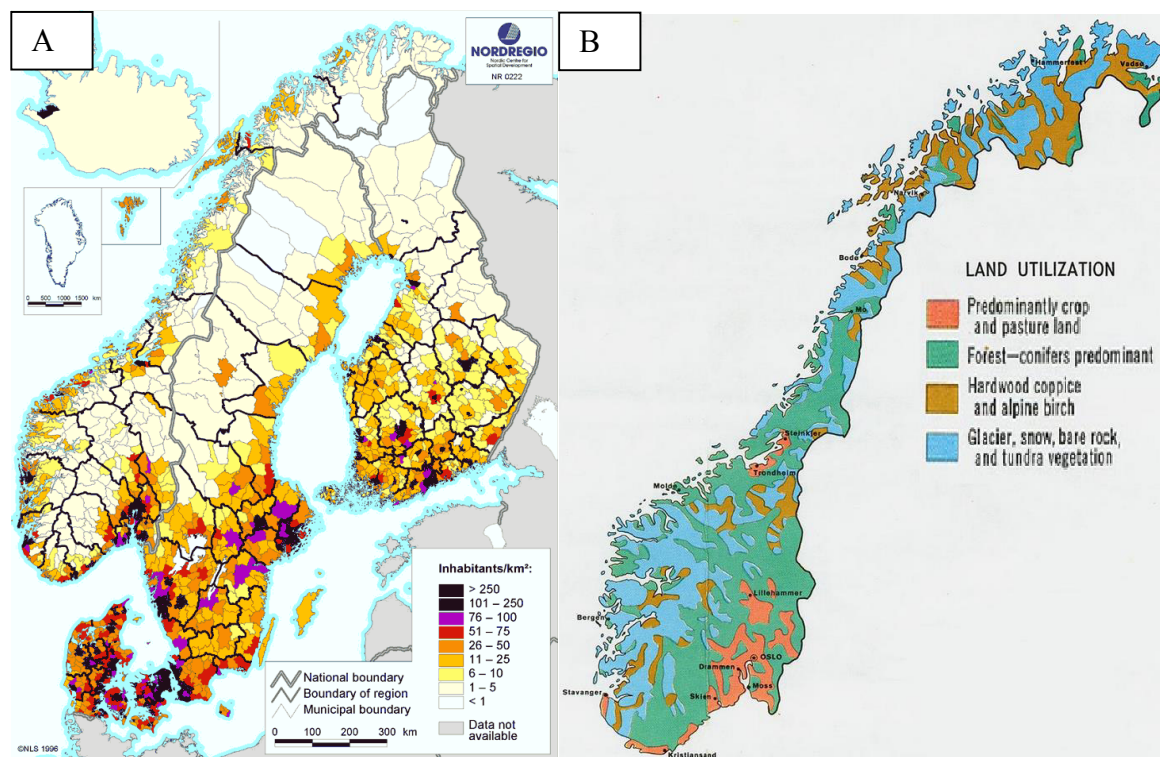


Figure 2: A) Population density in Scandinavia. B) Land utilization in Norway (NationMaster, 2016)

Impacts by industrial activities can have a much larger scale. Even though Norway itself has exclusively hydro-powerplants (except for Svalbard), the high density of coal-powerplants in middle Europe is capable of transporting pollutants such as  $\text{NO}_x$  and  $\text{SO}_2$  to Norway (Figure 3). Another example of industrial impacts on Norway are mining and metal processing sites, which are accumulated on the Russian site of the Eastern Finnmark and which may affect local ionic compositions (Figure 3; Reimann, 1996). In general, the South of Norway, and potentially the Eastern Finnmark can be affected by industrial pollutants from neighbouring countries, while the rest of Norway is mainly affected by local sources.



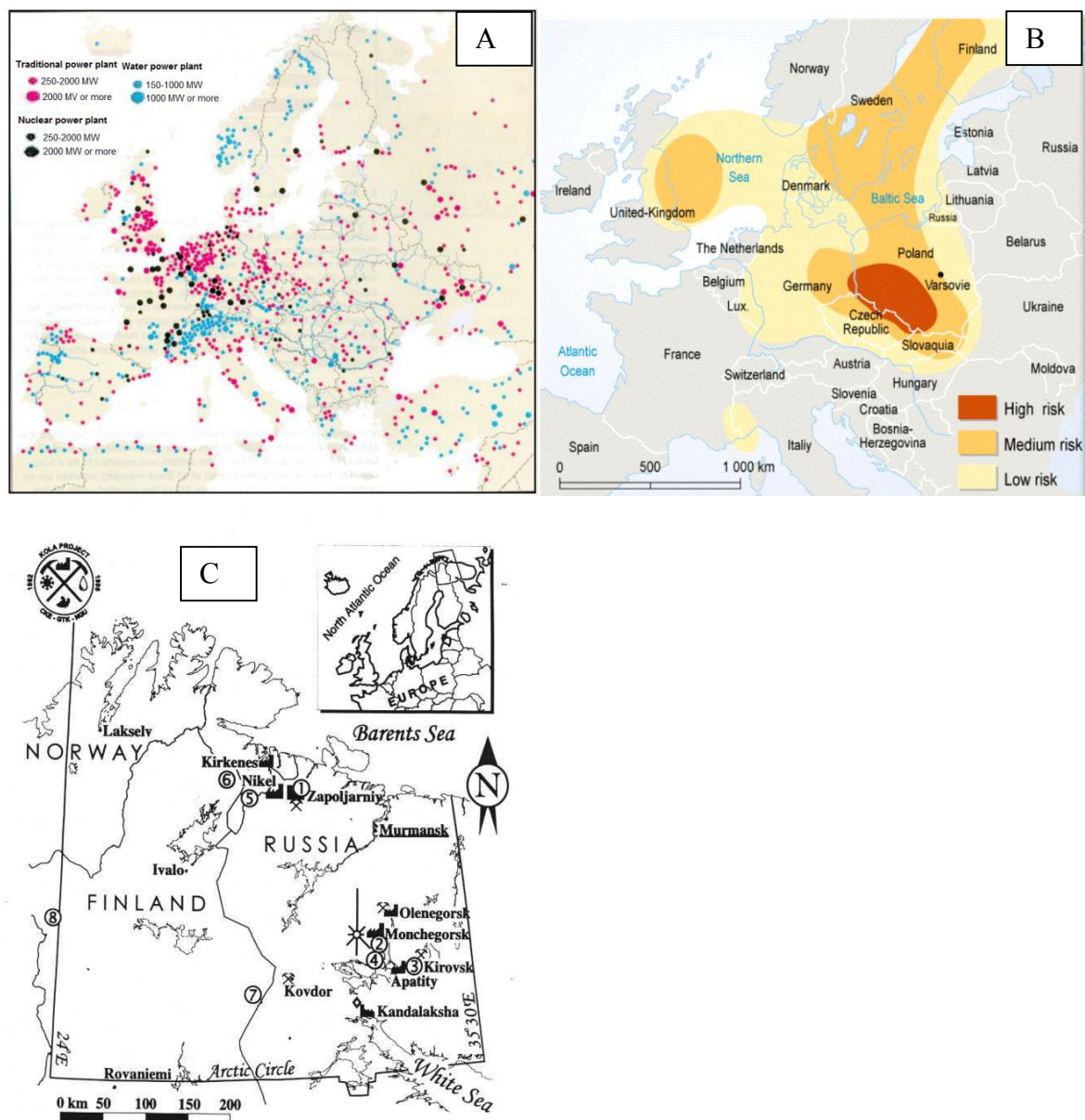


Figure 3: A) Densities of traditional, hydroelectric, and nuclear power plants located in Europe. B) Map showing the areas of acid rain risk ranging from low to high. Note: the general South-eastern trend of high risk away from the industrial heartland of Europe. (Hansen, 2001), C) industrial sites in Russia on the Eastern Finnmork (Reimann, 1996).

### 1.1.2 Geology

Surface stones can consist of both, bedrock and quaternary deposits. The bedrock in Norway consists mainly of hard slow weathering granite and gneiss (Figure 4) (Chapter 1.3.3). Thus, only small amounts of ions can be introduced into the lakes via weathering of these bedrocks (e.g. Sverdrup and Warfvinge, 1993).

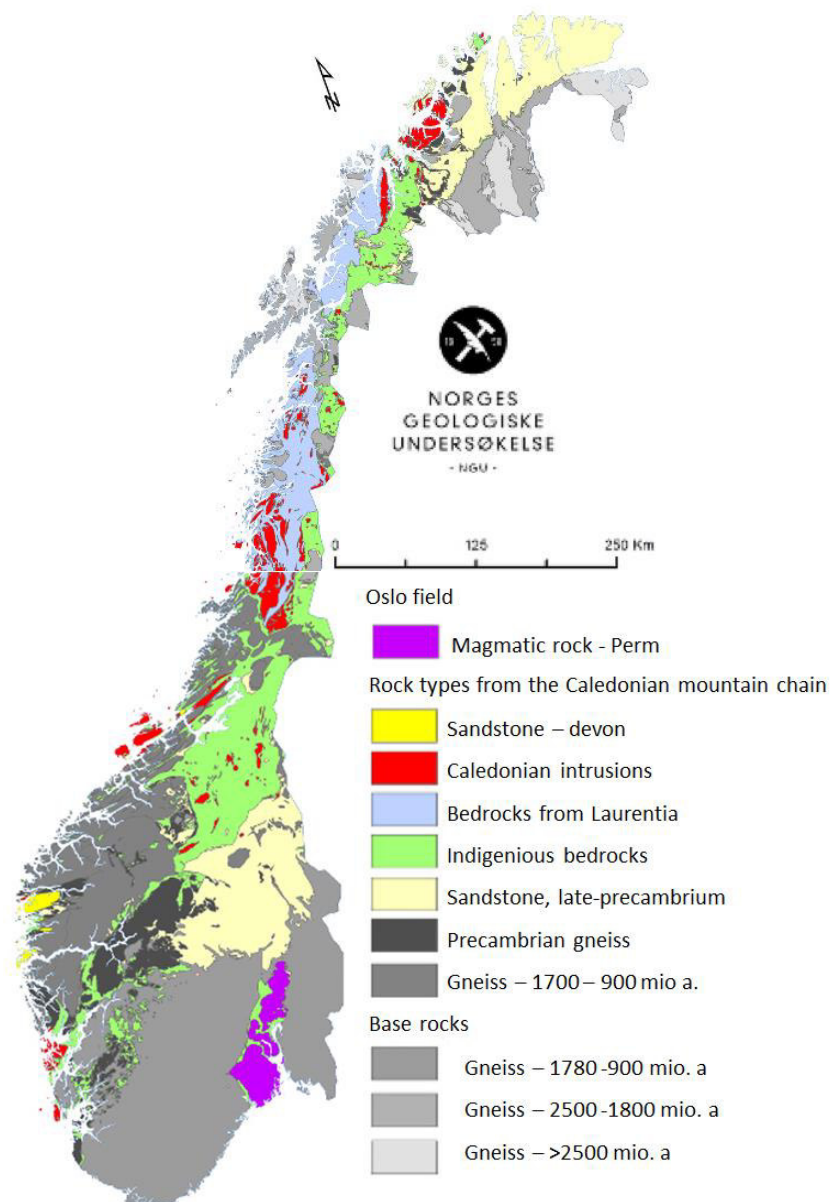


Figure 4: Geological bedrock in Norway, (modified after Nordgulen, 2015).

This yields in a low ionic strength of the lakes in Norway and particularly in the lakes chosen to be in the current study (Henriksen et al., 1989). Thus, even small changes in acidity can have large impacts (Henriksen et al., 1989). The lakes of the presented datasets are diluted and in general are low in ionic content (Skjelkvåle and Wright, 1998).

The glacier ice cap retreated from the little ice age, and the following post glacial rebound occurred firstly in the South of Norway and in lower latitudes at the coast. (Reviewed by Jørgensen et al., 1997). The abrasion by the glacier decreased the thickness quaternary deposits in these areas (Figure 5), and the buffer against acidification is lower. In regions with thin deposits, the retention time for ions is lower, and the drainage from lakes faster in most Norwegian regions (Skjelkvåle and Wright, 1998).



Figure 5: Quaternary deposits in Norway (NGU, 2016)

In the forest areas quaternary deposits and organic matter coverage is thicker, and lead to a longer retention time and higher buffer capacity in the catchment areas (Skjelkvåle and Wright, 1998). The ability for humus and soil particles to bind ions makes these areas more resistant to acidic changes than the mountain lakes with thin deposits (Skjelkvåle and Wright, 1998).

### 1.1.3 Hydrology

Hydrological regimes have important controls for ionic compositions and dynamics. The water balance equation summarizes the possible flows of water (Equation 1; Kane and Yang, 1998; Tollan, 2002). The different parts of the equation can have different importance in different regions in Norway. While snowfall and snow melt play an important role in higher altitudes, rain is more important in lower altitudes (Kane and Yang, 1998). Permafrost may be an additional limitation to water flow in different areas of Norway (Kane and Yang, 1998). The importance of many hydrological flows is controlled by

temperature and altitude (Kane and Yang, 1998).

#### Equation 1: Water balance equation

$$P_r + P_s = E_s + E_f + Q_s + Q_g + \Delta M_i + \Delta M_s + \Delta M_m + \Delta M_g$$

$P_r$  Precipitation as rain

$P_s$  precipitation as snow

$E_s$  Evapotranspiration

$E_f$  Evaporation

$Q_s$  Drainage, surface water

$Q_g$  Drainage, groundwater

$\Delta M_i$  Change storage capacity in snow and ice

$\Delta M_s$  Change in storage capacity of lakes

$\Delta M_m$  Change in storage capacity soil water

$\Delta M_g$  Change in storage capacity in ground water

Precipitation shows generally a west to east gradient with highest precipitation in the west (1500 – 4000 mm) and lowest in the East (500 – 1500 mm). In southern Norway, this gradient is separated by the drainage divide in the middle, causing a rain shadow effect on the east (Hanssen-Bauer and Forland, 2000). The mountains themselves have the highest precipitation (4000 mm), but also a

high amount of snowfall and lower temperatures. Runoff follows the same pattern without the extreme values. Snowfall is measured to have the biggest values in areas with high mountains. The flow rates are important for the distribution in fluvial waters, and in Norway they are typically at their highest in the spring and fall, and lowest during winter and summer (Saltveit & Heggenes 2000). During this period, the ionic composition of the runoff that is flushed into the lakes changes (Lewis and Grant, 1979).

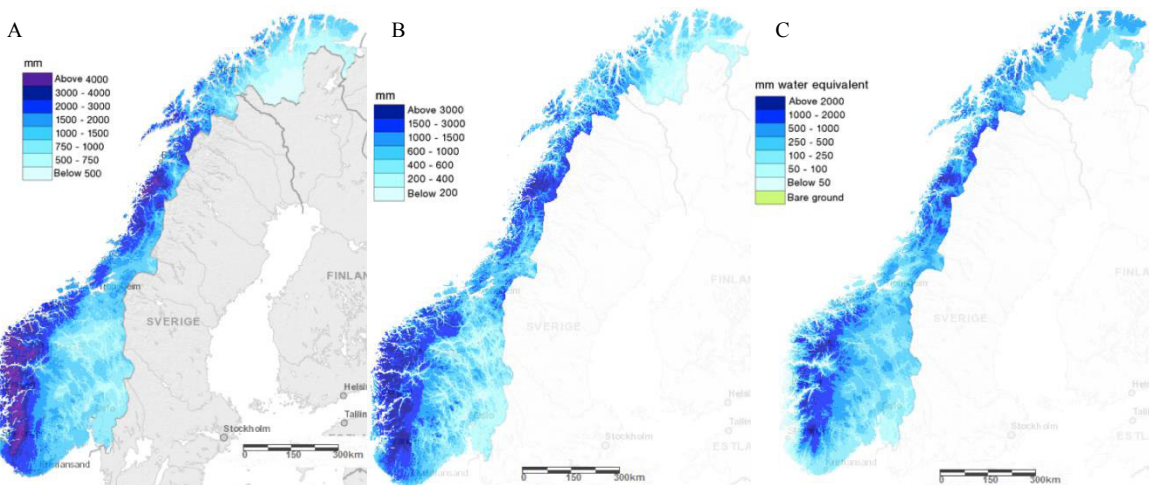


Figure 6: Mean annual levels for the normal period 1971-2000, A) Precipitation, B) Runoff and C) snowfall (senorge.no, 2016).

The highest temperatures are measured at the south- and west coasts of Norway (Figure 7). In the North, a gradient from higher temperatures at the coast, caused by the extensions of the Gulf Stream, to lower temperatures further inland are measured.

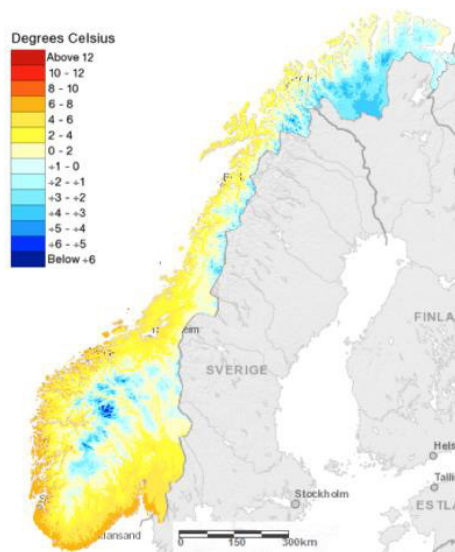


Figure 7. Mean temperature for 1971-2000.

The residence time or water age for a lake explains how long it takes for the water or a substance to get through the system. This depends on the lakes area and how fast water flows in or out of the lake (Tollan, 2002).

The retention time of water and ions is affected by many aspects of the hydrological regimes, and is a part of the total water balance equation that enlightens the complexity of water flow, e.g. in a catchment area (Tollan, 2002).

## 1.2 Ion chemistry and composition

Freshwater consists mainly of four major cations; calcium ( $\text{Ca}^{2+}$ ), magnesium ( $\text{Mg}^{2+}$ ), sodium ( $\text{Na}^+$ ) and potassium ( $\text{K}^+$ ), and four major anions; bicarbonate ( $\text{HCO}_3^-$ ), carbonic acid ( $\text{CO}_3^-$ ) sulphate ( $\text{SO}_4^{2-}$ ) and chloride ( $\text{Cl}^-$ ) (Wetzel 2001, reviewed by: Schwoerbel et al., 2005, Stumm and Morgan, 1996; Drever, 1988; VanLoon and Duffy, 2011). Other important ions in surface waters are the dissolved nutrients nitrogen (nitrite:  $\text{NO}_2^-$ , nitrate:  $\text{NO}_3^-$ , ammonium:  $\text{NH}_4^+$ ) and phosphorus (phosphate:  $\text{PO}_4^{2-}$ ), and suspended and dissolved organic matter (e.g. carbonic acids). Furthermore, trace metal ions can be found (e.g. aluminium: Al), both in suspended and dissolved forms (reviewed by: Schwoerbel et al., 2005). The composition of organic and inorganic ions in a lake determines the water quality.

The ionic strength in Norwegian lakes is usually low outside rural areas, due to low nutrient inputs by agriculture or sewage, and low weathering rates due to the hard bedrock and low thickness of deposits (reviewed by e.g.: Livingstone, 1963). The ionic composition varies a lot, depending on the biota and the bedrock in the catchment area (Livingstone, 1963). Even though the human population density and agricultural intensity in Norway is rather low, local and long-range anthropogenic impacts, such as pollution from rural areas and agriculture play a role for what kind of ions are found in the lake. (Skjelkvåle et al., 2007). The ionic strength is often measured by the electrical conductivity due to the good electrical conductivity of ions compared to pure water. (VanLoon and Duffy, 2011).

### 1.2.1 Inorganic ions

Inorganic ions can originate from different sources, such as the sea, soil, or stones. Sodium chloride ( $\text{NaCl}$ ) usually comes from sea water. Its input is particularly high after storm episodes (sea-salt episodes, Chapter 1.4.3). Another source of Na may be weathering of silicates (e.g. feldspar  $\rightarrow$  kaolinite)(Drever, 1997, VanLoon and Duffy, 2011; Chapter 1.3.3). Anthropogenically  $\text{NaCl}$  may be introduced by road salts (Blomqvist, 2001).

$\text{K}^+$ ,  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$  are base cations (BC) known as micronutrients for plant growth (Drever, 1997). They originate mainly from the weathering of the terrestrial environment, making soil in the catchment area, which may be enhanced by anthropogenic acidification (chapter 1.3.3) or climate change (Chapter 1.3.3). When plants take up these nutrients they are contributing to natural acidification, and change the composition (Drever, 1997). Furthermore Ca and Mg can bind bicarbonate and form precipitates, which reduces the bicarbonate buffer capacity (ANC)(Schwoerbel et al., 2005).

Examples of metal ions are iron (Fe) and Al. They mainly originate from weathering processes (Drever, 1997, VanLoon and Duffy, 2011)and can be introduced by flushing. Acid rain and climate change can accelerate the release of these ions. Anthropogenic pollution, such as mining or

disturbance of Al rich bedrock or minerals when building roads, (Drever, 1997) releases more of these ions. While Fe is an important micronutrient for plants and algae (VanLoon and Duffy, 2011), Al can become toxic in acidic conditions (Chapter 1.3.2.4). Al can be altered by oxidation e.g. during dry periods, while Fe, is in oxidised form in water and contribute to colouring of waters (Chapter 1.5.1).

Bicarbonate ( $\text{HCO}_3^-$ ) is another ion, which reaches the lakes directly from the atmosphere and from the catchment area.  $\text{HCO}_3^-$  can originate from the dissolution of  $\text{CO}_2$  in the lake (atmospheric input), or via weathering calcite containing rocks (Wetzel, 2001; Allan and Castillo, 2007). Bicarbonate plays an important role for buffering the lakes at high pH (Chapter 0).

$\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  can originate from natural sources such as weathering and sea-salt, but are usually increased by anthropogenic activities. Industrial combustion of  $\text{SO}_2$  and  $\text{NO}_x$ , can react with oxygen and water to  $\text{SO}_4^{2-}$  and  $\text{NO}_3^-$  before it reaches the lakes and catchments as precipitation (Nestaas, 2009; Schwoerbel et al., 2005). Both ions can be used as nutrients by plants and bacteria. An additional source of  $\text{NO}_3^-$  may be by nitrification of ammonium ( $\text{NH}_4^+$ )(reviewed by: Callisto et al., 2014)(Chapter 1.5.2).

$\text{NH}_4^+$  usually comes from degradation processes of organic matter, or excretion by fish (Schwoerbel et al., 2005). However, due to the Haber Bosch procedure, it became possible to fix  $\text{N}_2$  and use it as fertilizer (Galloway et al., 2004). Hence, agriculture is a main source of ammonium (Chapter 1.5.2).. Another ion which is important as nutrient and which is often introduced in high levels via agriculture is  $\text{PO}_4^{2-}$  (reviewed by: Schwoerbel et al., 2005).

Silicium (Si) originates mainly from weathering processes of rocks like feldspar (Ulrich, 1983)and has important implications for the buffer capacity of a catchment area (Silicate buffer).

Many ions dissolve in water due to its bipolar character, which allows ionic bonds with both, cations and anions. However, the solubility of a certain ion is dependent on; i) the distance of two ions from each other, ii) the type of ion, and iii) on the dielectrical constant (Equation 2)(Schwoerbel et al., 2005).

**Equation 2: Solubility of ions in water (Schwoerbel et al., 2005).**

$$K = \frac{e^+ \cdot e^-}{a^2}$$

$$K = \frac{1}{D} - \frac{e^+ \cdot e^-}{a^2}$$

$a$ : Distance from each other

$e^-$ : valency of anions

$e^+$ :valency of cations

D: dielectric constant

Examples of solubility of different ions are given in Table 1.

**Table 1: Solubility for typical salts in water ( g 100 ml<sup>-1</sup>) (Schwoerbel et al., 2005).**

<b>Substance</b>	<b>(g 100ml<sup>-1</sup>)</b>
NaCl	35.9
Na <sub>2</sub> CO <sub>3</sub>	21.6
MgCl <sub>2</sub> * H <sub>2</sub> O	53.4
MgCO <sub>3</sub>	0.011
Ca(OH) <sub>2</sub>	0.118
CaCO <sub>3</sub> (Calcite)	1.4*10 <sup>-3</sup>

### 1.2.2 Organic ions

Other kind of ions are related to organic matter. Due to different charges in organic matter (e.g. carbonic acids, NH<sub>4</sub><sup>+</sup>), the surplus charge is usually negative and the main organic matter compound in lakes is humus, which is the mixture of high molecular weight organic compounds and refractory organic matter. In the current study, total organic carbon (TOC) is measured as a proxy for humus. Since TOC is also known to be the main compound absorbing light, it is also used as a proxy for light absorbance or brownification (Wetzel, 2001). TOC comes mainly from detritus of living biota, e.g. forest leaves and branches, flowers and grass, in the catchment area, and in little (dust) or no degree from air (Steinberg, 2003).

For a few years, more specific measurements of the organic matter quality have been done. Since the quality and degradability of organic matter increases with its nitrogen and phosphorous content, the total nitrogen (TOT-N) and the total phosphorous (TOT-P) concentrations have been measured. Additionally, the TOT-N and TOT-P ratio can be used to assess the limiting nutrient after Redfield (optimum 16 :1)(Dodds and Smith, 2016).

### 1.2.3 Cation exchange

Cation exchange is the process by which cations bound to a certain anion are exchanged against other cations (Hillel, 2012). This may lead to different retention times for different ions. In Norwegian lakes, dangerous substances such as Al and other metals or H<sup>+</sup> can be remobilized from the soil via cation exchange processes. The process is mostly happening in the pore water.

Cation exchange processes are possible due to different binding behaviours of different cations. In fact, ions with a higher valence charge and a smaller hydrated radius form stronger bonds to other ions (vanLoon and Duffy, 2011). The strength of the bonds is usually in the following order: Al<sup>3+</sup> > Ca<sup>2+</sup> > Mg<sup>2+</sup> > K<sup>+</sup> = NH<sub>4</sub><sup>+</sup> > Na<sup>+</sup> (vanLoon and Duffy, 2011). H<sup>+</sup> is difficult to place in this order, since H<sup>+</sup> dominated systems lead to degradation of organic matter and colloid (particles ≤1 μm) formation which can have different affinities to certain anions. In fact, Al<sup>3+</sup> and H<sup>+</sup> become more important at lower pH (Hillel, 2012).

**Table 2: Cation exchange capacities (CEC) in different minerals and organic matter (VanLoon and Duffy, 2011)**

Cation exchange is most effective if the ions are close to the particle surfaces (VanLoon and Duffy, 2011). The ionic charge around a soil particle is decreasing with increasing distance from the particle surface area (vanLoon and Duffy, 2011). Another factor that determines the effectiveness of cation exchange is the type of soil particle and its surface charges. The binding strength of different anions can be as follows:  $\text{PO}_4^{2-} > \text{As}^- > \text{SO}_4^{2-} > \text{Cl}^- = \text{NO}_3^-$ , at normal pH (Hillel, 2012).

Some surface areas are permanently negatively charged, like clay minerals; others, like humus, can have a mix of both charges (Hillel, 2012). Thereby different Composition and thicknesses of the soil will effects the cation exchange capacity (CEC).

The technical definition of CEC is given in Equation 3 as the as the equivalent sum of cations.

**Equation 3: Definitions of cation exchange capacity (VanLoon and Duffy, 2011)**

$$\text{CEC} = \sum \text{Na}^+, \text{K}^+, \text{NH}_4^+, \text{Ca}^{2+}, \text{Mg}^{2+}, \text{Al}^{3+}, \text{H}^+, \dots$$

One example of cation exchange processes is nutrient acquisition of plants via their roots from the surrounding soil. Thereby, they take up nutrient cations ( $\text{Ca}^{2+}$ ,  $\text{Mg}^{2+}$ ) and releases  $\text{H}^+$ , making the surrounding soil more acidic.

### 1.3 Buffer systems, capacity and critical limits

#### 1.3.1 Buffer capacity (ANC, Alkalinity)

The buffer capacity is the quantification of a buffer system to pH changes after addition of acids. The resistance to pH changes can be achieved by different systems; i) an equilibrium system between a weak acid and its related base (e.g. bicarbonate buffer system); or ii) cation exchange processes (e.g. Silicate buffer). In general, the increase of  $\text{H}^+$  in the systems after addition of strong acids (e.g.  $\text{H}_2\text{SO}_4$ ,  $\text{HNO}_3$ ) will be lower than expected without the buffer systems (vanLoon and Duffy, 2011). The buffer capacity a lake have is dependent on the catchment areas characteristic (Figure 4, Figure 6).

Anion neutralisation capacity (ANC) and alkalinity (ALK) are two measures of buffer capacities. The technical definition of ANC is the sum of Bicarbonate, and other ions ( $\text{A}^-$ ) minus  $\text{H}^+$ , and Al cations (Equation 4). Due to the low concentrations of most ions compared to bicarbonate in natural systems in Norway  $\text{A}^-$  and  $\text{Al}^{n+}$  is set to 0 (Garmo et al., 2014).

**Equation 4: Simplified general ANC calculation, for natural Norwegian systems (GARMO ET AL., 2014).**

$$\text{ANC} = [\text{HCO}_3^-] + [\text{A}^-] - [\text{H}^+] - [\text{Al}^{n+}]$$

$$\text{ANC} = [\text{HCO}_3^-] - [\text{H}^+]$$

The system is in ionic balance when the sum of strong anions and the sum of base cations are the same (Equation 13). By solving this equation towards ANC ( $\text{HCO}_3^- - \text{H}^+$ ), we can calculate the ANC by using only the base cations and strong anions by subtracting the sum of measured anions



from the sum of measured base cations (Equation 5). Positive ANC values indicate a surplus of cations and bicarbonate; negative values indicate that there are more H<sup>+</sup> than bicarbonate.

#### Equation 5: The relationship between ionic balance and ANC

$\Sigma$  charge of cations [ $\mu\text{ekv } L^{-1}$ ] =  $\Sigma$  charge of anions [ $\mu\text{ekv } A^{-1}$ ]

$$\Sigma [H^+] + [Al^{n+}] + [Ca^+] + [Mg^{2+}] + [Na^+] + [K^+] + [NH_4^+] = \Sigma [Cl^-] + [SO_4^{2-}] + [NO_3^-] + [HCO_3^-] + [A^-]$$

$$ANC = ([Ca^{2+}] + [Mg^{2+}] + [Na^+] + [K^+] + [NH_4^+]) - ([Cl^-] + [SO_4^{2-}] + [NO_3^-])$$

$$ANC = \Sigma \text{ base cations} - \Sigma \text{ strong acid anions}$$

Alkalinity is another measure for the buffer capacity in liquids. It includes the carbonate buffer system, weak bases and potential organic acids as buffer. The sum of the buffer systems minus the sum of H<sup>+</sup> defines the Alkalinity (Equation 6) and can directly be measured by titrations with HCl (Wright, 1983; Garmo et al., 2014).

#### Equation 6: Definition of Alkalinity

$$\text{Alkalinity} = \text{Alk} = [HCO_3^-] + [CO_3^{2-}] + [OH^-] + [OA^-] - [H^+]$$

The input of seawater, or lime directly to the lake is a common method to increase the buffer capacity. Conversely, seawater in the catchment area may lead to a reduction of the buffer capacity in lakes, due to cation exchange processes.

(Chapter 1.4.3)

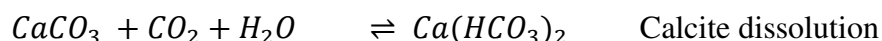
### 1.3.2 Buffer systems

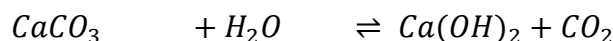
There are five ways for the lakes' catchments to buffer against acidity depending on the pH: i) Bicarbonate buffer (pH > 8 to 6.2); ii) Silicate buffers (pH 6.2 to 5.0); iii) Cation exchange buffer (pH 5.0 to 4.2); iv) Aluminium buffer (pH 4.2 to 2.8); and v) Iron buffer (pH 3.8 to 2.4) (reviewed by Norton, 1989). The use and effectiveness of these buffer systems depend on the availability of the buffers and the pH. When the lake system is gaining more H<sup>+</sup> than the buffer system can neutralize the buffer capacity is reached and the acidification accelerates.

#### 1.3.2.1 Bicarbonate buffer

The bicarbonate buffer system in a lake is formed via CO<sub>2</sub> input from the atmosphere or via lime inputs after weathering processes. CO<sub>2</sub> dissolves in water via hydrolysis and forms carbonic acid (H<sub>2</sub>CO<sub>3</sub>) (Equation 7). H<sub>2</sub>CO<sub>3</sub> is then converted to its related bases (bicarbonate (HCO<sub>3</sub><sup>-</sup>), and carbonate (CO<sub>3</sub><sup>2-</sup>) (Wetzel, 2001)). In the end a buffer system of CO<sub>2</sub>, HCO<sub>3</sub><sup>-</sup> and CO<sub>3</sub><sup>2-</sup> is formed. Due to the formation of carbonic acid, CO<sub>2</sub> dissolution in water can lead directly to acidification, but once in the system its corresponding bases can act as an efficient buffer system.

#### Equation 7: Hydrolysis and dissociation of ions when CO<sub>2</sub> enters from the atmosphere to the water body





The equilibrium between the three parts of the buffer system depends on the pH. At low pH values, most compounds of the carbonate buffer are available as  $\text{CO}_2$ , while higher pH values convert more  $\text{CO}_2$  to  $\text{CO}_3^{2-}$ . Hence, a low pH can reduce the buffer capacity of the system by transforming the buffers into a gas.

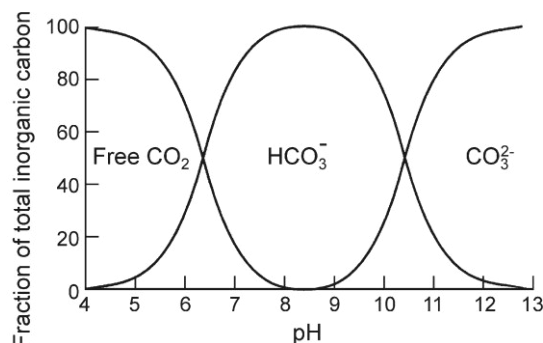


Figure 8: Influence of pH on the relative proportions of inorganic carbon species in solution (Allan, modified from Wetzel, 2001).

The carbonate buffer can be attacked by  $\text{Ca}^{2+}$  and  $\text{Mg}^{2+}$ , which can be released by weathering processes. Thereby carbonate gets bound to the cations and is not available to neutralize free  $\text{H}^+$  anymore. In soils that are poor in bases, such as sandstone, the carbonate buffer can be depleted by  $\text{Mg}^{2+}$  and  $\text{Ca}^{2+}$  release alone (reviewed by: Schwoerbel et al., 2005).

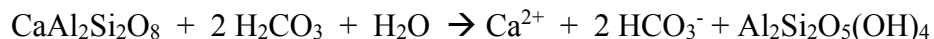
The ratio of the different carbon species in the water solution is changed when the pH concentration changes. The diffusion of  $\text{CO}_2$  also increases with temperature and pressure and decreases with higher salinity. Both  $\text{CO}_2$  and  $\text{O}_2$  dissolve more easily in water with a temperature at  $0^\circ\text{C}$  than at  $30^\circ\text{C}$  (Table 3, Schwoerbel et al., 2005).

Table 3: Solubility of different gases under atmospheric pressure at different water temperatures.

Gas	Partial-pressure %	0 °C	10 °C	20 °C	30 °C
$\text{O}_2$	20.99	14.5	11.1	8.9	7.2
$\text{N}_2$	78.0	22.4	17.5	14.2	11.9
$\text{CO}_2$	0.33	1.005	0.70	0.51	0.38

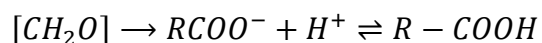
### 1.3.2.2 Silicate buffer

Between a pH of 6.2 and 5, silicate is the only available buffer system. The silicate buffer system is related to weathering of silicate minerals (Ulrich, 1983). Thereby alkali and earth alkali cations are released from the silicate lattice and can be replaced by  $\text{H}^+$  (Cation exchange) (Ulrich, 1983). An example of the principle is the weathering of Ca-feldspar to kaolinite (Equation 8). The process may be rather indirect and cations may first be exchanged with clay particles. The  $\text{H}^+$  taken up in this process is usually included in silicic acid and eventually converted into  $\text{SiO}_2$  and  $\text{H}_2\text{O}$  (Ulrich, 1983).

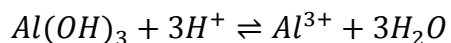
**Equation 8: Weathering of Ca-feldspar to kaolinite.****1.3.2.3 Cation Exchange buffer (Humus buffer)**

Organic soil particles (especially humus) are known to act as cation exchange buffer systems. Thereby, mainly  $\text{Ca}^{2+}$  and free  $\text{H}^+$  can be exchanged (Ulrich, 1983).

Under neutral or acidic pH, humus particles usually have a net negatively charged surface so that  $\text{H}^+$  or other cations can bind and cation exchange processes are possible (Equation 9). In fact, if there is more humus in a catchment area, the CEC and the buffer capacity of the catchment increases. This means that the lake in the catchment area is more adaptable to changes, and has a better ability to resist acidification.

**Equation 9: Humus buffer in soil surface****1.3.2.4 Aluminium buffer**

Aluminium comes primary from silicates, as clay minerals (Equation 10). The Al buffer system is most effective when; i) the pH range is between 5 and 4.2, ii) the exchangeable fraction of Ca drops below 5 to 10 % and iii) aluminium rises to 90%. If  $\text{H}^+$  stays attached to clay minerals for a week or month, clay will release Al from swollen intermediate layers into the pore water (Ulrich, 1983).

**Equation 10: Release of Al-ion when silicates are protolyzed****1.3.2.5 Iron buffer**

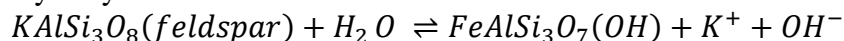
At pH values below 3.2 Fe starts dissolving in concentrations and becomes important for the ecological system (Ulrich, 1983). The direct effects are the release of a variety of heavy metals and nutrient deficiency (Ulrich, 1983). Due to the extremely low pH necessary for the iron buffer system, it is only observed for certain times with high  $\text{H}^+$  production in the soil (Ulrich, 1983).

**1.3.3 Weathering (trend in base cations)**

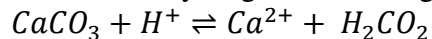
Weathering processes can release a variety of ions via the degradation of rocks and bedrock material. It can be caused by physical or chemical mechanisms.

For the release of BC to the lakes, chemical weathering is the most important. Five types of chemical weathering can be differentiated; i) the splitting of water in the crystal structure of the mineral (hydrolysis), ii) Acid-base reactions, iii) redox reactions, iv) dissolution and recrystallization (isomorph substitution), v) hydration reactions, vi) chelation reactions (vanLoon and Duffy, 2011).

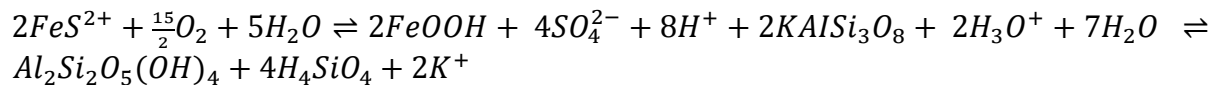
Hydrolysis:  $\text{K}^+$  is released from bedrock with water



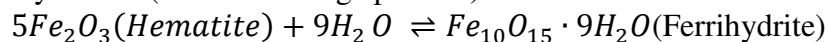
Acid dissolution in calcium carbonate rich alkaline areas can release more cations to the water. The acids may originate from e.g. acid rain, or plant cation exchange.



Oxidation



Hydration (cation exchange process):



Acid precipitation and sea-salt events both accelerate weathering significantly via cation exchange processes. Thereby  $Na^+$  from sea-salt or  $H^+$  from the acid rain can replace the base cations of the rocks. The BC can then bind ions, such as bicarbonate in the soil and thereby reduce the buffer capacity. Hence, weathering can increase lake acidification (vanLoon and Duffy, 2011; Schuiling and Krijgsman, 2006). Another dangerous compound that can be released from the bedrock via cation exchange processes is Al.

Areas close to the coast or above sea level are all influenced by physical weathering and erosion such as: i) weather impacts as snow and ice which lead to thawing and freezing between summer and winter months, ii) precipitation and runoff (particularly in spring and autumn), and iii) glacial abrasion (vanLoon and Duffy, 2011). Bedrock that is not hard enough cannot resist these weather impacts. Therefore, they are grained into smaller pieces and release cations and nutrients to the environment quicker. After the softest material weathers completely, harder material can still be degraded by weathering. As an example, water can run through thin cracks and reach harder bedrock. When the water then freezes in winter, a mechanically expansion of the crack is the consequence (vanLoon and Duffy, 2011). Each time the crack expands may lead to smaller rock fragments separating from the bedrock. In some cases, it can lead to rock avalanches (vanLoon and Duffy, 2011).

#### 1.3.4 Critical limits

Critical limits are the limits of chemical parameters for the survival and health of economically, or ecologically important and vulnerable species populations in an ecosystem. Especially, the ANC value has been found to be an important indicator for fish survival. The higher the ANC value, the better the fish survival (e.g. Hesthagen et al., 2008; Austnes and Lund, 2014). A critical load is defined as the maximum concentration of a pollutant an ecosystem can receive without negative effects (Austnes and Lund, 2014).

Before a system becomes too acidic, each country defines which ecosystem is most sensitive to acidification. In Germany for example forests are the most sensitive systems, while in Norway freshwater systems are most sensitive.

For pH values below 5.4 can be considered to be harmful for fishes, and for inorganic Aluminium  $50 \mu\text{g l}^{-1}$  (Figure 9). However the values are different for different fish species (Kroglund et al., 2002) and depend on different other ions, such as Ca and TOC. Since Norwegian lakes contain >2500 fish populations and of the rivers there are > 21 which has populations of Salmon, the limits

should be rather conservative (Kroglund et al., 2002). In the current study ANC values of  $5 \mu\text{eq l}^{-1}$  are used as threshold to differentiate very poorly buffered lakes and rather well buffered lakes (ANC lakes) as described by Haaland et al.,(2009).

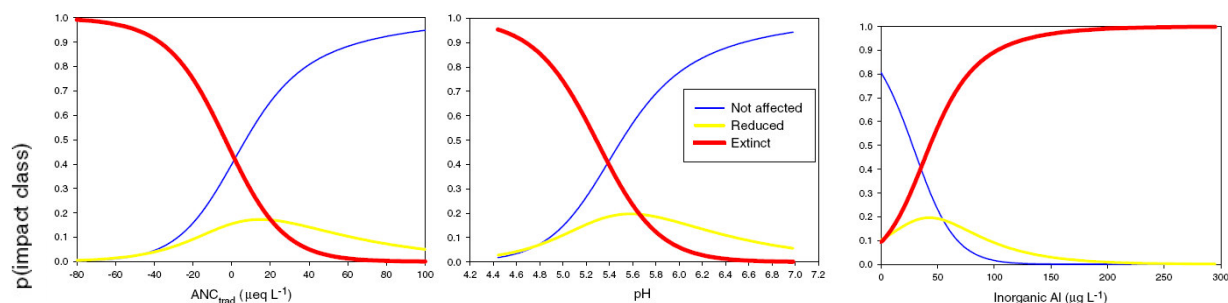


Figure 9: critical limits of ANC, pH and inorganic Al for a brown trout population (Hesthagen et al., 2008).

After the Water Framework Directive classification the critical loads of ANC depend mainly on Ca and TOC concentrations (Austnes and Lund, 2014). Since ANC is basically defined as the surplus of bicarbonate to  $\text{H}^+$ , a high concentration of Ca would bind and take out more bicarbonate. Hence, the critical ANC load must be higher if Ca concentrations increase (Austnes and Lund, 2014). TOC in freshwater usually introduces humic acids, which act as an acids themselves and reduce the buffer capacity. The good-moderate boundary values for 2009 and 2013 are given in Table 4 (Austnes and Lund, 2014).

Table 4. Good-moderate boundary values of ANC in 2009 and 2013 at different Ca and TOC concentrations as defined by Austnes and Lund (2014).

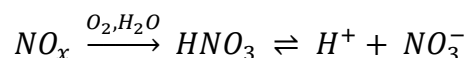
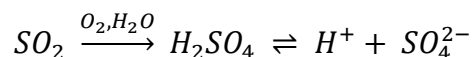
Ca ( $\text{mg l}^{-1}$ )	TOC ( $\text{mg l}^{-1}$ )	ANC 2009 ( $\mu\text{eq l}^{-1}$ )	ANC 2013 ( $\mu\text{eq l}^{-1}$ )
<0.25	<2	20	0
0.25–0.5	<2	20	5
0.5–0.75	<2	20	10
0.75–1.0	<2	20	20
1–4	<2	20	20
<0.25	2–5	25	5
0.25–0.5	2–5	25	10
0.5–0.75	2–5	25	15
0.75–1.0	2–5	25	25
1–4	2–5	30	30
<0.25	>5	35	10
0.25–0.5	>5	35	15
0.5–0.75	>5	35	20
0.75–1.0	>5	35	30
1–4	>5	40	30

## 1.4 Lake Acidification

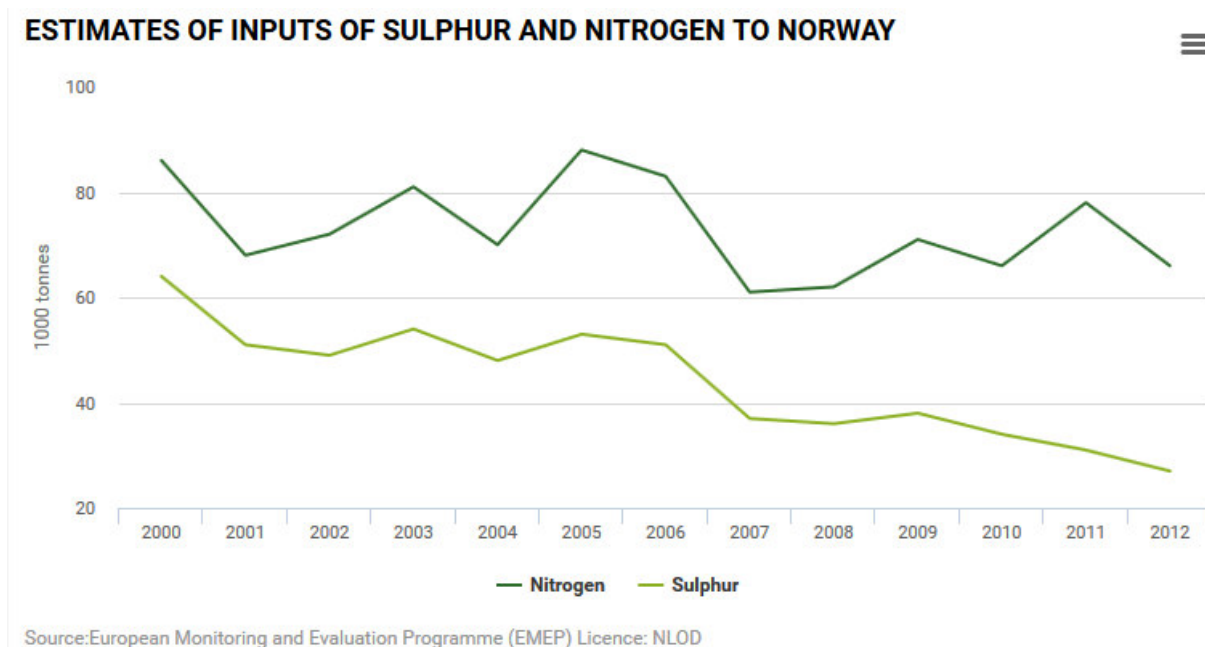
### 1.4.1 Acid rain

Air pollutants from the industry were first recognized as discoloured snow and waters in early 1900 (Dahl, 1926), with following death of fishes. The pollutants were found to come via wind systems mainly from Great Britain, Germany, Poland and Russia to Norway (Figure 3). The release of  $\text{SO}_2$  and  $\text{NO}_x$  from power plants and factories in the 70's have been found to be the main source of the strong acids reaching Norway (Nestaas, 2009). In the atmosphere, these compounds react with oxygen and water and become strong acids which precipitate and reach catchment areas and lakes (Equation 11; Schwoerbel et al., 2005). Acid rain is a term used for both dry deposits and wet deposits that contain a higher than normal content of nitric ( $\text{HNO}_3$ ) and sulphuric ( $\text{H}_2\text{SO}_4$ ) acids that comes from the lower troposphere (Nestaas, 2009).

**Equation 11. Reaction of  $\text{SO}_2$  and  $\text{NO}_x$  in the atmosphere to strong acids**



Recently the  $\text{SO}_2$  emissions in Europe were drastically reduced due to advanced technologies for emission cleaning and changing power sources away from sulphur rich coal power plants. Figure; Benum, 2015). Also within Norway, local pollution sources decreased. In 1976 local pollutant from Norwegian industry and stone mining consisted of 114 000 tons of  $\text{SO}_2$ . This was 72% of the total pollutants. In 1993 the  $\text{SO}_2$  concentrations of the mining outlets decreased down to 23 000 tons. Also heavy metal concentrations like mercury cadmium, zinc, and lead (Norwegian Environment Agency, 2015).



**Figure 10. Decrease of sulphur and nitrogen emissions reaching Norway (European Monitoring and Evaluation Programme)**

Acid rain appears to threaten the biodiversity in lakes and rivers seriously. In 1990 overall, > 9000 fish stocks were lost and > 5000 fish stocks were magnificently depleted after acidification. Recent data are lacking, but a study in 2008 estimated that the affected areas decreased from 20.000 km<sup>2</sup> in 1990 to 13.000 km<sup>2</sup> in 2006, which equals a reduction of 38% (Norwegian Environment Agency, 2015).

Lime has been used in some waters that are highly acidic to reverse and neutralize the processes. This was considered a first aid help for the most severely affected lakes (Lawrence et al., 2016; Norwegian Environment Agency, 2015). However, the aim of liming should not be the neutralization of naturally acidic environments and the hazard for indigenous species should be considered (Lawrence et al., 2016). Already in 1987 several methods if liming were developed to save lakes and water courses. In 1988 14 mio. NOK of tax money was dedicated to restore freshwater systems via liming.



**Figure 11 Lime treatment of a Norwegian lake (Norwegian Environment Agency, 2015)**

Nowadays, considerable resources are still used to keep the pH-value Norwegian waters at a level that biota can live and thrive in. Recently, 78-88 million NOK were spent annually for

liming rivers and lakes (Norwegian Environment Agency, 2015). However, anthropogenic acidification appears to become less of a problem due to the reduction of emissions. Thus, the focus should go to the understanding and countermeasures of natural and potentially climate change related sources of  $H^+$ , such as sea-salt events and afforestation.

#### 1.4.2 Aluminium remobilization

After the first fish deaths in southern Norway it appeared that the Al remobilization in the catchment areas and subsequent transport into the freshwater systems was the main cause. The input of strong acids by acid rain is thought to be the main reason for Al remobilization (Cronan and Schofield, 1979).

Three aluminium species are differentiated in the current thesis (Røgeberg and Henriksen, 1985); i) reactive aluminium (Al.R), or non-labile monomeric aluminium; ii) labile monomeric aluminium (LAL), which most toxic and can bind to the negatively charged fish mucus; and iii) non labile Aluminium (Al.II), which is usually bound in complexes is unreactive, and often positively related to TOC.

Different species of aluminium have different affinities to fish gills and organs, disturbing its normal functions (Schwoerbel et al., 2005). This can eventually lead to death. Only the positively charged fractions of Al are considered to be harmful for fishes. To figure out how much Al cations are potentially released in acidic environments, measurements based on technical cation exchange with  $Na^+$  can be conducted. After the cation exchange only inorganic fraction of Al that are positively charged are retained (LAL), while the neutral and organically bound fractions is not retained (Al.II). The fraction that is not retained can be measured directly. The inorganic form, that is retained, is measured as a difference between the organic forms and an untreated sample (Al.R).  $(Al.II/Al.R)$  (Røgeberg and Henriksen, 1985).

While inorganic base cations are important nutrients that contribute to the living biota, Al in bioavailable forms is toxic for fishes. In fact, the main problem for biotic life in acidic lakes is the remobilization  $Al_3^+$  when the pH decreases (Figure 12). Al is only bioavailable in acidic conditions. In some streams and rivers acidification and Al-toxicity is the reason for important organisms, such as fishes, to disappear (reviewed by Schofield, 1976)? Thereby, metals can be remobilized after acidification of the lake.



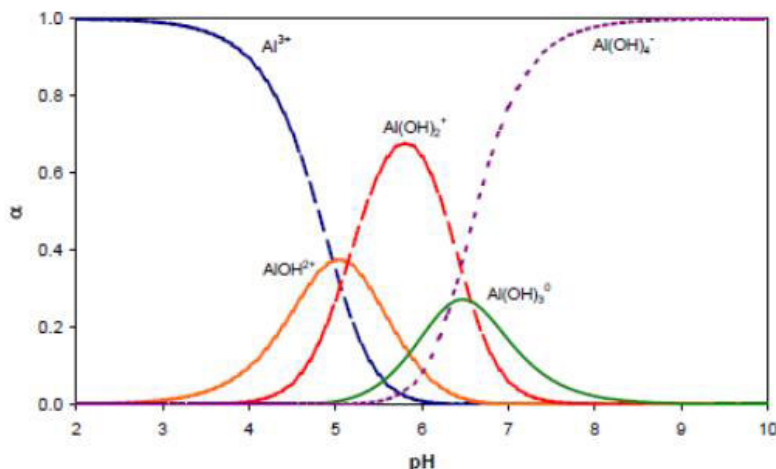


Figure 12. Aluminium speciation at different pH levels (Holt, 2002).

The mobilization of Al is also dependent on different other water quality indications (hardness, alkalinity). Although there are several methods to mobilize metals, cation exchange processes with sea-salt (NaCl) is most important to remobilize metals like Aluminium. Other process for heavy metal remobilization is the formation of aqueous complexes, the formation of chlorocomplexes, and colloid dispersion, (Bauske and Goetz, 1993; Bäckström et al., 2004; Dupuis et al., 2015). Aqueous complexes do not adsorb easily to soil particles when in solution and get transported easily into the lakes (Dupuis, 2015).

#### 1.4.3 Sea-salt episodes

Besides acid rain events, sea-salt episodes can enhance the mobilization of metals and H<sup>+</sup> from the catchment areas (Gagkas et al., 2008). In a changing climate with reduced anthropogenic SO<sub>2</sub> and NO<sub>x</sub> emissions, this process may, in fact, become more important than acid rain.

Sea-salt episodes are widespread along the coastal areas of Norway, and can be measured by chloride concentrations and conductivity in the water (Figure 13) (Bäckström et al., 2004; Wright et al., 1988). The best empirical evidence is the Na : Cl ratio. When the ratio of Na : Cl is below the ratio in seawater cation exchange in the catchment soil is the most probable cause.

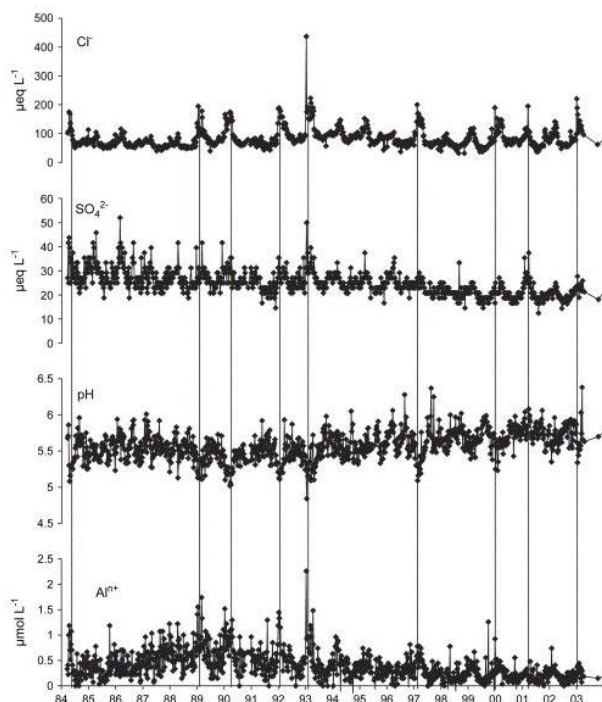


Figure 13. Cl, SO<sub>4</sub>, pH and Al measurements at the west coast of Norway. Sea-salt events are indicated by vertical lines (Gagkas et al., 2008).

The main contributor of Cl in a lake is from sea-salt that is carried with precipitation, or through dry depositions (Garmo et al., 2014). Cl is considered a conservative ion, which is not taken up by plants and does not adsorb to humus and soil particles (Skartveit et al., 1980, Norton et al., 1987, Wright et al., 1988). Hence, Cl flushes into the water body without much retardation. Na, however, is easily adsorbed to soil particles. It is retained in the catchment area for a longer time than Cl. Thus, Cl exchanges Na with other cations, such as H<sup>+</sup> (Skartveit et al., 1980, Norton et al., 1987, Wright et al., 1988) or Al (Wiklander 1975; Norrström and Jacks, 1998) and base cations like potassium (K), magnesium (Mg) and calcium (Ca) (Dupuis, 2015). This process changes the Na:Cl ratio after reaching the lakes.

The water running through the soil in the catchment area will also bring particles and ions to the water body, changing the ionic composition of the lake. In sea-salt events the catchment soil gets less acidic and the runoff becomes more acidic (Wiklander, 1975).

Close to the coast it is natural to have a higher content of sea-salt, which contributes to a higher amount of NaCl, than in inland areas. A storm/ sea-salt episode peaks this concentration, and increases the conductivity measurement due to the addition of salts in the water, but it also causes the ratio of Na:Cl to increase (Gagkas et al., 2008). This is caused by an increased drainage of Na from the catchment soil. As the soil is saturated, cations previously retained by the CEC in the soil are flushed to the lake body, together with the ones coming in by the storm event.

An example of a major storm event in Norway is documented from January 1993. Right after the storm, fish deaths in several freshwater systems in the southwest were observed. It has been found

that acidification and the following Al mobilization were the main causes for the fish death (Hindar et al., 1995). The sudden runoff and introduction of sea-salt was described as the reason for the acidification in the first place. In addition the vegetation type in the catchment was found to play an important role in acidification events after sea-salt episodes. The most dramatic acidification was found in catchments with dense forests (Hindar et al., 1995). In respect to pH and Al, the lakes recovered after 3-4 months. However, the Na : Cl ratios indicated, however, that some effects lasted for >2 years (Hindar et al., 1995).

## 1.5 Other anthropogenic impacts on lake water quality

### 1.5.1 Organic matter/ Brownification

Organic matter is a broad term describing all material of organic origin. This includes dissolved organic matter (DOM) and particulate organic matter (POM)(reviewed by: Schwoerbel et al., 2005). The sum of organic matter can be defined by the total organic carbon (TOC) content. Important organic substances are humic substances, which have complex chemical structures and more labile organic matter which may be used for remineralization processes (reviewed by: Schwoerbel et al., 2005). The current chapter focusses on humic substances or humus, due to its important role in cation exchange processes and buffer capacities in Norwegian lakes and catchment areas. In the current study TOC is used as a proxy for humus in lakes.

Humus can be an important buffer in catchment soils via cation exchange processes. It can bind  $H^+$  and reactive aluminium species, and thus protect the lakes. Another impact of humic substances in lakes is brownification.

Due to recovery of acid rain effects (increasing pH) the solubility of TOC has increased and the lability decreased. This leads to increased colouration of the water, called brownification. The effect has been observed by e.g. Erlandsson et al., (2008); and reviewed by Graneli (2012). An increase in TOC is also thought to be due to decline in  $SO_2$  in acid rain (Evans, 2005; Monteith et al., 2007). The dissolved organic carbon (DOC) (Monteith et al., 2007), acidity and  $SO_4$  content of lakes can suppress the amount of DOC, which means that a decline in these factors will lead to a higher concentration of DOC (Evans, 2006). However, humus in the lakes themselves is known to reduce the buffer capacity, since it is an acid itself (Austnes, 2014).

The buffer capacity is another factor controlling the solubility of TOC. A catchment area that has a high ANC has lower DOC concentrations (Evans, 2006). Therefore, the increases in TOC are thought to be a sign of recovery from previous acidifications in catchment areas where the ANC was low. Iron is another important contributor to colour changes, and allows more colouration than possible with TOC alone (Kritzberg and Ekström, 2012).

Climate change, with increased temperatures (Weyhenmeyer and Karlsson, 2009) precipitation and runoff (Hongve et al., 2004; Erlandsson et al., 2008), or permafrost thawing (Akselsson et al., 2013) is also thought to be a factor that increases TOC in the waters. Near the coast changes in sea-salt episodes and sea-salt intrusion is a factor which may play an additional role. Changes in land use and wood processing can be another factor increasing TOC release (Akselsson et al., 2013).

The Impacts of brownification for the biota are rather negative, due to the absorbance of light. Light is scattered and absorbed by particles of nonorganic and organic matter, and the depth of the euphotic zone is dependent on the colouration of the water (reviewed by Graneli, 2012). Light is an important key factor for primary production and the temperature gradient in the lake. Light gives the energy to drive primary production, and heat the water. Conversely, microbial activities may be enhanced by the import of organic matter. Eventually, phototrophic processes are negatively affected and microbial heterotrophic processes are enhanced, which leads to increased net CO<sub>2</sub> emissions (Tranvik et al., 2009). The CO<sub>2</sub> can then increase acidification, or can be released into the atmosphere and accelerate climate warming. Additional negative effects include the reduced biodiversity and fish stocks (Karlsson et al., 2009).

In recent years, the amount of DOC has increased (reviewed by Graneli, 2012). Looking back at periods in Norway with high acidification the lakes were clearer and were called Clearwater lakes. Now the opposite is observed; a brownification of lakes. A lowered deposition of industrial sulphate is the most likely explanation for the recent brownification. In fact, Scandinavian lakes in pre-industrial times are thought to be brown-water lakes (Cunningham et al., 2011).

### 1.5.2 Eutrophication

Phosphate, ammonium, and nitrate are important inorganic nutrients. They can originate from anthropogenic sources, such as waste and sewage in rural areas, or agricultural activities in the catchment (reviewed by: Callisto et al., 2014). This contributes to important nutrition for algal growth. Phosphorus is usually considered the limiting factor in freshwater lakes and most important component for eutrophication (Schindler 1977; Sharpley et al., 1994). Eutrophication usually yields in an increased algal biomass, which absorbs most light in the surface layer of the lake and has similar effects as brownification via allochthonous TOC input (reviewed by: Callisto et al., 2014), including the implications for ionic compositions.

However, an increase in phosphorous can yield in nitrogen limitations in freshwater lakes. In this case, the only phytoplanktons that are capable of using elemental N<sub>2</sub> as their nitrogen source are cyanobacteria. Since many fast growing cyanobacteria species in freshwater lakes are known to produce cyanotoxin, a surplus of phosphate may lead to toxic algal blooms (Anderson et al., 2002). In addition to a potential toxicity of new dominant algal species, the food web may be altered by altered competition and predator-prey dynamics due to a different dominant players in the food web (reviewed by: Callisto et al., 2014).

Increased primary production also leads to increased microbial respiration, especially below the euphotic zone. This process may quickly consume all oxygen, which is crucial for higher organisms, such as fish (Corell, 1999). In case of complete oxygen consumption in the sediments alternative electron acceptors may be used. Thereby toxic sulphide or reduced heavy metals, or orthophosphate may be released (Chen et al., 2016).

A higher biomass in the euphotic zone can also absorb more sunlight and, thereby, increase the temperature of the surface layer. This will lead to a higher temperature and density difference between the surface layer and deeper layers, which means a more stable stratification (reviewed by: Wetzel, 2001).

In regard to acidification,  $\text{NO}_3$  from acid rain is known as one reason, due to its conversion to  $\text{HNO}_3$ .  $\text{NH}_4$  may have the same effect. Since  $\text{NH}_4$  is largely used for microbial nitrification processes,  $\text{NO}_3$  is the final product, which can become acidic ( $\text{HNO}_3$ ) (reviewed by: Callisto et al., 2014).

However, since the lakes of this study were chosen for monitoring of lake acidification, the lakes were chosen in regions with little anthropogenic import of phosphate and nitrogen (Garmo et al., 2014).

### 1.5.3 Climate change

According to the latest IPCC report (Stocker et al., 2014; Pachauri et al., 2014), the global climate is rapidly changing, mainly due to anthropogenic greenhouse gas emissions. An increasing temperature ( $\sim 2\text{--}4^\circ\text{C}$ ), changing precipitation patterns, increased  $\text{CO}_2$  levels and water acidification, increasing storm events, and northward migration of species, are some of the observed consequences of global warming (Stocker et al., 2014; Pachauri et al., 2014).

Between 1900 and 2008 the annual mean temperature in Norway increased by  $0.9^\circ\text{C}$  (Climate change – Norway; Ministry of the Environment of Norway, 2009). Higher temperatures themselves can already decrease gas solubility (Table 3), and lead to changing forestation, which can be important for brownification (Akselsson et al., 2013).

The changes in Norway will appear as more storms, especially in the East and South (Gregow et al., 2011). Mean geostrophic wind speeds are thought to increase all over Northern Europe by 5 – 10%. Extreme geostrophic wind speeds are predicted to increase in the south and west of Norway and to decrease over the Norwegian Sea (Gregow et al., 2011). Thus, it is hard to predict what the implications for sea-salt episodes will be. Sea-salt episodes are known to increase the ionic strength in catchment areas and to yield in increased cation exchange processes. In already acidic catchments this could cause a sudden flush of  $\text{H}^+$  into the lakes (Skjelkvåle et al., 2007; Laudon, 2008).

Precipitation has been increasing in Norway since 1900. The largest increase (19-22%) has been found in the western and north-western parts of Norway (Ministry of the Environment of Norway, 2009). Increased temperature can enhance nitrification processes and the leaching of nitrate (van Breemen et al., 1998; Wright and Jenkins, 2001). Eventually, the nitrate can end up in the lakes and lead to acidification and eutrophication.

Higher temperatures and precipitation rates will affect weathering processes and production of ions and drainage of ions via increased runoff in the catchment areas. Higher erosion of coastal areas with more frequent sea-salt episodes will increase the sea-salt input into lakes close to the coast.

Increased  $\text{CO}_2$  levels can lead to acidification, but can also increase the carbonate buffer capacity (Stocker et al., 2014; Pachauri et al., 2014). The northward migration of species may lead to increased biomass in form of conifer forests in the North, which would increase the TOC contents and humus loads, which can act as buffer-, or acid. The thawing of permafrost may lead to less surface runoff and an increasing role of groundwater flow. Additionally, thawed permafrost soil may supply additional organic matter (Semiletov et al., 2016).

Increased temperatures will decrease the solubility of gases such as oxygen and  $\text{CO}_2$ , which are crucial for biological processes and fishes, but which may also counteract the acidification by  $\text{CO}_2$ .

## 1.6 Objectives

In the current study I will use a long-term dataset from 1986 to 2014 of the ionic composition in Norwegian lakes, which are rather unaffected by local anthropogenic disturbances, to study the effects of reduced anthropogenic SO<sub>2</sub> and NO<sub>x</sub> emissions in comparison to climate change on different processes that control the ionic composition of the studied lakes. I will investigate whether...

- 1) ...weathering processes are more increasing because of climate change, or decreasing because of a rising pH. Most likely regional differences can be observed.
- 2) ... the anthropogenic impacts of NO<sub>x</sub> and SO<sub>2</sub> emissions on lake acidity, TOC, and Al speciation continue to decrease since the last study.
- 3) ... sea-salt episodes are increasing in frequency or magnitude, because of climate change, and what the implications for the ionic composition in Norwegian lakes are.
- 4) ... the increase in pH can be related to changing buffer systems.
- 5) ... the pH increase and potential climate change and land use change leads to an increased brownification in Norwegian lakes.

Due to the decreased emissions of SO<sub>2</sub> and NO<sub>x</sub> in middle Europe, I expect a large change in ionic composition in Norwegian lakes between 1986 and 2014. I expect climate change to play an important role in delaying the recovery processes expect that lakes that are influenced by sea-salt episodes (West coast), and lakes in catchments with high humus contents and good buffer capacities (e.g. Southeast Norway) will recover less than lakes that were heavily impacted by anthropogenic acid rain deposition (Southern Norway and Eastern Finnmark). Different ionic compositions may be observed due to changing sea-salt events and TOC increase by climate-related processes.

## 2 Methods

## 2.1 Dataset

The dataset used in this thesis is composed of the reference lakes of the Norwegian environment agency in Norway (Garmo et al., 2014). Different cations: Mg, Ca, K, Na, NH<sub>4</sub>, anions: Cl, SO<sub>4</sub>, NO<sub>3</sub>, PO<sub>4</sub>, aluminium species: labile Al (LAL), reactive Al (Al.R), and unreactive Al (Al.iL), organic matter (TOC, TOTN, TOTP), pH, alkalinity and conductivity were measured as described by Garmo et al. (2014). In addition, ANC, ANCO<sub>aa</sub> (ANC calculated with OA), and different sea-salt corrected ion concentrations were calculated as described later. The measurements were done by the Norwegian institute for water research (NIVA) from 1986 to 2014. The lakes were sampled every year after the autumn circulation in order to measure representative values for the whole lake.

The dataset is a part of a larger monitoring program by the Norwegian environment agency (MD). The program is called *Overvåking av langtransportert forurenset luft og nedbør* (Monitoring of long-range transported acidified air and precipitation) and started already in 1980. Precursor of this monitoring program was the *Sur nedbørs virkning på skog og fisk* (Sour precipitation effects on forests and fish) - project. The MD administrates different departments that each are experts in their field, and responsible for monitoring different areas of expertise affected by long distant air carried and precipitated pollutants. Each department is responsible for different smaller categories in this program Norwegian institute for air research, (NILU) is monitoring atmospheric deposition, NIVA and the Norsk instiutt for naturorskning (NINA) are monitoring water chemistry. In addition, the Laboratorium for ferskvannøkologi og innlandsfiske (LFI) in Bergen monitors biological factors of 84 Norwegian lakes and two rivers. In addition, water chemistry is monitored at 6 field research stations. From starting with 1000 lakes, the dataset is now reduced. The initial dataset contains 87 lakes and 2 rivers, and 6 research stations but due to missing data only 77 lakes are analyzed in the current study. The chemical parameters and metadata are shown in the appendix (Appendix)

The 77 lakes from this dataset is a part of the “100-lake” - research, to see changes in acidification. The number of lakes is reduced from 100 originally to 77 due to alkalisation with adding lime in some of the lakes to sustain life and biota. Each year a set of five reports are made under this project.

All lakes in the dataset are located above the marine limit, and are chosen for the reason that they are not directly influenced by any local human activities, such as farming. Hence the nitrogen and phosphorous concentrations are low (Garmo, 2014). The global human disturbances, such as atmospheric deposition of different ions are however present. Due to the low ionic strength in these lakes, even small changes in acidity can be detected (Henriksen et al., 1989).

A table of the median values of each lake is shown in the Appendix (Table 12). The working can be found in the latest MD report (Garmo, 2014). Also the classification of lakes are shown (Figure 15), together with ionic budget (Table 13)

### 2.1.1 Ionic budget

As a quality control the ionic budget was calculated as described by Garmo et al.,(2014). The surplus of anions or cations can be expressed as the ionic budget, or ionic charge. The relation between cations and anions are approximately in a ratio 1:1 in lakes. The NVE used the

calculations of the ionic budget to control the quality of the measured ion concentrations by the MD. In general a balance between anions and cations is expected.

The concentration of main cations and anions in the dataset is measured with a Dionex DX 320 duo ICP-MS, the concentration of bicarbonate is calculated by titrating the solution to pH 4.5.

The main cations used for calculating ionic budget are Ca, Mg, Na, K, SO<sub>4</sub>, CL, NO<sub>3</sub> and HCO<sub>3</sub>. The primary data are given in mg l<sup>-1</sup>. Thus, the measured main cations units have to be converted from mg l<sup>-1</sup> to mmol l<sup>-1</sup> or meq l<sup>-1</sup> in order to account for different atomic weights and charges (Equation 13).

**Equation 12: Calculation of equivalent values of ions**

$$\frac{\text{Concentration of the ions}}{\text{molecular weight of the ion}} \cdot \text{valency}$$

The ionic balance/ionic charge is calculated in two ways depending on what measured parameters, and how much TOC and LAL there is in the lake measurements. Prior to calculating ionic balances, it is crucial to analyse all main chemical parameters. The parameter concentrations are in  $\mu\text{ekv L}^{-1}$ . The difference in ionic charge is calculated by the sum of anions minus the sum of cations (Equation 13).

**Equation 13: Calculations of ionic charge**

$$\sum_c ([Ca^{2+}] + [Mg^{2+}] + [Na^+] + [K^+] + [H^+]) - \sum_a ([Cl^-] + [NO_3^-] + [SO_4^{2-}] + [ALK])$$

$\sum_c$ : sum cations

$\sum_a$ : sum anions

If the lake contains labile aluminium, (LAL), ammonium (NH<sub>4</sub><sup>+</sup>), and total organic carbon (TOC), these are taken into the calculation; The sum of cations include LAL and NH<sub>4</sub><sup>+</sup>, the sum of anion include organic anions (OA<sup>-</sup>).

The OA<sup>-</sup> is calculated using an empirical equation (Equation 14) with TOC concentrations (MD, 2014).

**Equation 14: Empirical equation for calculating the content of OA<sup>-</sup>**

$$OA^- = 4.7 - 6.87 * \exp^{-0.322 * TOC} * TOC$$

For quality control, measurements with ionic budgets > 10 %, or > 10  $\mu\text{ekv l}^{-1}$ , are checked and possibly reanalysed (MD, 2014).



### 2.1.2 Non-sea-salt ions

Even though some Cl can come from the soil deposition or bedrock, this is ruled out by choosing lakes with low contribution from these sources for the dataset. Therefore the chloride measured from the output is assumed to be the same as the chloride input from sea-salt. In addition, it is assumed that the ratio between chloride and the other ions in seawater has the same ratio as Cl and other ions in the lake (Table 5). That way, measuring the content of chloride and subtracting the chloride:cation ratio in seawater, non-marine base cations can be calculated (Equation 15).

**Table 5: See water ratios between Cl and each base cation.**

<b>Cation</b>	<b>Ratio per chloride (r)</b>
Ca <sup>2+</sup>	0.037
Mg <sup>2+</sup>	0.196
Na <sup>+</sup>	0.859
K <sup>+</sup>	0.018
SO <sub>4</sub> <sup>2-</sup>	0.103

**Equation 15: Calculation of non-sea-salt cations**

$$Cat' = Cat - r * Cl$$

*Cat'*: Non – marine cation

*Cat*: Cation measured

*r*: Sea water to cation ratio

*Cl*: Chloride measured

## 2.2 Clustering of the lakes

For the final evaluation of the lakes, all lakes were clustered after a) regions and b) lake type. The clusters are shown in the appendix (Table 11).

Ten regions are previously chosen after biogeographical, meteorological and chemical (extend of acidification) characteristics by Garmo et al.,(2014). The ten regions are located in I) Østlandet - North. II) Østlandet - South. III) Mountain regions – South in Norway. IV) Sørlandet - East. V) Sørlandet - West. VI) Vestlandet -South. VII) Vestlandet - North. VIII) Midt-Norge - Middel IX) Nord-Norge - North. and X) Øst-Eastern Finnmark (Figure 14), with number of lakes in each region (Table 6).



Figure 14: Regional division of Norway based on extent of acidification, meteorology and biogeography. The numbers refers to biological monitoring locations in 1999. (Garmo et al., 2014)

Table 6: Regions names and number for each of the 10 regions (Garmo et al., 2014).

Region-nr.	Region	"100-sjøer"
I	Østlandet – Nord	1
II	Østlandet – Sør	15
III	Høgfjellet i Sør-Norge	3
IV	Sørlandet – Øst	14
V	Sørlandet – Vest	11
VI	Vestlandet – Sør	3
VII	Vestlandet – Nord	5
VIII	Midt-Norge	10
IX	Nord-Norge	5
X	Øst-Finnmark	11
Total		78

The three lake types were clustered after their ionic composition (median values for all years) as described by Haaland et al.,(2009). Lakes with the highest buffer capacity were characterized by their ANC values. Lakes with an ANC value  $> 5 \mu\text{eq}$  were called ANC lakes (Haaland, 2009). They are characterized by a surplus of cations, and bicarbonate which have buffering capacities. The remaining lakes were clustered into Cl lakes (Cl:SO<sub>4</sub><sup>2-</sup> ratio  $> 2$ ) and SO<sub>4</sub><sup>2-</sup> lakes (Cl: SO<sub>4</sub><sup>2-</sup> ratio  $\leq 2$ ) (Haaland, 2009). Cl lakes are influenced by sea-salt events and have a high ionic strength, while SO<sub>4</sub><sup>2-</sup> lakes are influenced by acidic precipitation connected to industrial activities.

## 2.3 Statistical analyses

All statistical analyses and data visualizations were done using R v.3.1.0 (R Core team, 2014).

General relationships between parameters were tested via scatter plots and via linear regression models of log-transformed data, using the function “lm”. Prior to the linear model the distribution of the parameters was evaluated via histograms. After the test Quantile-Quantile and residuals vs. fitted plots were used to evaluate whether a linear model was appropriate. In addition, Mann-Kendall tests were performed to test for monotonous relations between different parameters and the tau values are presented in heat maps.

The parameters of each cluster of lakes were visualized in boxplots showing the median, quantiles and outliers. The non-parametric Kruskal Wallis test was applied to test whether the differences between the clusters are significant. The tests were done for the first year of measurements (1986) and the last year in the dataset (2014) and the different years were compared for general trends.

For testing the magnitude of different trends over time the Theill-sens slope, with its 95% confidence intervals, was calculated using the “sens.slope” function with 95% confidence intervals of the package “trend” (Pohlert, 2016). The statistical significance (p values) and the rank correlations coefficient (tau) for monotonous changes over time were calculated via the Mann-Kendall test using the “kendall” function of the package “Kendall” (McLeod, 2011). For the evaluation of time trends, the data were normalized as the proportional change from the first year of measurements (% change since 1986). Only lakes or cluster of lakes with more than 10 years of measurements were included in the analysis.

For testing general trends of the different clusters, time trend analyses were done on median values of the parameters in each cluster. For testing whether the trends are significantly different between the clusters, the sens slope was calculated separately for each lake. Kruskal Wallis tests were then applied to test whether the differences between the clusters are significantly different.

## 2.4 GIS visualizations

All geographical information system (GIS) visualizations were done using the “sp” package (Roger et al., 2013) in R v.3.1.0 (R Core team, 2014). Different median parameters and elevations in the different lakes are displayed in colour gradients significant time trends (sen’s slope), lake types, and regions are indicated as different colours.

# 3 Results

## 3.1 Lake geography and types

As shown in Figure 15B, the ten regions are characterized by different environmental and geographical features.

Lakes in regions one to three are often quite far away from the coast in the Southeast of Norway. Regions four and five are mainly at the south coast and partly in the mountains (Figure 1). Regions six and seven are at the west coast. Regions eight and nine are in the north-west. Region ten is located in Eastern Finnmark, close to the Russian border.

The lake types (2.2) are shown in Figure 15A. All the Chloride lakes (n=13) are located at the south and west coast. Most Sulphate lakes (n=12) are in the south, a bit further inland, with one exception in Region ten. Six of the Sulphate lakes are located in the mountains (elevation  $\geq 500$  m.a.s.l.). Most lakes (n=54) are ANC lakes (defined after median values for all years) and distributed all over Norway.

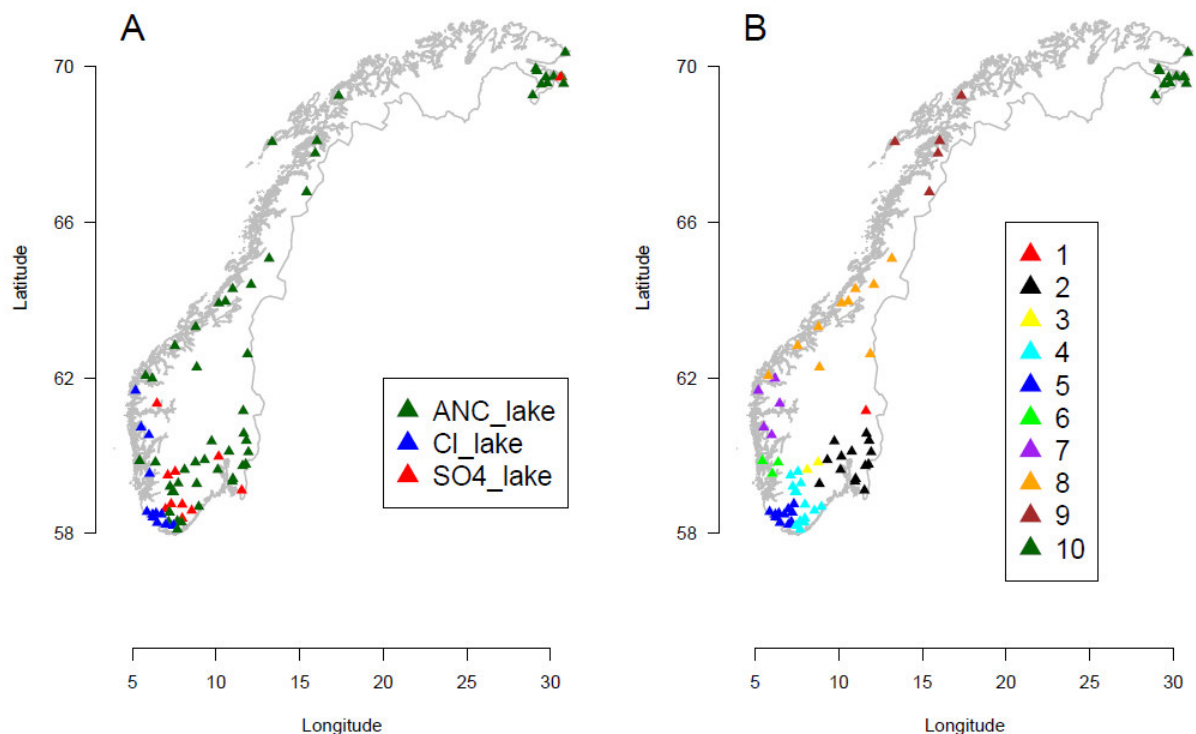


Figure 15: A) location of the different lake types using the median values of all years for its calculation (ANC lake, Cl lake and SO4 lake) and B) location of the lakes in the different regions.

### 3.2 Regression analysis

Generalized correlation tests on the median values of the lakes were done via Mann-Kendall correlations. The correlations of all ions, including  $H^+$  and alkalinity are shown in Figure 16A. The correlations between; pH, Cl,  $SO_4$ ,  $NO_3$ , ANC, and the Al-species L.R, and AL.II are shown in Figure 16B. The strength of the correlation (tau) is displayed in **Error! Reference source not found.**; Red colours indicate positive correlations, and blue colours indicate negative correlations.

Lakes with high  $H^+$  concentrations have mostly higher sulphate and nitrate concentrations, and lower alkalinity, Mg, and K concentrations. The ions with highest correlations are, from highest to lowest; Na and Cl > Na and Mg = Cl and Mg. All other ions are positively correlated with  $NO_3$  followed by Ca having the weakest correlations to other ions. Sulphate is less correlated to the sea-salt ions Na and Cl than to the other ions.

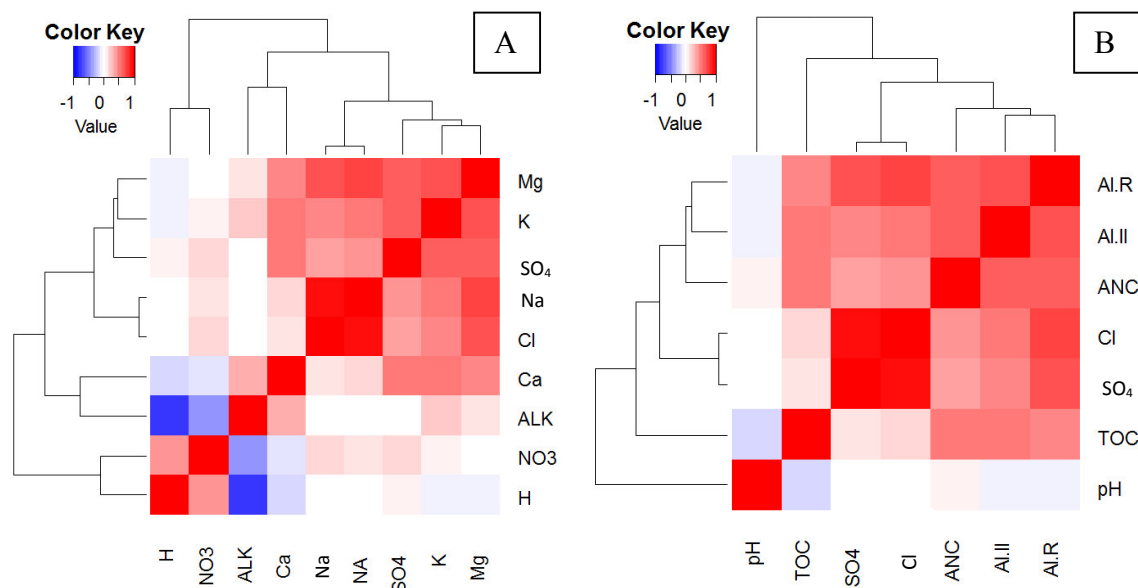


Figure 16: Heat map of the correlation coefficients between different parameters (tau value in the Mann-Kendall test).

Reactive (Al.R) and non-labile (Al.II) Al species are positively related to each other, to ANC, SO<sub>4</sub>, and Cl, but negatively to pH. Reactive Al seems to be more correlated to SO<sub>4</sub> and Cl, while Al.II is more correlated to TOC. TOC is negatively correlated to pH, nearly unrelated to Cl and SO<sub>4</sub> and positively to ANC, and Al.

For a closer detail on the correlations and for testing for linearity of the relationships, the most important parameters for acidification are shown as scatter plots in Figure 17.

General linear regression models were applied, but the p-values should be considered carefully due to the lack of independence in a time series. However, the  $r^2$  value can be used as an additional indication for possible linear relationships. The focus of interpretations is on the visual interpretation of the scatter plots.

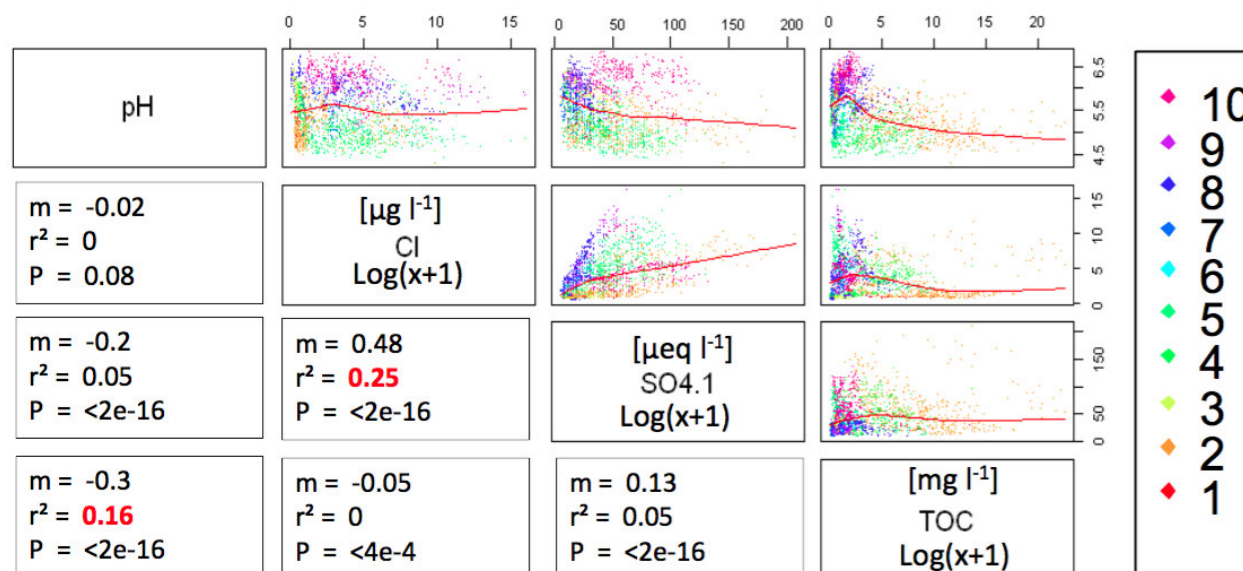


Figure 17: Scatter plots showing the relations between pH, Cl, non-sea-salt SO<sub>4</sub> and TOC. The red line indicates a moving average; the colours represent the different regions as shown in the legend. A linear model was applied to test for the strength (slope =  $m$ ) and linearity ( $r^2$ ) of potential relationships. The  $p$  value is a rough estimate due to non-independent samples (time series). The necessary transformations are shown below the parameter, if it was necessary to give the data a normal distribution. SO<sub>4</sub> is given in equivalents due its calculation, which has to be done with equivalent units.

The relationship between pH, Cl, SO<sub>4</sub>, and TOC are shown in Figure. It appears pH is not directly related to any of the parameters. Cl and SO<sub>4</sub> are correlated, but the increase of Cl per increase of SO<sub>4</sub> is different in different regions (differences in natural sea-salt fluctuations vs. acid rain decline). The Cl to SO<sub>4</sub> ratio in the west and North is generally higher than in the south, with eastern Finnmark as an exception with relatively low Cl to SO<sub>4</sub> ratios. For TOC it appears that higher TOC values are possible, if the anion concentrations are low.

The relationship between ANC as proxy for the buffer capacity, TOC as indication for organic matter input and brownification, and the labile aluminium species and the aluminium species bound to TOC are shown in Figure 18.

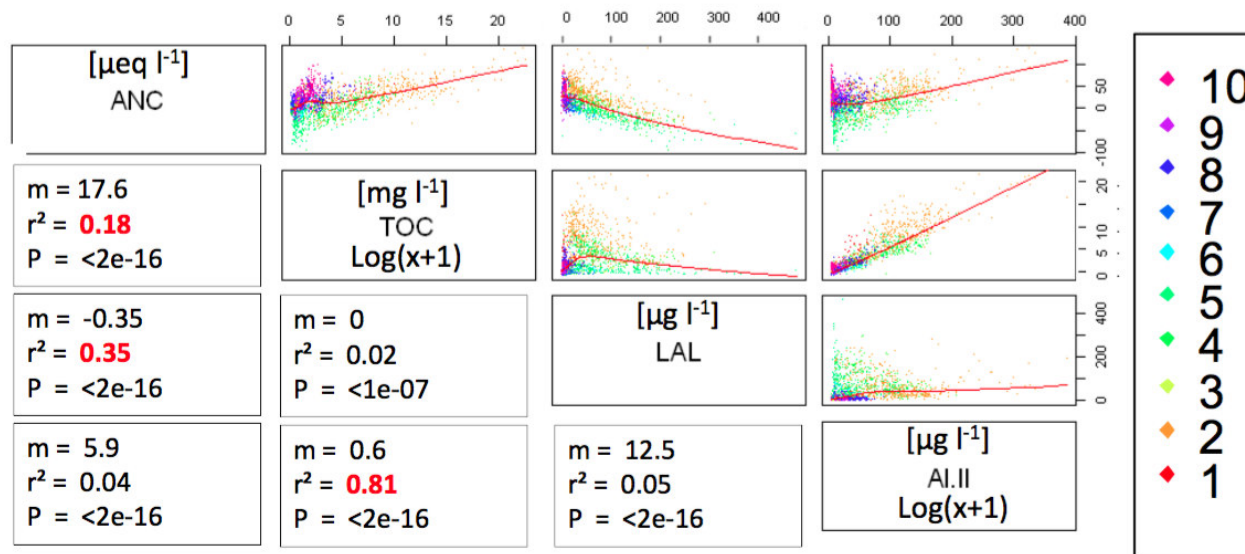


Figure 18: Scatter plots showing the relations between ANC, TOC, LAL, and Al.II. The red line indicates a moving average; the colours represent the different regions as shown in the legend. A linear model was applied to test for the strength (slope =  $m$ ) and linearity ( $r^2$ ) of potential relationships. The  $p$  value is a rough estimate due to non-independent samples (time series). The necessary transformations are shown below the parameter, if it was necessary to give the data a normal distribution.  $\text{ANC}_{\text{COAA}}$  is not shown since ANC and  $\text{ANC}_{\text{COAA}}$  ovary per definition.

ANC has a positive relationship with TOC and a negative relationship with LAL. Bound aluminium (Al.II) does not seem to be affected directly by the buffer capacity. TOC and Al.II have a strongly positive relation. High LAL concentrations are only possible if the TOC concentrations and fraction of bound aluminium is low. For region two a negative relation between TOC and Al.II and between Al.II and LAL appears.

The relationships between; pH and LAL, pH and Latitude, and elevation and conductivity are shown in Figure 19. The pH is mostly above 5.5 if the LAL concentrations are below  $25 \mu\text{g l}^{-1}$  and below 5.5 if the LAL concentrations are higher. An opposing pattern is observed for the relation with latitude. Below  $62^\circ\text{N}$  the pH has a range between 4.5 and 6 with a slight increase towards the North. Further north the pH stays mostly above 6. The conductivity generally decreases with elevation.

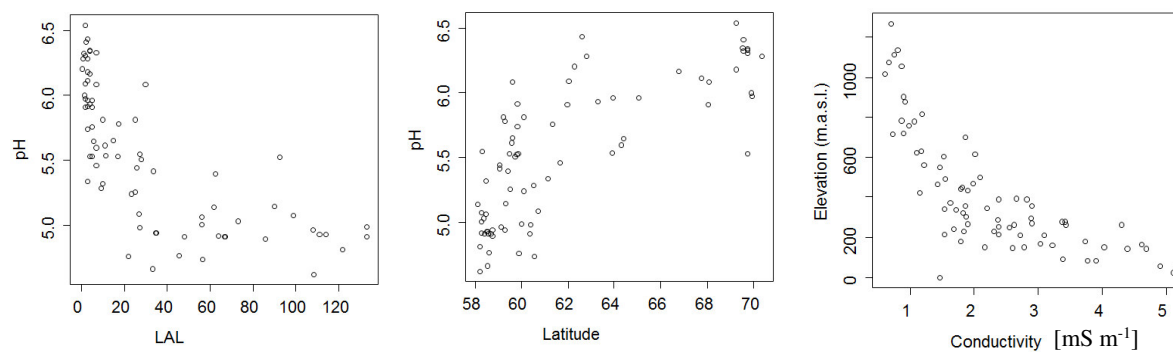


Figure 19: Scatter plots, showing the relationships between A) pH and LAL  $[\mu\text{g l}^{-1}]$ , B) pH and latitude ( $^\circ\text{N}$ ), and C) elevation and conductivity.

### 3.3 Chemical measurements in the different lakes, regions and lake types

#### 3.3.1 Differences between the regions, and lake types

The measurements of all parameters are summarized in Table 7 and Table 8, by their median values of all years in the ten regions and the three lake types.

**Table 7** Median values for all data between 1986 and 2014 for different chemical parameters for the different lake types and regions. The yellow and red bars indicate the relative magnitude of the parameters compared to the other regions and lake types.

lake type	COND mS m <sup>-1</sup>	ALK mmol l <sup>-1</sup>	TOTP µg l <sup>-1</sup> P	PO4.P µg l <sup>-1</sup> P	TOTN µg l <sup>-1</sup> N	NH4 µg l <sup>-1</sup> N	TOC mg l <sup>-1</sup> C
ANC lake	1.95	0.04	4	1	151	8	2.365
Cl lake	3.10	0.03	3	1	311	23	0.9
SO4 lake	1.80	0.03	3	1	253	8	3.1
Region 1	1.21	0.04	8	2	240	5	6.6
Region 2	1.94	0.04	6	1	320	14	8.7
Region 3	0.82	0.05	2	1	142	4	1.0
Region 4	2.03	0.03	4	1	280	19	3.0
Region 5	3.14	0.02	4	1	345	22	1.4
Region 6	2.34	0.03	2	1	198	9	1.7
Region 7	1.20	0.03	2	1	132	4	0.6
Region 8	1.98	0.04	2	1	115	10	1.8
Region 9	1.84	0.05	2	1	95	6	1.0
Region 10	2.61	0.05	3	1	95	4	1.4
lake type	AlR µg l <sup>-1</sup>	AlI µg l <sup>-1</sup>	LAL µg l <sup>-1</sup>	ANC µeq. l <sup>-1</sup>	ANC <sub>OAA</sub> µeq. l <sup>-1</sup>	ionic budget µeq. l <sup>-1</sup>	
ANC lake	45	28	15	23.5	11	71	
Cl lake	90	13	62	-15	-20	11	
SO4 lake	122	40	69	-3	-11	30	
Region 1	47	41	3	37	13.5	85	
Region 2	167	106	48	34	2	80	
Region 3	46.5	15.5	19	21	16.5	74	
Region 4	125	44	58	2	-8	40	
Region 5	137	23	63.5	-17	-24.5	10	
Region 6	67	34.5	22	5.5	0.5	46	
Region 7	24	10	7	-1	-2	39	
Region 8	25	20	4	20	14	65	
Region 9	18	14	4	23	19	71	
Region 10	10	9	3	30	24	98	



Table 8 Median values for all data between 1986 and 2014 for different chemical parameters for the different lake types and regions. In mass per volume, as well as equivalents per volume for calculations of the sea-salt corrected cations and direct comparisons (\*) the yellow and red bars indicate the relative magnitude of the parameters compared to the other regions and lake types.

lake type	Ca mg l <sup>-1</sup>	Mg mg l <sup>-1</sup>	K mg l <sup>-1</sup>	Na mg l <sup>-1</sup>	SO <sub>4</sub> mg l <sup>-1</sup>	NO <sub>3</sub> -N μg l <sup>-1</sup> N	Cl mg l <sup>-1</sup>	pH
ANC lake	0.74	0.33	0.19	1.78	1.80	26.00	2.82	5.80
Cl lake	0.42	0.37	0.17	2.93	2.20	195.00	5.18	5.04
SO4 lake	0.37	0.15	0.11	1.02	1.60	70.00	1.60	5.10
Region 1	0.98	0.15	0.17	0.56	1.70	5.50	0.50	5.34
Region 2	1.05	0.32	0.22	1.06	2.69	47.50	1.20	5.11
Region 3	0.74	0.11	0.11	0.41	1.20	53.50	0.41	6.05
Region 4	0.50	0.26	0.13	1.55	1.89	87.00	2.43	5.21
Region 5	0.43	0.36	0.17	2.90	2.38	195.00	5.07	4.90
Region 6	0.58	0.31	0.14	2.31	1.53	94.50	3.95	5.39
Region 7	0.24	0.17	0.09	1.26	0.81	75.00	2.04	5.50
Region 8	0.53	0.32	0.17	2.36	1.20	23.00	3.86	5.97
Region 9	0.52	0.27	0.28	2.11	1.13	16.00	3.50	6.10
Region 10	1.13	0.51	0.21	2.22	3.00	4.00	3.32	6.30

lake type	Ca μeq. l <sup>-1</sup>	Mg μeq. l <sup>-1</sup>	K μeq. l <sup>-1</sup>	Na μeq. l <sup>-1</sup>	SO <sub>4</sub> μeq. l <sup>-1</sup>	NO <sub>3</sub> -N μeq. l <sup>-1</sup> N	Cl μeq. l <sup>-1</sup>	H+
ANC lake	37	27	5	77	80	37	2	2
Cl lake	21	30	4	127	146	46	14	9
SO4 lake	18	12	3	44	45	33	5	8
Region 1	48.5	12	4	24.5	14	35	0	5
Region 2	52	26	6	46	34	56	3	8
Region 3	36.5	9	3	18	11.5	25	4	1
Region 4	25	21	3	67	69	39	6	6
Region 5	21	30	4	126	143	49.5	14	13
Region 6	28.5	26	4	100	111.5	31.5	7	4
Region 7	12	14	2	54.5	57.5	17	5	3
Region 8	26	26	4	102.5	109.5	25	2	1
Region 9	26	22	7	92	99	24	1	1
Region 10	56	42	5	97	94	62	0	1

lake type	Ca* μeq. l <sup>-1</sup>	Mg* μeq. l <sup>-1</sup>	K* μeq. l <sup>-1</sup>	Na* μeq. l <sup>-1</sup>	SO <sub>4</sub> * μeq. l <sup>-1</sup>
ANC lake	33	9	9	3	28
Cl lake	16	2	3.5	1	27.5
SO4 lake	17	5	5	2	28
Region 1	48	9	12	4	34
Region 2	50	16	16	5	48
Region 3	36.5	6	6	3	24
Region 4	23	5	6	2	31
Region 5	16	3	6	2	33
Region 6	24.5	4	4	2	20
Region 7	11	2	3	1	12
Region 8	20.5	4	6	2	11
Region 9	20	3	9	2	12
Region 10	53	22	9	3	50

ANC lakes have generally higher conductivities and lower nitrogen levels than Cl and SO<sub>4</sub> lakes. Al-concentrations are general lower and most is bound as Al.II. The pH is magnificently (about 0.7) higher and most ions did not originate from sea salt.

Cl lakes have the highest conductivity (from sea salts) and the highest nitrogen concentrations. The TOC concentrations and ANC values are lowest. Ion concentrations are highest in this lake type, but the non-sea salt ions have the lowest concentrations.

SO<sub>4</sub> lakes are more comparable to ANC lakes than to Cl lakes, and appear to have concentrations between these two lake types. Different are the exceptionally high concentrations of aluminium in all forms.

pH and alkalinity are generally increasing from region one to ten with an exceptionally high median in region three which includes two lakes above 1000 m.a.s.l.. Regions one, two, four, and five are fairly similar in pH and alkalinity. The conductivity is highest within regions five and ten, with a gradient of decreasing conductivity towards the neighbouring regions. Phosphorous compounds are similar in all regions with an exception in region one, which however, only contains one lake. Nitrogen concentrations are highest in regions four and five and relatively high in region two. TOC concentrations are highest in region two.

As for nitrogen, Al concentrations are highest in region two, four and five. However, only in region two, the concentration of bound aluminium (Al.II) is considerably higher than in the other regions. This indicates that most Al in this region is bound, while most aluminium in region four and five are labile and reactive.

The ANC and ANC<sub>OAA</sub> values are highest in region one and ten with a gradient towards region five with the lowest values. Region two and ten appear to have the highest ion concentrations, which are supported by the conductivity. Region five is another region with high concentrations of certain ions. SO<sub>4</sub> is enriched in region one, two and ten (>30% of the anions; acid rain). In region two, Ca and K reach higher concentrations than in the other regions (K ~4% of cations, Ca 30-50% of cations). Most ions in region two (~80%) and partly in region ten (~50%) are not related to sea-salt. In region five, Na (64% of cations), Cl (63% of anions) and NO<sub>3</sub> (6% of anions) are enriched and most ions are related to sea-salt (76%). Region ten is most similar to region two, but more ions originate from sea-salt (30% more). Enriched ions in region ten are Ca (25% of cations), Mg (25% of cations), and K (2.5% of cations), whereby Ca and Mg come mainly from non-sea-salt sources, while K mainly comes from sea-salt.

### 3.3.2 Geographical differences between the lakes

The location and magnitude of the chemical parameters of interest are shown in Figure 20. The highest buffer capacities are found in Eastern Finnmark (Region ten) and in the southeast furthest away from the coastline. In addition, one lake near Trondheim (Lundalsvatnet) has been found to have a high ANC value. The lowest ANC values are found in region five and partly in region four in the southwest of Norway.

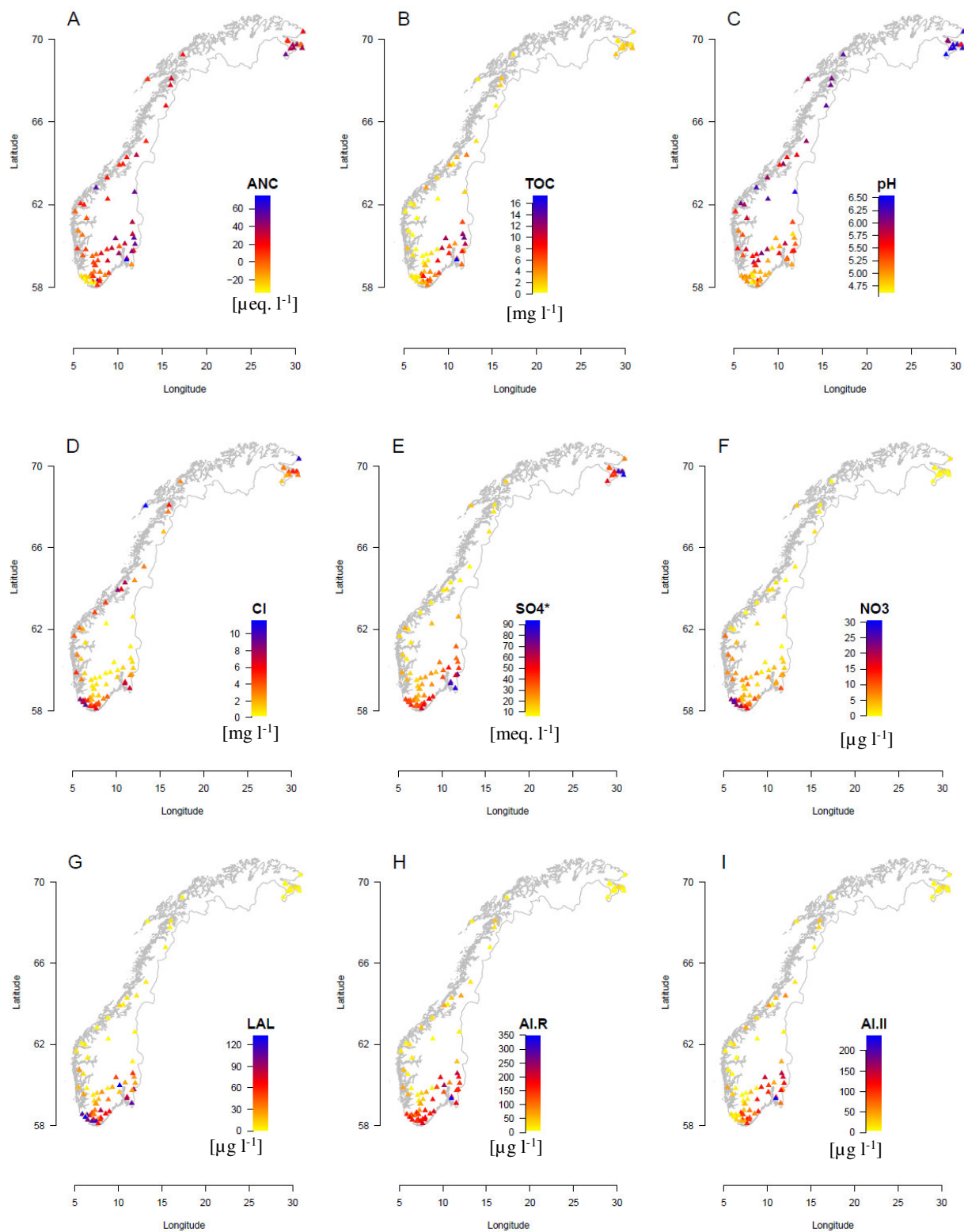


Figure 20: Magnitude of A) ANC [μeq l<sup>-1</sup>], B) TOC [mg l<sup>-1</sup>], C) pH, D) Cl [mg l<sup>-1</sup>], E) non sea-salt SO<sub>4</sub> [meq l<sup>-1</sup>], F) NO<sub>3</sub> [μg l<sup>-1</sup>], G) LAL [μg l<sup>-1</sup>], H) ALR [μg l<sup>-1</sup>], and I) Al.II [μg l<sup>-1</sup>], for each lake and its geographic location.

The highest values of TOC have been measured in the southeast (regions one and two), furthest away from the coast (median = 8.3 mg l<sup>-1</sup>). (The remaining lakes have TOC values below 8.4 mg l<sup>-1</sup> (median = 1.4 mg l<sup>-1</sup>). The pH values show a distinct gradient from low values in the South (approx. 5) to higher values (approx. 6.5) in the North. One exception for an acidic lake (pH = 5.5) in Eastern Finnmark, is Lille Djupvatnet in Sør-Varanger.

Chloride concentrations are highest close to the sea, and lowest in the inland at elevations below 500 m. The highest concentrations have been measured in the Lofoten in Storvatn (11 mg l<sup>-1</sup>). Non-sea SO<sub>4</sub> is highest in the far south (Region five) and far north (Region ten). These regions are closest to the border to countries which are known as sources for sulphur-dioxide pollution, like middle-Europe and Russia. Additionally two lakes near Oslo are found to have exceptionally high non-sea-salt sulphate concentrations (Holvatn and Ravnsjoen, SO<sub>4</sub>\* > 82 µeq l<sup>-1</sup>). The same lakes also have the highest nitrate concentrations.

Different aluminium species behave similarly as shown in Figure 20G-I. Generally, the highest Al concentrations have been measured in the South. However, in region five, around Kristiansand, more LAL have been found than anywhere else in Norway. In fact, most Al in the southwest is not bound as Al.II (II:R ratio approx. 0.05), while more Al in the southeast is bound as Al.II (II:R ratio approx. 0.9)

### 3.4 Time trends

#### 3.4.1 Differences between 1990 and 2014

The first year of the time series (1986) and the last year of measurements (2014) were compared in order to evaluate potential recovery of the lakes.

As shown in Table 9, in 1986 most lakes (53) were sulphate lakes with a Cl : SO<sub>4</sub> ratio below 2 and ANC values ≤ 5. In 2014 no sulphate lakes, after this definition, is left. Instead, ANC lakes increased in number from 21 lakes in 1986 to 64 lakes in 2014. Chloride lakes stayed approximately the same over the study period.

**Table 9: Number of different lake types in 1990 and 2014.**

Lake type	# of lakes in 1986	# of lakes in 2014
ANC lake (ANC >5 ueq)	21	64
Cl lake (Cl / SO <sub>4</sub> > 2)	4	12
SO <sub>4</sub> lake (Cl / SO <sub>4</sub> ≤ 2)	53	0

The supposedly most important parameters for acidification and Al remobilization are displayed as boxplots for 1986 and 2014 (Figure 21, Figure 22). Significantly different lake types (Kruskall Wallis test, p<sub>cor</sub> < 0.05) are marked in red.

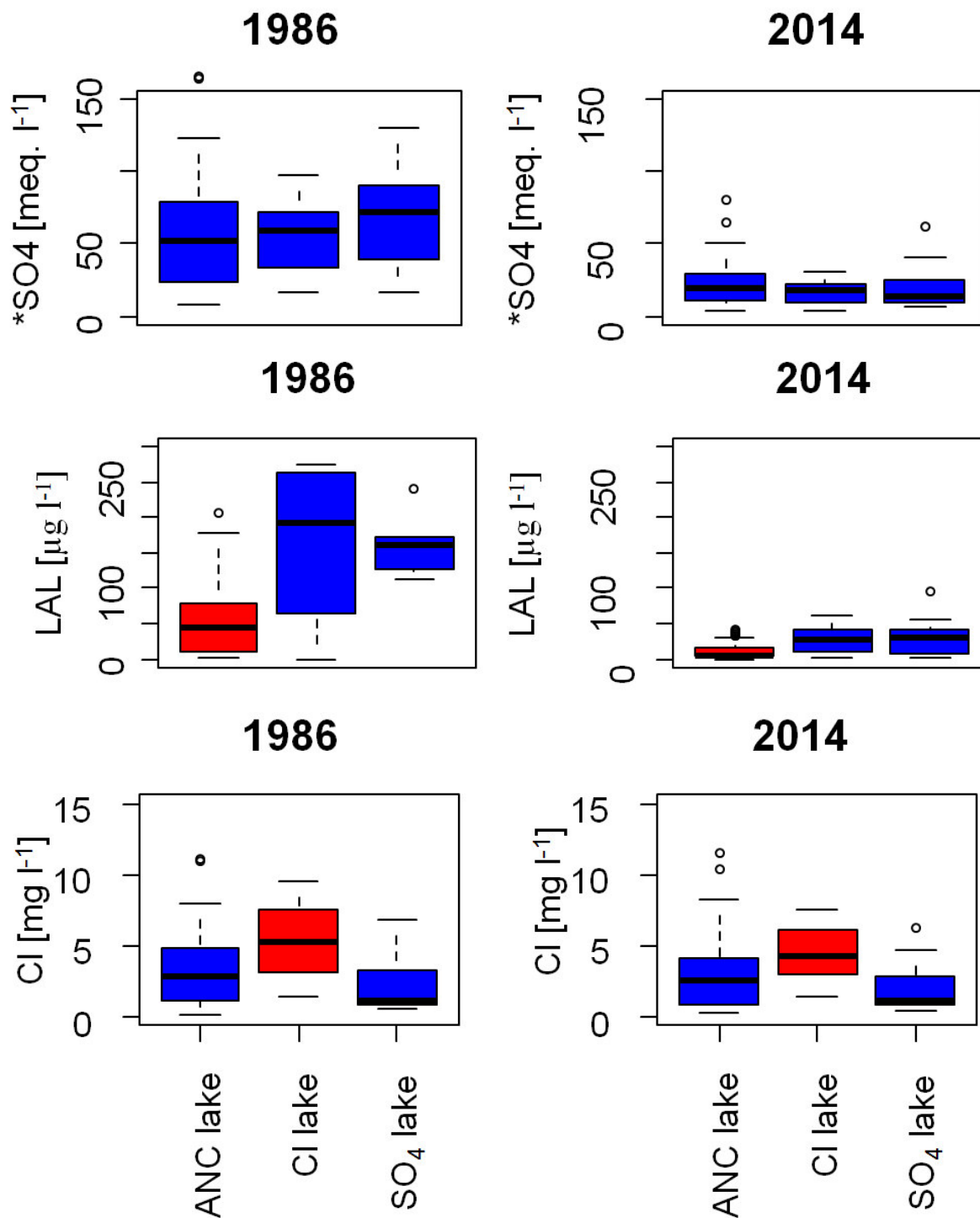


Figure 21 Boxplots, showing the median, quantiles, interquantiles and outlier values of non-sea-salt SO<sub>4</sub> [meq l<sup>-1</sup>], Cl [mg l<sup>-1</sup>] in the different lake types in 1986 and 2014. Significant different lake types are shown in red (Kruskal Wallis test,  $p < 0.05$ )

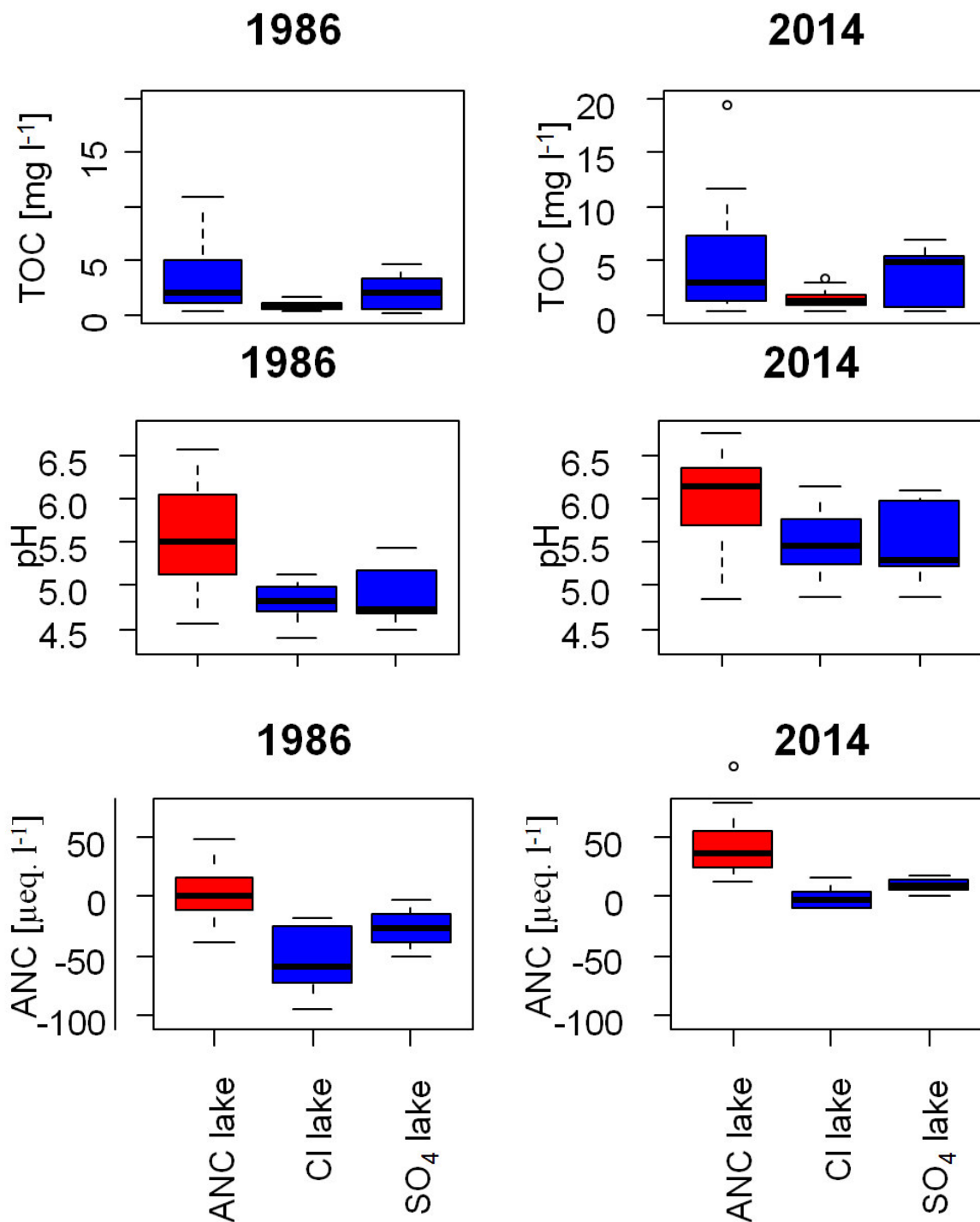


Figure 22: Boxplots, showing the median, quartiles, interquartile and outlier values of TOC [mg l<sup>-1</sup>], pH, and ANC [µeq l<sup>-1</sup>] in the different lake types in 1986 and 2014. Significant different lake types are shown in red (Kruskall Wallis test,  $p < 0.05$ )

Generally, pH and TOC values increased from 1986 to 2014, while Cl, non-sea-salt SO<sub>4</sub>, LAL decreased. The general Cl concentration stayed rather constant. Total SO<sub>4</sub> concentrations are similar in all lake types and decreased by a factor of approx. 1.5. LAL concentrations decreased in all lake types by a factor of approx. 2. The pH increased by approx. 0.25 in Cl lakes, but not much in ANC lakes, which already had a high pH in 1986.

### 3.4.2 Mann-Kendall test and Theill-Sens slope

For estimating the trends over the whole time period, Mann-Kendall statistics and the sens slope of the proportional change was calculated for each lake and the different lake types.

As shown in Figure 23 and Figure 24, most lakes did change significantly in ANC, TOC, pH, Cl, non-sea-salt SO<sub>4</sub>, NO<sub>3</sub>, LAL, Al.R, and All values. Most changes did usually occur in the South. ANC, TOC and pH are generally increasing, while non-sea-salt SO<sub>4</sub>, Al.R and LAL are decreasing and NO<sub>3</sub>, Cl and Al.II do not show any significant trends.

ANC and ANC<sub>OAA</sub> are significantly increasing all over Norway. Four exceptions with non-significant ANC increase are Stigebotvatn, Songevatn, Grovliatnet and Grytsjøen. TOC increases in most lakes, especially in the South. Only in Oddmundalsvatn and Skjerivatnet TOC values decrease significantly. In Eastern Finnmark, only L. Djupvatnet and Langvatnet showed a significant increase in TOC. Similar patterns are apparent in pH with significant increases all over Norway. Cl is mainly decreasing in the very South and in the inland of Southern Norway and one lake in the middle of Norway. Non-sea-salt SO<sub>4</sub> is decreasing in all lakes, except for Svartdalsvatnet and Stigebotvatn. Nitrate is decreasing in most Norwegian lakes.

The reactive aluminium species LAL- and Al.R lakes are decreasing in most Norwegian lakes, but mainly in the South. Inorganic bound Al (Al.II) is mainly increasing in the South, but decreasing in the far North and Southwest. One exception is Langvatnet in region ten with a significant increase. Non-sea-salt Ca and Mg are significantly decreasing in the Southeast and significantly increasing in the West. Ca shows most significant changes. Non-sea-salt Na is increasing in 24 lakes all over Norway, mainly further inland. Non-sea-salt K is decreasing in the Southeast and in a few lakes in the middle and far North of Norway. Four lakes further inland show an increase in non-sea-salt

K.

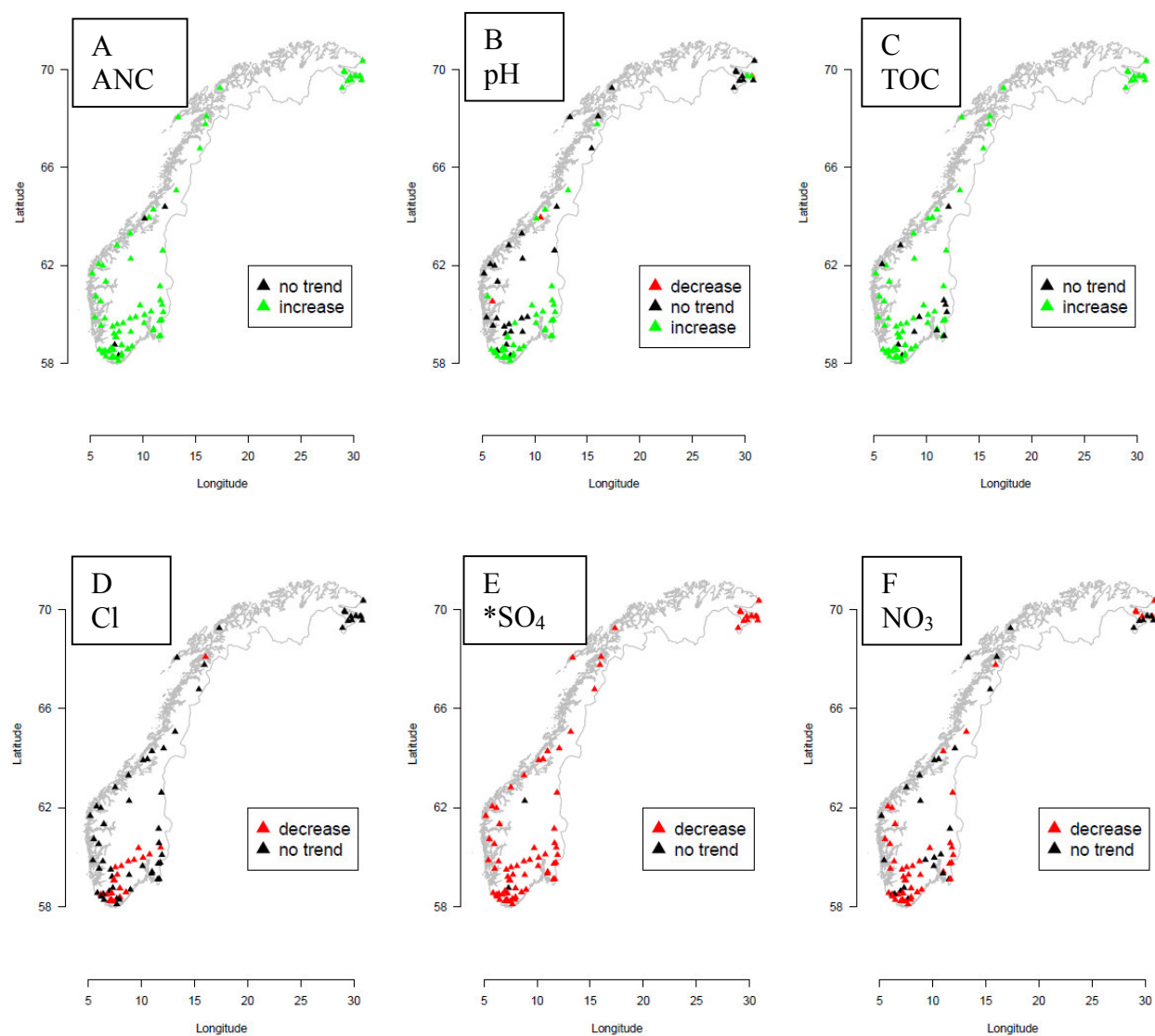
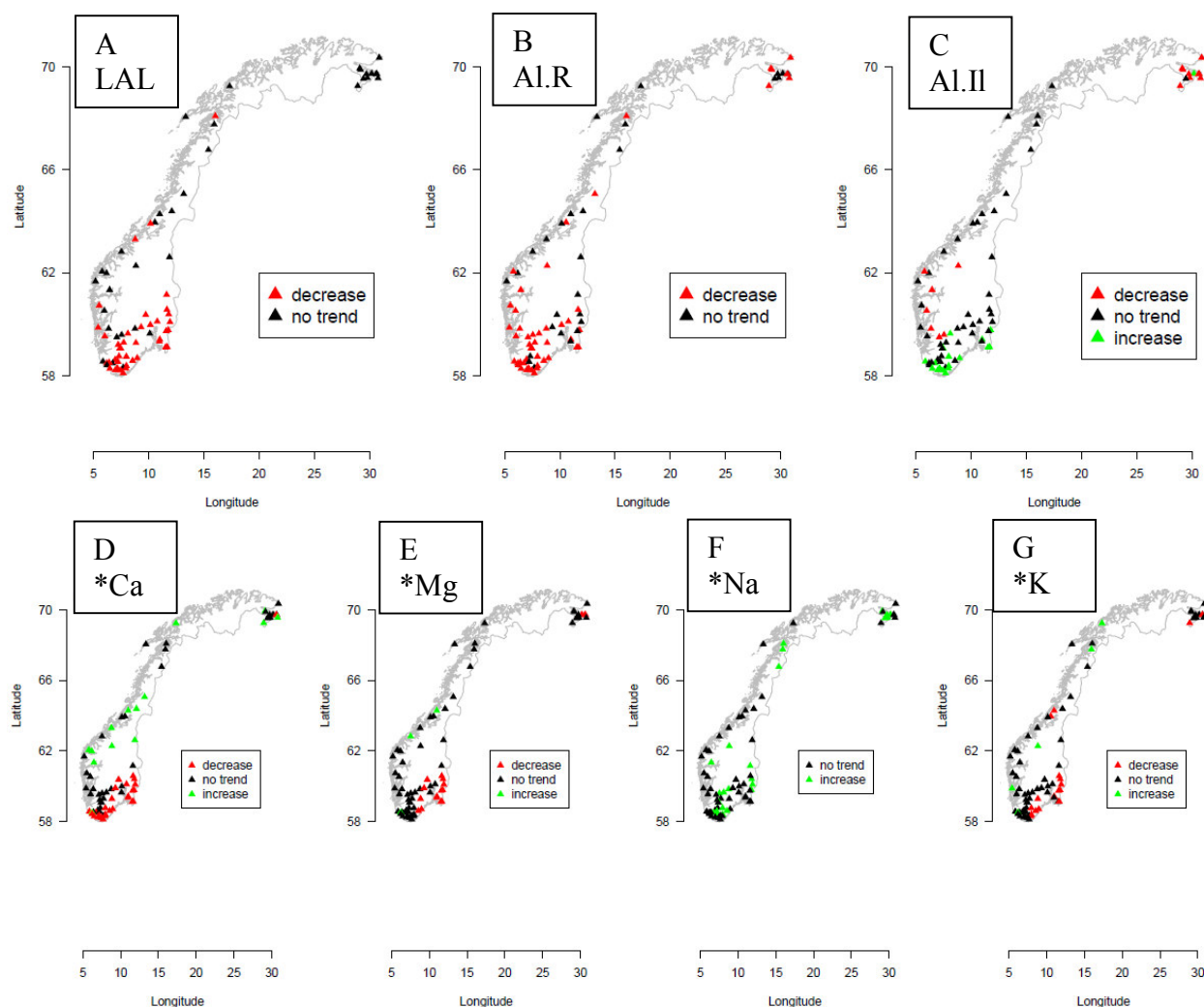


Figure 23 Significant trends of A) ANC, B) TOC, C) pH, D) Cl, E) non sea-salt SO<sub>4</sub>, F) NO<sub>3</sub> for each lake and its geographic location. Only significant changing lakes ( $p < 0.05$ ) are shown. A significant increase ( $\tau > 0$ ) is shown in green, and a significant decrease ( $\tau < 0$ ) in red. The values are based on the Mann-Kendall test with false discovery rate corrected p-values for multiple testing.





**Figure 24: Significant trends of A) LAL, B) Al.R, and C) Al.II, the non-sea-salt cations: D) Ca, K) Mg, E)Na, F)K for each lake and its geographic location. Only significant changing lakes ( $p < 0.05$ ) are shown. A significant increase ( $\tau > 0$ ) is shown in green, and a significant decrease ( $\tau < 0$ ) in red. The values are based on the Mann-Kendall test with false discovery rate corrected p-values for multiple testing.**

A closer look on the trends between the different lake types revealed significant trends in different lake types (Table 10). Mg,  $\text{SO}_4$ ,  $\text{NO}_3$ ,  $\text{H}^+$ , conductivity, Al.R, LAL and  $\text{ANC}_{\text{OAA}}$  are significantly decreasing in all lake types. Additional ions (Ca, Na, and Cl) are mainly decreasing in Cl lakes. ANC values are decreasing in Cl and  $\text{SO}_4$  lakes and highly increasing in ANC lakes. In fact, the ANC increase in ANC lakes is the largest trend of all parameters in all lake types. pH, TOC, and Al.II concentrations are increasing in all lake types.

**Table 10: Theill Sen slopes estimates (% a<sup>-1</sup>) for the different lake types. Insignificant trends are shown in grey, significant increases in green and significant decreases in red (Mann Kendall test, p<0.05, fdr corrected). Ions are given in mass per volume units (row 1), and as sea-salt corrected values in equivalents (row 2 and 3).**

lake type	Ca	Mg	K	Na	SO <sub>4</sub>	NO <sub>3</sub> -N	Cl	pH
ANC lake	0.08	-0.27	-0.17	0.22	-2.13	-2.50	-0.14	0.33
Cl lake	-0.48	-0.90	-0.56	-0.82	-2.43	-1.61	-0.83	0.50
SO <sub>4</sub> lake	-0.47	-1.03	-0.40	-0.92	-2.49	-1.72	-1.84	0.39
lake type	Ca*	Mg*	K*	Na*	SO <sub>4</sub> *			
ANC lake	0.12	0.00	0.00	5.51	-2.54			
Cl lake	-0.71	0.00	0.00	-4.17	-2.80			
SO <sub>4</sub> lake	-0.42	0.00	0.00	7.08	-2.49			
lake type	COND	ALK	TOTP	TOTN	TOC			
ANC lake	-0.86	0.69	0.00	-0.22	1.20			
Cl lake	-1.43	0.60	0.00	-0.21	3.62			
SO <sub>4</sub> lake	-2.12	0.45	0.00	-0.14	4.65			
lake type	AI/R	AI/II	LAL	ANC	ANC <sub>OOA</sub>			
ANC lake	-1.26	1.33	-2.84	83.33	-23.87			
Cl lake	-2.87	3.21	-2.91	-3.11	-2.81			
SO <sub>4</sub> lake	-1.69	4.59	-3.29	-5.70	-3.85			

A closer look on the dynamics of TOC, Cl and SO<sub>4</sub> in the different lake types reveal that all parameters are changing quicker in Cl and SO<sub>4</sub> lakes than in ANC lakes (Figure 25). SO<sub>4</sub> lakes have the largest spread of the data. A closer look on the residuals shows that a hidden cycle of TOC and Cl concentrations, especially in SO<sub>4</sub> lakes, but also in Cl lakes, is visible in 1987, 1989, 1993, 1997, 2000, 2003, 2008, and 2012.

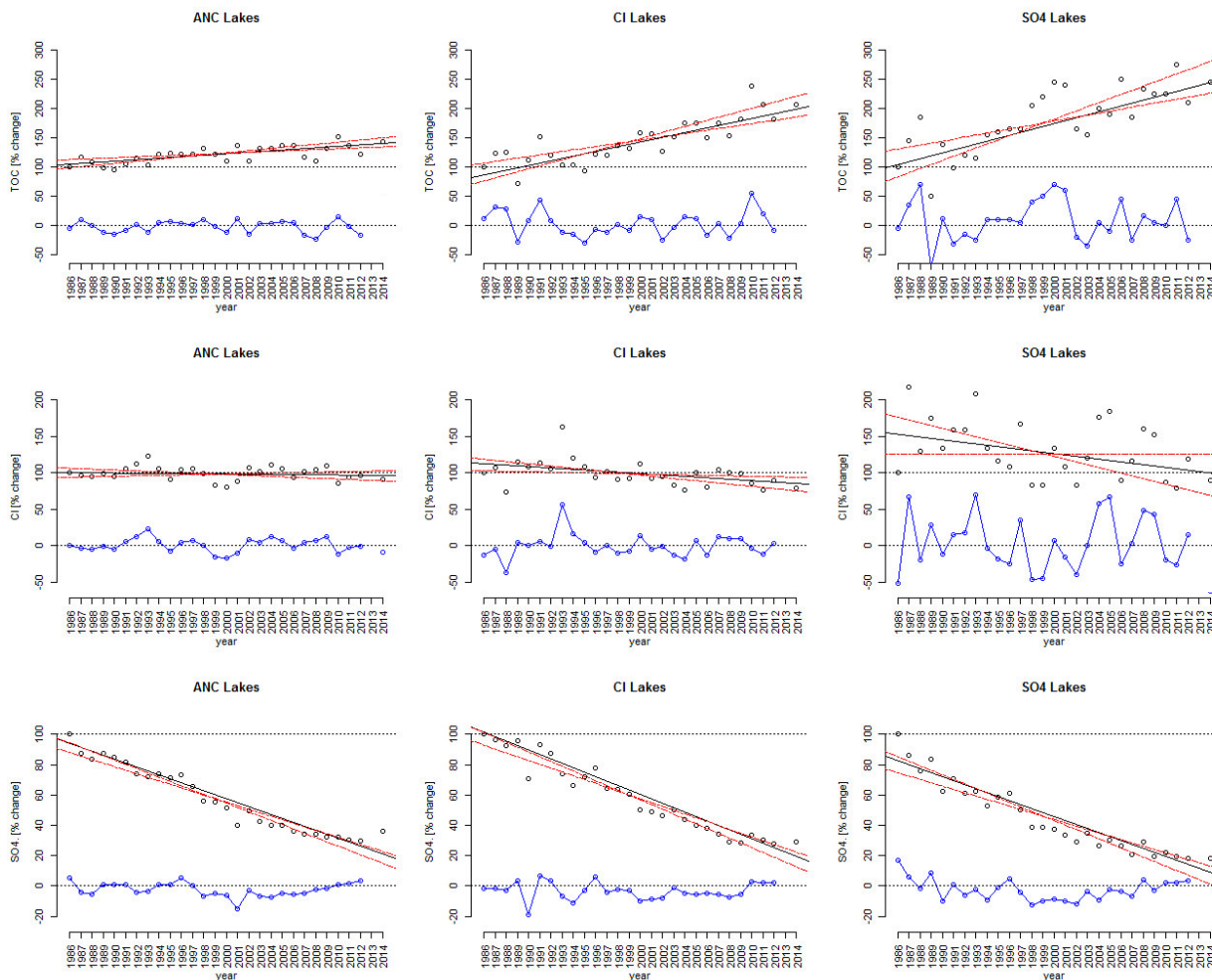


Figure 25: Theil Sens slope estimates (black line) and 95% confidence intervals (red lines) of the median values for the different lake types for TOC, Cl and SO<sub>4</sub>. The residuals are shown as blue lines.

The dynamics of Al,II, LAL and pH are shown in a time series in Figure 26. Al,II and pH are generally increasing, while LAL is decreasing. The most pronounced increase of LAL is observed in SO<sub>4</sub> lakes, while the most pronounced decrease of Al,II is observed in ANC lakes. The residuals show again patterns that deviate from linear trend. The Al,II concentrations for example decreased rapidly after 1991 and stayed low until recent measurements. In CI and SO<sub>4</sub> lakes the Al,II decrease is also highest between approx. 1995 and 2005. LAL shows similar periodic increases and decreases as TOC and Cl with peaks in 1987-1988, 1990-1991, 2000-2001, 2004-2006, and 2011.

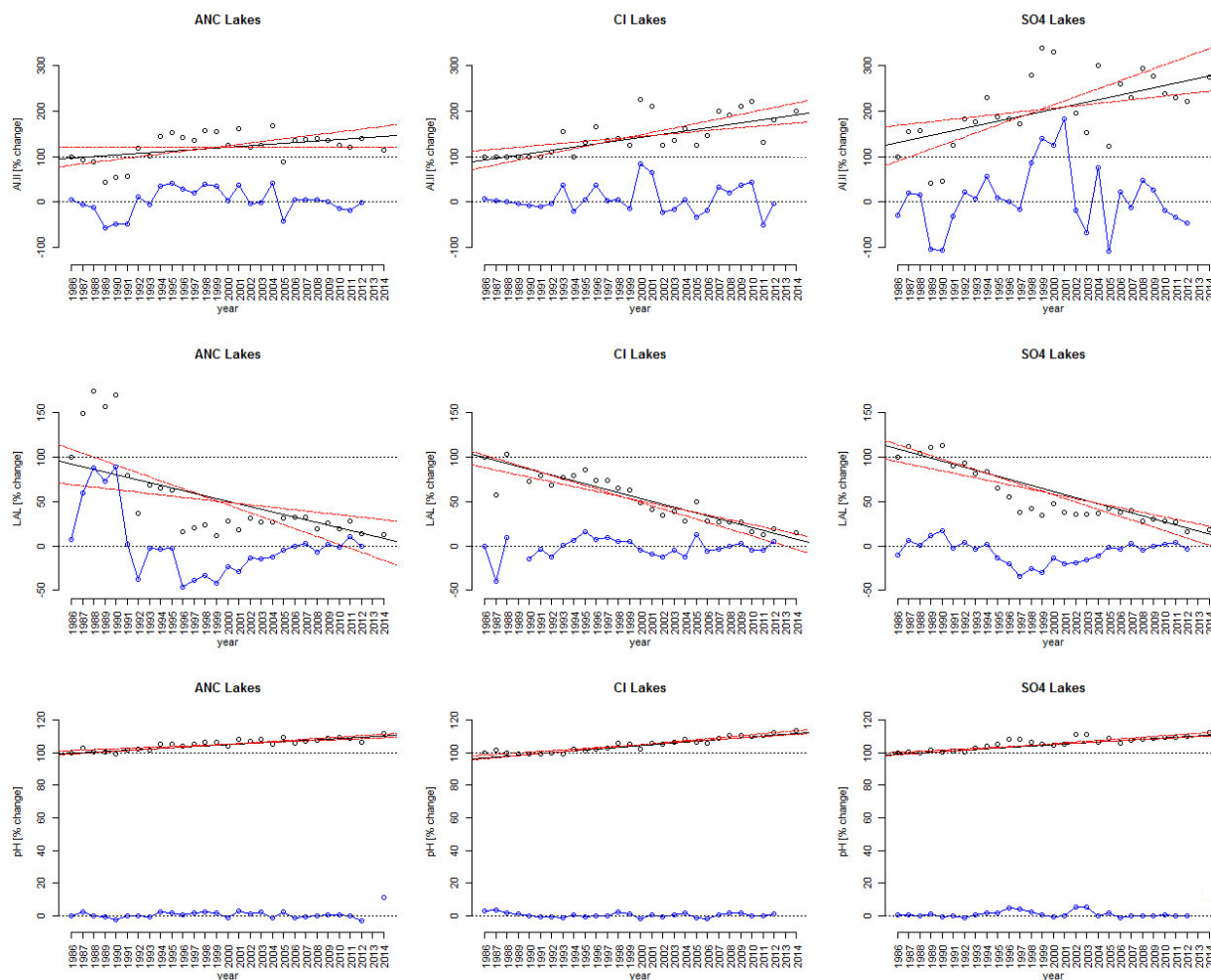


Figure 26: Theil Sens slope estimates (black line) and 95% confidence intervals (red lines) of the median values for the different lake types for AIJ, LAL and pH. The residuals are shown as blue lines.

## 4 Discussion

### 4.1 Geographic differences

The ten regions are characterized by different environmental and geographical features. In general, the western regions are characterized by mountains with thin soil deposits and high precipitation rates, while the eastern regions have thicker soil deposits, more forests and less precipitation.

Region one to three are quite far away from the coast in the area of high forestation (Figure 2B). The elevation, precipitation and runoff are low in these regions (Figure 6) and the population density is high (Figure 2). In fact, region two is in the Oslo region and has the highest population of Norway. The wind patterns in these areas bring air borne pollutants mainly from middle Europe (Figure 3).

Regions four and five are mainly at the south coast and partly in the mountains (Figure 1). The coast regions are characterized by high temperatures and moderate precipitation, while the regions in the mountains have higher precipitation, and lower temperatures. The regions are closest to the influence of air-borne pollutants from middle Europe and Great Britain (Figure 3).

Regions six and seven are at the west coast, which have high population densities and the highest precipitation and runoff, high temperatures and low forestation due to the mountains close to the coast.

Regions eight and nine are in the north-west. In these areas, the temperature is lower and more snowmelt occurs. In general the precipitation and runoff is high. The population densities in the northern countries are much lower than in the south.

Region ten is located in Eastern Finnmark, close to the Russian border. The population density is very low and the climate dry and cold. On the Russian side of the border, several local pollution sources in form of mines, and power plants are known (Figure 2; Reimann, 1996).

All the chloride lakes (n=13) are located at the south and west coast. This is the area with most precipitation, runoff (Figure 6) and storm episodes. Most sulphate lakes (n=12) are in the south, a bit further inland, with one exception in region ten. Six of the sulphate lakes are located in the mountains (elevation  $\geq 500$  m.a.s.l.). Most lakes (n=54) are ANC lakes and distributed all over Norway.

Based on the geographic distribution of different chemical lake characteristics, following regions can be separated:

- 1) **Southeast:** In the Southeast, the TOC concentrations are highest, the sea-salt impact is low (Cl), the buffer capacity is high (ANC) and the non-sea-salt SO<sub>4</sub> concentrations are high. Al concentrations in this region are also high, but a large fraction of it is not bound. (regions one and two could be clustered in here)
- 2) **Southwest:** In the Southwest, the lowest ANC values and lowest pH-values have been measured. The Cl concentrations are generally higher than the others. Non-sea-salt SO<sub>4</sub> concentrations are high, and nitrate concentrations is the highest here. Al concentrations in this region are also high, and most of it is not bound. (regions five is most characteristics, but region four near the coast could be clustered in here)
- 3) **Western coast:** The ANC values are relatively high, the TOC and SO<sub>4</sub> concentrations are low, and the chloride concentrations are high for the lakes located below 500 m.a.sl.. (regions six to nine are located here)
- 4) **Eastern Finnmark:** The ANC values are high, the pH is highest in this area, and SO<sub>4</sub> – values are relatively high. (Only region ten is located here).
- 5) **Mountains:** the altitudes are high, Cl concentrations are low and the pollutions by SO<sub>4</sub>, NO<sub>3</sub>, and Al are low.

Cl:SO<sub>4</sub>-ratios can be used to assess whether anthropogenic sulphate introduction or Cl introduction by sea-salt are more important in acidification, cation exchange processes, and ANC reduction. The Cl:SO<sub>4</sub>-ratio is highest in the west and north, indicating a higher importance of sea-salt while the Cl:SO<sub>4</sub>-ratio in the South and Eastern Finnmark is lower, indicating higher impacts of anthropogenic SO<sub>2</sub> emission.

Cl concentrations are in general lower in higher elevations. This indicates that the transport of sea spray is limited to lower altitudes. This could indicate that increased sea-salt events by increased storm events, may not have a large impact on mountain lakes. However, increased runoff and potentially weathering related to higher precipitation may play an additional role in determining the ionic composition of mountain lakes in a changing climate.

Three regions with exceptionally high ionic strength are apparent; regions 2, 5 and 10. Region 5 is characterised by high sea-salt concentrations (Na, Cl and NO<sub>3</sub>). This indicates high impacts of sea-salt events and impacts on anthropogenic high NO<sub>x</sub> emission. In regions 1 and 2 more ions are unrelated to sea-salt. SO<sub>4</sub> and base cations are enriched. This indicates high impacts on anthropogenic SO<sub>2</sub> emissions and higher weathering rates, which may be related to bed rock acidification

#### 4.2 General trends

The trends in the present dataset are overall comparable to previous studies in Norwegian lakes (Skjelkvåle et al., 2001; Skjelkvåle et al., 2007) and other European surface waters (Prechtel et al., 2001). The main difference is the finding of more significant changes (Skjelkvåle et al., 2001; Skjelkvåle et al., 2007). Due to a dataset with a longer time series, which allows a higher statistical resolution with low p-values, which allows the detection of even small changes?

The general trends of decreasing SO<sub>4</sub>, and a small decrease in NO<sub>3</sub> are found to be similar to earlier studies. Most striking is the increased trend of NO<sub>3</sub> decrease, which may be caused by the longer time series, which allows detecting also small trends. The recovery from acidification is indicated by increased ANC, alkalinity and pH in the current thesis, as well as in earlier studies.

In general, more lakes recovered since 2004, which can be seen as an increased number of ANC lakes and lack of SO<sub>4</sub> lakes in 2014. Furthermore, there are more lakes with a significant increase in ANC, TOC and pH, indicating that the recovery is still happening. This indicates that more lakes recovered since the last publication about the 1000 Norwegian lake dataset in 2007 (Skjelkvåle et al., 2007).

In 2007 no NO<sub>3</sub> trends or regional patterns of change have been found, while more trends and first regional pattern become apparent in the current thesis (Skjelkvåle et al., 2007). Wright et al., (2001) proposed that long term trends in NO<sub>3</sub> concentrations may be masked by climate effects which could cause noise in the signal, by biogenic nitrate release in autumn, and by the saturation of catchments with NO<sub>3</sub> in the 1980s. However, the current dataset has a sufficient long time series to show, for the first time, significant decreases of nitrates in most Norwegian lakes.

As predicted by Skjelkvåle et al (2001, 2007), the SO<sub>4</sub> decrease and ANC increase continued in the last decade after the full implementation of the Gothenburg protocol. The aim of the Gothenburg protocol is an 80% sulphate and 50% N, by 2010, relative to 1980. (Shapiro, 2000). 20 % decrease of SO<sub>4</sub> has already been registered in the 1980's, and 40% in the 1990's. (Skjelkvåle et al 2001). Now it appears that the remaining reduction of NO<sub>x</sub> and SO<sub>2</sub> emissions were achieved and had an effect on the chemical recovery of the lakes. Chloride concentration continued to decrease, with two new sea-salt events since 2004, and TOC continues to increase. NO<sub>3</sub> is decreasing in the long run. The effect of climate change is apparent, as increased weathering and base cation release at the west coast.

Whether the biota has recovered at the same speed and whether climate change had or will have additional effects will be discussed later and should be studied in the future.

#### 4.3 Acid rain, and ANC changes

As shown via the time trends of pH, many lakes, especially in the South, recovered significantly from the effects of acid rain since 1986.

The increasing pH and decreasing H<sup>+</sup> concentrations are accompanied by decreasing SO<sub>4</sub> concentrations. Since SO<sub>4</sub> is the main reason for anthropogenically caused acid rain in Norway, it appears that the reduction of coal power plants and the development of cleaner technologies in Europe allowed the recovery of Norwegian lakes. The fact that mostly lakes close to pollution sources in Europe or Russia had significant increases in pH and a significant decrease in non-sea-salt SO<sub>4</sub> shows that the lakes recover towards a natural situation, typical for pristine Norwegian lakes.

Additionally, the buffer capacity (ANC) appears to be restored in the most impacted lakes from the 70s. In fact, no sulphate lakes were present in 2014 anymore, and the number of ANC lakes increased. Increasing buffer systems by increasing TOC and the restoration of the bicarbonate buffer system with increasing pH may help the natural restoration of the studied lakes. Even though restoring seems to take longer than previously expected (Skjelkvåle et al., 2007). However, since TOC concentrations are increasing all over Norway and Ca concentrations increase in mid Norway the good-moderate boundary values of ANC are just reached, but still close to the limit (Austnes, 2014; Table 4).

Figure 27 shows the proposed impacts and interactions of different ions with the buffer capacity (ANC) and pH of the lakes, and what the causes and consequences may be.

A positive relationship of H<sup>+</sup> with SO<sub>4</sub> and NO<sub>3</sub> shows that SO<sub>4</sub> and NO<sub>3</sub> are the main drivers of acidification. Base cations have a negative relationship with H<sup>+</sup>, this leads to higher ANC values at a higher pH. Thereby, base cations appear to be important for the ANC buffer system. Increased base cation release by weathering may be one explanation for the ANC recovery. The increased ANC may also be attributed to an increasing Na:Cl ratio or to non-sea-salt SO<sub>4</sub> reduction (Skjelkvåle et al., 2007). In 2007 the increasing ANC has been found to be caused largely by a gradual decrease in Cl and no trend in Na, which increases the Na:Cl ratio. The use of the more extended dataset of the current study confirms this hypothesis and even shows an increase of non-sea-salt Na in lakes with decreasing Cl concentration, which are located away from the coast. The areas which are significantly increasing in Na lie in the south east. Since there is less sea-salt events, the Na that is retained from previous sea-salt events is now slowly coming into the lakes, while no more Cl is introduced away from the coast. This finding may indicate an accelerated increase of the Na:Cl ratio, which may be a cause for the increased buffer capacity (ANC).

Establishing balance in the Na: Cl ratio is assumed to take >2 years (Hindar 1995, Skjelkvåle et al., 2007) and could explain a delayed increase in ANC after sea-salt depositions.

In areas with strong non-sea-salt SO<sub>4</sub> decrease the role of the Na:Cl ratio is less pronounced and the increase of ANC is rather related to the decrease of SO<sub>4</sub>, while the opposite is true for areas with little SO<sub>4</sub> decrease (Skjelkvåle et al., 2007)

The TOC increase is an additional factor which may explain the ANC increase. However, the contribution of OA to the ANC increase is rather low compared to SO<sub>4</sub> (Hruska et al., 2003).

TOC increase in UK (Evans and Monteith 2001), is most likely related to recent changes in climate, but may also be related to acidification recovery. With increasing pH, the TOC is less charged, so that the CEC increases and TOC does not coagulate that easily, but stays in solution and leads to brownification of the water (Skjelkvåle et al., 2007).

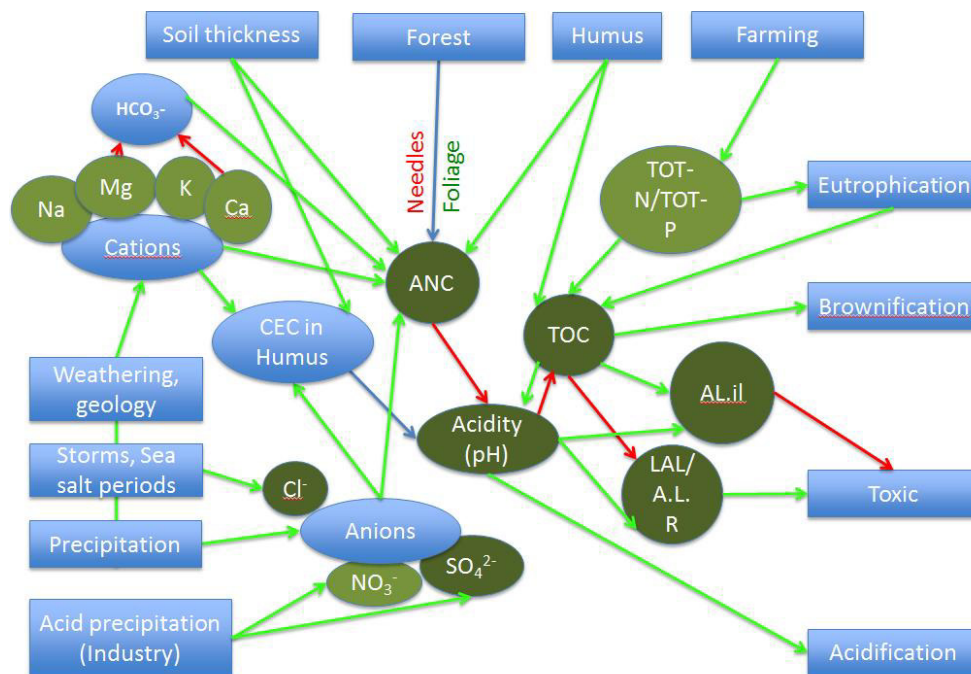


Figure 27. External Influences on and outcome of parameter changes in the water body. The green circles are measured parameters. Green arrows indicate positive correlations and red arrows negative correlations (Made by Hunskaar, based on information from several textbooks: e.g. Schwoerbel, 2005; VanLoon and Duffy, 2011, Wetzel 1997).

#### 4.4 Sea-salt episodes

Chloride can originate from both sea-salt, and old marine deposits. The highest input of sea-salt to freshwater lakes is during sea-salt episode, which can be related to climate fluctuations such as storms and the North Atlantic Oscillations (Evans et al., 2001).

Skjelkvåle et al., (2007) observed a decrease in Cl mainly in the south. Our dataset with the later years shows a similar decrease. In contrast to the proposal of Skjelkvåle et al.,(2007) that no trends in chloride from sea-salt would show in a longer time period, a significant decrease is still found in a 28 years' time series, in many lakes in the south and in lakes with low ionic strength (SO<sub>4</sub> lakes). This indicates that the long term trend of Cl is really a decrease and not only related to a sampling time during the descending period after a sea-salt event. Skjelkvåle et al.,(2007) proposed that high initial concentrations of Cl in the 1990s are responsible for the decreasing trends. In 1989, 1990, and 1993 unusually strong storms, causing sudden fish deaths have been described (Hindar et al.,1994). Skjelkvåle et al.,(2007) found that Cl is more decreasing in the south, and that difference in ionic composition in the different regions may be a reason. They proposed that there are more decreasing Cl-events in the east, caused by less rain and thicker soil. Less precipitation leads to less ions flushing away, and thicker soil leads to longer residence times of the ions. This means that there can be higher concentrations, and higher absolute changes in concentrations in the east compared to the west.

This may show that there is really a long term trend in sea-salt episodes decrease. But two other explanations can be proposed; i) Old marine deposits may become depleted in some areas; or ii) the use of road salt might be reduced. Sea-salt episodes did not seem to increase since 1986 and they did not seem to stop the recovery of acidified lakes considerably. Sea-salt events were



detected as periodic Cl peaks in the residual plots of the sens slope. The Cl peaks usually fit to previously describe sea-salt events (Figure 13). The Cl peaks did not increase in frequency, but decreased in magnitude in the last years. Sea-salt events related Cl peaks appear to be of highest magnitude in the most poorly buffered SO<sub>4</sub> lakes, followed by Cl lakes. In ANC lakes the Cl pattern are hardly visible, due to the high buffer capacity. The same pattern has been observed for the general decrease of Cl, which is only significant in SO<sub>4</sub> and Cl lakes.

In the coming 50 to 100 years the changes in Norway will appear as more storms, especially in the East and South (Gregow et al., 2011). Mean geostrophic wind speeds are thought to increase all over Northern Europe by 5 – 10%. Extreme geostrophic wind speeds are predicted to increase in the South and West of Norway and to decrease over the Norwegian Sea (Gregow et al., 2011). Marine deposits should not play a role since these lakes are chosen above the marine limit. Road salt use on Norwegian roads is increasing in the later years. This might have an impact on sea-salt calculations, and ionic content, also in the snow melt periods.

The ions with the highest correlation in the lake water are Na and Cl, which indicates that Na is mainly introduced by sea-salt. However, negative values in the sea-salt corrected calculations of Na show that an over/ or under estimation of certain sea-salt corrected ions are made, thus there must be other sources for Na and possibly other cations that are currently categorized as sea-salt ions than Sea-salt itself.

Even though anthropogenic sources are low after the monitoring design (Henriksen et al., 1989), potential impacts of road salts should not be ignored. Road salts in Norway consist mainly of NaCl, rarely mixed with Mg, and may in some cases have an impact on the ionic composition on lakes (Blomqvist, 2001). The consequence of road salt input would, in fact, be an underestimation of Na and an overestimation of other cations. In this case other ions like Al and Cu might also be introduced (Zehetner et al., 2009). Negative values of chloride might also show that the calculation of Non-sea-salt ions assumptions should be carefully interpreted since different ions have different retention time in the catchment area, which alters the Cl : cation ratio in the lake compared to the rain.

#### 4.5 Aluminium trends

Aluminium concentrations are generally constant, but the Al speciation changes. Generally LAL and Al/R are decreasing especially in the south, while Al/II is increasing in the far South, and decreasing in the west coast and Eastern Finnmark, at the same lakes as Al/R is decreasing.

TOC is most positively related to ANC and Al/II, and negatively to pH, which means that TOC is important to bind Al and that this process could be reduced by high pH and low ANC values. Thus, I propose that the reactive and labile Al can be bound by TOC and therefore loses its toxicity. Increasing TOC concentrations can explain the increase in Al.II, with bounding of labile aluminium species to TOC (reviewed by e.g. vanLoon and Duffy, 2011).

Furthermore, the increased pH is known to lead to a decreased mobility of Al, a minor role of the aluminium buffer system, and lower fish toxicity. The Al concentrations follow, however, the pattern of sea-salt events. This can be explained by cation exchange processes and periodic flushing of remobilized Al from the catchment into the lakes.

Different trends of Al<sub>II</sub> in the South and the West of Norway can be explained by different processes that determine the Al cycle and the mechanism behind the decrease in reactive Al species in these areas. In the South, East, and Eastern Finnmark charged Al species are mainly bound to TOC, and start coagulating in a rapidly increasing pH after reduction of anthropogenic acidification. At the west coast increased precipitation can explain the removal of not only reactive and labile Al, but also of Al<sub>II</sub>. Thereby, increased precipitation can lead to flushing of ions from the catchment and to increased weathering. Increased weathering in a higher pH, leads then to an increasing role of Silicate and cation exchange buffer systems.

#### 4.6 Brownification

Generally, TOC concentrations increase all over Norway. During sea-salt episodes TOC coming from the catchment appears to be flushed into the lakes together with Cl coming from the sea. This means that the H<sup>+</sup> introduced via sea-salt events can be partly neutralized by the co-introduced TOC, and that the introduced TOC can flocculate, which may prevent severe brownification events together with the sea-salt events.

#### 4.7 Weathering and base cation exchange

Changes in weathering process can be studied via trends in non-marine base cation concentrations. General west-east gradient in Norway include gradients from high to low precipitation, (Henriksen et al., 1998), from mountain areas with thin soil covers towards forests with thicker soil. The gradient is also visible as more Clearwater lakes in the west and more brown waters in the east (Henriksen et al., 1998).

Skjelkvåle et al., (2007) found another gradient from low base cation concentrations and alkalinity in the West compared to the East. In the west high precipitation can wash out more base cations after weathering processes. The same is true in the current dataset. However; the base cation concentrations are increasing in the West and decreasing in the East, which means that the gradient weakens.

As for Al different processes lead the changes in weathering in the East and in the West. In the South east and Eastern Finnmark the highest reduction in acidification and ionic strength is observed due to their location closer to middle European and Russian industry sites. This means that less base cations released by acid dissolution (weathering) when the lakes recover. Thereby, lower concentration of strong acid anions leads to lower cation concentrations. As described by Skjelkvåle et al., (2001), a decrease in cations is correlated to a decrease in SO<sub>4</sub> in Norwegian lakes.

In the North and West coast of Norway, pH does not increase that much and even an increasing weathering is observed. I propose, that more precipitation at the west and north West coast leads to increased physical and chemical weathering; hydrolysis and dissolution and consequently to higher base cation concentrations.

The observed reduction in SO<sub>4</sub> anions has to be compensated by reductions in base cations or the ANC will increase. The F-factor is usually used to show this compensation. The F-factor shows how much of the decrease in SO<sub>4</sub> is compensated with the decrease in base cations (Mg + Ca) (Henriksen, 1984, Skjelkvåle, 2001)

$$F - \text{factor} = \frac{(|\text{Mg}| + |\text{Ca}|)}{(|\text{SO}_4|)}$$

A low F-factor means that base cations can compensate for a small proportion of SO<sub>4</sub>-change, which means a high increase in ANC.

The average F-factor in the extended dataset used in this thesis is 0.45 (only lakes with significant trends in Mg, Ca and H<sup>+</sup> are considered), which means that 45% of the SO<sub>4</sub> reduction can be compensated by base cation reduction, which means that a larger fraction is compensated by an increase in ANC.

In a time series between 1990-1999, the F-factor was 0.65 (Skjelkvåle, 2001), meaning that base cations played a more important role during this time. And the ANC-increase appears to have become more important during the recent years.

This means that the base cation concentration have increased more in the recent years than in the beginning. This may be related to increase weathering because of increased precipitation.

#### 4.8 Buffer system changes

Alkalinity is significantly increasing in 16 lakes in the current extended dataset. The alkalinity increase is less than found by Skjelkvåle et al., (2007). An increase in alkalinity is usually attributed to an increase in bicarbonate which may act as a buffer system. However, the bicarbonate buffer system only plays an important role at pH>6.2, most lakes still have a rather acidic pH of around 5.5 up to 6.5, which indicates that the bicarbonate buffer system did not replace the other buffer systems, yet. Median pH and alkalinity comparisons show that the alkalinity increases significantly in lakes with a higher pH. This indicates that the alkalinity increases most if the pH allows an effective bicarbonate buffer system. In some lakes a pH above 5.5 is reached and the bicarbonate buffer system might work, but increase in TOC and increase in Al contributes to the increase in ALK, (Skjelkvåle, 2001, B). Similar trends are observed in ANC values, which are by definition the surplus of bicarbonate to H<sup>+</sup>. Since the ANCOAA value, which includes TOC in the calculations, is significantly increasing in 73 lakes, it appears that the humus buffer system is still more important than the bicarbonate buffer systems.

The buffer system that recovers most seems to be the ANC-cation exchange system and not the bicarbonate buffer system. With changing pH the dominant buffer systems seem to change from Aluminium (between pH 2.8 and 4.2) and cation-exchange (between pH 4.2 and 5) buffer systems to cation-exchange, silicate (between pH6.2 and 5), and bicarbonate buffer systems (reviewed by Norton, 1989, Ulrich, 1983). In the end increasing CO<sub>2</sub> levels, caused by climate change may increase the Alkalinity of the lakes even more and increase the buffer capacity by strengthening the role of the bicarbonate buffer system. However, the acidification related to increased CO<sub>2</sub> levels, may counteract this trend.

#### 4.9 Nutrient change

The nutrients PO<sub>4</sub>, NH<sub>4</sub>, TOT-N and TOT-P stay rather stable over the measured time period in the measured lakes, with only a few lakes showing significant decreases or increases in TOT-N. Since the study lakes were chosen in areas that are not impacted by local pollution (Henriksen, 1989), such as agriculture, the finding of no change in nutrients supports the robustness of the monitoring setup. However, increased TOC indicates that natural sources of organic matter

increase, most likely because of the increase in pH and the decrease in ionic strength and hence increased solubility of natural organic matter (measured as TOC) (Monteith et al., 2007; Haaland et al., 2010).  $\text{NO}_3$  is a nutrient that can be transported via the atmosphere (as  $\text{NO}_x$ ), which is significantly reduced in many lakes. This may have implications on the nitrogen cycle and may lead eventually to nitrogen limitations. This could then lead to cyanobacteria dominated lakes, which can cause toxic algal blooms (reviewed by: Callisto et al., 2014). Whether the reduction of anthropogenic  $\text{NO}_x$  is sufficient to cause this effect remains, however, speculative and should be investigated in later studies and modelling approaches.

#### 4.10 Climate change

Since industrial emissions of  $\text{SO}_2$  and  $\text{NO}_x$  are decreasing, climate-related factors can become more important in delaying lake recovery (Skjelkvåle et al., 2003; Akselsson et al., 2013). Hence, climate change should be considered as the main threat to Norwegian lakes, in the future

Sea-salt events were proposed to increase in magnitude and frequency due to increasing storm events in Norway in a changing climate. Sea-salt episodes are known to increase the ionic charge in catchment areas and to yield in increased cation exchange processes. In already acidic catchments this could cause a sudden flush of  $\text{H}^+$  into the lakes (Skjelkvåle et al., 2007; Laudon, 2008). However, the data of the thesis cannot support this hypothesis. Sea-salt ions are usually decreasing over time and the magnitude, or frequency of sea-salt events did not increase. Due to the slow change and low frequency of these events, this finding should be interpreted carefully and further studies are necessary to be sure.

Increased runoff and permafrost thawing appears to lead to higher weathering. This effect is most apparent in regions that were not much affected by acid based weathering in the 1980s. In areas that were most affected by anthropogenic acidification the increase of pH leads to a decrease in weathering that cannot be compensated by increased weathering related to increased precipitation and temperature. In western Norway, where the precipitation is found to be most increasing, a positive trend in weathering can be observed, which is most likely directly related to climate change.

Permafrost thawing in the north and in the mountains, higher runoff and increased forestation can also accelerate the inflow of ions and TOC. The TOC concentrations are indeed increasing, which indicates that some of the factors play a role. This of these factors is most important in explaining the increasing brownification remains unclear. A multivariate model including forest extension data, permafrost, and runoff data for the studied catchments may help to quantify the importance of the difference processes that may increase brownification. So far, the increasing pH appears to play an important role.

A changing buffer system by increasing  $\text{CO}_2$  levels can be another effect of climate change, but acidification caused by the same compound may counteract this trend. However, increased temperatures also decrease the solubility of gases such as oxygen and  $\text{CO}_2$ , which are crucial for biological processes and fishes, and which play an important role in the carbonate buffer system.

Increased nitrification processes by increased temperatures (van Breemen et al., 1998; Wright and Jenkins, 2001) do not seem to play a role on the general ionic composition of lakes. Most likely the lack of agricultural input in the study lake and the saturation of the catchment with industrial

$\text{NO}_3$  can explain the lack of increasing nitrification. A potential increase in natural nitrification would be much lower than the reduction of nitrate by reducing  $\text{NO}_x$  emissions. Direct measurements of nitrification rates could help to clarify the role of climate change on natural nitrification processes.

## 5 Conclusion

The present thesis showed, quite clearly, how climate change can counteract the success of decreased  $\text{NO}_x$  and  $\text{SO}_2$  emissions on the trends in ionic compositions of Norwegian lakes. The joined European efforts of the Gothenburg protocol lead to a significant recovery of lakes in the most impacted areas since the 1980s. Namely, the pH increased and the buffer capacity (ANC) recovered, which yielded in a reduction of toxic Al species (LAL, Al/R) concentrations and weathering rates (BC).

The changes in ionic compositions were most drastic in the South and North, where industrial impacts from Europe are highest. Also lakes with lower buffer capacities are more vulnerable. However, most lakes reached the ANC limit and became ANC lakes in recent years. This shows that lakes in initially highly impacted areas are still recovering.

However, in lakes where the impact of acid rain in the 1980s was rather low, the ionic composition appears to change because of climate change related processes, such as increasing temperature, precipitation, and thawing of permafrost. Most pronounced is the increased weathering rate (BC), which counteracts the trend of lake recoveries in the south. The release of Ca by weathering may reduce the buffer capacity of the lakes via binding of bicarbonate. How far the increased  $\text{CO}_2$  concentrations will have an impact on the bicarbonate buffer system remains unclear.

Another process that might decrease the pH in the future is the increase of TOC concentrations all over Norway. Whether the increase is more related to climate change related issues, such as permafrost thawing or increasing biomass production, or to the increasing pH, remains speculative. Increasing frequencies and magnitudes of sea-salt episodes could be another climate change associated acidification process. In spite of the expectations of increasing storm events, no such increase in sea-salt episodes has been observed, yet. Climate change will most likely have most drastic effects in northern Norway due to thawing permafrost and increasing wood growth.

## 6 Outlook

The current study shows that the reduction of rather local anthropogenic  $\text{SO}_2$  and  $\text{NO}_x$  emissions in Europe was successful in reducing lake acidification in Norway, but that the increasing emissions of  $\text{CO}_2$  worldwide becomes a new issue for changing ionic compositions and potentially acidification in Norwegian lakes. A worldwide framework for reducing  $\text{CO}_2$  emissions, as strict as the Gothenburg protocol would be a way to counteract the effects. The Kyoto protocol may be a start, but it appears that the goals of reducing  $\text{CO}_2$  emissions are too low. Even though effects of climate change due to anthropogenic  $\text{CO}_2$  emissions are hard to stop; a worldwide effort could reduce the effects and may prevent more severe acidification events.

An extension of the 1000 lake dataset in the future with a new focus on the climate change based change of the ionic compositions can help to understand and proof the described mechanisms in more detail. Thereby, climate variables, such as runoff, CO<sub>2</sub> concentrations, temperature, and permafrost thawing should be quantified and included in later analysis. Modelling of the already available dataset and extrapolations of the results for the future may help to quantify the effects.

### 7 Acknowledgements

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## 8 References

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## 9 Appendix

### 9.1 Appendix A: Supplementary Figures

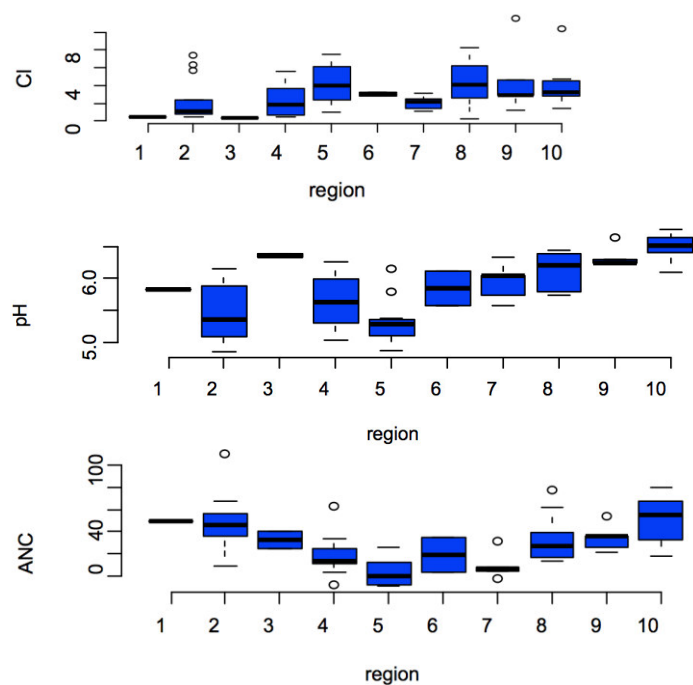


Figure 28: Magnitudes of different parameters in different regions. Cl in mg l<sup>-1</sup>, pH, ANC in μeq l<sup>-1</sup>

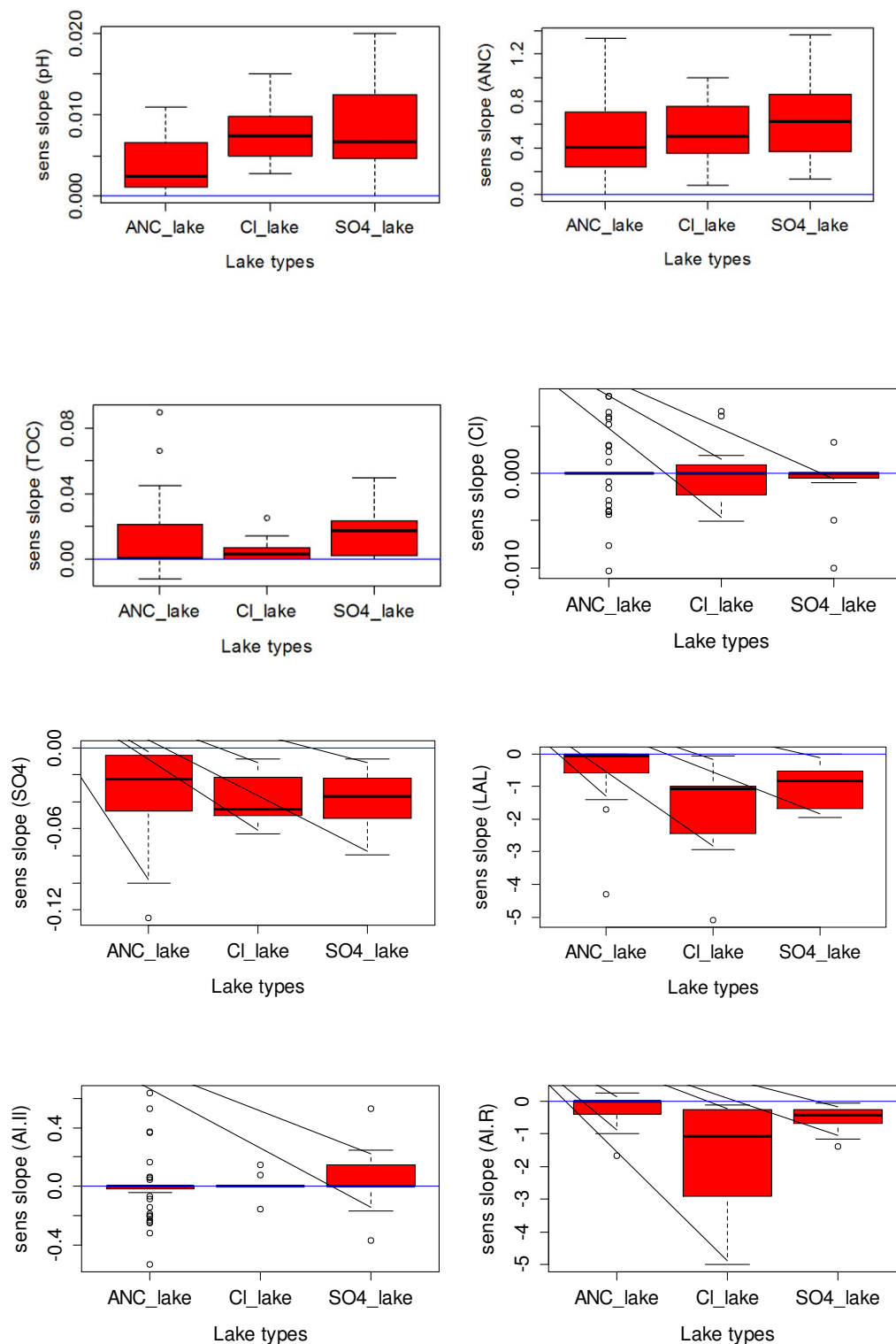


Figure 29: Sens slope of TOC, SO<sub>4</sub> and Al.II in different microhabitats as % change per year.

## 9.2 Appendix B: Supplementary tables

**Table 11: Metadata of the studied lakes, with NVE identification numbers, state, municipality number, Region, Station code, Station, or lake name, coordinates, elevation (m.a.s.l.), lake area (km<sup>2</sup>) and catchment area (km<sup>2</sup>), and Lake type.**

NVE no.	State	Municipality no.	Municipality no.	Region	Station code	Station Name	Latitude	Longitude	elevation	Lake area	Catchment area	Lake type
282	Hedmark	Åmot	429	1	429-601	Holmsjøen	61.15	11.62	559	1.15	5.9	ANC_lake
331	Østfold	Halden	101	2	101-605	Holvatn?	59.11	11.53	161	1.15	9.35	SO4_lake
5844	Østfold	Sarpsborg	105	2	105-501	Isebakkjern	59.34	10.97	60	0.3	6.6?	ANC_lake
5828	Østfold	Våler	137	2	137-501	Ravnsjøen	59.41	11	82	0.3	2.85	ANC_lake
3238	Akershus	Aurskog-Høland	221	2	221-605	Store_Lyseren	59.78	11.77	229	0.51	3.37	ANC_lake
3259	Akershus	Aurskog-Høland	221	2	221-607	Holvatn	59.74	11.58	214	0.42	4.95	ANC_lake
5114	Oslo	Oslo	301	2	301-605	Langvatn	60.11	10.77	342	0.56	3.57	ANC_lake
368	Hedmark	Kongsvinger	402	2	402-604	Storbørja	60.09	11.93	301	1.15	29.2	ANC_lake
3838	Hedmark	Nord-Odal	418	2	418-603	Skurvsjøen	60.57	11.65	432	0.43	20.7	ANC_lake
281	Hedmark	Grue	423	2	423-601	Meitsjøen	60.39	11.81	358	1.02	20.35	ANC_lake
7272	Buskerud	Flå	615	2	615-604	Langtjern(LAE01)	60.37	9.73	0	0	4.8	ANC_lake
5269	Buskerud	Modum	623	2	623-603	Breidlivatnet	59.98	10.15	632	0.3	1.54	SO4_lake
5961	Buskerud	Flesberg	631	2	631-607	Skakktjern	59.89	9.31	547	0.08	4.6	ANC_lake
5742	Vestfold	Sande	713	2	713-601	Øyvannet(Store)	59.64	10.1	442	0.33	5.53	ANC_lake
14367	Telemark	Nome	819	2	819-501	Nedre_Furovatn	59.28	8.84	605	0.1	2.7	ANC_lake
69	Telemark	Hjartdal	827	3	827-601	Heddersvatnet	59.83	8.76	1136	1.83	11.65	ANC_lake
13194	Telemark	Vinje	834	3	834-614	Stavsvatn	59.64	8.11	1053	0.4	2.43	ANC_lake
14277	Telemark	Fyresdal	831	4	831-501	Brårvatn	59.29	7.73	902	1.25?	4	ANC_lake
1094	Telemark	Tokke	833	4	833-603	Skurevatn	59.59	7.55	1269	1.08	7.75	SO4_lake
9534	Aust-Agder	Tvedestrand	914	4	914-501	Sandvatn	58.69	8.96	150	0.32	2.75	ANC_lake
10127	Aust-Agder	Froland	919	4	919-606	Hundevatn	58.59	8.54	286	0.32	2.3	SO4_lake
10926	Aust-Agder	Iveland	935	4	935-7?938	Grunnevatn	58.39	7.97	250	1	3.3	SO4_lake
9219	Aust-Agder	Bygland	938	4	938-66	Grimsdalsvatn	58.75	7.97	463	0.31	8.3	SO4_lake
15177	Aust-Agder	Valle	940	4	940-502	Myklevatn	59.07	7.38	785	0.6	32.7	ANC_lake
14534	Aust-Agder	Valle	940	4	940-527	Skammevatn	59.21	7.24	1074	0.68	8.4	ANC_lake
15100	Aust-Agder	Valle	940	4	940-501	Tjurrmovatn	59.07	7.46	720	0.75	6.8	ANC_lake
13592	Aust-Agder	Bykle	941	4	941-24	Bånevatn	59.5	7.11	1115	1.46	16.9	SO4_lake

**Table 10 continued**

11078	Vest-Agder	Vennesla	1014	4	1014-12	Songevatn	58.32	7.68	268	0.25	9.3	ANC_lake
11147	Vest-Agder	Vennesla	1014	4	1014-25	Drivenesvatn	58.29	7.93	168	0.22	11.5	ANC_lake
11592	Vest-Agder	Søgne	1018	4	1018-4	Kleivsetvatn	58.11	7.68	83	0.57	17.2	ANC_lake
11373	Vest-Agder	Marnardal	1021	4	1021-14	Homestadvatn	58.21	7.45	278	0.62	3?	Cl_lake
1431	Vest-Agder	Flekkefjord	1004	5	1004-13	St.Eitlandsvt	58.49	6.74	392	1.15	6.3	Cl_lake
21797	Vest-Agder	Flekkefjord	1004	5	1004-15	Botnevatn	58.28	6.48	56	0.6	8	Cl_lake
1174	Vest-Agder	Åseral	1026	5	1026-210	Stigebottsvatn	58.76	7.31	814	0.93	7.3?	SO4_lake
11292	Vest-Agder	Lyngdal	1032	5	1032-14	Troldevatn	58.23	6.99	278	0.22	1	Cl_lake
10305	Vest-Agder	Hægebostad	1034	5	1034-8	Trollselvatn	58.55	7.21	617	0.25	3.5	ANC_lake
11095	Vest-Agder	Hægebostad	1034	5	1034-19	I.Espelandsvatn	58.3	7.16	391	0.28	10	ANC_lake
1373	Vest-Agder	Kvinesdal	1037	5	1037-17	Heievatn	58.63	6.97	500	0.31	12.5	SO4_lake
21186	Rogaland	Eigersund	1101	5	1101-43	Glypstadvatn	58.49	6.2	261	0.34	2?	Cl_lake
21438	Rogaland	Sokndal	1111	5	1111-3	Ljosvatn	58.42	6.21	150	0.22	1.1	Cl_lake
21049	Rogaland	Lund	1112	5	1112-15	Gjuvvatn	58.52	6.41	389	0.35	2.4	Cl_lake
1545	Rogaland	Hå	1119	5	1119-602	Homsevatn	58.56	5.86	142	0.67	8.7	Cl_lake
22548	Rogaland	Vindafjord	1154	6	1154-601	Røyrvatn	59.54	6.02	230	0.42	16.3	Cl_lake
23386	Hordaland	Etne?	1211	6	1211-601	Vaulavatn	59.83	6.37	879	1.12	25.75	ANC_lake
22101	Hordaland	Fitjar	1222	6	1222-502	Ø.Steindalsvatn	59.87	5.42	262	0.25	3.3	ANC_lake
26511	Hordaland	Samnanger	1242	7	1251-601	Oddmundalsvatn	60.53	5.98	760	0.32	5.72	Cl_lake
26267	Hordaland?	Lindås?	1263	7	1263-601	Båtevatn	60.73	5.51	451	0.42	2.77	Cl_lake
28197	Sogn_og_Fjordane	Flora	1401	7	1401-501	Langevatn	61.67	5.18	470	0.67	2.67	Cl_lake
1651	Sogn_og_Fjordane	Balestrand	1418	7	1418-601	Nystølsvatn	61.34	6.46	715	1.25	21.45	SO4_lake
1935	Sogn_og_Fjordane	Eid	1443	7	1443-501	Movatn	61.98	6.18	422	1.05	20	ANC_lake
34660	Oppland	Lesja	512	8	512-601	Svartdalsvatnet	62.27	8.84	1018	0.6	49.9	ANC_lake
31186	Møre_og_Romsdal	?Molde	1502	8	1502-602	Lundalsvatnet	62.82	7.53	254	0.3	5.65	ANC_lake
31047	Møre_og_Romsdal	Vanylven	1511	8	1511-601	Blæjevatnet	62.05	5.78	700	0.55	1.93	ANC_lake
36436	Møre_og_Romsdal	Aure?	1569	8	1569-601	Skardvatnet	63.3	8.78	346	0.52	3.75	ANC_lake
36727	Sør-Trøndelag	Åfjord?	1630	8	1630-603	Skjerivatnet	63.96	10.56	357	0.88	3.25	ANC_lake
36780	Sør-Trøndelag	Åfjord?	1630	8	1630-601	Grovlivatnet	63.91	10.16	180	1.03	10.4	ANC_lake
35326	Sør-Trøndelag	Røros	1640	8	1640-603	Tufsingn	62.61	11.88	781	1.38	5.15	ANC_lake



**Table 10 continued**

40844	Nord-Trøndelag	Namdalseid	1725	8	1725-3-14	Bjørfarvatnet	64.28	10.99	263	1.01	3.8	ANC_lake
716	Nord-Trøndelag	Namsskogan	1740	8	1740-602	Storgåsvatnet	65.06	13.17	493	2.77	10.85	ANC_lake
40322	Nord-Trøndelag	Grong	1742	8	1742-501	Grytsjøen	64.39	12.09	372	0.45	10	ANC_lake
806	Nordland	Saltdal	1840	9	1840-601	Kjemåvatn	66.77	15.41	626	2.6	33	ANC_lake
45724	Nordland	Sørfold	1845	9	1845-601	Tennvatn	67.76	15.93	339	2.62	5.18	ANC_lake
1001	Nordland	Tysfjord	1850	9	1850-603	Kjerrvatn	68.08	16.03	209	1.4	6.62	ANC_lake
48048	Nordland	Flakstad	1859	9	1859-601	Storvatn	68.05	13.35	25	1.1	6.2	ANC_lake
50879	Troms	Tranøy	1927	9	1927-501	Kapervann	69.24	17.33	214	0.67	18	ANC_lake
2430	Eastern Finnmark	Vardø	2002	10	2002-501	Oksevatn	70.35	30.88	143	2.73	9.9	ANC_lake
2437	Eastern Finnmark	Sør-Varanger	2030	10	2030-503	Skaidejavri	69.93	29.11	322	1.85	7.3	ANC_lake
2456	Eastern Finnmark	Sør-Varanger	2030	10	2030-619	Følvatnet	69.25	28.93	177	2.57	11.8	ANC_lake
2474	Eastern Finnmark	Sør-Varanger	2030	10	2030-607	StValvatnet	69.72	30.66	157	3.6	19.58	ANC_lake
63664	Eastern Finnmark	Sør-Varanger	2030	10	2030-504	Råtjern	69.88	29.19	264	0.7	2.47	ANC_lake
64193	Eastern Finnmark	Sør-Varanger	2030	10	2030-614	Langvatnet	69.73	30.19	90	0.32	3	ANC_lake
64217	Eastern Finnmark	Sør-Varanger	2030	10	2030-612	L.Djupvatnet	69.71	30.59	211	0.4	1.98	SO4_lake
64278	Eastern Finnmark	Sør-Varanger	2030	10	2030-625	Holmvatnet	69.71	29.72	146	0.92	3.07	ANC_lake
64684	Eastern Finnmark	Sør-Varanger	2030	10	2030-501	Bårjasjavri	69.56	29.81	150	0.45	7.25	ANC_lake
64713	Eastern Finnmark	Sør-Varanger	2030	10	2030-603	Otervatnet	69.55	30.78	293	0.18	1.48	ANC_lake
64799	Eastern Finnmark	Sør-Varanger	2030	10	2030-624	Ulekristajav	69.53	29.45	242	0.17	1.2	ANC_lake

Table 12: measurement data of different parameters for the different lakes (given as NVE ID number).

NVE no.	pH	KOND mS/m	ALK mmol/l	Ca mg/l	Mg mg/l	K mg/l	Na mg/l	Cl mg/l	SO4 mg/l	NO3-N µg/l N	TOTP µg/l P	PO4-P µg/l P	TOTN µg/l N	NH4 µg/l N	TOC mg/l C	Al/R µg/l	Al/II µg/l	LAL µg/l
282	5.335	1.21	0.04	0.975	0.15	0.17	0.56	0.5	1.7	5.5	8	2	240	5	6.6	47	41	3
331	4.965	4.62	0.026	1.01	0.69	0.36	4.325	6.99	5.15	145.5	4	1	405	41	5.25	190	68.5	108
5844	5.145	5.145	0.05	1.99	0.905	0.65	4.855	7.24	4.61	71	14	2	550	56	17.4	349	237	90
5828	5.395	3.905	0.041	1.205	0.66	0.345	3.96	6.05	4.75	84.5	5	1	310	35	4.6	180	84.5	63
3238	5.52	2.305	0.0405	1.095	0.42	0.34	1.64	2.29	3.9	89	3	1	273	5	3.6	155	48	93
3259	5.505	2.38	0.047	1.305	0.48	0.4	1.76	2.6	3.1	69.5	5	2	355	15	8.3	153	120	28
5114	5.81	1.54	0.046	1.035	0.245	0.195	1.07	1.16	2.55	52	4	1	235	8	3.7	83	41.5	25
368	5.24	1.87	0.0405	1.08	0.39	0.22	1.145	1.25	2.15	48	7	1	325	25	11	144	120	24
3838	4.735	1.895	0.0165	0.66	0.22	0.17	0.865	0.81	1.65	22	7	1	300	6	12.6	221	150	57
281	4.98	1.855	0.031	1.095	0.36	0.235	0.96	1.04	1.8	36.5	8	1	350	12	13.4	171	139	28
7272	4.905	1.465	0.028	1.025	0.155	0.08	0.565	0.49	1.45	12	5.5	1	285	2	11.6	186	145	48
5269	4.985	1.175	0.029	0.385	0.13	0.12	0.595	0.6	1.65	28	9.5	1	317	44	5.95	239	92	133
5961	4.76	1.46	0.017	0.76	0.15	0.08	0.43	0.48	0.9	11	6	1	262.5	2	10.8	135	112	22
5742	5.65	1.78	0.047	1.26	0.3	0.25	1.16	1.4	2	77	8	2	350	22	7.9	122	105	15
14367	4.94	1.52	0.027	0.855	0.175	0.115	0.57	0.81	1.35	30	5	1	310	9	9.05	175	120	35
69	5.915	0.805	0.0415	0.635	0.11	0.13	0.35	0.41	1.25	97	2	1	160	4	0.81	15	10	3
13194	6.085	0.865	0.0505	0.82	0.11	0.075	0.44	0.41	1.15	29	2	1	128	4	1.47	65	34	30
14277	5.78	0.89	0.0365	0.49	0.12	0.1	0.59	0.81	1.16	105	2	1	192.5	7	0.89	39	11	18
1094	5.61	0.7	0.035	0.35	0.09	0.05	0.39	0.53	1	101.5	1	1	147	1	0.33	35	5.5	11
9534	4.91	2.785	0.0265	0.8	0.415	0.245	2.34	3.78	2.69	85.5	5	2	400	22	7.05	198	113	67
10127	4.905	2.375	0.024	0.63	0.37	0.24	1.73	2.76	2.8	146	3	1	385	31	4.4	162	59.5	67
10926	5.03	2.545	0.03	0.725	0.355	0.19	2.185	3.65	2.555	76.5	3	1	305	21	4.69	188	93	73
9219	4.89	1.43	0.0235	0.33	0.115	0.09	0.815	1.25	1.4	43	3.5	1	250	16	4.15	165	64.5	86
15177	5.435	0.86	0.0355	0.42	0.105	0.06	0.58	0.8	1.005	32	4	1	160	4	2.5	62	34	26
14534	5.81	0.66	0.0375	0.34	0.07	0.05	0.53	0.7	0.8	43	2	1	91.5	4	0.44	22	9	10
15100	5.415	0.885	0.034	0.32	0.11	0.06	0.67	1	1.05	25	3	1	175	10	2.25	72	32.5	34
13592	5.53	0.75	0.032	0.245	0.085	0.07	0.525	0.88	0.8	86	1	1	117	2	0.26	16	5	5

**Table 11 Cont.**

11078	5.545	2.895	0.047	1.225	0.455	0.63	2.62	4.19	2.4	103	22	11	440	50	7.6	158	119	28
11147	5.005	3.03	0.03	0.97	0.395	0.33	2.775	4.39	3.07	130	17	8	430	24	5.65	184	93	56
11592	5.135	3.765	0.035	1.195	0.555	0.37	3.785	6.46	3.65	212.5	5	1	445	26	4.7	199	109	62
11373	4.81	3.42	0.0195	0.52	0.38	0.19	3.22	5.46	2.6	195	4.5	2	380	36	2.7	174	39	122
1431	5.06	2.66	0.025	0.41	0.35	0.17	2.74	4.8	2.2	165	3	1	288	26	0.95	101	11.5	56
21797	5.07	4.9	0.0275	0.79	0.705	0.31	5.435	9.5	3.5	295	6	2	410	22	1.3	173	24	99
1174	4.94	1.19	0.023	0.24	0.11	0.06	0.7	1.1	1	76	5	1	236	3	2.4	77	32	35
11292	4.62	3.365	0.012	0.305	0.34	0.17	2.925	5.07	2.3	300	3	1	470	23	2.45	148	36.5	109
10305	4.66	2.015	0.0125	0.355	0.19	0.07	1.255	1.83	1.185	54	8.5	1	347.5	10	8.4	136	95	33
11095	4.915	2.385	0.027	0.565	0.255	0.15	2.245	3.21	2.125	81.5	7	1	325	19	5.15	166	89.5	64
1373	4.765	2.085	0.0185	0.37	0.19	0.1	1.555	2.5	1.545	60.5	6	1	285	8	5.6	160	102	46
21186	5.32	4.305	0.033	1.055	0.725	0.425	4.505	8.05	3.2	425	3	1	550	16	0.79	64	10	10
21438	4.905	4.025	0.022	0.485	0.52	0.2	4.285	7.55	2.8	332.5	3	1	390	24	0.67	186	10	133
21049	4.925	2.82	0.022	0.33	0.37	0.14	2.84	5.01	2.15	195	2	1	295	24	0.89	148	18	114
1545	4.925	4.385	0.022	0.555	0.59	0.22	4.62	8.5	3	302.5	3	1	445	47	1.1	159	17	111
22548	5.25	1.835	0.031	0.41	0.275	0.12	1.8	3.02	1.39	106	2	1	185	10	1.3	58	26.5	25
23386	5.74	0.91	0.034	0.31	0.12	0.12	0.8	1.24	0.8	82	2	1	128	1	0.4	12	10	3
22101	5.53	2.62	0.037	0.685	0.365	0.16	2.865	4.95	1.725	90.5	2	1	205	8	2.3	67	46	17
26511	5.28	0.975	0.029	0.16	0.11	0.05	0.81	1.39	0.6	86	2	1	132	4	0.36	15	7.5	9
26267	5.08	1.805	0.026	0.18	0.22	0.1	1.715	2.9	1.115	107	2	1	165	2	0.59	50	11	27
28197	5.455	1.98	0.031	0.41	0.29	0.09	2.2	3.88	1.115	126	3	1	190	4	0.7	19	10	7
1651	5.76	0.725	0.034	0.245	0.08	0.07	0.61	0.98	0.62	53.5	2	1	89	4	0.34	16	7	5
1935	5.905	1.15	0.042	0.345	0.17	0.12	1.255	2.04	0.8	16.5	2	1	93	7	1.3	28	20.5	5
34660	6.205	0.6	0.0475	0.49	0.06	0.16	0.3	0.22	0.89	30	1.5	1	74	3	0.36	7	6	0.4
31186	6.28	2.38	0.063	0.72	0.43	0.22	2.9	4.5	1.05	5	3	1.5	136	9	3.4	42	37	3
31047	6.09	1.855	0.0435	0.62	0.26	0.15	2.01	3.43	1.505	30.5	1.5	1	71	2	0.35	9.5	7.5	2
36436	5.93	2.2	0.0415	0.4	0.35	0.15	2.775	4.63	1.1	13	2	1	95	17	1.65	26	21	4
36727	5.96	2.9	0.041	0.52	0.455	0.165	3.625	6.23	1.45	27.5	2	1	104	9	1.1	16	13	3
36780	5.535	3.735	0.036	0.52	0.61	0.22	4.87	8.43	1.7	25.5	2	1	141	16	3	63	49.5	12
35326	6.43	1.065	0.0675	0.69	0.24	0.23	0.81	0.73	1.1	30	2	1	122	63	1.82	14	11	3

<b>Table</b>	<b>11</b>	<b>Cont.</b>																	
40844	5.595	3.43	0.037	0.5	0.61	0.185	4.55	7.8	1.575	29	3	1	131	10	2.9	41	34	7	
716	5.96	1.55	0.0395	0.33	0.23	0.1	1.835	3.19	0.72	17.5	2	1	89	10	1.1	22	15.5	5	
40322	5.64	1.62	0.04	0.52	0.27	0.06	1.73	2.64	0.69	1	4	1	163.5	2	4.74	67	59	6	
806	6.165	1.095	0.05	0.455	0.14	0.12	1.125	1.58	0.8	27	2	1	77	10	0.65	13	10	4	
45724	6.115	1.715	0.049	0.48	0.26	0.37	1.875	3.1	0.9	16	3	1	104	2	1.84	25	21	3	
1001	6.08	2.72	0.049	0.63	0.415	0.39	3.295	5.58	1.265	12.5	2	1	105	3	2.2	45	33	7	
48048	5.91	5.11	0.041	0.63	0.89	0.285	6.545	11.6	2.45	37.5	2	1	101	11	0.95	15	12	2	
50879	6.18	1.54	0.048	0.33	0.21	0.13	1.95	2.9	1.1	1	2	1	53	6	0.92	15	12	3	
2430	6.28	4.69	0.055	0.8	0.96	0.28	5.79	10.3	2.8	12	2	1	101.5	2	1.1	8	5	1	
2437	5.975	1.825	0.04	0.66	0.33	0.12	1.77	3.03	1.96	25	1	1	72	14	0.73	10	9	2	
2456	6.54	1.78	0.0735	1.28	0.39	0.23	1.18	1.4	2.77	3	3	1	118	1	2.1	8	5.5	1.9	
2474	6.305	3.215	0.0545	1.33	0.725	0.285	3	4.9	4.42	24	3	1	89	2	1.1	10	8.5	2	
63664	5.995	1.9	0.041	0.72	0.355	0.15	1.84	3.18	2.2	6	2	1	65	2	0.91	10	7.5	1.5	
64193	6.33	3.385	0.057	1.32	0.755	0.235	3.265	5.45	4.75	8	3	1	117	8	2.4	28	19.5	7	
64217	5.53	3.09	0.033	1	0.625	0.21	2.925	4.99	4.46	1.5	3	1	51	4	0.62	18	8.5	4	
64278	6.34	2.6	0.0565	1.21	0.51	0.215	2.41	4.05	3.05	9	1	1	94	4	1.4	12	9	4	
64684	6.41	2.16	0.065	1.16	0.44	0.2	1.81	2.9	2.4	1	3	1	116.5	2	2.1	10	8	2.1	
64713	6.325	2.885	0.0595	1.39	0.775	0.21	2.22	3.16	5	1	6	2	139	4	2.4	10	8.5	1	
64799	6.35	1.67	0.056	1.04	0.33	0.19	1.28	1.95	2.3	1	2	1	91.5	11	1.7	12	10	4	

Table 13: measured parameters for all lakes converted into sea-salt corrected concentrations (blue and marked with an \*), and converted to equivalents (red)

NVE	Mg*	Na*	K*	SO4*	H+	Ca	Mg	K	Na	Cl	SO4	NO3	Al/R	ANC	TOC	ANCOAA	Ionic budget
no.	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	%
282	9	12	4	34	5	48.5	12	4	24.5	14	35	0	5	37	22.5	13.5	48
331	17.5	20	5.5	87	11	50	57	9	188	197	108	10.5	20	-11	17.5	-26	0
5844	37.5	37.5	13.5	80	7	99	74.5	17	211	204	96	5	36	75	59	22	25
5828	20	25.5	6	82.5	4	60	54	9	173	171	99	6	19	21	16	6	8
3238	22	17	8	74.5	3	54.5	34.5	9	71.5	65	81	6	16	15	12	2	12
3259	25	15	9	56.5	3	65	39	10	77	73	64.5	5	16	44	28.5	17.5	27
5114	13	18	4	50	2	51.5	20.5	5	46.5	33	53	4	8.5	33	13	20.5	28
368	25	19	5	41.5	6	53.5	32	6	49.5	35.5	45	3	15	52	37	18.5	43
3838	14	18	4	32	18.5	33	18	4	37.5	23	34	2	23	32	42.5	-14.5	47
281	24	17	5.5	34.5	10	54.5	30	6	42	29	37	3	18	57	46	8.5	52
7272	10.5	12	2	29	12.5	51	12.5	2	24.5	14	30	1	19	46	39	3	56
5269	7.5	11	3	33	10.5	19	11	3	26	17	34	2	25	2	20	-17	24
5961	9	7	2	17	17	38	12	2	19	14	19	1	14	33	37	-7	61
5742	17	17	6	38	2	63	25	6	50	39	42	6	13	41	27	18	40
14367	10.5	5	3	25.5	11.5	42.5	14.5	3	25	23	28	2	18	28	31	-4	45
69	6	4	3	25	1	31.5	9	3	15	11.5	26	7	1.5	12	3	8.5	25
13194	6	8.5	2	23	1	41	9	2	19	11.5	24	2	7	26	5	21	48
14277	5	6	2	22	2	24	10	3	26	23	24	8	4	6	3	2	15
1094	4	4	1	18.5	2.5	17	7	1	17	15	21	7.5	3.5	-1.5	1	-5	2
9534	14	12	4	46.5	12	40	34.5	6	102	107	56	6	20.5	10	24	-16.5	13
10127	16	7.5	5	50.5	12.5	31.5	30.5	6	75.5	78	58	10.5	16.5	-8	15	-25	6
10926	7.5	5.5	3	44.5	9.5	36	29.5	5	95	103	53	5.5	19.5	-1	16	-16	8
9219	3	5	2	26	13	16	9.5	2	35.5	35.5	29	3	17	-3	14	-19	11
15177	4	5.5	1	19	4	21	8.5	2	25	23	21	2.5	6	10	9	1.5	23
14534	2	5	1	15	2	17	6	1	23	20	17	3	2	7	1	6	18
15100	3	5	1	19	4	16	9	2	29	28	22	2	7	6	7.5	-3	13
13592	2	2	1	13	3	12	7	2	23	24.5	17	6	2	-1	1	-2	-1

Table 12 Cont.

11078	13.5	11	14	38	3	61	37.5	16	114	118	50	7.5	16.5	48	26	23	24
11147	8	13	6	52	10	48	32.5	8	121	124	64	9	19	19	19.5	-3.5	10
11592	10	11	6	57	7	59.5	45.5	9	165	182	76	15	21	11	16	-8	4
11373	1	7	2	37	15	26	31	5	140	154	54	14	18	-29	9	-38	-2
1431	2	4	2	30	9	20	29	4	119	135	46	12	10	-18	3	-21	-7
21797	3	6.5	3.5	45.5	8.5	39	58	8	237	268	73	21	18	-29	4	-32.5	-3
1174	2	4	1	18	11	12	9	2	30	31	21	5	8	-6	8	-13	11
11292	0	7	2	34	24	15	28	4	127	143	48	21	15.5	-34	8.5	-42	-7
10305	5	10.5	1	20.5	22	17.5	16	2	54.5	51.5	24.5	4	14	9.5	29	-19	29
11095	2	15	2	36	12	28	21	4	97.5	90.5	44.5	5.5	17	6.5	17.5	-13	14
1373	2	10.5	1	25.5	17	18	16	2.5	67.5	71	32	4.5	16.5	-1	19	-17.5	11
21186	15	-1	7	41.5	5	52.5	59.5	11	196	227	67	30.5	7	-16	3	-18.5	0
21438	1	4	1	37	12.5	24	43	5	187	213	58	24	19	-33	2	-35	-9
21049	3	3.5	1	29.5	12	16	30	4	124	142	45	14	15.5	-25	3	-29.5	-8
1545	3	9.5	1	40	12	27.5	49	6	201	240	62	21.5	16.5	-29	4	-31.5	-9
22548	5	3	2	19	6	20	22.5	3	78.5	85	29	7.5	6	1.5	4	-3.5	7
23386	3	4	2	13	2	15	10	3	35	35	17	6	1	7	1	4.5	11
22101	3	7.5	2	21	3	34	30	4	125	140	35.5	6.5	7	11	8	2.5	7
26511	1	1.5	1	9	5	8	9	1	35.5	39	12	6	1.5	-4	1	-5	3
26267	2	3	1	15	8.5	9	18	3	74.5	82	23.5	8	5.5	-8	2	-10	0
28197	2	1.5	0	13	3.5	20	24	2	95.5	110	23.5	9	2	-0.5	2	-2.5	2
1651	1	3	1	10	2	12	7	2	27	27.5	13	4	2	2.5	1	2	11
1935	3	8	2	10.5	1	17	14	3	54.5	57.5	17	1	3	17	4	13	16
34660	4	7	4	18	1	24	5	4	13	6	18.5	2	1	16	1	15	44
31186	10	12	3	9	1	36	35	6	126	127	22	0	4	56	12	44	27
31047	2	4.5	2	21	1	31	21	4	87	96.5	31	2	1	14	1	13	10
36436	4	6	1.5	9	1	20	29	4	121	131	23	1	3	17	5.5	12	11
36727	3.5	5	1	12	1	26	37.5	4	158	176	30	2	2	16	4	11	8
36780	2.5	6	1	10.5	3	26	50	6	212	238	35	2	6.5	12	10	3	8
35326	16	17.5	6	21	0	34	20	6	35	20.5	23	2	1	51	6	45	52

Table 12 Cont.

40844	5	1.5	1	9.5	3	25	50	5	198	220	32.5	2	4	17	10	6.5	9
716	2	2	1	6.5	1	16	19	3	80	90	15	1	2	13	4	9	11
40322	6	10	0	7	2	26	22	2	75	74	14	0	7	34	16	18	31
806	2	11.5	2	11	1	22.5	12	3	49	44.5	17	2	1	23	2	20.5	27
45724	3	6	8	10.5	1	23.5	21	9	81.5	87	19	1	2.5	27	6	19	21
1001	2.5	6.5	7	10	1	31	34	10	144	158	26.5	1	5	29	7	21.5	16
48048	8	0.5	1	16.5	1	31	73	7	285	327	51	3	2	11	3	7.5	4
50879	2	16	2	14	1	16	17	3	85	82	23	0	2	20	3	17	14
2430	21	2	2	27	1	40	79	7	252	291	58	1	1	23	4	18	8
2437	10.5	4	1.5	31.5	1	33	27	3	77	85.5	41	2	1	11	2	8.5	9
2456	24	18	5	53.5	0	64	32	6	51.5	39	57.5	0	1	52	7	43	37
2474	32.5	12.5	5	78.5	0.5	66.5	59.5	7	131	138	92	2	1	27	4	23	12
63664	11	4.5	2	36.5	1	36	29	4	80	89.5	46	0.5	1	12	3	8.5	9
64193	31.5	11	3.5	84.5	0	65.5	62.5	6	142	154	99	1	3	37	8	30.5	8
64217	24	7	3	78	3	50	51.5	5	127	141	93	0	2	-4.5	2	-7	1
64278	19	9.5	3.5	52	0	60.5	42	5.5	105	114	63.5	1	1	39	5	33.5	16
64684	19	10	4	42	0	58	36	5	79	82	50	0	1	43	7	33	26
64713	46	20.5	4	93.5	0	69	63.5	5	97	89	104	0	1	39	8	25	18
64799	16	11	4	42	0	52	27	5	56	55	48	0	1	35	6	29	26

Table 14: Tau values (correlation coefficients) of the Mann-Kendall statistics. The 30% of most decrease are marked in red and the 30% of most increase in green. The units indicate, which units were used for the statistical tests

NVE no.	pH	KOND mS m <sup>-1</sup>	ALK mmol l <sup>-1</sup>	Ca mg l <sup>-1</sup>	Mg mg l <sup>-1</sup>	K mg l <sup>-1</sup>	Na mg l <sup>-1</sup>	Cl mg l <sup>-1</sup>	SO4 mg l <sup>-1</sup>	NO3N µg N l <sup>-1</sup>	TOTP µg P l <sup>-1</sup>	PO4-P µg P l <sup>-1</sup>	TOT-N µg N l <sup>-1</sup>	NH4 µg N l <sup>-1</sup>	TOC mg C l <sup>-1</sup>	Al.R µg l <sup>-1</sup>	Al.II µg l <sup>-1</sup>	LAL µg l <sup>-1</sup>
1431	0.6	-0.56	0.12	-0.44	-0.42	-0.22	-0.27	-0.35	-0.66	-0.56	0.02	NA	-0.48	NA	0.59	-0.64	0.46	-0.12
2179	0.6																	
7	5	-0.28	0.27	-0.36	-0.24	-0.24	-0.12	-0.16	-0.75	-0.48	0.03	NA	-0.29	NA	0.63	-0.70	0.63	-0.58
1107	0.2																	
8	4	-0.16	0.17	-0.06	-0.10	0.15	-0.07	-0.05	-0.37	-0.07	0.07	NA	0.15	NA	0.09	-0.24	-0.10	-0.20
1114	0.5																	
7	7	-0.50	0.06	-0.33	-0.34	-0.36	-0.19	-0.26	-0.65	-0.41	0.01	NA	-0.05	NA	0.57	-0.41	0.30	-0.54
331	0.4																	
2	2	-0.58	-0.02	-0.50	-0.51	-0.18	-0.19	-0.23	-0.74	-0.24	0.14	NA	0.39	NA	0.42	-0.45	0.30	-0.66
1159	0.5																	
2	2	-0.43	0.19	-0.30	-0.28	-0.25	-0.17	-0.21	-0.67	-0.40	-0.01	NA	-0.07	NA	0.55	-0.50	0.37	-0.51
1137	0.6																	
3	0	-0.53	0.27	-0.42	-0.38	-0.40	-0.28	-0.30	-0.66	-0.50	0.03	NA	0.07	NA	0.46	-0.51	0.29	-0.36
1174	0.1																	
3	-0.15	0.06	-0.04	-0.02	-0.02	0.03	-0.05	-0.24	-0.13	NA	NA	-0.18	NA	0.13	-0.16	0.05	-0.31	
1129	0.6																	
2	4	-0.48	0.22	-0.54	-0.45	-0.18	-0.26	-0.31	-0.76	-0.52	0.09	NA	-0.38	NA	0.63	-0.67	0.39	-0.51
1109	0.5																	
5	1	-0.50	-0.02	-0.38	-0.42	-0.25	-0.21	-0.30	-0.71	-0.33	-0.01	NA	0.46	NA	0.61	-0.50	0.28	-0.63
1030	0.5																	
5	5	-0.54	-0.04	-0.22	-0.38	-0.22	-0.21	-0.34	-0.69	-0.29	-0.07	NA	0.00	NA	0.56	-0.20	0.07	-0.42
1373	0.5																	
5	-0.44	0.08	-0.25	-0.34	-0.17	-0.15	-0.19	-0.66	-0.17	0.03	NA	0.01	NA	0.27	-0.47	-0.02	-0.47	
5844	0.1																	
9	-0.39	-0.06	-0.44	-0.34	-0.04	-0.04	-0.10	-0.66	-0.14	0.13	NA	0.23	NA	0.47	-0.20	0.21	-0.42	
2118	0.7																	
6	-0.62	0.29	0.42	0.24	0.45	-0.23	-0.32	-0.78	0.08	-0.02	NA	0.17	NA	0.46	-0.67	0.04	-0.36	
2143	0.6																	
8	9	-0.50	0.40	-0.45	-0.26	-0.33	-0.09	-0.11	-0.73	-0.58	0.10	NA	-0.68	NA	0.40	-0.71	0.02	-0.22
2104	0.3																	
9	4	-0.24	0.20	-0.21	-0.12	0.10	-0.11	-0.14	-0.39	-0.23	0.01	NA	-0.21	NA	0.26	-0.37	0.19	-0.54



1545	0.5																		
	9	-0.40	0.25	-0.52	-0.24	-0.08	-0.18	-0.20	-0.73	-0.46	0.04	NA	-0.48	NA	0.42	-0.68	0.37	-0.22	
2254	0.6																		
	8	2	-0.45	0.17	0.07	-0.23	0.04	-0.19	-0.21	-0.73	-0.45	0.21	NA	-0.08	NA	0.24	-0.63	0.00	-0.62
	0.2																		
3555	5	-0.48	-0.36	-0.54	-0.50	-0.53	-0.17	-0.24	-0.70	-0.58	-0.13	NA	-0.06	NA	0.58	-0.45	0.49	-0.61	
2338	0.5																		
	6	5	-0.41	0.21	0.26	0.01	-0.39	0.02	-0.06	-0.58	-0.52	0.00	NA	-0.21	NA	0.26	-0.38	-0.31	-0.01
2210	0.5																		
	1	6	-0.27	-0.02	0.02	-0.01	0.23	-0.12	-0.07	-0.71	-0.10	-0.11	NA	0.13	NA	0.26	-0.60	0.07	-0.56
2332	0.5																		
	8	9	-0.39	0.01	-0.01	-0.03	-0.16	-0.02	-0.03	-0.64	-0.53	0.20	NA	-0.30	NA	0.16	-0.43	-0.42	NA
2651	0.7																		
	1	0	-0.30	-0.28	0.06	0.08	-0.13	0.10	0.07	-0.65	-0.44	0.11	NA	-0.38	NA	-0.41	-0.54	-0.43	NA
2626	0.6																		
	7	5	-0.55	0.31	-0.01	-0.23	-0.29	-0.23	-0.26	-0.74	-0.36	0.09	NA	-0.08	NA	0.27	-0.53	0.20	-0.42
	0.3																		
5828	7	-0.50	-0.06	-0.53	-0.50	-0.48	-0.02	-0.09	-0.78	-0.38	0.16	NA	0.53	NA	0.59	-0.27	0.45	-0.56	
<b>Table 13 Cont.</b>																			
2819	0.3																		
	7	3	-0.12	-0.04	0.00	0.02	-0.08	-0.02	-0.05	-0.34	-0.15	0.02	NA	-0.26	NA	0.04	-0.20	-0.10	-0.23
	0.6																		
1651	3	-0.40	0.27	0.41	0.06	0.05	0.10	-0.10	-0.72	-0.51	0.11	NA	-0.17	NA	0.05	-0.47	-0.37	-0.24	
	0.4																		
1935	2	-0.11	0.12	0.27	-0.02	-0.03	0.04	-0.04	-0.58	-0.41	-0.05	NA	0.29	NA	0.17	-0.12	0.14	-0.09	
3118	0.1																		
	6	6	-0.06	0.01	0.15	0.03	-0.05	-0.04	-0.10	-0.35	0.01	-0.02	NA	0.09	NA	0.12	-0.09	0.03	-0.09
3104	0.2																		
	7	0	-0.35	0.24	0.45	-0.16	-0.18	-0.02	-0.17	-0.56	-0.56	0.07	NA	-0.53	NA	-0.18	-0.30	-0.48	0.12
3643	0.4																		
	6	2	-0.01	0.19	0.32	0.11	-0.22	-0.03	-0.01	-0.37	0.01	0.16	NA	0.32	NA	0.17	0.02	0.23	-0.30
3678	0.4																		
	0	4	-0.18	0.00	0.09	-0.04	-0.29	-0.12	-0.16	-0.59	-0.14	0.00	NA	0.08	NA	0.42	-0.26	0.04	-0.34
3672	0.5																		
	7	7	-0.02	0.13	0.26	0.19	-0.34	0.03	-0.01	-0.51	-0.23	-0.05	NA	-0.11	NA	-0.35	-0.29	-0.18	0.01
3532	0.3																		
	6	5	-0.22	0.44	0.43	0.15	0.11	0.30	0.19	-0.73	-0.57	0.06	NA	-0.34	NA	-0.12	-0.24	-0.17	0.09
4084	0.3																		
	4	5	-0.12	-0.15	0.24	0.15	-0.37	-0.08	-0.11	-0.36	-0.45	-0.02	NA	0.17	NA	0.41	-0.26	0.13	-0.24
	0.7																		
716	2	-0.11	0.41	0.32	-0.02	0.01	0.03	-0.08	-0.68	-0.50	0.15	NA	0.16	NA	0.45	-0.38	0.18	-0.18	

4032	0.1																		
2	7	-0.01	0.00	0.27	0.11	-0.20	0.06	0.03	-0.19	0.10	0.05	NA	0.00	NA	0.06	-0.05	0.04	-0.02	
	0.4																		
806	1	-0.24	0.21	0.19	-0.20	0.13	-0.08	-0.18	-0.33	-0.24	0.02	NA	0.21	NA	0.16	-0.12	-0.06	0.03	
4572	0.5																		
4	3	-0.27	0.33	0.16	-0.11	0.22	-0.06	-0.26	-0.67	-0.47	0.06	NA	0.12	NA	0.34	-0.12	-0.09	0.10	
	0.3																		
1001	5	-0.41	0.33	0.06	-0.24	0.00	-0.23	-0.31	-0.71	-0.27	0.11	NA	0.23	NA	0.15	-0.43	-0.10	-0.37	
4804	0.5																		
8	4	-0.11	0.43	0.22	0.10	0.15	0.06	-0.09	-0.45	-0.40	0.09	NA	0.20	NA	0.22	-0.06	0.09	0.03	
5087	0.3																		
9	6	-0.07	0.20	0.23	0.05	0.18	-0.01	-0.05	-0.35	-0.05	0.22	NA	0.31	NA	0.16	-0.05	0.11	0.04	
	0.2																		
2430	7	-0.21	0.38	0.12	0.05	-0.28	-0.06	-0.09	-0.59	-0.40	-0.02	NA	-0.06	NA	-0.22	-0.37	-0.49	NA	
6468	0.3																		
4	5	-0.09	0.43	0.23	-0.02	0.12	0.09	-0.12	-0.52	-0.29	0.10	NA	0.18	NA	0.19	-0.27	-0.39	-0.20	
	0.6																		
2437	2	-0.12	0.20	0.40	0.17	-0.22	0.49	0.25	-0.76	-0.48	-0.13	NA	-0.38	NA	-0.19	-0.28	-0.46	0.00	
6366	0.6																		
4	4	-0.23	0.19	0.19	0.03	0.08	0.47	0.24	-0.76	-0.56	-0.02	NA	0.12	NA	-0.06	-0.39	-0.55	0.10	
6471	0.4																		
3	0	-0.06	0.31	0.31	0.15	0.03	-0.05	-0.12	-0.34	-0.15	0.00	NA	-0.04	NA	-0.04	-0.37	-0.44	0.11	
	0.3																		
2474	4	-0.20	0.11	0.03	-0.28	0.09	0.14	0.10	-0.71	-0.36	-0.07	NA	0.08	NA	-0.48	-0.17	-0.62	-0.01	
6421	0.7																		
7	3	-0.72	0.19	-0.44	-0.55	-0.58	0.03	-0.10	-0.74	-0.53	-0.03	NA	0.15	NA	0.30	-0.62	-0.39	-0.03	
6419	0.3																		
3	3	-0.35	0.16	-0.20	-0.47	-0.17	0.00	-0.16	-0.45	-0.07	-0.01	NA	0.39	NA	0.34	0.08	0.27	-0.13	
	0.5																		
2456	3	-0.18	0.59	0.29	0.09	-0.23	0.46	0.27	-0.74	-0.33	-0.07	NA	0.05	NA	-0.27	-0.52	-0.61	NA	
6479	0.4																		
9	5	-0.26	0.39	0.07	-0.29	0.14	0.01	-0.16	-0.63	-0.18	0.02	NA	0.31	NA	0.11	-0.09	-0.19	-0.12	
6427	0.3																		
8	3	-0.22	0.28	0.22	0.03	-0.02	0.16	-0.04	-0.52	-0.27	-0.01	NA	0.11	NA	-0.07	-0.21	-0.44	-0.02	
	0.6																		
3238	9	-0.76	0.12	-0.43	-0.49	-0.31	0.12	-0.12	-0.80	-0.55	0.02	NA	0.10	NA	0.60	-0.68	0.40	-0.80	
	0.5																		
3259	0	-0.62	0.37	-0.52	-0.56	-0.40	-0.20	-0.17	-0.79	-0.44	-0.03	NA	0.18	NA	0.46	-0.25	0.11	-0.49	
<b>Table 13 Cont.</b>																			
	0.6																		
5114	3	-0.67	0.39	-0.37	-0.39	-0.31	-0.22	-0.36	-0.78	-0.17	0.10	NA	0.24	NA	0.41	-0.43	0.24	-0.48	

368	0.1	-0.56	-0.10	-0.27	-0.36	-0.45	0.06	-0.13	-0.75	-0.45	-0.10	NA	0.08	NA	0.37	-0.23	-0.02	-0.34
3838	0.2	-0.48	-0.14	-0.45	-0.48	-0.35	-0.07	-0.19	-0.76	-0.47	-0.08	NA	0.11	NA	0.36	-0.40	-0.08	-0.51
281	0.1	-0.59	-0.27	-0.42	-0.47	-0.37	-0.08	-0.30	-0.73	-0.48	0.06	NA	0.29	NA	0.45	-0.17	0.00	-0.40
282	0.3	-0.48	-0.08	0.23	-0.10	-0.07	0.15	-0.26	-0.72	-0.39	-0.07	NA	0.25	NA	0.33	-0.25	0.04	-0.28
3466	0.5	0.16	0.34	0.36	0.15	0.46	0.23	-0.16	-0.10	-0.17	0.04	NA	0.04	NA	0.08	-0.56	-0.49	NA
5269	0.3	-0.31	0.07	-0.12	-0.15	-0.12	-0.21	-0.29	-0.55	0.21	0.12	NA	0.20	NA	0.39	-0.28	0.20	-0.33
5961	0.1	-0.32	0.04	-0.30	-0.38	-0.20	-0.15	-0.43	-0.61	-0.32	-0.04	NA	-0.36	NA	0.10	-0.25	-0.18	-0.37
5742	0.2	-0.34	0.18	0.02	-0.13	0.02	-0.17	-0.21	-0.46	-0.13	0.12	NA	0.50	NA	0.34	-0.12	-0.04	-0.23
1436	0.1	-0.43	-0.13	-0.33	-0.35	-0.52	-0.05	-0.16	-0.74	-0.40	-0.09	NA	-0.26	NA	0.24	-0.33	-0.01	-0.29
69	0.5	-0.47	0.35	0.07	-0.31	-0.21	-0.31	-0.45	-0.61	-0.41	-0.01	NA	-0.31	NA	0.21	-0.35	-0.15	-0.14
1427	0.3	-0.48	0.09	-0.06	-0.31	-0.23	-0.31	-0.41	-0.50	-0.36	-0.08	NA	-0.11	NA	0.19	-0.37	0.21	-0.51
1094	0.7	-0.72	0.08	0.07	-0.48	-0.05	-0.12	-0.35	-0.75	-0.57	0.02	NA	-0.37	NA	0.14	-0.76	-0.42	NA
1319	0.5	-0.61	0.43	0.03	-0.52	-0.19	-0.24	-0.55	-0.82	-0.35	0.21	NA	-0.19	NA	0.29	-0.52	0.34	-0.69
9534	0.4	-0.44	0.01	-0.52	-0.48	-0.49	-0.13	-0.16	-0.70	-0.53	0.07	NA	-0.18	NA	0.49	-0.36	0.28	-0.39
1012	0.6	-0.74	0.22	-0.53	-0.50	-0.57	-0.31	-0.38	-0.77	-0.66	-0.06	NA	-0.66	NA	0.54	-0.65	0.21	-0.73
1092	0.4	-0.55	-0.12	-0.44	-0.37	-0.49	-0.20	-0.28	-0.74	-0.22	0.01	NA	0.18	NA	0.59	-0.35	0.44	-0.39
9219	0.6	-0.63	0.09	-0.54	-0.57	-0.41	-0.26	-0.41	-0.76	-0.53	0.07	NA	-0.02	NA	0.45	-0.42	0.30	-0.57
1510	0.6	-0.54	0.03	-0.13	-0.45	-0.32	-0.22	-0.30	-0.79	-0.33	0.07	NA	0.09	NA	0.41	-0.37	0.32	-0.40
1517	0.6	-0.58	0.18	-0.04	-0.44	-0.23	-0.12	-0.30	-0.71	-0.51	-0.04	NA	-0.13	NA	0.37	-0.45	0.01	-0.53
1453	0.3	-0.34	0.16	-0.02	-0.15	-0.14	-0.12	-0.15	-0.47	-0.31	0.02	NA	-0.14	NA	0.21	-0.31	-0.07	-0.36
1359	0.7	-0.62	0.07	0.15	-0.36	-0.46	-0.19	-0.23	-0.80	-0.54	-0.07	NA	-0.53	NA	-0.09	-0.61	-0.54	NA
2	2	-0.62	0.07	0.15	-0.36	-0.46	-0.19	-0.23	-0.80	-0.54	-0.07	NA	-0.53	NA	-0.09	-0.61	-0.54	NA

7272	0.3	-0.63	6	0.07	-0.39	-0.56	-0.28	-0.27	-0.49	-0.72	-0.47	0.01	NA	-0.19	NA	0.34	-0.20	-0.01	-0.36
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Table 14 continued (blue columns are sea-salt corrected, red columns are converted into equivalents)

NVE no.	SO4.						Al.R.			TOC.			Ca.	Mg.	NA.	K.	SO4.
	H.	Ca.1	Mg.1	K.1	NA..2	Cl.1	1	NO3	1	ANC	1	ANC <sub>OOA</sub>					
	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$	$\mu\text{eq l}^{-1}$
1431	-0.65	-0.44	-0.42	-0.11	-0.27	-0.35	-0.66	-0.56	-0.57	0.59	0.61	0.59	-0.40	-0.06	0.36	-0.03	-0.66
21797	-0.67	-0.38	-0.23	-0.20	-0.11	-0.16	-0.75	-0.48	-0.71	0.51	0.62	0.50	-0.35	-0.07	0.15	0.02	-0.69
11078	-0.25	-0.07	-0.10	0.15	-0.07	-0.05	-0.37	-0.06	-0.24	0.25	0.12	0.22	-0.06	-0.07	0.02	0.16	-0.37
11147	-0.58	-0.34	-0.33	-0.39	-0.19	-0.26	-0.65	-0.41	-0.44	0.61	0.58	0.49	-0.34	-0.18	0.26	-0.30	-0.64
331	-0.42	-0.52	-0.51	-0.12	-0.19	-0.22	-0.74	-0.24	-0.46	0.55	0.41	0.49	-0.51	-0.65	-0.01	0.03	-0.77
11592	-0.53	-0.31	-0.28	-0.29	-0.17	-0.22	-0.67	-0.40	-0.49	0.51	0.57	0.44	-0.29	-0.17	0.09	-0.18	-0.67
11373	-0.61	-0.43	-0.39	-0.43	-0.27	-0.30	-0.66	-0.52	-0.52	0.57	0.48	0.58	-0.40	-0.01	0.17	-0.09	-0.61
1174	-0.14	-0.06	-0.02	-0.03	0.03	-0.05	-0.24	-0.13	-0.15	0.22	0.13	0.22	-0.04	0.05	0.11	-0.07	-0.21
11292	-0.64	-0.53	-0.46	-0.19	-0.25	-0.31	-0.75	-0.53	-0.69	0.66	0.65	0.62	-0.49	-0.04	0.23	0.04	-0.72
11095	-0.53	-0.39	-0.43	-0.26	-0.21	-0.29	-0.71	-0.36	-0.51	0.58	0.62	0.53	-0.35	0.03	0.31	-0.03	-0.70
10305	-0.55	-0.24	-0.37	-0.22	-0.22	-0.33	-0.70	-0.36	-0.23	0.65	0.57	0.60	-0.23	0.03	0.38	-0.07	-0.69
1373	-0.54	-0.26	-0.34	-0.20	-0.15	-0.20	-0.66	-0.18	-0.50	0.51	0.27	0.53	-0.24	-0.15	0.20	0.03	-0.67
5844	-0.20	-0.44	-0.34	-0.04	-0.04	-0.10	-0.66	-0.19	-0.20	0.54	0.45	0.44	-0.46	-0.48	0.18	-0.03	-0.70
21186	-0.76	0.44	0.18	0.47	-0.23	-0.32	-0.77	0.08	-0.68	0.59	0.41	0.56	0.53	0.47	0.13	0.50	-0.77
21438	-0.71	-0.43	-0.25	-0.39	-0.08	-0.11	-0.73	-0.59	-0.71	0.55	0.40	0.55	-0.52	-0.12	0.19	-0.15	-0.74
21049	-0.35	-0.21	-0.13	0.09	-0.11	-0.14	-0.40	-0.24	-0.36	0.31	0.25	0.31	-0.17	0.03	0.16	0.14	-0.39
1545	-0.60	-0.51	-0.25	-0.12	-0.17	-0.20	-0.73	-0.47	-0.70	0.57	0.45	0.56	-0.50	-0.11	0.06	0.08	-0.71
22548	-0.63	0.03	-0.23	-0.02	-0.20	-0.23	-0.73	-0.48	-0.68	0.58	0.16	0.60	0.13	0.11	0.10	0.28	-0.71
3555	-0.26	-0.55	-0.50	-0.54	-0.18	-0.23	-0.71	-0.59	-0.46	0.66	0.59	0.56	-0.56	-0.54	0.20	-0.50	-0.73
23386	-0.54	0.26	0.01	-0.24	0.01	-0.06	-0.59	-0.54	-0.31	0.46	0.34	0.46	0.27	0.09	0.16	-0.28	-0.56
22101	-0.58	0.02	-0.04	0.25	-0.13	-0.06	-0.71	-0.11	-0.59	0.57	0.33	0.55	0.07	0.07	-0.12	0.29	-0.65
23328	-0.62	-0.03	-0.03	-0.09	-0.02	-0.03	-0.64	-0.55	-0.46	0.46	0.02	0.46	-0.04	-0.03	0.12	-0.21	-0.60
26511	-0.70	0.09	0.08	-0.19	0.09	0.07	-0.65	-0.50	-0.44	0.58	-0.30	0.59	0.02	-0.01	-0.01	-0.09	-0.69
26267	-0.66	-0.01	-0.24	0.07	-0.24	-0.27	-0.73	-0.39	-0.63	0.53	0.32	0.54	0.00	0.01	0.04	-0.10	-0.76
5828	-0.47	-0.54	-0.51	-0.48	-0.01	-0.09	-0.78	-0.39	-0.27	0.66	0.60	0.59	-0.57	-0.62	0.15	-0.54	-0.78
28197	-0.32	0.01	0.02	-0.08	-0.03	-0.04	-0.34	-0.12	-0.18	0.29	-0.04	0.29	0.03	0.07	0.04	-0.07	-0.36
1651	-0.59	0.41	0.06	0.21	0.11	-0.05	-0.72	-0.53	-0.47	0.60	-0.13	0.61	0.42	0.12	0.28	0.07	-0.71

1935	-0.40	0.28	-0.02	0.10	0.04	-0.04	-0.60	-0.31	-0.14	0.54	0.14	0.52	0.31	0.15	0.24	-0.02	-0.68
<b>Table 13</b>	<b>Cont</b>																
31186	-0.18	0.14	0.02	-0.08	-0.04	-0.10	-0.37	0.03	-0.13	0.32	0.14	0.33	0.20	0.28	0.13	-0.04	-0.29
31047	-0.19	0.47	-0.18	-0.22	-0.02	-0.18	-0.56	-0.61	-0.11	0.44	-0.22	0.45	0.46	-0.02	0.08	-0.07	-0.59
36436	-0.32	0.33	0.11	-0.19	-0.03	-0.01	-0.40	-0.01	0.12	0.36	0.20	0.36	0.32	0.20	0.06	-0.17	-0.54
36780	-0.41	0.09	-0.04	-0.32	-0.13	-0.16	-0.59	-0.28	-0.24	0.22	0.39	0.20	0.15	0.10	0.03	-0.15	-0.71
36727	-0.49	0.29	0.18	-0.32	0.03	-0.02	-0.52	-0.18	-0.26	0.37	-0.39	0.36	0.25	0.26	0.09	-0.37	-0.65
35326	-0.19	0.46	0.13	-0.01	0.32	0.13	-0.73	-0.62	-0.29	0.59	-0.12	0.56	0.41	0.04	0.13	0.06	-0.72
40844	-0.38	0.24	0.16	-0.27	-0.08	-0.11	-0.36	-0.49	-0.25	0.38	0.44	0.35	0.29	0.44	0.15	-0.31	-0.52
716	-0.55	0.35	-0.02	-0.10	0.03	-0.07	-0.69	-0.58	-0.49	0.45	0.34	0.42	0.34	0.11	0.26	0.01	-0.77
40322	-0.13	0.28	0.11	-0.27	0.06	0.03	-0.17	0.15	-0.13	0.24	0.07	0.28	0.29	0.14	0.08	-0.22	-0.35
806	-0.23	0.20	-0.18	0.12	-0.07	-0.18	-0.39	-0.22	-0.20	0.46	0.10	0.47	0.24	0.13	0.28	0.17	-0.28
45724	-0.42	0.15	-0.08	0.25	-0.05	-0.26	-0.70	-0.32	-0.10	0.54	0.34	0.51	0.21	0.14	0.37	0.34	-0.71
1001	-0.20	0.05	-0.26	0.01	-0.23	-0.31	-0.71	-0.06	-0.49	0.48	0.13	0.47	0.13	0.12	0.34	0.10	-0.72
48048	-0.47	0.20	0.11	0.16	0.06	-0.09	-0.45	-0.30	-0.12	0.40	0.34	0.40	0.23	0.20	0.20	0.28	-0.61
50879	-0.26	0.22	0.04	0.18	-0.02	-0.05	-0.39	-0.06	-0.15	0.38	0.09	0.38	0.28	0.18	0.09	0.30	-0.32
2430	-0.23	0.13	0.05	-0.20	-0.07	-0.09	-0.59	-0.53	1.00	0.27	-0.36	0.30	0.14	0.01	-0.10	-0.15	-0.63
64684	-0.26	0.23	-0.02	0.11	0.09	-0.11	-0.52	1.00	-0.12	0.60	0.10	0.61	0.23	-0.04	0.44	-0.01	-0.53
2437	-0.36	0.41	0.17	0.04	0.50	0.24	-0.77	-0.58	-0.09	0.58	-0.21	0.58	0.37	-0.07	0.28	-0.20	-0.78
63664	-0.36	0.17	0.01	-0.05	0.47	0.26	-0.76	-0.55	1.00	0.57	0.03	0.55	0.15	-0.19	0.19	0.09	-0.78
64713	-0.39	0.32	0.15	0.03	-0.06	-0.12	-0.34	-0.21	0.03	0.44	0.00	0.43	0.34	0.23	0.18	0.06	-0.32
2474	-0.34	0.03	-0.29	0.09	0.15	0.12	-0.72	-0.33	-0.08	0.44	-0.46	0.47	0.00	-0.35	-0.09	0.04	-0.74
64217	-0.75	-0.42	-0.56	-0.53	0.00	-0.10	-0.74	-0.25	-0.70	0.47	0.29	0.47	-0.41	-0.50	0.12	-0.54	-0.73
64193	-0.34	-0.21	-0.47	-0.21	0.00	-0.16	-0.45	-0.09	0.07	0.60	0.31	0.57	-0.20	-0.52	0.43	-0.05	-0.43
2456	1.00	0.30	0.09	-0.18	0.46	0.31	-0.74	0.00	1.00	0.58	-0.37	0.59	0.29	0.06	0.23	-0.46	-0.75
64799	-0.37	0.09	-0.30	0.08	0.00	-0.17	-0.63	1.00	-0.14	0.54	0.10	0.54	0.09	-0.20	0.35	0.11	-0.61
64278	-0.24	0.23	0.02	-0.15	0.16	-0.07	-0.50	-0.32	-0.19	0.35	-0.02	0.34	0.23	0.04	0.30	-0.20	-0.55
3238	-0.69	-0.43	-0.48	-0.34	0.12	-0.09	-0.80	-0.55	-0.69	0.71	0.64	0.68	-0.44	-0.53	0.35	-0.34	-0.81
3259	-0.51	-0.53	-0.57	-0.41	-0.19	-0.18	-0.79	-0.48	-0.25	0.65	0.48	0.63	-0.53	-0.59	0.08	-0.41	-0.80
5114	-0.61	-0.38	-0.42	-0.22	-0.21	-0.35	-0.78	-0.18	-0.43	0.66	0.46	0.65	-0.36	-0.26	0.25	-0.23	-0.79
368	-0.14	-0.27	-0.36	-0.47	0.05	-0.14	-0.76	-0.43	-0.22	0.57	0.37	0.45	-0.27	-0.40	0.34	-0.40	-0.77

3838	-0.25	-0.46	-0.47	-0.37	-0.06	-0.22	-0.76	-0.47	-0.43	0.60	0.36	0.53	-0.45	-0.43	0.16	-0.39	-0.76
<b>Table 13</b>	<b>Cont</b>																
281	-0.08	-0.42	-0.47	-0.35	-0.09	-0.32	-0.74	-0.51	-0.21	0.58	0.46	0.47	-0.41	-0.42	0.29	-0.41	-0.74
282	-0.37	0.22	-0.05	0.11	0.15	-0.25	-0.73	-0.27	-0.26	0.66	0.32	0.62	0.25	-0.12	0.34	-0.11	-0.74
34660	-0.45	0.36	0.16	0.39	0.22	-0.28	-0.14	-0.23	1.00	0.57	-0.01	0.56	0.36	0.26	0.38	0.40	-0.07
5269	-0.37	-0.14	-0.15	-0.12	-0.21	-0.29	-0.55	0.19	-0.30	0.51	0.40	0.43	-0.11	-0.08	0.01	-0.04	-0.56
5961	-0.17	-0.31	-0.38	-0.23	-0.15	-0.41	-0.63	-0.17	-0.25	0.37	0.12	0.31	-0.31	-0.30	0.15	-0.16	-0.62
5742	-0.27	0.01	-0.14	0.04	-0.17	-0.19	-0.47	-0.12	-0.13	0.38	0.35	0.33	0.02	-0.02	-0.01	0.06	-0.47
14367	-0.20	-0.33	-0.36	-0.51	-0.05	-0.17	-0.74	-0.42	-0.33	0.53	0.24	0.47	-0.33	-0.36	0.15	-0.50	-0.75
69	-0.39	0.06	-0.31	-0.20	-0.29	-0.44	-0.60	-0.45	-0.38	0.48	0.15	0.47	0.08	0.02	0.28	-0.17	-0.60
14277	-0.36	-0.06	-0.31	-0.15	-0.29	-0.41	-0.50	-0.39	-0.36	0.43	0.22	0.42	-0.03	0.05	0.27	-0.30	-0.50
1094	-0.74	0.08	-0.31	0.03	-0.17	-0.34	-0.75	-0.58	-0.75	0.70	-0.07	0.72	0.07	0.00	0.49	1.00	-0.73
13194	-0.57	0.01	-0.51	-0.20	-0.26	-0.55	-0.82	-0.42	-0.52	0.69	0.27	0.70	0.05	-0.05	0.56	-0.09	-0.82
9534	-0.42	-0.52	-0.48	-0.48	-0.14	-0.16	-0.70	-0.53	-0.36	0.53	0.52	0.47	-0.55	-0.41	0.11	-0.42	-0.70
10127	-0.70	-0.54	-0.50	-0.53	-0.31	-0.37	-0.76	-0.68	-0.65	0.73	0.54	0.71	-0.53	-0.52	0.36	-0.60	-0.77
10926	-0.45	-0.44	-0.40	-0.50	-0.19	-0.28	-0.74	-0.29	-0.34	0.66	0.61	0.62	-0.43	-0.05	0.35	-0.41	-0.71
9219	-0.65	-0.55	-0.58	-0.46	-0.24	-0.40	-0.76	-0.54	-0.45	0.77	0.44	0.67	-0.52	-0.23	0.45	-0.31	-0.76
15100	-0.65	-0.14	-0.45	-0.29	-0.23	-0.31	-0.79	-0.39	-0.37	0.70	0.41	0.67	-0.10	-0.08	0.38	0.00	-0.76
15177	-0.63	-0.03	-0.45	-0.08	-0.14	-0.29	-0.72	-0.54	-0.44	0.68	0.40	0.63	-0.02	-0.11	0.25	-0.19	-0.72
14534	-0.35	-0.03	-0.16	-0.02	-0.12	-0.15	-0.47	-0.34	-0.33	0.43	0.16	0.42	-0.02	0.02	0.19	-0.12	-0.44
13592	-0.69	0.13	-0.36	-0.15	-0.20	-0.23	-0.79	-0.58	-0.59	0.59	-0.18	0.58	0.21	-0.05	0.24	-0.19	-0.73
7272	-0.37	-0.40	-0.55	-0.26	-0.30	-0.47	-0.72	-0.42	-0.20	0.51	0.35	0.41	-0.39	-0.53	0.02	-0.26	-0.72

Table 15: P values after the Mann Kendall test and units used for the test. Due to the high number of statistical tests the p-values are uncorrected for multiple testing and should be interpreted carefully. Green fields indicate p values < 0.05.

NVE no.	pH	COND mS m <sup>-1</sup>	ALK mmol l <sup>-1</sup>	Ca mg l <sup>-1</sup>	Mg mg l <sup>-1</sup>	K mg l <sup>-1</sup>	NA..1 mg l <sup>-1</sup>	Cl mg l <sup>-1</sup>	SO4 mg l <sup>-1</sup>	NO3.N µg N l <sup>-1</sup>	TOTP µg P l <sup>-1</sup>	PO4.P µg P l <sup>-1</sup>	TOTN µg N l <sup>-1</sup>	NH4 µg N l <sup>-1</sup>	TOC mg C l <sup>-1</sup>	Al.R µg l <sup>-1</sup>	Al.II µg l <sup>-1</sup>	LAL µg l <sup>-1</sup>
1431	0.0																	
2179	0	0.00	0.55	0.00	0.00	0.11	0.04	0.01	0.00	0.00	0.88	NA	0.00	NA	0.00	0.00	0.00	0.39
7	0	0.03	0.13	0.01	0.07	0.09	0.38	0.23	0.00	0.00	0.82	NA	0.06	NA	0.00	0.00	0.00	0.00
1107	0.0																	
8	7	0.24	0.34	0.67	0.49	0.28	0.61	0.72	0.01	0.60	0.61	NA	0.33	NA	0.49	0.07	0.45	0.14
1114	0.0																	
7	0	0.00	0.75	0.01	0.01	0.01	0.16	0.05	0.00	0.00	0.94	NA	0.77	NA	0.00	0.00	0.02	0.00
331	0	0.00	0.94	0.00	0.00	0.20	0.16	0.09	0.00	0.07	0.44	NA	0.01	NA	0.00	0.00	0.02	0.00
1159	0.0																	
2	0	0.00	0.30	0.02	0.04	0.06	0.21	0.11	0.00	0.00	0.97	NA	0.65	NA	0.00	0.00	0.00	0.00
1137	0.0																	
3	0	0.00	0.13	0.00	0.01	0.00	0.04	0.02	0.00	0.00	0.86	NA	0.67	NA	0.00	0.00	0.03	0.01
1174	0.3																	
3	0.26	0.74	0.76	0.89	0.88	0.84	0.84	0.73	0.08	0.32	NA	NA	0.25	NA	0.34	0.22	0.72	0.03
1129	0.0																	
2	0	0.00	0.23	0.00	0.00	0.18	0.06	0.02	0.00	0.00	0.55	NA	0.01	NA	0.00	0.00	0.00	0.00
1109	0.0																	
5	0	0.00	0.95	0.00	0.00	0.06	0.11	0.03	0.00	0.01	0.96	NA	0.00	NA	0.00	0.00	0.03	0.00
1030	0.0																	
5	0	0.00	0.87	0.10	0.01	0.11	0.11	0.01	0.00	0.03	0.67	NA	1.00	NA	0.00	0.13	0.63	0.00
1373	0.0																	
0	0.00	0.67	0.06	0.01	0.23	0.27	0.15	0.00	0.21	0.82	NA	0.96	NA	0.04	0.00	0.88	0.00	
5844	0.1																	
5	0.00	0.66	0.00	0.01	0.78	0.76	0.48	0.00	0.30	0.47	NA	0.13	NA	0.00	0.13	0.12	0.00	
2118	0.0																	
6	0	0.00	0.12	0.00	0.08	0.00	0.08	0.02	0.00	0.57	0.95	NA	0.28	NA	0.00	0.00	0.80	0.02
2143	0.0																	
8	0	0.00	0.03	0.00	0.06	0.02	0.52	0.40	0.00	0.00	0.54	NA	0.00	NA	0.00	0.00	0.91	0.10
2104	0.0																	
9	1	0.07	0.31	0.13	0.37	0.49	0.43	0.30	0.00	0.08	0.97	NA	0.18	NA	0.05	0.01	0.16	0.00
1545	0	0.00	0.19	0.00	0.07	0.54	0.19	0.13	0.00	0.00	0.79	NA	0.00	NA	0.00	0.00	0.01	0.09
2254	0.0																	
8	0	0.00	0.31	0.64	0.09	0.80	0.15	0.12	0.00	0.00	0.24	NA	0.60	NA	0.08	0.00	0.99	0.00



3555	0.0	6	0.00	0.05	0.00	0.00	0.00	0.20	0.08	0.00	0.00	0.50	NA	0.73	NA	0.00	0.00	0.00	0.00	
2338	0.0	6	0	0.00	0.16	0.05	0.94	0.01	0.90	0.68	0.00	0.00	1.00	NA	0.16	NA	0.05	0.01	0.03	0.97
2210	0.0	1	0	0.05	0.91	0.93	0.94	0.09	0.37	0.61	0.00	0.44	0.57	NA	0.41	NA	0.05	0.00	0.61	0.00
2332	0.0	8	0	0.00	1.00	0.97	0.83	0.27	0.90	0.84	0.00	0.00	0.27	NA	0.04	NA	0.24	0.00	0.01	NA
2651	0.0	1	0	0.03	0.15	0.66	0.56	0.38	0.48	0.63	0.00	0.00	0.57	NA	0.01	NA	0.00	0.00	0.00	NA
2626	0.0	7	0	0.00	0.09	0.98	0.10	0.04	0.09	0.05	0.00	0.01	0.61	NA	0.61	NA	0.05	0.00	0.17	0.00
5828	0.0	0	0.00	0.70	0.00	0.00	0.00	0.90	0.49	0.00	0.00	0.38	NA	0.00	NA	0.00	0.04	0.00	0.00	
2819	0.0	7	1	0.37	0.86	1.00	0.88	0.59	0.88	0.75	0.01	0.26	0.92	NA	0.10	NA	0.81	0.14	0.51	0.15
1651	0.0	0	0.00	0.07	0.00	0.67	0.74	0.45	0.48	0.00	0.00	0.49	NA	0.27	NA	0.74	0.00	0.01	0.13	
<b>Table 14</b>	<b>Cont.</b>																			
1935	0.0	0	0.43	0.44	0.05	0.92	0.86	0.76	0.75	0.00	0.00	0.76	NA	0.06	NA	0.21	0.38	0.30	0.53	
3118	0.2	6	3	0.64	0.97	0.27	0.85	0.71	0.78	0.46	0.01	0.95	0.92	NA	0.58	NA	0.37	0.50	0.87	0.52
3104	0.1	7	5	0.01	0.11	0.00	0.25	0.20	0.88	0.20	0.00	0.00	0.69	NA	0.00	NA	0.19	0.03	0.00	0.56
3643	0.0	6	0	0.94	0.22	0.02	0.45	0.11	0.81	0.93	0.01	0.97	0.33	NA	0.03	NA	0.22	0.88	0.10	0.03
3678	0.0	0	0	0.18	1.00	0.52	0.78	0.04	0.36	0.24	0.00	0.32	1.00	NA	0.60	NA	0.00	0.05	0.75	0.01
3672	0.0	7	0	0.93	0.41	0.05	0.17	0.01	0.85	0.93	0.00	0.09	0.77	NA	0.48	NA	0.01	0.03	0.20	0.94
3532	0.0	6	1	0.11	0.00	0.00	0.29	0.45	0.03	0.17	0.00	0.00	0.78	NA	0.03	NA	0.40	0.08	0.22	0.50
4084	0.0	4	1	0.39	0.33	0.07	0.27	0.01	0.57	0.43	0.01	0.00	0.94	NA	0.28	NA	0.00	0.06	0.36	0.08
716	0.0	0	0.41	0.01	0.02	0.88	0.97	0.84	0.55	0.00	0.00	0.35	NA	0.31	NA	0.00	0.01	0.19	0.21	
4032	0.2	2	1	0.94	1.00	0.04	0.44	0.16	0.64	0.85	0.16	0.50	0.76	NA	1.00	NA	0.64	0.72	0.79	0.88
806	0.0	0	0.08	0.17	0.15	0.15	0.37	0.56	0.18	0.02	0.08	0.94	NA	0.17	NA	0.24	0.40	0.70	0.84	

4572	0.0																		
4	0	0.05	0.03	0.24	0.41	0.10	0.65	0.05	0.00	0.00	0.74	NA	0.46	NA	0.01	0.37	0.51	0.46	
	0.0																		
1001	1	0.00	0.03	0.68	0.07	1.00	0.09	0.02	0.00	0.04	0.52	NA	0.13	NA	0.27	0.00	0.48	0.01	
4804	0.0																		
8	0	0.42	0.00	0.11	0.44	0.26	0.64	0.50	0.00	0.00	0.61	NA	0.19	NA	0.10	0.65	0.50	0.86	
5087	0.0																		
9	1	0.60	0.18	0.09	0.73	0.19	0.94	0.71	0.01	0.75	0.22	NA	0.05	NA	0.24	0.73	0.43	0.80	
	0.0																		
2430	4	0.11	0.01	0.38	0.71	0.04	0.64	0.52	0.00	0.00	0.94	NA	0.71	NA	0.10	0.01	0.00	NA	
6468	0.0																		
4	1	0.50	0.00	0.08	0.91	0.40	0.50	0.40	0.00	0.05	0.51	NA	0.22	NA	0.17	0.05	0.01	0.22	
	0.0																		
2437	0	0.38	0.19	0.00	0.23	0.13	0.00	0.06	0.00	0.00	0.42	NA	0.01	NA	0.15	0.04	0.00	1.00	
6366	0.0																		
4	0	0.09	0.23	0.17	0.85	0.60	0.00	0.07	0.00	0.00	0.91	NA	0.44	NA	0.68	0.01	0.00	0.59	
6471	0.0																		
3	0	0.64	0.03	0.02	0.27	0.83	0.71	0.37	0.01	0.31	1.00	NA	0.80	NA	0.81	0.01	0.00	0.49	
	0.0																		
2474	1	0.15	0.47	0.85	0.04	0.52	0.28	0.44	0.00	0.01	0.66	NA	0.62	NA	0.00	0.22	0.00	0.98	
6421	0.0																		
7	0	0.00	0.24	0.00	0.00	0.00	0.85	0.48	0.00	0.00	0.87	NA	0.33	NA	0.02	0.00	0.01	0.88	
6419	0.0																		
3	1	0.01	0.26	0.13	0.00	0.23	0.99	0.23	0.00	0.61	0.98	NA	0.01	NA	0.01	0.55	0.05	0.36	
	0.0																		
2456	0	0.18	0.00	0.03	0.51	0.12	0.00	0.05	0.00	0.02	0.64	NA	0.73	NA	0.06	0.00	0.00	NA	
6479	0.0																		
9	0	0.05	0.01	0.61	0.04	0.34	0.94	0.23	0.00	0.26	0.91	NA	0.05	NA	0.42	0.54	0.18	0.40	
6427	0.0																		
8	1	0.11	0.05	0.10	0.83	0.89	0.24	0.76	0.00	0.04	0.98	NA	0.52	NA	0.61	0.14	0.00	0.91	
	0.0																		
3238	0	0.00	0.42	0.00	0.00	0.02	0.38	0.40	0.00	0.00	0.95	NA	0.52	NA	0.00	0.00	0.00	0.00	
	0.0																		
3259	0	0.00	0.02	0.00	0.00	0.00	0.14	0.20	0.00	0.00	0.88	NA	0.24	NA	0.00	0.06	0.41	0.00	
	0.0																		
5114	0	0.00	0.01	0.01	0.00	0.03	0.09	0.01	0.00	0.22	0.62	NA	0.12	NA	0.00	0.00	0.07	0.00	
	0.3																		
368	5	0.00	0.59	0.04	0.01	0.00	0.68	0.36	0.00	0.00	0.59	NA	0.63	NA	0.01	0.08	0.88	0.01	
<b>Table 14 Cont.</b>	0.0																		
3838	5	0.00	0.44	0.00	0.00	0.01	0.61	0.16	0.00	0.00	0.68	NA	0.51	NA	0.01	0.00	0.57	0.00	

281	0.4 4	0.00	0.15	0.00	0.00	0.01	0.54	0.02	0.00	0.00	0.74	NA	0.06	NA	0.00	0.20	0.99	0.00
282	0.0 1	0.00	0.62	0.09	0.51	0.62	0.26	0.06	0.00	0.00	0.73	NA	0.10	NA	0.01	0.07	0.78	0.04
3466	0.0 0	0.23	0.03	0.01	0.29	0.00	0.09	0.26	0.48	0.22	0.86	NA	0.81	NA	0.55	0.00	0.00	NA
5269	0.0 1	0.02	0.72	0.40	0.27	0.41	0.12	0.03	0.00	0.11	0.49	NA	0.19	NA	0.00	0.03	0.14	0.01
5961	0.2 1	0.02	0.82	0.02	0.01	0.17	0.26	0.00	0.00	0.02	0.83	NA	0.02	NA	0.44	0.06	0.19	0.01
5742	0.0 4	0.01	0.22	0.92	0.33	0.92	0.21	0.12	0.00	0.32	0.50	NA	0.00	NA	0.01	0.39	0.78	0.09
1436	0.1 7	0.00	0.45	0.01	0.01	0.00	0.75	0.24	0.00	0.00	0.64	NA	0.09	NA	0.08	0.01	0.97	0.03
69	0.0 0	0.00	0.02	0.63	0.04	0.13	0.02	0.00	0.00	0.00	1.00	NA	0.04	NA	0.11	0.01	0.28	0.34
1427	0.0 7	0.00	0.57	0.65	0.03	0.11	0.02	0.00	0.00	0.01	0.71	NA	0.51	NA	0.16	0.00	0.14	0.00
1094	0.0 0	0.00	0.63	0.63	0.00	0.73	0.38	0.01	0.00	0.00	0.94	NA	0.01	NA	0.28	0.00	0.01	NA
1319	0.0 4	0.00	0.00	0.82	0.00	0.16	0.07	0.00	0.00	0.01	0.24	NA	0.22	NA	0.03	0.00	0.01	0.00
9534	0.0 0	0.00	0.97	0.00	0.00	0.00	0.32	0.22	0.00	0.00	0.73	NA	0.23	NA	0.00	0.01	0.04	0.00
1012	0.0 7	0.00	0.24	0.00	0.00	0.00	0.02	0.01	0.00	0.00	0.78	NA	0.00	NA	0.00	0.00	0.11	0.00
1092	0.0 6	0.00	0.54	0.00	0.01	0.00	0.14	0.04	0.00	0.10	1.00	NA	0.25	NA	0.00	0.01	0.00	0.00
9219	0.0 0	0.00	0.63	0.00	0.00	0.00	0.06	0.00	0.00	0.00	0.69	NA	0.92	NA	0.00	0.00	0.03	0.00
1510	0.0 0	0.00	0.89	0.34	0.00	0.03	0.11	0.03	0.00	0.01	0.72	NA	0.60	NA	0.00	0.00	0.02	0.00
1517	0.0 7	0.00	0.35	0.78	0.00	0.11	0.40	0.03	0.00	0.00	0.84	NA	0.39	NA	0.01	0.00	0.96	0.00
1453	0.0 4	0.01	0.29	0.89	0.29	0.33	0.40	0.29	0.00	0.02	0.95	NA	0.38	NA	0.13	0.02	0.63	0.01
1359	0.0 2	0.00	0.70	0.29	0.01	0.00	0.17	0.10	0.00	0.00	0.75	NA	0.00	NA	0.50	0.00	0.00	NA
7272	0.0 1	0.00	0.70	0.00	0.00	0.05	0.04	0.00	0.00	0.00	1.00	NA	0.21	NA	0.01	0.13	0.97	0.01

Table 14 continued. (blue columns are sea-salt corrected, red columns are converted into equivalents)

NVE no.	H+	Ca.1	Mg.1	K.1	NA..2	Cl.1	SO4.1	NO3	Al.R.1	ANC	TOC.1	ANC <sub>COA</sub>	Ca.	Mg.	NA.	K.	SO4.
	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>
1431	0.00	0.00	0.00	0.51	0.05	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.68	0.01	0.86	0.00
21797	0.00	0.01	0.10	0.18	0.41	0.22	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.64	0.26	0.91	0.00
11078	0.07	0.64	0.49	0.27	0.63	0.72	0.01	0.66	0.09	0.06	0.39	0.10	0.68	0.64	0.91	0.23	0.01
11147	0.00	0.01	0.01	0.01	0.16	0.05	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.19	0.05	0.04	0.00
331	0.00	0.00	0.00	0.44	0.15	0.09	0.00	0.10	0.00	0.00	0.00	0.00	0.00	0.00	0.97	0.87	0.00
11592	0.00	0.02	0.04	0.04	0.21	0.11	0.00	0.00	0.00	0.00	0.00	0.00	0.03	0.22	0.52	0.20	0.00
11373	0.00	0.00	0.00	0.00	0.04	0.03	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.94	0.21	0.54	0.00
1174	0.31	0.67	0.88	0.84	0.85	0.75	0.08	0.36	0.28	0.10	0.35	0.11	0.79	0.77	0.43	0.67	0.12
11292	0.00	0.00	0.00	0.19	0.07	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.79	0.09	0.82	0.00
11095	0.00	0.00	0.00	0.08	0.12	0.03	0.00	0.01	0.00	0.00	0.00	0.00	0.01	0.86	0.02	0.83	0.00
10305	0.00	0.08	0.01	0.14	0.10	0.01	0.00	0.01	0.11	0.00	0.00	0.00	0.10	0.82	0.00	0.66	0.00
1373	0.00	0.06	0.01	0.20	0.28	0.15	0.00	0.20	0.00	0.00	0.05	0.00	0.08	0.28	0.14	0.85	0.00
5844	0.14	0.00	0.01	0.78	0.76	0.48	0.00	0.18	0.13	0.00	0.00	0.00	0.00	0.00	0.17	0.85	0.00
21186	0.00	0.00	0.21	0.00	0.08	0.02	0.00	0.57	0.00	0.00	0.01	0.00	0.00	0.00	0.35	0.00	0.00
21438	0.00	0.00	0.06	0.01	0.54	0.42	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.36	0.16	0.30	0.00
21049	0.01	0.13	0.34	0.54	0.43	0.31	0.00	0.08	0.01	0.02	0.09	0.02	0.24	0.83	0.24	0.35	0.00
1545	0.00	0.00	0.06	0.41	0.20	0.13	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.45	0.65	0.60	0.00
22548	0.00	0.85	0.09	0.90	0.14	0.09	0.00	0.00	0.00	0.00	0.28	0.00	0.36	0.45	0.49	0.07	0.00
3555	0.05	0.00	0.00	0.00	0.18	0.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.14	0.00	0.00
23386	0.00	0.06	0.94	0.12	0.97	0.65	0.00	0.00	0.04	0.00	0.02	0.00	0.05	0.56	0.24	0.07	0.00

22101	0.00	0.89	0.81	0.09	0.33	0.64	0.00	0.42	0.00	0.00	0.02	0.00	0.60	0.62	0.40	0.05	0.00
23328	0.00	0.87	0.82	0.60	0.88	0.82	0.00	0.00	0.00	0.00	0.93	0.00	0.80	0.86	0.38	0.18	0.00
26511	0.00	0.51	0.58	0.23	0.50	0.62	0.00	0.00	0.00	0.00	0.05	0.00	0.91	0.94	0.95	0.59	0.00
26267	0.00	0.95	0.08	0.70	0.07	0.05	0.00	0.01	0.00	0.00	0.03	0.00	1.00	0.98	0.81	0.55	0.00
5828	0.00	0.00	0.00	0.00	0.93	0.49	0.00	0.00	0.05	0.00	0.00	0.00	0.00	0.00	0.27	0.00	0.00
28197	0.02	0.97	0.89	0.63	0.87	0.78	0.01	0.41	0.23	0.03	0.81	0.03	0.83	0.60	0.79	0.67	0.01
1651	0.00	0.00	0.69	0.19	0.42	0.75	0.00	0.00	0.00	0.00	0.43	0.00	0.00	0.43	0.04	0.68	0.00
1935	0.01	0.04	0.88	0.53	0.79	0.81	0.00	0.04	0.36	0.00	0.34	0.00	0.02	0.31	0.08	0.93	0.00

Table 14 continued

31186	0.25	0.31	0.92	0.60	0.75	0.46	0.01	0.86	0.40	0.02	0.34	0.01	0.15	0.04	0.33	0.78	0.03
31047	0.26	0.00	0.19	0.16	0.90	0.19	0.00	0.00	0.51	0.00	0.17	0.00	0.00	0.89	0.56	0.69	0.00
36436	0.04	0.02	0.45	0.21	0.84	0.97	0.00	0.96	0.46	0.01	0.17	0.01	0.02	0.17	0.66	0.26	0.00
36780	0.00	0.50	0.79	0.03	0.35	0.23	0.00	0.08	0.12	0.10	0.01	0.13	0.27	0.48	0.87	0.33	0.00
36727	0.00	0.03	0.19	0.03	0.82	0.91	0.00	0.26	0.08	0.01	0.01	0.01	0.07	0.06	0.53	0.01	0.00
35326	0.24	0.00	0.36	0.96	0.02	0.36	0.00	0.00	0.06	0.00	0.44	0.00	0.00	0.82	0.35	0.74	0.00
40844	0.01	0.08	0.25	0.07	0.56	0.43	0.01	0.00	0.12	0.00	0.00	0.01	0.04	0.00	0.28	0.04	0.00
716	0.00	0.01	0.92	0.54	0.82	0.60	0.00	0.00	0.00	0.00	0.03	0.00	0.02	0.45	0.06	0.98	0.00
40322	0.38	0.04	0.42	0.09	0.67	0.84	0.20	0.37	0.38	0.07	0.62	0.04	0.03	0.33	0.56	0.15	0.01
806	0.16	0.14	0.22	0.44	0.60	0.18	0.01	0.14	0.18	0.00	0.53	0.00	0.08	0.37	0.04	0.28	0.04
45724	0.01	0.26	0.59	0.09	0.71	0.05	0.00	0.04	0.56	0.00	0.02	0.00	0.12	0.32	0.01	0.02	0.00
1001	0.21	0.73	0.06	0.97	0.08	0.02	0.00	0.75	0.00	0.00	0.39	0.00	0.36	0.38	0.01	0.51	0.00
48048	0.00	0.16	0.43	0.30	0.65	0.50	0.00	0.05	0.44	0.00	0.03	0.00	0.11	0.14	0.13	0.06	0.00
50879	0.11	0.12	0.78	0.23	0.91	0.71	0.01	0.77	0.35	0.00	0.58	0.00	0.04	0.21	0.53	0.04	0.02
2430	0.14	0.34	0.71	0.20	0.61	0.52	0.00	0.00	1.00	0.04	0.02	0.03	0.31	0.94	0.45	0.32	0.00
64684	0.11	0.10	0.91	0.50	0.51	0.41	0.00	1.00	0.46	0.00	0.50	0.00	0.09	0.80	0.00	0.95	0.00
2437	0.02	0.00	0.23	0.80	0.00	0.08	0.00	0.00	0.59	0.00	0.19	0.00	0.01	0.61	0.04	0.20	0.00
63664	0.02	0.22	0.98	0.78	0.00	0.06	0.00	0.00	1.00	0.00	0.87	0.00	0.28	0.19	0.18	0.59	0.00
64713	0.01	0.02	0.27	0.88	0.69	0.37	0.01	0.21	0.90	0.00	1.00	0.00	0.01	0.09	0.19	0.69	0.02
2474	0.03	0.85	0.04	0.59	0.27	0.39	0.00	0.03	0.68	0.00	0.00	0.00	1.00	0.01	0.53	0.80	0.00
64217	0.00	0.00	0.00	0.00	1.00	0.47	0.00	0.12	0.00	0.00	0.06	0.00	0.00	0.00	0.40	0.00	0.00
64193	0.03	0.12	0.00	0.16	1.00	0.23	0.00	0.58	0.62	0.00	0.03	0.00	0.15	0.00	0.00	0.72	0.00

2456	1.00	0.03	0.53	0.27	0.00	0.03	0.00	1.00	1.00	0.00	0.02	0.00	0.04	0.66	0.09	0.00	0.00
64799	0.02	0.53	0.03	0.63	1.00	0.21	0.00	1.00	0.38	0.00	0.53	0.00	0.53	0.14	0.01	0.48	0.00
64278	0.13	0.09	0.92	0.35	0.24	0.60	0.00	0.04	0.25	0.01	0.91	0.01	0.09	0.76	0.03	0.20	0.00
3238	0.00	0.00	0.00	0.02	0.38	0.52	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.02	0.00
3259	0.00	0.00	0.00	0.00	0.15	0.19	0.00	0.00	0.07	0.00	0.00	0.00	0.00	0.00	0.55	0.00	0.00
5114	0.00	0.01	0.00	0.14	0.11	0.01	0.00	0.21	0.00	0.00	0.00	0.00	0.01	0.07	0.07	0.14	0.00
368	0.30	0.05	0.01	0.00	0.71	0.31	0.00	0.00	0.11	0.00	0.01	0.00	0.04	0.00	0.01	0.01	0.00
3838	0.06	0.00	0.00	0.01	0.69	0.12	0.00	0.00	0.00	0.00	0.01	0.00	0.00	0.00	0.25	0.01	0.00

Table 14 continued

281	0.57	0.00	0.00	0.02	0.53	0.02	0.00	0.00	0.14	0.00	0.00	0.00	0.00	0.00	0.03	0.00	0.00
282	0.01	0.11	0.72	0.47	0.30	0.07	0.00	0.08	0.06	0.00	0.02	0.00	0.07	0.43	0.01	0.49	0.00
34660	0.00	0.01	0.28	0.01	0.11	0.06	0.31	0.12	1.00	0.00	0.97	0.00	0.01	0.08	0.01	0.01	0.60
5269	0.01	0.31	0.31	0.45	0.12	0.04	0.00	0.19	0.03	0.00	0.00	0.00	0.45	0.58	0.97	0.82	0.00
5961	0.22	0.02	0.01	0.12	0.28	0.00	0.00	0.28	0.07	0.01	0.40	0.02	0.02	0.04	0.28	0.30	0.00
5742	0.06	0.95	0.33	0.78	0.21	0.15	0.00	0.39	0.36	0.00	0.01	0.01	0.88	0.88	0.97	0.71	0.00
14367	0.14	0.01	0.01	0.00	0.73	0.21	0.00	0.01	0.01	0.00	0.08	0.00	0.02	0.01	0.26	0.00	0.00
69	0.01	0.68	0.04	0.21	0.03	0.00	0.00	0.00	0.01	0.00	0.35	0.00	0.56	0.92	0.04	0.28	0.00
14277	0.01	0.67	0.03	0.36	0.04	0.00	0.00	0.01	0.01	0.00	0.14	0.00	0.86	0.73	0.05	0.05	0.00
1094	0.00	0.57	0.04	0.84	0.23	0.01	0.00	0.00	0.00	0.00	0.69	0.00	0.62	1.00	0.00	1.00	0.00
13194	0.00	0.97	0.00	0.19	0.07	0.00	0.00	0.00	0.00	0.00	0.06	0.00	0.73	0.78	0.00	0.54	0.00
9534	0.00	0.00	0.00	0.00	0.28	0.22	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	0.44	0.00	0.00
10127	0.00	0.00	0.00	0.00	0.02	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.01	0.00	0.00
10926	0.00	0.00	0.00	0.00	0.15	0.04	0.00	0.04	0.01	0.00	0.00	0.00	0.00	0.71	0.01	0.00	0.00
9219	0.00	0.00	0.00	0.00	0.08	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.11	0.00	0.04	0.00
15100	0.00	0.33	0.00	0.06	0.09	0.02	0.00	0.01	0.01	0.00	0.00	0.00	0.50	0.58	0.01	1.00	0.00
15177	0.00	0.82	0.00	0.62	0.32	0.04	0.00	0.00	0.00	0.00	0.00	0.00	0.92	0.45	0.07	0.22	0.00
14534	0.02	0.85	0.27	0.90	0.40	0.29	0.00	0.02	0.02	0.00	0.31	0.00	0.91	0.92	0.19	0.43	0.00
13592	0.00	0.34	0.02	0.37	0.14	0.10	0.00	0.00	0.00	0.00	0.26	0.00	0.14	0.73	0.08	0.23	0.00
7272	0.01	0.00	0.00	0.10	0.03	0.00	0.00	0.01	0.14	0.00	0.01	0.00	0.00	0.00	0.89	0.08	0.00

Table 16: Theill-sens slope estimates for each lake. The changes are the unit given in the header per year.

NVE no.	pH	KOND mS m <sup>-1</sup>	ALK mmol l <sup>-1</sup>	Ca mg l <sup>-1</sup>	Mg mg l <sup>-1</sup>	K mg l <sup>-1</sup>	Na mg l <sup>-1</sup>	Cl mg l <sup>-1</sup>	SO4 mg l <sup>-1</sup>	NO3.N µg N l <sup>-1</sup>	TOTP µg P l <sup>-1</sup>	PO4.P µg P l <sup>-1</sup>	TOTN µg N l <sup>-1</sup>	NH4 µg N l <sup>-1</sup>	TOC mg C l <sup>-1</sup>	Al.R µg l <sup>-1</sup>	Al.II µg l <sup>-1</sup>	LAL µg l <sup>-1</sup>
1431	0.02	-0.05	0.00	0.00	0.00	0.00	-0.01	-0.04	-0.08	-3.65	0.00	NA	-2.73	NA	0.05	-4.29	0.44	-1.07
21797	0.03	-0.04	0.00	0.00	0.00	0.00	-0.02	-0.04	-0.09	-5.05	0.00	NA	-2.50	NA	0.05	-8.87	1.14	-9.25
11078	0.02	-0.01	0.00	0.00	0.00	0.00	-0.01	0.00	-0.06	-0.34	0.00	NA	3.33	NA	0.03	-1.74	-0.75	-1.13
11147	0.02	-0.04	0.00	-0.01	0.00	0.00	-0.02	-0.04	-0.11	-2.74	0.00	NA	-0.63	NA	0.14	-2.86	2.02	-4.48
331	0.01	-0.06	0.00	-0.01	-0.01	0.00	-0.01	-0.04	-0.18	-1.00	0.00	NA	2.78	NA	0.14	-2.77	1.62	-4.65
11592	0.02	-0.05	0.00	-0.01	0.00	0.00	-0.02	-0.05	-0.13	-4.13	0.00	NA	-0.91	NA	0.14	-3.53	2.50	-5.75
11373	0.02	-0.06	0.00	-0.01	0.00	0.00	-0.03	-0.05	-0.11	-4.00	0.00	NA	0.71	NA	0.08	-4.52	0.97	-4.70
1174	0.00	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.04	-0.90	NA	NA	-1.63	NA	0.02	-0.73	0.04	-2.00
11292	0.02	-0.07	0.00	0.00	0.00	0.00	-0.02	-0.04	-0.09	-9.29	0.00	NA	-7.14	NA	0.08	-5.89	1.00	-6.05
11095	0.02	-0.04	0.00	-0.01	0.00	0.00	-0.02	-0.04	-0.09	-1.68	0.00	NA	3.33	NA	0.14	-2.20	1.59	-3.70
10305	0.01	-0.05	0.00	0.00	0.00	0.00	-0.01	-0.03	-0.08	-1.58	0.00	NA	0.00	NA	0.11	-0.54	0.29	-1.51
1373	0.01	-0.04	0.00	0.00	0.00	0.00	-0.01	-0.02	-0.07	-1.00	0.00	NA	0.00	NA	0.06	-2.78	-0.24	-2.98
5844	0.01	-0.06	0.00	-0.03	-0.01	0.00	-0.01	-0.03	-0.26	-0.64	0.08	NA	5.00	NA	0.35	-2.65	3.15	-5.25
21186	0.05	-0.03	0.00	0.01	0.00	0.01	-0.01	-0.03	-0.07	1.40	0.00	NA	2.50	NA	0.02	-5.45	0.00	-2.00
21438	0.02	-0.06	0.00	0.00	0.00	0.00	-0.01	-0.02	-0.09	-9.30	0.00	NA	-10.63	NA	0.02	-12.35	0.00	-6.61
21049	0.02	-0.03	0.00	0.00	0.00	0.00	0.00	-0.01	-0.06	-1.50	0.00	NA	-1.00	NA	0.02	-5.59	0.38	-6.36
1545	0.02	-0.06	0.00	-0.01	0.00	0.00	-0.02	-0.05	-0.08	-6.42	0.00	NA	-7.27	NA	0.02	-9.20	0.31	-5.07
22548	0.02	-0.02	0.00	0.00	0.00	0.00	-0.01	-0.02	-0.04	-2.33	0.00	NA	-0.59	NA	0.01	-2.43	0.00	-2.26
3555	0.01	-0.05	0.00	-0.02	-0.01	-0.01	-0.01	-0.05	-0.19	-3.00	0.00	NA	-0.63	NA	0.30	-2.82	4.31	-8.56
23386	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.02	-1.65	0.00	NA	-1.39	NA	0.01	-0.50	-0.10	0.00
22101	0.03	-0.02	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.06	-0.48	0.00	NA	0.56	NA	0.02	-1.38	0.17	-1.53
23328	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	-2.38	0.03	NA	-1.20	NA	0.00	-1.50	0.00	NA
26511	0.02	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.02	-1.71	0.00	NA	-2.26	NA	-0.01	-0.57	-0.16	NA
26267	0.02	-0.02	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.03	-1.47	0.00	NA	-0.42	NA	0.01	-1.63	0.05	-1.33
5828	0.02	-0.05	0.00	-0.02	-0.01	0.00	0.00	-0.01	-0.21	-2.16	0.00	NA	6.67	NA	0.19	-2.08	2.80	-5.30
28197	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	-0.60	0.00	NA	-1.67	NA	0.00	-0.37	0.00	-0.25

Table	15	Cont.																	
1651	0.02	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.02	-1.20	0.00	NA	-0.43	NA	0.00	-0.67	-0.15	-0.18
1935	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.02	-0.50	0.00	NA	1.17	NA	0.01	-0.09	0.18	0.00
31186	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.01	0.00	0.00	NA	0.54	NA	0.01	-0.16	0.00	0.00
31047	0.01	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.02	-1.00	0.00	NA	-1.50	NA	0.00	-0.07	-0.15	0.00
36436	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	NA	0.94	NA	0.01	0.00	0.15	-0.12
36780	0.01	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.03	-0.02	-0.06	0.00	NA	0.42	NA	0.03	-0.35	0.10	-0.42
36727	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.33	0.00	NA	-0.58	NA	-0.01	-0.30	-0.07	0.00
35326	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	-1.15	0.00	NA	-0.92	NA	0.00	-0.22	-0.16	0.00
40844	0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.01	-0.89	0.00	NA	0.60	NA	0.03	-0.24	0.22	-0.19
716	0.02	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.02	-0.81	0.00	NA	0.25	NA	0.01	-0.28	0.16	-0.08
40322	0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	NA	0.00	NA	0.01	-0.07	0.05	0.00
806	0.01	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.57	0.00	NA	0.80	NA	0.00	-0.07	0.00	0.00
45724	0.02	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.02	-0.50	0.00	NA	0.50	NA	0.02	-0.08	-0.09	0.06
1001	0.01	-0.02	0.00	0.00	0.00	0.00	0.00	-0.01	-0.04	-0.03	-0.26	0.00	NA	0.71	NA	0.01	-0.79	-0.22	-0.53
48048	0.01	-0.01	0.00	0.00	0.00	0.00	0.01	-0.01	-0.02	-0.50	0.00	0.00	NA	0.55	NA	0.00	0.00	0.00	0.00
50879	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	0.00	0.00	NA	0.90	NA	0.01	0.00	0.00	0.00
2430	0.01	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.05	-0.90	0.00	NA	-0.10	NA	0.00	0.00	0.00	NA
64684	0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.01	-0.05	0.00	0.00	0.00	NA	0.50	NA	0.01	-0.09	-0.11	0.00
2437	0.01	0.00	0.00	0.00	0.00	0.00	0.01	0.01	-0.03	-0.83	0.00	0.00	NA	-1.25	NA	0.00	-0.13	-0.15	0.00
63664	0.02	0.00	0.00	0.00	0.00	0.00	0.01	0.01	-0.04	-0.47	0.00	0.00	NA	0.21	NA	0.00	-0.10	-0.20	0.00
64713	0.02	0.00	0.00	0.01	0.00	0.00	0.00	-0.01	-0.05	0.00	0.00	0.00	NA	0.00	NA	0.00	-0.17	-0.20	0.00
2474	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.01	-0.05	-0.63	0.00	0.00	NA	0.07	NA	-0.02	0.00	-0.15	0.00
64217	0.03	-0.03	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.09	-0.10	0.00	0.00	NA	0.60	NA	0.01	-1.56	-0.07	0.00
64193	0.02	-0.02	0.00	-0.01	-0.01	0.00	0.00	-0.02	-0.09	-0.05	0.00	0.00	NA	2.25	NA	0.03	0.09	0.47	-0.10
2456	0.01	0.00	0.00	0.01	0.00	0.00	0.01	0.00	-0.05	-0.10	0.00	0.00	NA	0.08	NA	-0.01	-0.13	-0.19	NA
64799	0.01	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.06	0.00	0.00	0.00	NA	0.75	NA	0.00	0.00	-0.04	0.00
64278	0.01	0.00	0.00	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	-0.33	0.00	NA	0.38	NA	0.00	-0.08	-0.14	0.00
3238	0.02	-0.04	0.00	-0.01	-0.01	0.00	0.00	0.00	-0.14	-3.00	0.00	0.00	NA	0.47	NA	0.08	-4.09	1.30	-5.57
3259	0.02	-0.05	0.00	-0.02	-0.01	-0.01	-0.01	-0.01	-0.17	-2.21	0.00	0.00	NA	1.50	NA	0.13	-0.91	0.65	-1.60
5114	0.03	-0.03	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.12	-0.63	0.00	0.00	NA	1.93	NA	0.06	-1.63	0.73	-1.90



Table	15	Cont.																
368	0.00	-0.03	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.10	-1.00	0.00	NA	0.36	NA	0.14	-0.86	-0.08	-0.73
3838	0.01	-0.03	0.00	-0.01	0.00	0.00	0.00	0.00	-0.10	-0.74	0.00	NA	0.59	NA	0.15	-2.50	-0.41	-2.62
281	0.00	-0.03	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.11	-1.09	0.00	NA	1.67	NA	0.20	-0.75	0.00	-1.26
282	0.02	-0.02	0.00	0.00	0.00	0.00	0.00	0.00	-0.07	-0.29	0.00	NA	2.27	NA	0.10	-0.47	0.09	-0.33
34660	0.01	0.00	0.00	0.01	0.00	0.00	0.00	0.00	0.00	-0.37	0.00	NA	0.09	NA	0.00	-0.17	-0.13	NA
5269	0.01	-0.02	0.00	0.00	0.00	0.00	0.00	-0.01	-0.10	1.00	0.00	NA	2.19	NA	0.08	-2.53	1.24	-4.08
5961	0.00	-0.01	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.08	-0.33	0.00	NA	-2.50	NA	0.04	-1.34	-1.25	-1.00
5742	0.02	-0.02	0.00	0.00	0.00	0.00	-0.01	-0.01	-0.13	-0.70	0.00	NA	5.80	NA	0.16	-0.68	-0.31	-0.61
14367	0.01	-0.02	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.10	-0.78	0.00	NA	-2.31	NA	0.07	-2.12	0.00	-1.36
69	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.05	-2.11	0.00	NA	-2.12	NA	0.01	-0.33	-0.04	0.00
14277	0.03	-0.02	0.00	0.00	0.00	0.00	0.00	-0.01	-0.05	-2.24	0.00	NA	-0.58	NA	0.01	-2.00	0.06	-2.00
1094	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.03	-2.87	0.00	NA	-1.89	NA	0.00	-2.63	0.00	NA
13194	0.02	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.07	-1.34	0.00	NA	-1.13	NA	0.02	-1.67	0.67	-2.44
9534	0.01	-0.05	0.00	-0.02	-0.01	-0.01	-0.01	-0.03	-0.14	-3.28	0.00	NA	-1.82	NA	0.15	-2.62	1.89	-3.91
10127	0.02	-0.06	0.00	-0.01	-0.01	-0.01	-0.01	-0.02	-0.13	-6.00	0.00	NA	-5.92	NA	0.10	-4.32	0.98	-5.08
10926	0.02	-0.05	0.00	-0.01	0.00	0.00	-0.01	-0.03	-0.10	-1.00	0.00	NA	1.67	NA	0.11	-2.18	2.75	-3.00
9219	0.02	-0.05	0.00	-0.01	0.00	0.00	-0.01	-0.02	-0.09	-1.82	0.00	NA	0.00	NA	0.09	-2.76	1.45	-3.84
15100	0.03	-0.02	0.00	0.00	0.00	0.00	0.00	-0.01	-0.06	-1.00	0.00	NA	0.27	NA	0.04	-1.38	0.85	-2.23
15177	0.02	-0.02	0.00	0.00	0.00	0.00	0.00	-0.01	-0.05	-1.85	0.00	NA	-0.45	NA	0.03	-1.69	0.00	-1.46
14534	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	0.00	-0.03	-1.59	0.00	NA	-0.63	NA	0.00	-1.27	0.00	-0.71
13592	0.03	-0.01	0.00	0.00	0.00	0.00	0.00	-0.01	-0.03	-2.09	0.00	NA	-2.58	NA	0.00	-1.44	0.00	NA
7272	0.01	-0.04	0.00	-0.01	0.00	0.00	0.00	-0.01	-0.10	-0.50	0.00	NA	-1.20	NA	0.08	-1.33	-0.03	-1.49

Table 15 continued (red fields are converted units in equivalents, blue fields are sea-salt corrected values)

NVE	H.	Ca.1	Mg.1	K.1	NA..2	Cl.1	SO4.1	NO3	Al.R.1	ANC	TOC.1	ANC <sub>OOA</sub>	Ca.	Mg.	NA.	K.	SO4.
no.	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>	μeq l <sup>-1</sup>
1431	-0.57	-0.17	-0.27	0.00	-0.51	-1.19	-1.56	-0.25	-0.43	1.88	0.14	1.68	-0.11	0.00	0.39	0.00	-1.45
21797	-0.50	-0.25	-0.26	0.00	-0.93	-1.16	-2.00	-0.37	-0.94	2.33	0.17	2.08	-0.24	-0.05	0.22	0.00	-1.83
11078	-0.15	-0.11	-0.09	0.11	-0.20	-0.11	-1.34	0.00	-0.19	1.44	0.11	1.24	-0.08	-0.02	0.00	0.15	-1.27
11147	-0.50	-0.38	-0.30	-0.09	-0.65	-1.15	-2.28	-0.21	-0.30	2.35	0.43	1.71	-0.38	-0.11	0.50	-0.06	-2.08
331	-0.33	-0.71	-1.00	0.00	-0.56	-1.10	-3.77	-0.05	-0.27	2.09	0.46	1.58	-0.67	-0.85	0.00	0.00	-3.71
11592	-0.39	-0.57	-0.32	-0.10	-1.00	-1.30	-2.72	-0.29	-0.37	2.22	0.46	1.72	-0.50	-0.14	0.23	-0.04	-2.45
11373	-0.75	-0.28	-0.33	-0.02	-1.12	-1.42	-2.23	-0.27	-0.46	2.50	0.27	2.11	-0.22	0.00	0.28	0.00	-2.15
1174	-0.17	0.00	0.00	0.00	0.00	0.00	-0.82	-0.04	-0.04	0.85	0.02	0.55	0.00	0.00	0.00	0.00	-0.67
11292	-1.00	-0.22	-0.29	0.00	-1.04	-1.24	-2.00	-0.66	-0.63	2.55	0.29	2.28	-0.18	0.00	0.29	0.00	-1.83
11095	-0.50	-0.33	-0.29	0.00	-0.89	-1.26	-2.00	-0.12	-0.24	2.02	0.47	1.58	-0.27	0.00	0.58	0.00	-1.82
10305	-0.67	-0.13	-0.20	0.00	-0.31	-0.83	-1.67	-0.11	-0.05	2.00	0.37	1.55	-0.10	0.00	0.38	0.00	-1.55
1373	-0.59	-0.23	-0.21	0.00	-0.32	-0.60	-1.53	-0.08	-0.29	1.86	0.18	1.58	-0.17	-0.06	0.29	0.00	-1.39
5844	-0.18	-1.31	-0.88	0.00	-0.26	-0.80	-5.44	-0.05	-0.29	3.42	1.17	2.00	-1.29	-0.81	0.42	0.00	-5.48
21186	-0.44	0.41	0.08	0.16	-0.40	-0.83	-1.46	0.06	-0.59	2.14	0.00	2.07	0.49	0.30	0.19	0.16	-1.37
21438	-0.67	-0.22	-0.22	0.00	-0.44	-0.61	-1.83	-0.67	-1.26	2.50	0.04	2.43	-0.20	-0.08	0.26	0.00	-1.75
21049	-0.50	-0.09	-0.09	0.00	-0.18	-0.40	-1.30	-0.09	-0.58	1.52	0.05	1.44	-0.04	0.00	0.19	0.00	-1.16
1545	-0.55	-0.40	-0.31	0.00	-0.92	-1.61	-1.78	-0.45	-0.98	2.08	0.05	2.00	-0.31	-0.06	0.12	0.00	-1.67
22548	-0.30	0.00	-0.13	0.00	-0.33	-0.50	-0.90	-0.18	-0.25	1.27	0.00	1.20	0.04	0.04	0.04	0.00	-0.83
3555	-0.33	-0.83	-0.83	-0.18	-0.61	-1.38	-3.93	-0.21	-0.30	2.80	1.00	1.58	-0.74	-0.50	0.33	-0.14	-3.72
23386	-0.12	0.14	0.00	0.00	0.00	-0.02	-0.50	-0.11	0.00	0.86	0.00	0.78	0.15	0.00	0.07	0.00	-0.49
22101	-0.20	0.00	0.00	0.00	-0.40	-0.21	-1.26	0.00	-0.14	1.23	0.05	1.12	0.04	0.00	-0.19	0.05	-1.28
23328	-0.32	0.00	0.00	0.00	0.00	0.00	-0.57	-0.17	-0.14	0.75	0.00	0.75	0.00	0.00	0.00	0.00	-0.53
26511	-0.25	0.00	0.00	0.00	0.06	0.09	-0.42	-0.13	-0.05	0.62	0.00	0.63	0.00	0.00	0.00	0.00	-0.44
26267	-0.38	0.00	-0.08	0.00	-0.40	-0.43	-0.71	-0.10	-0.17	0.85	0.00	0.81	0.00	0.00	0.00	0.00	-0.67
5828	-0.18	-1.08	-0.71	-0.11	0.00	-0.42	-4.40	-0.15	-0.21	2.71	0.67	2.08	-1.08	-0.63	0.33	-0.10	-4.48
28197	-0.17	0.00	0.00	0.00	0.00	0.00	-0.55	0.00	0.00	0.50	0.00	0.60	0.00	0.00	0.00	0.00	-0.50
1651	-0.09	0.13	0.00	0.00	0.07	0.00	-0.39	-0.08	-0.05	0.78	0.00	0.81	0.14	0.00	0.09	0.00	-0.36
1935	0.00	0.09	0.00	0.00	0.06	0.00	-0.40	0.00	0.00	0.80	0.00	0.80	0.10	0.00	0.20	0.00	-0.36

Table	15	Cont.															
31186	0.00	0.08	0.00	0.00	-0.10	-0.40	-0.21	0.00	0.00	1.00	0.00	0.91	0.16	0.13	0.24	0.00	-0.17
31047	0.00	0.20	-0.05	0.00	0.00	-0.30	-0.33	-0.07	0.00	0.78	0.00	0.82	0.22	0.00	0.05	0.00	-0.30
36436	0.00	0.14	0.05	0.00	-0.07	0.00	-0.16	0.00	0.00	0.47	0.00	0.43	0.14	0.02	0.00	0.00	-0.21
36780	-0.04	0.00	0.00	0.00	-0.56	-0.95	-0.46	0.00	0.00	0.53	0.08	0.50	0.00	0.04	0.00	0.00	-0.33
36727	0.00	0.11	0.10	0.00	0.00	-0.02	-0.28	0.00	0.00	0.60	-0.04	0.65	0.10	0.12	0.11	0.00	-0.34
35326	0.00	0.22	0.00	0.00	0.10	0.00	-0.67	-0.07	0.00	1.00	0.00	1.00	0.20	0.00	0.00	0.00	-0.67
40844	-0.06	0.14	0.17	0.00	-0.28	-0.27	-0.20	0.00	0.00	1.13	0.11	1.00	0.14	0.29	0.31	0.00	-0.20
716	0.00	0.14	0.00	0.00	0.00	-0.12	-0.34	-0.04	0.00	0.67	0.00	0.62	0.14	0.00	0.13	0.00	-0.33
40322	0.00	0.24	0.07	0.00	0.18	0.03	-0.18	0.00	0.00	0.53	0.00	0.50	0.25	0.06	0.05	0.00	-0.28
806	0.00	0.20	0.00	0.00	-0.11	-0.33	-0.20	0.00	0.00	0.71	0.00	0.70	0.20	0.00	0.25	0.00	-0.11
45724	0.00	0.07	0.00	0.00	0.00	-0.38	-0.50	0.00	0.00	1.00	0.05	0.93	0.10	0.00	0.25	0.00	-0.43
1001	0.00	0.00	-0.15	0.00	-0.53	-1.00	-0.62	0.00	-0.09	0.86	0.00	0.91	0.06	0.04	0.29	0.00	-0.50
48048	0.00	0.05	0.13	0.00	0.24	-0.32	-0.49	0.00	0.00	1.25	0.00	1.23	0.05	0.18	0.39	0.00	-0.50
50879	0.00	0.11	0.00	0.00	-0.03	-0.15	-0.26	0.00	0.00	0.80	0.00	0.75	0.14	0.04	0.11	0.00	-0.25
2430	0.00	0.08	0.00	0.00	-0.39	-0.23	-0.98	-0.06	0.00	0.71	0.00	0.71	0.05	0.00	-0.16	0.00	-0.84
64684	0.00	0.18	0.00	0.00	0.12	-0.26	-1.00	0.00	0.00	1.84	0.00	1.78	0.20	0.00	0.44	0.00	-1.00
2437	0.00	0.22	0.00	0.00	0.59	0.28	-0.60	-0.04	0.00	1.13	0.00	1.16	0.20	0.00	0.17	0.00	-0.64
63664	0.00	0.06	0.00	0.00	0.50	0.25	-0.73	-0.02	0.00	1.00	0.00	1.00	0.04	0.00	0.11	0.00	-0.79
64713	0.00	0.33	0.15	0.00	-0.05	-0.31	-1.12	0.00	0.00	2.25	0.00	2.18	0.33	0.17	0.18	0.00	-1.09
2474	0.00	0.00	-0.17	0.00	0.24	0.25	-1.00	0.00	0.00	0.80	-0.04	0.92	0.00	-0.31	-0.07	0.00	-1.00
64217	-0.21	-0.24	-0.40	0.00	0.00	-0.18	-1.84	0.00	-0.14	1.12	0.00	1.10	-0.23	-0.40	0.09	0.00	-1.78
64193	0.00	-0.24	-0.50	0.00	0.00	-0.54	-1.86	0.00	0.00	1.80	0.11	1.67	-0.21	-0.43	0.50	0.00	-1.78
2456	0.00	0.27	0.00	0.00	0.25	0.10	-1.09	0.00	0.00	1.50	0.00	1.53	0.26	0.00	0.10	0.00	-1.10
64799	0.00	0.06	-0.15	0.00	0.00	-0.19	-1.25	0.00	0.00	1.46	0.00	1.45	0.05	-0.09	0.26	0.00	-1.18
64278	0.00	0.23	0.00	0.00	0.17	-0.03	-0.68	0.00	0.00	1.31	0.00	1.29	0.20	0.00	0.29	0.00	-0.69
3238	-0.14	-0.54	-0.47	-0.10	0.10	0.00	-3.00	-0.20	-0.42	2.25	0.27	1.94	-0.54	-0.50	0.17	-0.11	-3.00
3259	-0.14	-0.94	-0.67	-0.15	-0.26	-0.39	-3.46	-0.14	-0.09	2.00	0.40	1.68	-0.89	-0.60	0.08	-0.15	-3.36
5114	-0.12	-0.33	-0.18	0.00	-0.20	-0.38	-2.38	0.00	-0.17	2.00	0.20	1.78	-0.32	-0.08	0.14	0.00	-2.36
368	-0.05	-0.28	-0.25	-0.09	0.00	-0.17	-2.13	-0.06	-0.08	1.89	0.49	1.23	-0.26	-0.22	0.23	-0.08	-2.13
3838	-0.29	-0.33	-0.18	-0.05	0.00	-0.11	-2.07	-0.05	-0.23	1.67	0.50	1.00	-0.31	-0.15	0.08	-0.05	-2.07

Table	15	Cont.															
281	0.00	-0.47	-0.33	-0.05	-0.08	-0.32	-2.28	-0.08	-0.08	2.00	0.68	1.13	-0.41	-0.25	0.24	-0.08	-2.27
282	-0.14	0.20	0.00	0.00	0.00	-0.08	-1.40	0.00	-0.05	1.83	0.33	1.40	0.20	0.00	0.13	0.00	-1.38
34660	0.00	0.34	0.00	0.00	0.07	0.00	-0.07	0.00	0.00	0.79	0.00	0.76	0.34	0.00	0.14	0.00	0.00
5269	-0.38	-0.07	0.00	0.00	-0.19	-0.16	-2.00	0.00	-0.25	2.10	0.29	1.54	-0.04	0.00	0.00	0.00	-2.00
5961	-0.14	-0.28	-0.11	0.00	-0.07	-0.20	-1.67	0.00	-0.11	1.04	0.13	0.88	-0.25	-0.06	0.06	0.00	-1.62
5742	-0.08	0.00	-0.09	0.00	-0.32	-0.35	-2.67	0.00	-0.07	2.74	0.52	1.83	0.00	0.00	0.00	0.00	-2.59
14367	-0.17	-0.26	-0.14	-0.09	0.00	-0.13	-2.00	-0.02	-0.22	1.72	0.25	1.26	-0.25	-0.13	0.09	-0.10	-2.00
69	-0.06	0.00	-0.04	0.00	-0.14	-0.26	-1.09	-0.14	0.00	1.33	0.00	1.30	0.06	0.00	0.12	0.00	-1.05
14277	-0.15	0.00	-0.09	0.00	-0.08	-0.33	-1.13	-0.17	-0.22	1.31	0.00	1.25	0.00	0.00	0.10	0.00	-1.06
1094	-0.18	0.00	0.00	0.00	0.00	-0.21	-0.67	-0.19	-0.27	1.00	0.00	1.04	0.00	0.00	0.17	0.00	-0.60
13194	0.00	0.00	-0.10	0.00	-0.10	-0.42	-1.42	-0.09	-0.18	1.80	0.00	1.68	0.00	0.00	0.25	0.00	-1.38
9534	-0.37	-0.82	-0.60	-0.15	-0.39	-0.87	-2.94	-0.22	-0.30	1.89	0.52	1.42	-0.78	-0.33	0.20	-0.13	-2.78
10127	-0.56	-0.52	-0.45	-0.11	-0.33	-0.63	-2.64	-0.43	-0.46	2.33	0.33	2.00	-0.50	-0.33	0.18	-0.12	-2.57
10926	-0.50	-0.50	-0.25	-0.08	-0.39	-0.87	-2.17	-0.07	-0.21	1.78	0.36	1.32	-0.45	0.00	0.36	-0.06	-2.07
9219	-0.65	-0.25	-0.18	-0.06	-0.25	-0.67	-1.95	-0.12	-0.28	2.00	0.30	1.65	-0.22	-0.06	0.27	0.00	-1.82
15100	-0.25	0.00	-0.13	0.00	-0.18	-0.34	-1.23	-0.05	-0.12	1.40	0.13	1.24	0.00	0.00	0.18	0.00	-1.14
15177	-0.19	0.00	-0.11	0.00	-0.07	-0.20	-1.04	-0.12	-0.15	1.20	0.13	1.08	0.00	0.00	0.10	0.00	-1.00
14534	-0.08	0.00	0.00	0.00	-0.07	-0.10	-0.67	-0.10	-0.14	1.00	0.00	1.00	0.00	0.00	0.05	0.00	-0.59
13592	-0.21	0.04	0.00	0.00	-0.14	-0.20	-0.55	-0.15	-0.13	0.76	0.00	0.77	0.06	0.00	0.07	0.00	-0.47
7272	-0.39	-0.61	-0.20	0.00	-0.20	-0.20	-2.00	0.00	-0.13	1.38	0.29	1.00	-0.62	-0.20	0.00	0.00	-2.00

Table 17. Analysis method for the presented parameters (modified after Garmo et al., 2014)

abbreviation	variable name	unit	analysis method	instrument	detection limit
pH	pH		Potentiometric	Methrom Titrino E702 Sm	
Cond	Conductivity	mS m <sup>-1</sup> 25C	Electrometric	WTW LF 539 RS	0.2
Ca	Calcium	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.02
Mg	Magnesium	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.02
Na	Sodium	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.02
K	Potassium	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.02
Cl	Chloride	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.03
SO4	Sulphate	mg l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	0.04
NO3-N	Nitrate	µg N l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	1
NH4-N	Ammonium	µg N l <sup>-1</sup>	Ion chromatography	Dionex DX 320 duo	5
Alk	Alkalinity	mmol l <sup>-1</sup>	Potentiometric titration to pH 4.5 Oxidation of CO <sub>2</sub> with UV/persulphate and measurement with IR detector	Methron Titrino E702 SM	0.01
TOC	Total organic carbon	mg C l <sup>-1</sup>		Phoenix 8000	0.1
Al/R,	Reactive and non-labile Aluminium	µg l <sup>-1</sup>	automated photometry	Skalar SAN Plus Autoanalyser	5
Al/II	Reactive and non-labile Aluminium	µg l <sup>-1</sup>	automated photometry	Skalar SAN Plus Autoanalyser	5
LAL	Labile Aluminium	µg l <sup>-1</sup>	calculated as difference between Al/R and Al/II		
Tot-N	Total nitrogen	µg N l <sup>-1</sup>	automated photometry	S208 oxidation in autoclave Skalar SAN plus Autoanalyser	10



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