

# Critical loads and the MAGIC model. Evaluating the country-scale applications in Norway using data from the 2019 national lake survey



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<p>Summary</p> <p>The critical loads methodology used at the national scale in Norway and the MAGIC model as applied to the lakes in the national 1000-lake survey have been evaluated, making use of the data from the 2019 national lake survey. For the acid-sensitive lakes in southern/middle Norway the MAGIC simulations based on data from the 1995 survey gave an acceptable prognosis for the lakewater chemistry measured in the 2019 resurvey. Several potential improvements are suggested for a re-calibration of MAGIC using the 2019 data, including two-point calibration. This can be used to evaluate scenarios of changed acid deposition in concert with other environmental drivers, such as climate change and altered forestry practices. The future role of N deposition is of special interest. Most lakes in the areas with critical load exceedance were not acidified. Several possible explanations for this are given. Testing various alternative values for inputs and parameters in the critical loads and exceedance calculations showed that certain combinations of changes could have marked effects on the critical load exceedance. Two approaches to updating the critical loads are suggested: 1) Calculate critical loads for the 1000 lakes and transfer the results to the national grid. 2) Keep the current setup but updating some of the inputs. Independent of choosing 1) or 2), the FAB model parameters should be re-visited.</p>
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**Critical loads and the MAGIC model**  
Evaluating the country-scale applications in  
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survey

# Preface

This project has been funded by the Norwegian Environment Agency and is related to NIVA's role as National Focal Centre under the UNECE International Cooperative Programme on Modelling and Mapping of Critical Loads & Levels and Air Pollution Effects, Risks and Trends (ICP M&M). The work on critical loads and dynamic modelling in the context of acid deposition is central to this role, and the current report provides background for the future work. The report also has a wider scope, as both methods are and can be used in national projects on different scales and with various objectives.

Kari Austnes has been the project leader. She is the main author of the part on critical loads and has generally been responsible for collecting the various input data. Richard F. Wright is the main author of the MAGIC part. James E. Sample has conducted the critical loads and exceedance calculations, while François Clayer has been involved in the classification of acidification status and comparisons with critical load exceedance data. Thanks to Wenche Aas (NILU) for deposition data, Espen Lund for map layout and to Magnus Norling for input on Monte-Carlo routines.

The project has been conducted under the contract 20087253 from the Norwegian Environment Agency. The contact person has been Gunnar Skotte.

Oslo, 7<sup>th</sup> December 2020

*Kari Austnes*

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## Summary

Critical loads and dynamic modelling have been essential in understanding and mitigating the effects of acid deposition in surface waters. The objective of this study was to evaluate the critical loads methodology used at the national scale in Norway and the Model of Acidification of Groundwater In Catchments (MAGIC) as applied to the lakes in the national 1000-lake survey. The intent was to assess which parts of the methodologies are most important to improve and which parts *can* be improved in terms of new knowledge and data.

The 2019 re-survey of 1000 lakes in Norway provides a platform for the evaluation of the MAGIC model forecasts based on the 1995 survey. The comparison rests on the premise that the actual decrease in sulphur (S) and nitrogen (N) deposition 1995-2019 followed that of the forecast scenario used in MAGIC. For the acid-sensitive lakes in southern/middle Norway the MAGIC simulations based on data from the 1995 survey gave an acceptable prognosis for the lakewater chemistry measured in the 2019 resurvey. In Sørlandet the discrepancy in acid neutralising capacity (ANC) can be explained by the fact that seasalt deposition in 2019 was lower than expected. In Møre and Romsdal the discrepancy was due to an unexpected increase in calcium (Ca) concentrations. For the lakes in both Sørlandet and Møre and Romsdal the increase in Ca concentrations in the lakes cannot be explained by changes in sulphur or seasalt deposition. Some other factor such as year-to-year variations in temperature or precipitation must be behind the changes observed. Further research is needed to understand the observed increase in Ca concentration.

MAGIC can be re-calibrated to the 2019 lake data and thus produce a new platform for forecasting future changes in acidification of lakes in Norway. Re-calibration could make use of the most recent developments and improvements in the MAGIC model. MAGIC will be incorporated in the new MOBIUS modelling platform. The most obvious way to improve the calibration of MAGIC is to use data from more than one point in time. Several other potential improvements are suggested. Re-calibration of the 2019 dataset can be used to evaluate scenarios of changed acid deposition in concert with other environmental drivers, such as climate change and altered forestry practices. The future role of N deposition is of special interest because N deposition plays an increasingly important role as S deposition has decreased substantially over the past 30 years.

The critical loads methodology was evaluated by comparing the acidification status (according to the Water Framework Directive (WFD) classification) of the lakes in the 2019 1000-lake dataset with the critical load exceedance. Very few lakes were classified as acidified in 2019. Most lakes in the areas with critical load exceedance were not acidified. This mismatch could be due to the different nature of the data, not all lakes being acid sensitive, particularly high ANC in 2019 and somewhat lower actual deposition than the one used in the exceedance calculations. However, it could also be related to the WFD classification system and/or the inputs and fixed parameters used in the critical loads and exceedance calculations.

Testing various alternative values for inputs and parameters in the critical loads and exceedance calculations for the two different critical loads models (The Steady-State Water Chemistry (SSWC) model and the First-order Acidity Balance (FAB) model) showed that individual changes or certain combinations of changes in the values could have marked effects on the critical load exceedance. Comparing two methods for assigning pre-industrial base cation concentration to the critical loads

grid, both based on MAGIC modelling, indicated that the current approach is the best. If MAGIC output can be used directly this could be a better approach.

Two different approaches to updating the critical loads are suggested: 1) To calculate critical loads for the lakes in the national lake survey and transfer the results to the national grid. This would require a re-calibration of the MAGIC model to these lakes, but it would give a totally updated basis for the critical loads. 2) Updating the total organic carbon (TOC) and nitrate concentration using data from the national lake survey but continue using the same pre-industrial base cation concentration. Independent of choosing 1) or 2), the FAB parameters should be re-visited, in particular nitrogen immobilisation and denitrification. The land cover distribution could be updated, and the new discharge normal should be used when available.

# Sammendrag

Tittel: Tålegrenser og MAGIC-modellen. Evaluering av anvendelsen på land-skala i Norge ved hjelp av data fra den nasjonale innsjøundersøkelsen i 2019

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Forfatter(e): Kari Austnes, Richard F. Wright, James E. Sample, François Clayer

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Tålegrenser og dynamisk modellering har vært sentrale i å forstå og begrense effekter av sur nedbør i overflatevann. Formålet med denne studien var å evaluere tålegrensemetodikken som blir brukt på nasjonalt nivå i Norge og MAGIC-modellen (Model of Acidification of Groundwater In Catchments) slik den er benyttet for innsjøene i den nasjonale 1000-sjøers-undersøkelsen. Intensjonen var å vurdere hvilke deler av metodikken det er viktigst å forbedre og hvilke deler som *kan* forbedres med tanke på kunnskap og data.

Re-undersøkelsen av 1000 innsjøer i Norge i 2019 gir en basis for evaluering av MAGIC-modellframskrivinger basert på 1995-undersøkelsen. Sammenligningen bygger på premisset om at den faktiske nedgangen i svovel- (S) og nitrogenavsetning (N) 1995-2019 var den samme som i framtidsscenarioet som ble benyttet i MAGIC. For de forsuringfølsomme innsjøene i Sør-Norge gav MAGIC-simuleringene basert på data fra 1995-undersøkelsen en akseptabel prognose for innsjøkjemien målt i 2019. På Sørlandet kan avviket i syrenøytraliserende kapasitet (ANC) forklares med at sjøsaltavsetningen i 2019 var lavere enn forventet. I Møre og Romsdal skyldtes avviket en uventet økning i kalsiumkonsentrasjonen (Ca). Hverken for innsjøene på Sørlandet eller i Møre og Romsdal kan økningen i Ca-konsentrasjon i innsjøene forklares med endringer i svovel- eller sjøsaltavsetning. Andre faktorer som år-til-år-variasjon i temperatur eller nedbør må ligge bak de observerte endringene. Det trengs videre forskning for å forstå den observerte økningen i Ca-konsentrasjon.

MAGIC kan re-kalibreres til 2019-dataene og dermed danne en ny plattform for prognoser for framtidige endringer i forsuring av innsjøer i Norge. En rekalkibrering kan benytte de siste utviklingene og forbedringene av MAGIC-modellen. MAGIC vil bli integrert i den ny MOBIUS modellplattformen. Den mest åpenbare måten å forbedre kalibreringen av MAGIC, er å bruke data fra mer enn ett tidspunkt. Flere andre potensielle forbedringer er foreslått. Rekalkibrering av 2019-datasettet kan brukes til å evaluere scenarier for endret sur nedbør i samspill med andre miljødrivere, som klimaendringer og endrede skogbruksmetoder. Den framtidige rollen til N-avsetning er spesielt interessant, fordi N-avsetning spiller en stadig viktigere rolle siden S-avsetningen har avtatt betraktelig over de siste 30 årene.

Tålegrensemetodikken ble evaluert ved å sammenligne forsuringstatusen (i henhold til klassifiseringen under Vannforskriften) til innsjøene i 1000-sjøers-datasettet for 2019 med tålegrenseoverskridelsen. Veldig få innsjøer ble klassifisert som forsuret i 2019. De fleste av innsjøene med tålegrenseoverskridelse var ikke forsuret. Denne uoverensstemmelsen kan skyldes de ulike typene data, at ikke alle innsjøene er forsuringfølsomme, spesielt høy ANC i 2019 og noe lavere faktisk avsetning enn den som ble brukt i overskridelseberegningene. Den kan imidlertid også være



knyttet til klassifiseringssystemet under Vannforskriften og/eller data eller faste parametere benyttet i tålegrense- og overskridelseberegningene.

Testing av forskjellige alternative verdier for data og parametere i tålegrense- og overskridelseberegningene for de to forskjellige tålegrensemodellene (The Steady-State Water Chemistry (SSWC) model and the First-order Acidity Balance (FAB) model) viste at enkeltendringer eller spesielle kombinasjoner av endringer i verdiene kan ha betydelig effekt på tålegrenseoverskridelsene. Sammenligning av to metoder for å tilegne før-industriell basekationkonsentrasjon til tålegrenserutenettet, begge basert på MAGIC-modellering, antydte at den nåværende tilnærmingen er den beste. Hvis MAGIC-resultater kan brukes direkte, kunne dette være en bedre tilnærming.

To forskjellige tilnærminger til å oppdatere tålegrensene er foreslått: 1) Å beregne tålegrenser for innsjøene i den nasjonale innsjøundersøkelsen og overføre disse resultatene til det nasjonale rutenettet. Dette ville kreve en recalibrering av MAGIC for disse innsjøene, men det ville gi en totalt oppdatert basis for tålegrensene. 2) Oppdatere konsentrasjonen av totalt organisk karbon (TOC) og nitrat ved å bruke data fra den nasjonale innsjøundersøkelsen, men fortsette å bruke den samme før-industrielle basekationkonsentrasjonen. Uavhengig av valget av 1) eller 2) bør FAB-parameterene vurderes igjen, spesielt nitrogenimmobilisering og denitrifisering. Arealfordelingen kan oppdateres og den nye vannføringsnormalen bør benyttes når denne er tilgjengelig.

# 1 Introduction

Atmospheric transport of acidifying compounds from industrial emissions and the subsequent deposition of such compounds resulted in widespread acidification of surface waters in Europe and North America in the last century. Surface water acidification was one of the environmental impacts of air pollution that led to the signing of the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP)<sup>1</sup> in 1979.

The concept of critical loads has been central to the work on emission reductions under the LRTAP Convention (CLRTAP, 2015b) and is a basis of both the sulphur protocol (1994) and the multi-pollutant protocol (the Gothenburg protocol, 1999). Critical loads for acidification of surface waters quantify the acid deposition that an area can tolerate without negative effects on aquatic biota. Exceedance of the critical loads means that the systems receive a higher deposition load than they can tolerate. The exceedance depends both on the critical load and the deposition level. Critical loads are steady-state quantities (CLRTAP, 2017). This means that the exceedance reflects the conditions at the point in time at which the ecosystem has fully adapted to a given level of acid deposition. If the current deposition level no longer exceeds the critical load, it does not necessarily mean that the system is no longer acidified, as a new steady state may not yet have been reached.

Critical loads are useful in many respects, but to estimate the acidification at a certain point in time, dynamic models are needed. Model of Acidification of Groundwater In Catchments (MAGIC) is widely used to model surface water acidification (Cosby et al., 1985a; Cosby et al., 1985b; Cosby et al., 2001). The model is calibrated against measured soil and water chemistry and simulates historical and future water chemistry based on deposition scenarios.

The MAGIC model has many different uses, including evaluating the need for liming in the future (Bjerknes et al., 2004), assessing the effects of climate and forestry practices on acidification, modelling nitrogen processes (Oulehle et al., 2015), estimating the deposition level that will give acceptable conditions in a specific year (target loads) (Posch et al., 2019), making a MAGIC library providing MAGIC output for non-modelled lakes from similar MAGIC modelled lakes (Moldan et al., 2020), and estimating reference conditions (Wright and Cosby, 2012). The latter is a link to the critical loads, as the pre-industrial base cation concentration is an essential component in the estimation of critical loads.

In 2019 a national lake survey was conducted in Norway (Hindar et al., 2020). This was a re-sampling of the roughly 1000 statistically-selected lakes that were sampled in 1995 (Skjelkvåle et al., 1996; Henriksen et al., 1998). The lakes were selected by stratified random sampling, ensuring that lakes from all counties were included, but with half the lakes from southern Norway, one third from middle Norway and one sixth from northern Norway. The reasoning behind this was that the pollution pressure is larger in the south. A larger proportion of the lakes in the larger size classes were included, but given that there are generally more small lakes, the majority of the sampled lakes were small (about 40% < 0.1 km<sup>2</sup>). The MAGIC model has been calibrated for these lakes based on the 1995 data (Larssen et al., 2008a; Austnes et al., 2016).

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<sup>1</sup> <http://www.unece.org/environmental-policy/conventions/envlrtapwelcome/the-air-convention-and-its-protocols/the-convention-and-its-achievements.html>

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The objective of this study was to evaluate the critical loads methodology used at the national scale in Norway and the MAGIC model as applied to the lakes in the national 1000-lake survey. The data from the 2019 survey were useful in this context. The intent was to assess which parts of the methodology are most important to improve and which parts *can* be improved in terms of new knowledge and data. A central aspect was also how the 1000-lake data can be used to improve the critical loads and exceedance calculations. The International Cooperative Programme on Modelling and Mapping of Critical Levels and Loads and Air Pollution Effects, Risks and Trends (ICP M&M) of the LRTAP Convention issued a Call for Data 2019-2021, including a review of the steady-state critical loads. In light of the upcoming review of the Gothenburg protocol, the critical loads should be up to date and policy relevant. A revision of the Norwegian critical loads for surface waters could be a contribution to the Call for Data, even if it is not possible to reach the deadline 1<sup>st</sup> March 2021.

## 2 The MAGIC model and the 1000-lake survey

### 2.1 Background

The data from the 1995 1000-lake survey (Skjelkvåle et al., 1996) has been used to generate forecasts for future lake water chemistry based on various scenarios for future deposition of sulphur (S) and nitrogen (N). One of the forecast scenarios was the “current legislation emission scenario” (CLE) of the European Monitoring and Evaluation Programme (EMEP) of the LRTAP Convention. The MAGIC model was used, and the calibration proceeded using MAGIC in a batch mode. This was first done by Larssen et al. (2008a) and more recently updated by (Austnes et al., 2016). The resulting forecasts for lake water chemistry for the year 2019 can now be compared with the actual observed data from the 2019 re-survey of the lakes (Hindar et al., 2020). This is an exercise similar to that made by Helliwell et al. (2014) for MAGIC model forecasts for acidification and recovery of lakes in several regions of Europe.

### 2.2 Deposition 1995-2019 – measured and modelled

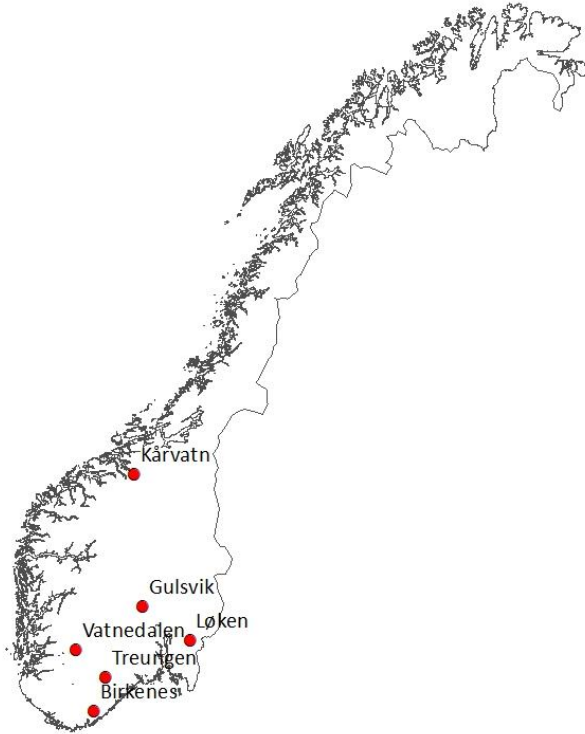
The first premise for the comparison of modelled with observed data for 2019 is that the CLE scenario agrees with the actual decline in S and N deposition over the period 1995-2019. Here we can use the observed deposition data from Norwegian Institute for Air Research (NILU) stations in Norway. In addition, the forecasts assume no systematic changes in the deposition of seasalts or in the amount of precipitation.

Six of the NILU stations in southern/middle Norway have observed deposition data starting in the 1970s: Birkenes, Vatnedalen and Treungen are in the region “Sørlandet”, Løken and Gulsvik (now Brekkebygda) in “Østlandet” and Kårvatn in “Møre and Romsdal” (**Figure 1**).

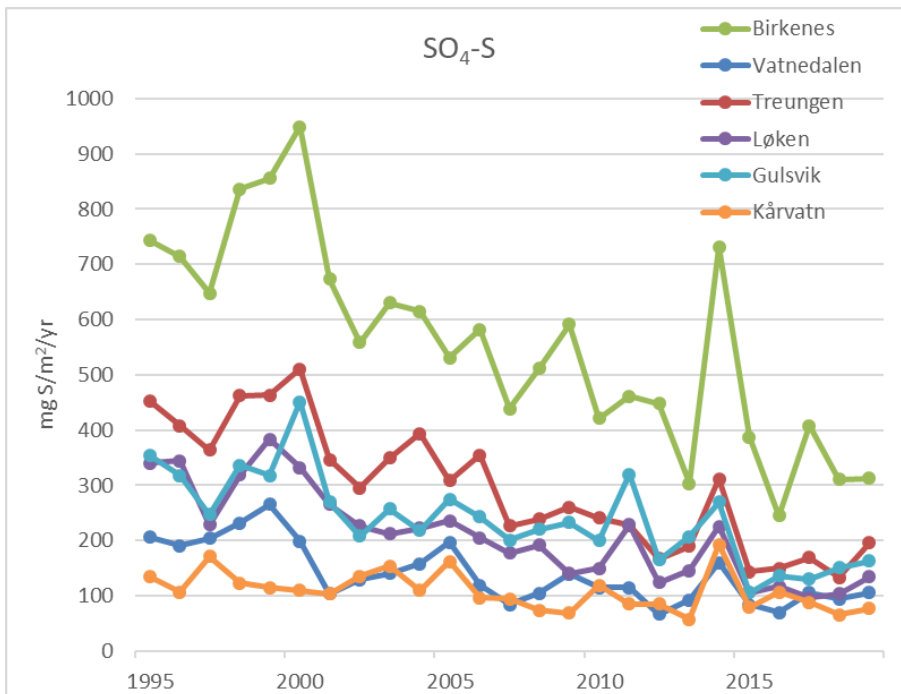
**Sulphur.** The measured S deposition (non-seasalt fraction) decreased by about 50-60% over the period 1995-2019 at the selected NILU stations (**Figure 2**).

Each of these stations lies within an EMEP grid cell, and the forecast EMEP S deposition for the period 1995 to 2019 can be compared with the actual measured deposition by NILU. For comparison purposes the NILU measured and the EMEP forecast deposition were each scaled to the value for the year 1995 (i.e. 1995=1.0). These comparisons show that the actual deposition decreased less than

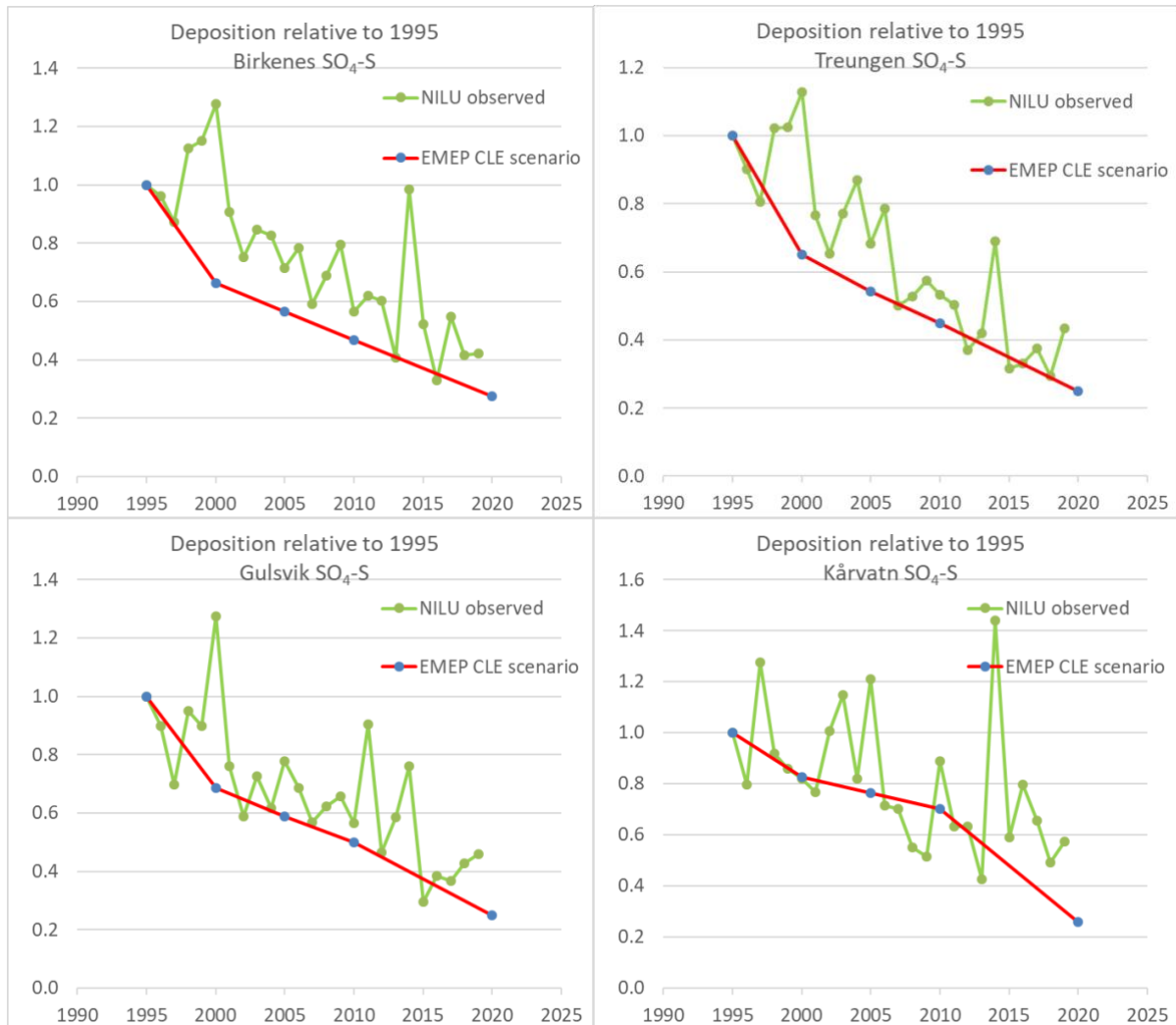
forecast by EMEP under the CLE scenario. The actual decrease in S deposition was about 50-60% whereas the EMEP deposition was forecast to decrease by about 70% (Figure 3).



**Figure 1.** Location of the six NILU stations used in the analysis. The station at Gulsvik was moved in the 1990s to nearby Brekkebygda.



**Figure 2.** Sulphur deposition (wet, SO<sub>4</sub>, non-seasalt fraction) measured at six NILU stations over the period 1995-2019. Data from Aas et al. (2020). The station at Gulsvik was moved in the 1990s to nearby Brekkebygda.

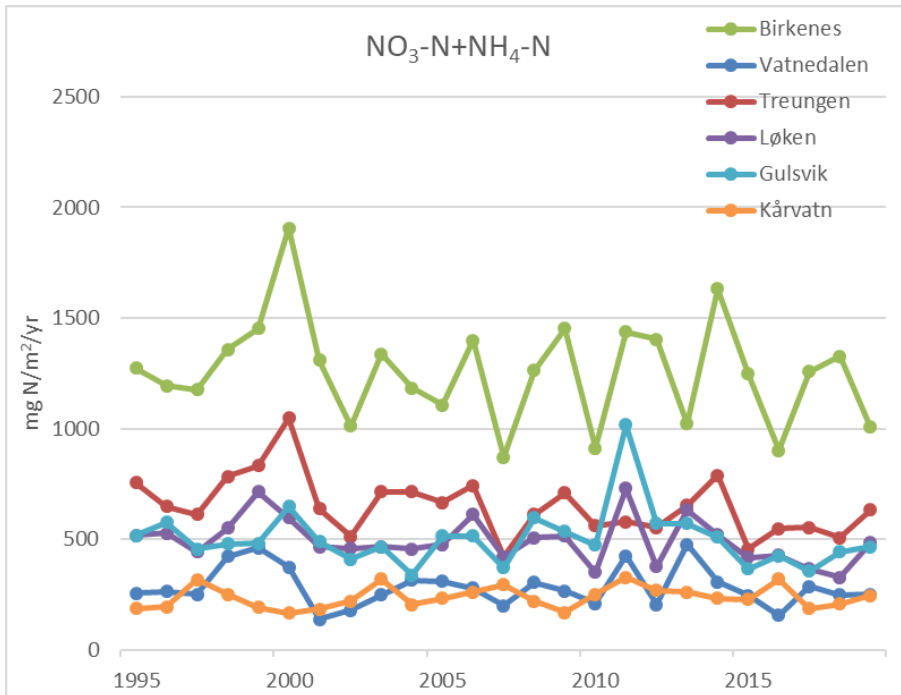


**Figure 3.** Sulphur deposition (wet,  $\text{SO}_4$ , non-seasalt fraction) measured and forecast (EMEP) for four stations in southern/middle Norway for the period 1995-2019. Values are scaled to the year 1995=1.0. NILU data from Aas et al. (2020). The station at Gulsvik was moved in the 1990s to nearby Brekkebygda. EMEP data courtesy of the Co-ordinating Centre for Effects (CCE) of the LRTAP Convention.

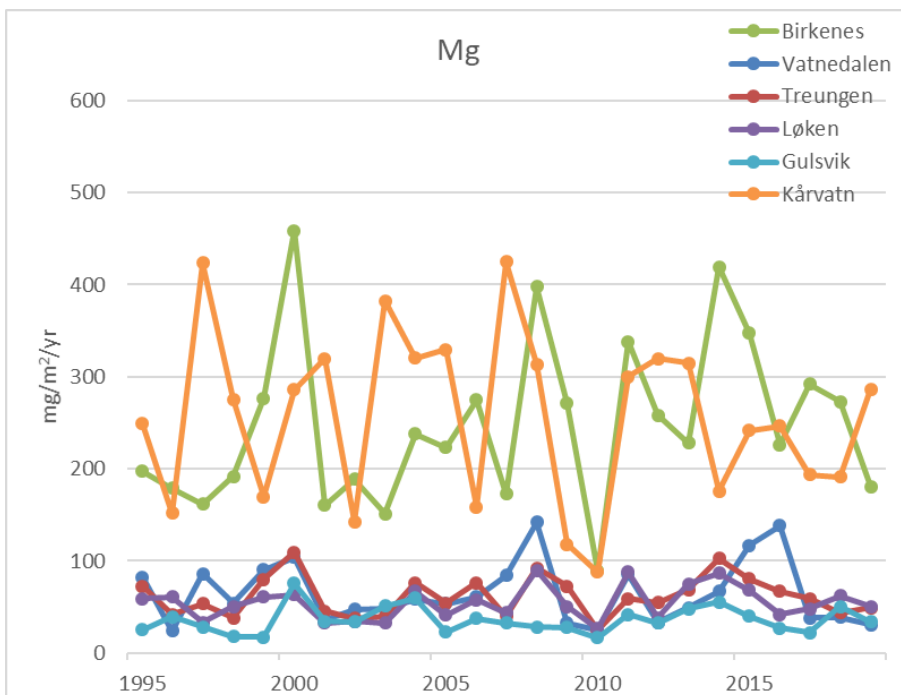
**Nitrogen.** Similarly, for N deposition ( $\text{NO}_3+\text{NH}_4$ ) the measured wet deposition at the selected NILU stations decreased by only 0-20% over the period 1995-2019, whereas the EMEP forecast for the CLE scenario was a 40-50% decrease (**Figure 4**).

**Seasalts (chloride).** There has apparently been no time-trend in the deposition of seasalts over the period 1995-2019 (**Figure 5**). NILU uses magnesium (Mg) deposition as the indicator for seasalts, as seasalt spray is the only significant source of Mg in deposition in Norway. We can assume that also chloride (Cl) deposition shows no change over time, although there are large year-to-year variations at stations near the coast, such as Birkenes.

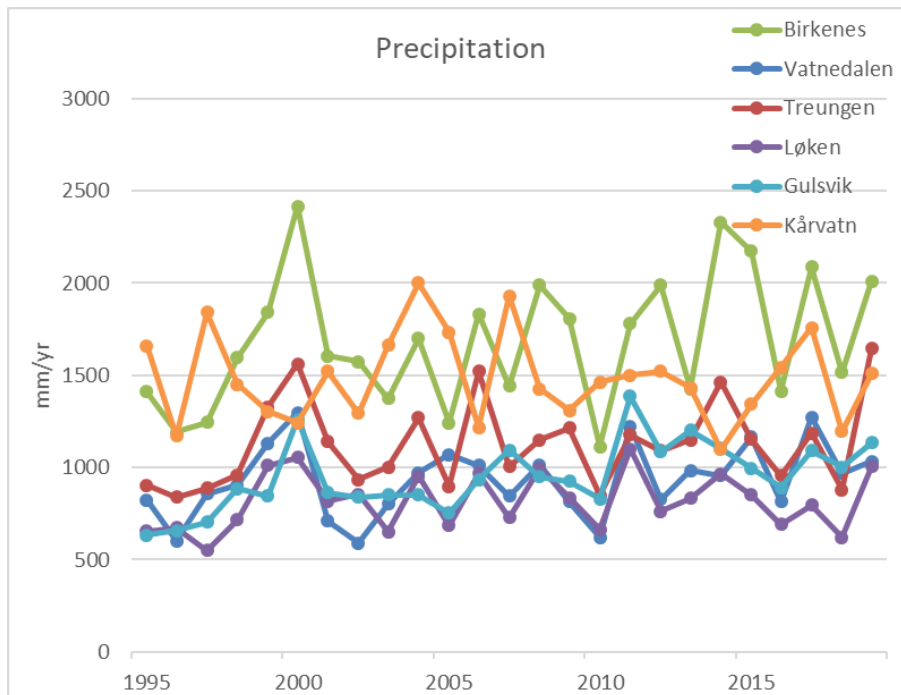
**Precipitation amount.** The amount of precipitation (mm/yr) measured at the six NILU stations also does not appear to have changed significantly over the period 1995-2019 (**Figure 6**). Again, there have been large year-to-year variations.



**Figure 4.** Nitrogen deposition (wet, NO<sub>3</sub>+NH<sub>4</sub>) measured at six NILU stations over the period 1995-2019. Data from Aas et al. (2020). The station at Gulsvik was moved in the 1990s to nearby Brekkebygda.



**Figure 5.** Magnesium wet deposition measured at six NILU stations over the period 1995-2019. NILU uses magnesium as a measure of seasalt deposition. Data from Wenche Aas (NILU). The station at Gulsvik was moved in the 1990s to nearby Brekkebygda.



**Figure 6.** Precipitation amount measured at six NILU stations over the period 1995-2019. Data from (Aas et al., 2020). The station at Gulsvik was moved in the 1990s to nearby Brekkebygda.

## 2.3 Lake chemistry 1995-2019. Observed and modelled by MAGIC

### 2.3.1 Selection of lakes

The 1995 survey was comprised of 1006 lakes, of which 990 were successfully calibrated by Larssen et al. (2008a), with an update by Austnes et al. (2016). The 2019 re-survey was comprised of 1000 lakes of which almost all had been sampled in 1995. Of these there were 962 lakes that had been sampled in 1995 and successfully calibrated by MAGIC. These 962 lakes form thus the basis for the comparison between observed and modelled lake water chemistry for lakes in Norway in 2019.

The MAGIC model is primarily intended to simulate the changes in lake water chemistry given changes in atmospheric deposition of major ions. Of central interest is the effect of deposition of sulphur and nitrogen compounds, but also of seasalt compounds such as chloride. Moderating factors include changes in precipitation amount, as this affects the concentration of all major components in surface waters.

The analysis of MAGIC simulated versus observed lake chemistry thus is relevant primarily in regions that receive significant amounts of S and N deposition, and for lakes that are inherently sensitive to acidification by acid deposition. We chose to focus our analysis on two regions in southern/middle Norway: Sørlandet and the county of Møre and Romsdal. Sørlandet comprises the four counties of Telemark, Aust-Agder, Vest-Agder and Rogaland (1995 county designation). Sørlandet is the region of Norway hardest impacted by acid deposition due to high levels of S and N deposition (for Norway). A large fraction of the lakes is sensitive to acid deposition. Møre and Romsdal county is located on the west coast in middle Norway. This region receives low levels of S and N deposition, and lakes are not significantly acidified. For both regions we included only lakes that had ANC < 100  $\mu\text{eq/l}$  in 1995 as

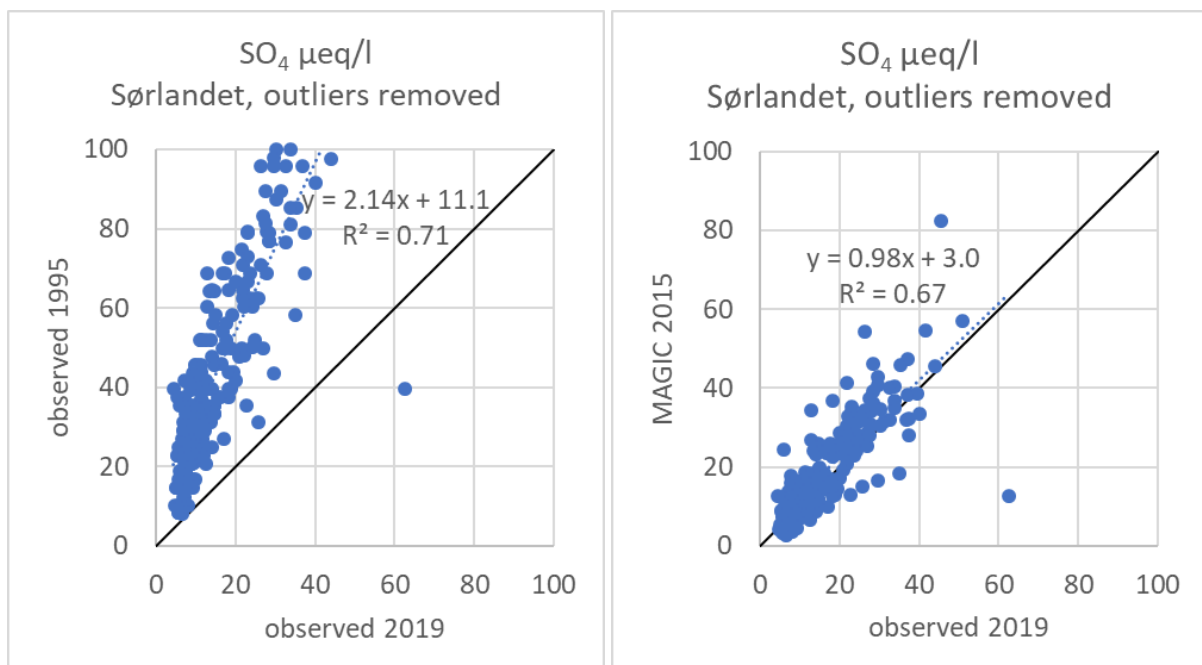
these are of most interest with respect to recovery from acid deposition. These are defined as sensitive.

The 1995 and 2019 datasets also include a number of outliers, that is lakes that have unusually high concentrations of one or more components. The inclusion of such lakes can skew the results of linear regression analyses. Together 8 outlier lakes in Sørlandet were excluded from further analysis (**Appendix A**). There were no outlier lakes in Møre and Romsdal.

The regions differ in the impact of acid deposition, but both regions experience changes in lake water chemistry over time due to other factors such as natural year-to-year fluctuations in seasalt deposition (reflecting frequency and severity of storms), precipitation amount and possible effects of climate change.

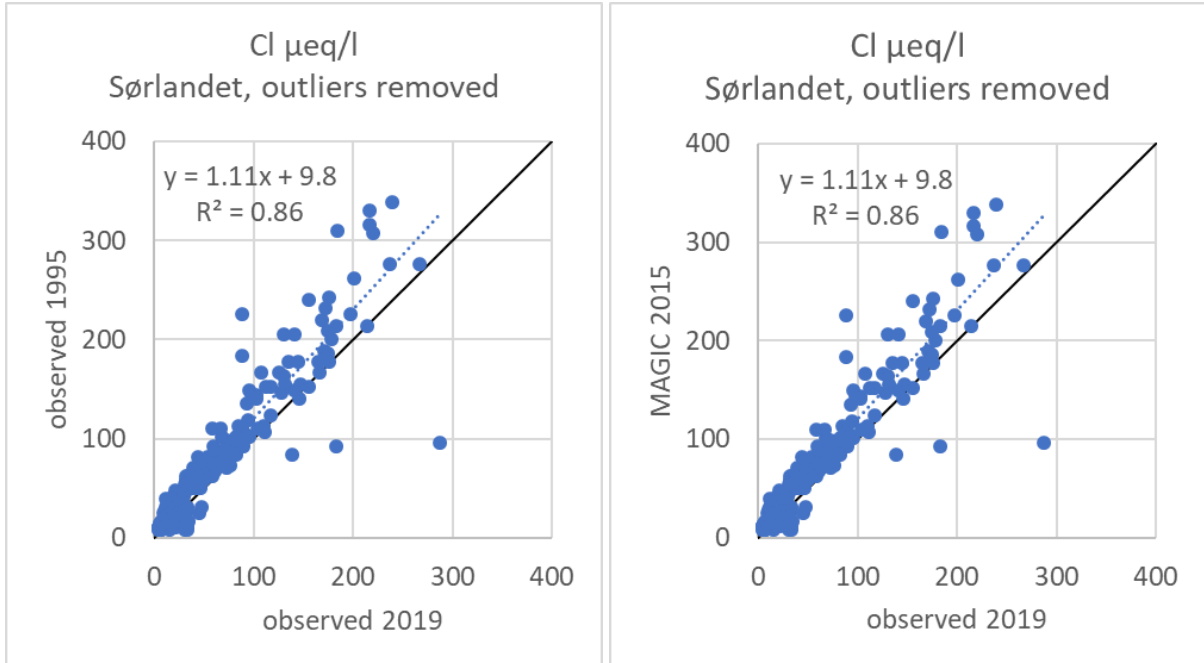
### 2.3.2 Sørlandet (southernmost Norway)

The 2019 1000-lake survey included 238 sensitive lakes in Sørlandet that had also been calibrated by MAGIC. The sulphate concentrations ( $\text{SO}_4$ ) forecast for the year 2019 by the MAGIC model for these lakes were on the average lower than the measured concentrations, but this is not unexpected as the EMEP prognosis for S deposition in 2019 was somewhat lower than the NILU measured deposition (**Figure 3**). The MAGIC forecast for 2015, in which the EMEP S deposition is slightly higher than in 2019, appears to fit the observed lake  $\text{SO}_4$  concentrations best (**Figure 7**). The linear regression of MAGIC 2015 on observed 2019 is highly significant and falls very close to the 1:1 line. As S deposition is an important driver of changes in lake chemistry over the period 1995-2019, we chose the MAGIC simulations for the year 2015 to be the best estimate of the expected concentrations in lakes in 2019. There were a few lakes that lie far from the 1:1 line; for these lakes one or more of the assumptions used in MAGIC to estimate the  $\text{SO}_4$  deposition may not be valid.

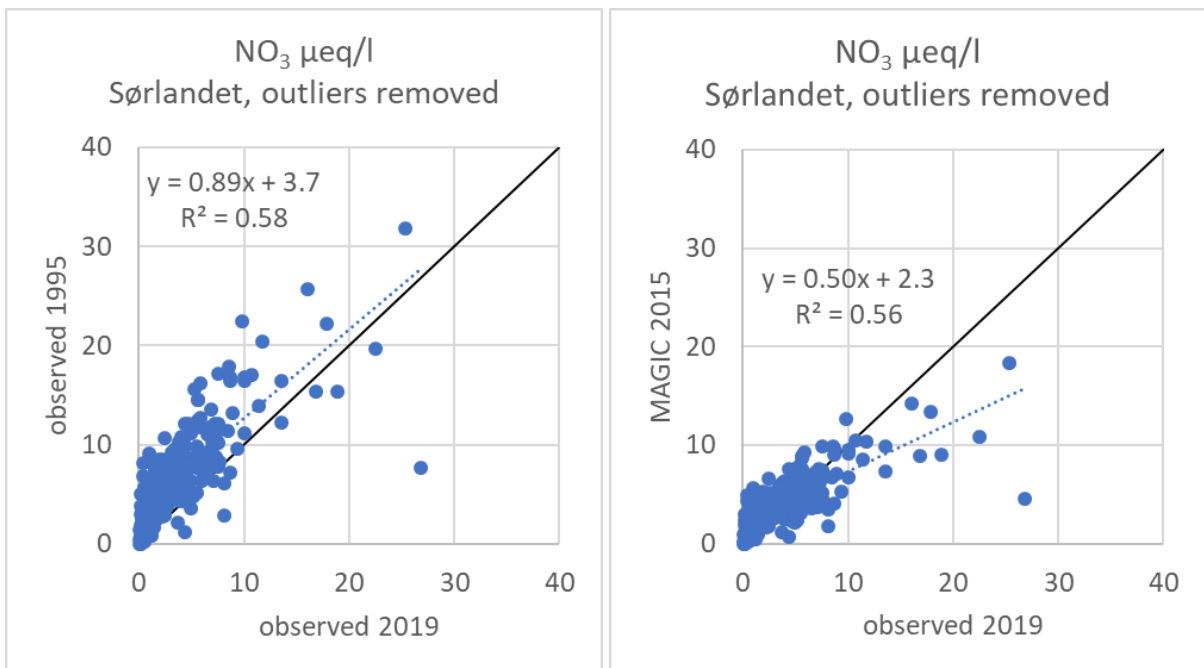


**Figure 7.**  $\text{SO}_4$  concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.





**Figure 8.** Cl concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.



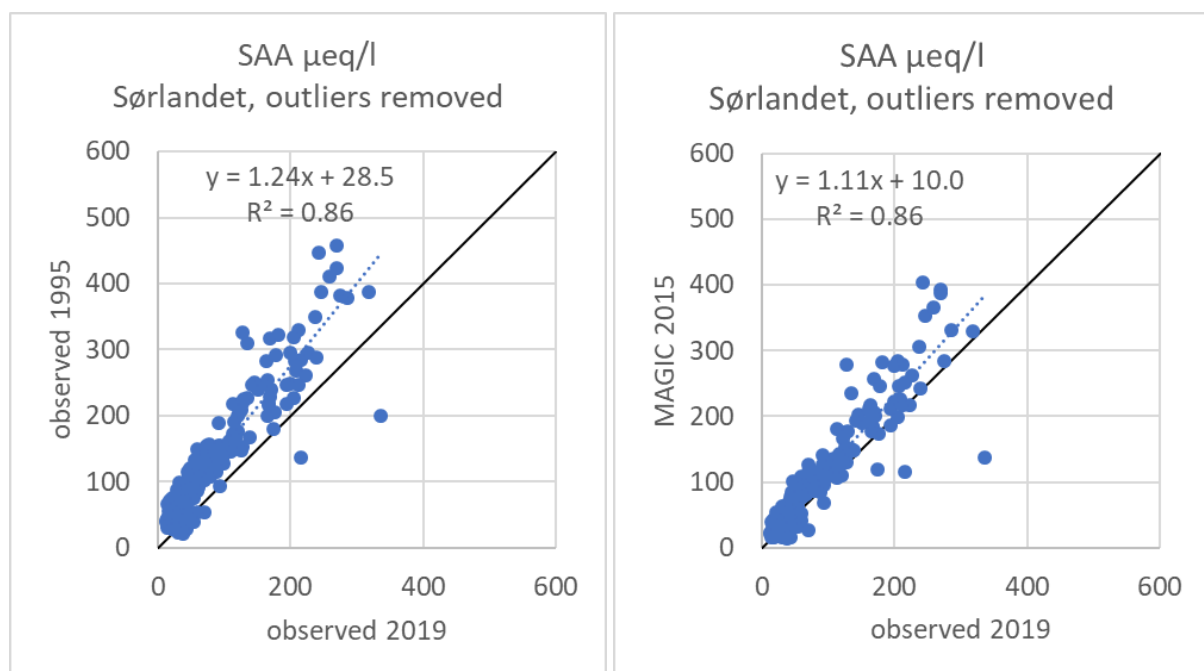
**Figure 9.** NO<sub>3</sub> concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

Chloride is the other strong acid anion of importance. Its major source is seasalt spray blown into the atmosphere and transported inland. Concentrations of Cl are highest in lakes near the coast and there is a strong gradient inland. The MAGIC simulations assumed that the future annual Cl deposition from 1995 to 2019 would remain at the same level as the annual Cl flux measured in the lakes in 1995. The observed data from 2019 showed that on average there was about 11% lower

concentrations of Cl in the 238 lakes relative to the data from 1995 (**Figure 8**). This was most probably due to lower amounts of seasalt deposition in 2019 relative to 1995. The measured data from the NILU deposition stations showed large year-to-year fluctuations in seasalt deposition (**Figure 5**: NILU uses Mg as a surrogate for Cl; deposition of both comes nearly entirely from seasalts).

Nitrate ( $\text{NO}_3$ ), the third acid anion, is presently at much lower concentrations than either  $\text{SO}_4$  or Cl.  $\text{NO}_3$  concentrations were generally about 10% lower in the lakes in Sørlandet in 2019 relative to 1995 (**Figure 9**). The change, however, amounted to only a few  $\mu\text{eq/l}$  in most lakes. The decrease can be ascribed to lower deposition of inorganic N (**Figure 4**) with about the same percent retention in the catchment-lake systems (Hindar et al., 2020).

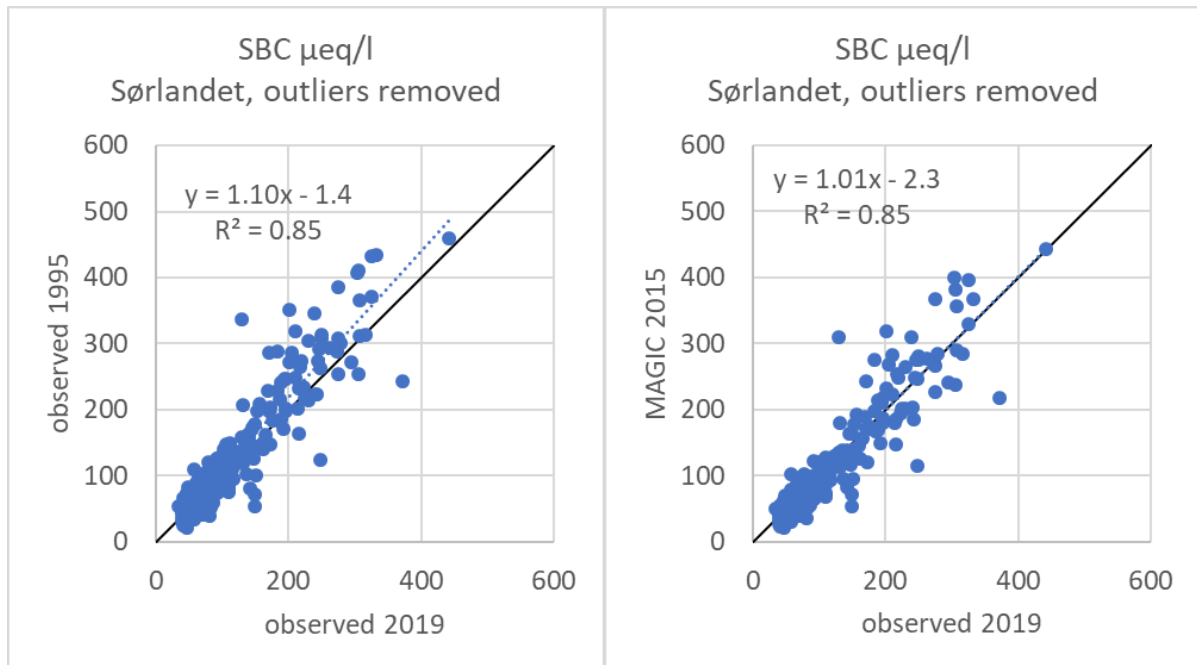
The sum of concentrations of  $\text{SO}_4$ , Cl and  $\text{NO}_3$  comprise the strong acid anions (SAA). In the 238 Sørlandet lakes SAA concentrations were higher in 1995 relative to 2019 (**Figure 10**). The decline in SAA is due in part to the decrease in  $\text{SO}_4$  concentrations (due in turn to lower S deposition) and in part to the lower Cl concentrations (due in turn to lower seasalt deposition) in 2019. As the MAGIC simulated  $\text{SO}_4$  concentrations in 2015 closely agree with the observed concentrations in 2019, and the  $\text{NO}_3$  concentrations are negligible, the scatter plot and linear regression of SAA concentrations of MAGIC 2015 on observed 2019 were nearly identical with that of Cl (**Figure 8**).



**Figure 10.** SAA (sum strong acid anions) concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

The strong acid anions  $\text{SO}_4$  and Cl are highly mobile in soil and largely independent of one another. The flux of these anions transports cations from the soil to lakewater. A fraction of the cations accompanying  $\text{SO}_4$  and Cl is comprised of the base cations Ca, Mg, Na and K (SBC=sum base cations) and a fraction is comprised of the acid cations  $\text{H}^+$  and inorganic-Al (often termed labile aluminium LAL). These six cations participate in cation-exchange reactions in the soil, which are then reflected in the concentrations in lake water. The 10-15% decrease in SAA concentrations in 2019 relative to

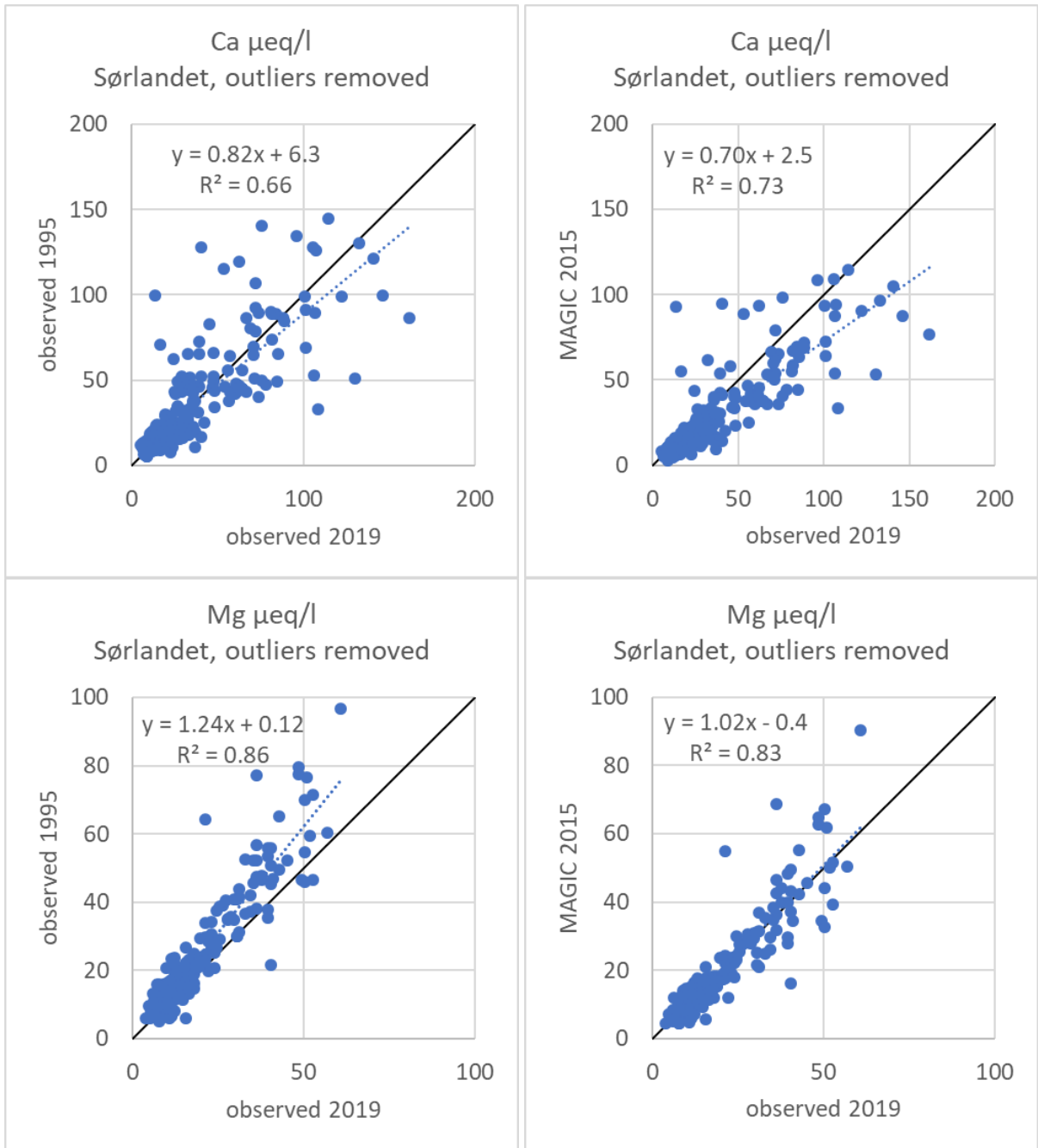
1995 can thus be expected to be manifest as lower concentrations of all cations, base cations as well as acid cations. The data show that SBC decreased by about 10% from 1995 to 2019 (left-hand panel, **Figure 11**). This implies that the acid cations also decreased over this period. The MAGIC simulations of SBC concentrations for 2015 agreed very well with the observed in 2019 (right-hand panel, **Figure 11**).



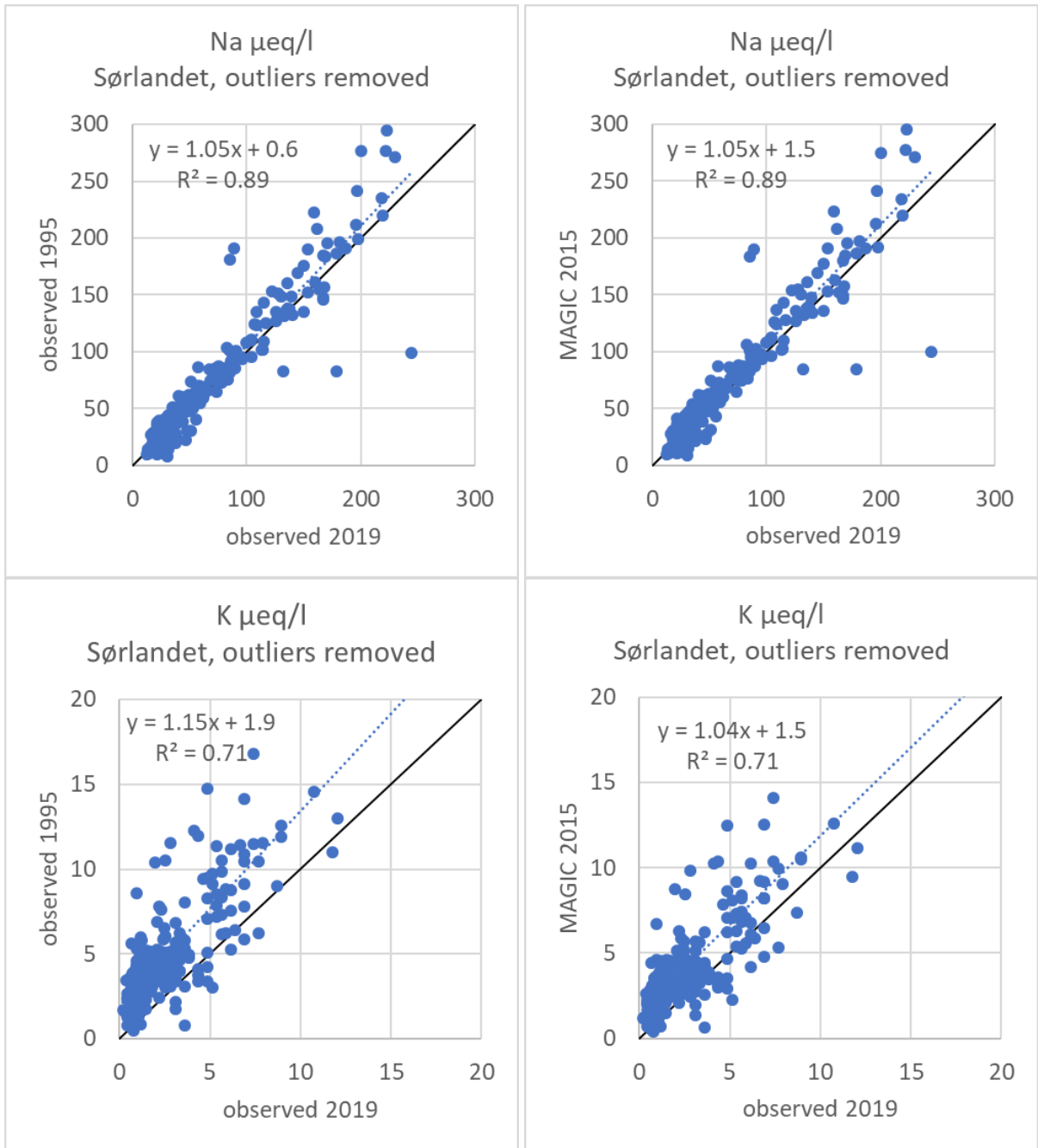
**Figure 11.** SBC (sum base cations) concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

Of the four base cations the concentrations of Ca showed increased concentrations between 1995 and 2019, while the concentrations of Mg, sodium (Na) and potassium (K) decreased (**Figure 12**, **Figure 13**). K concentrations were very low and contribute little to SBC. There is no obvious explanation for the higher concentrations of Ca observed in the lakes in Sørlandet in 2019 relative to 1995. The decrease in S deposition should have resulted in a small decrease in Ca concentration to compensate the decrease in  $\text{SO}_4$  concentrations. This is simulated by MAGIC. Some other factor must be invoked, such as year-to-year variations in weather conditions. Perhaps the dry summer of 2018 in southernmost Norway affected the Ca concentrations in lakewater in 2019. The other two dominant base cations, Mg and Na, show small but significant decreased concentrations in 2019 relative to 1995. This can be ascribed to lower seasalt deposition in 2019.

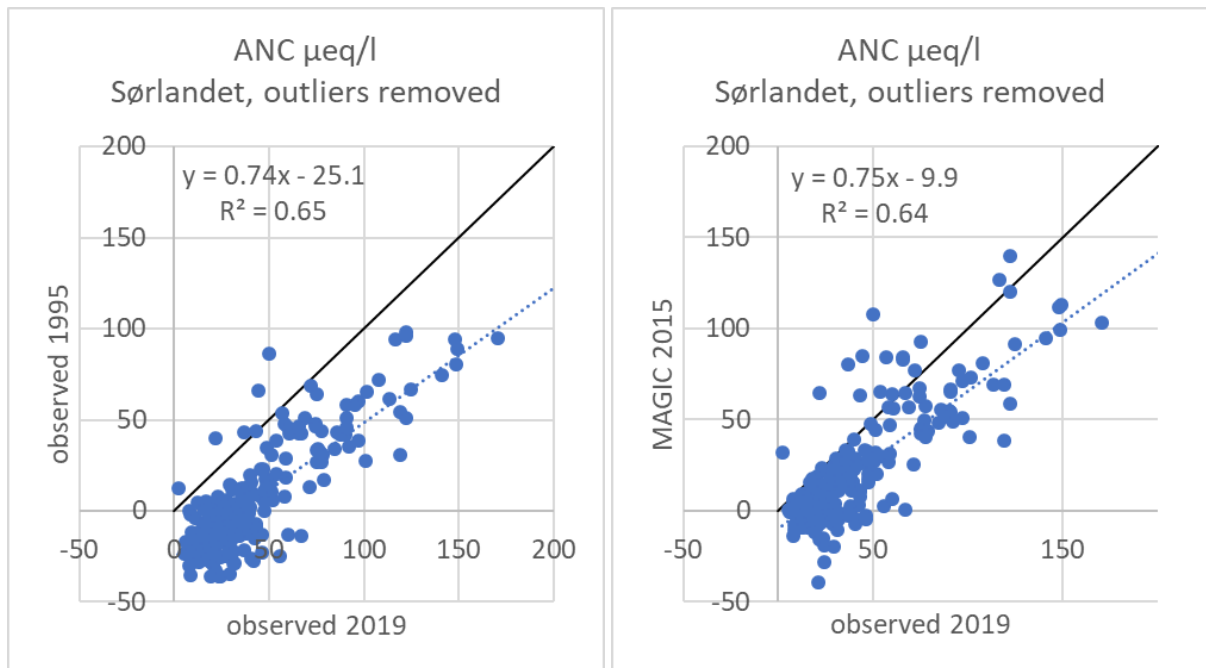
The difference between SBC and SAA is the ANC (acid neutralising capacity). This parameter is used as a measure of acidification of surface waters and low ANC is associated with damage to fish populations and other aquatic organisms. The observed data show that ANC has increased significantly between 1995 and 2019 (**Figure 14**). The lakes are recovering from acidification. The MAGIC simulations also forecast an increase in ANC but less than that measured. The discrepancy between the modelled and measured ANC in 2019 can be explained by the fact that seasalt deposition and thus the SAA concentrations in 2015 were lower than expected.



**Figure 12.** Ca and Mg concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panels) and simulated by MAGIC for the year 2015 (right-hand panels) relative to the observed values in 2019.



**Figure 13.** Na, and K concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panels) and simulated by MAGIC for the year 2015 (right-hand panels) relative to the observed values in 2019.

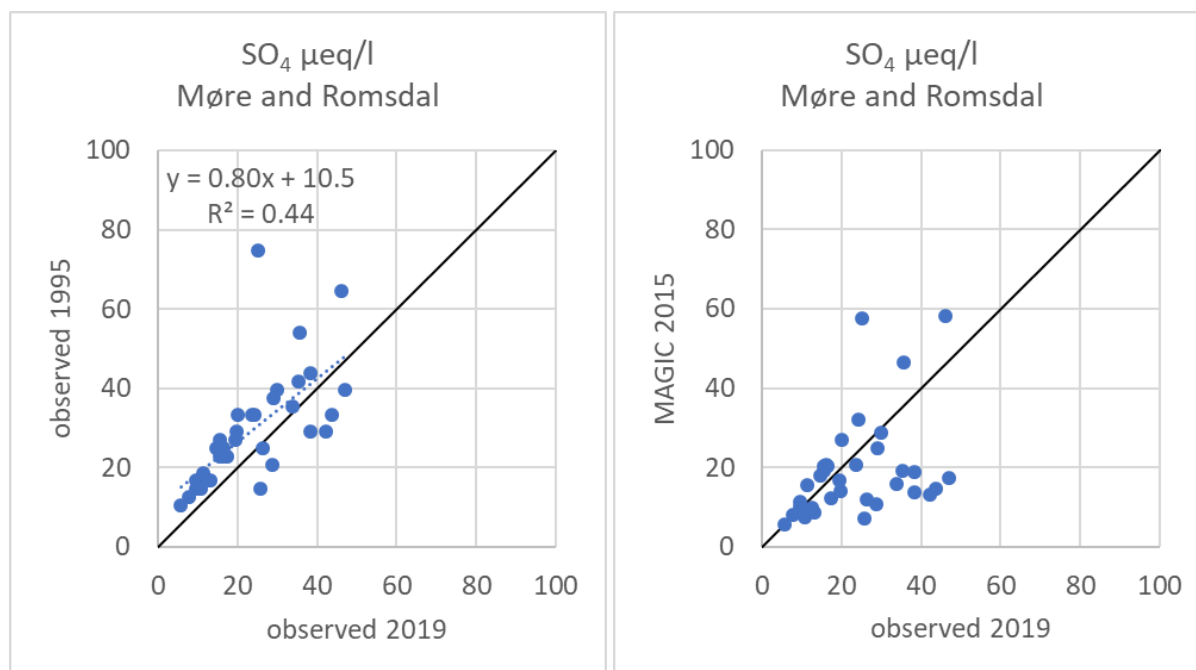


**Figure 14.** ANC (acid neutralising capacity) concentrations in 238 lakes in Sørlandet measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

### 2.3.3 Møre and Romsdal (west coast, middle Norway)

The 2019 1000-lake survey included 34 sensitive lakes in Møre and Romsdal that had also been calibrated by MAGIC. The region receives low levels of S and N deposition. Deposition of S has decreased about 45% over the period 1995 to 2019, from low levels to even lower levels (**Figure 2**). Deposition of N has not changed significantly.

The observed data show a small decrease from 1995 to 2019 in  $\text{SO}_4$  concentrations of about  $10 \mu\text{eq/l}$  in most of these lakes (**Figure 15**). A small number show an increase in  $\text{SO}_4$  concentrations and fall below the 1:1 line. The comparison of MAGIC simulations with the measured 2019 concentrations of  $\text{SO}_4$  in 2019 reveals two groups of lakes, one group falls close to the 1:1 line while the other group falls below the 1:1 line. The linear regression of MAGIC 2015 on observed 2019 for the 34 lakes is not significant.



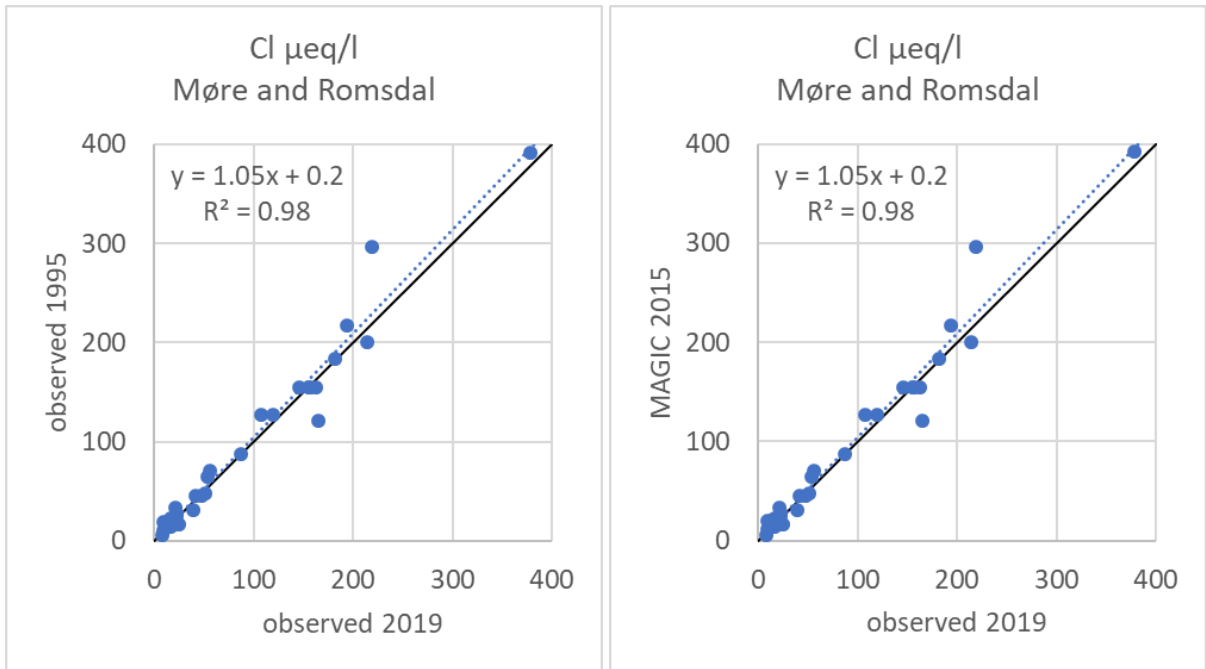
**Figure 15.** SO<sub>4</sub> concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019. The linear regression of MAGIC 2015 on observed 2019 (right-hand panel) is not significant.

Cl concentrations showed a very slight decrease between 1995 and 2019 (**Figure 16**). As the MAGIC simulations assumed no change in Cl deposition, the simulated and observed Cl concentrations in 2019 agreed very well.

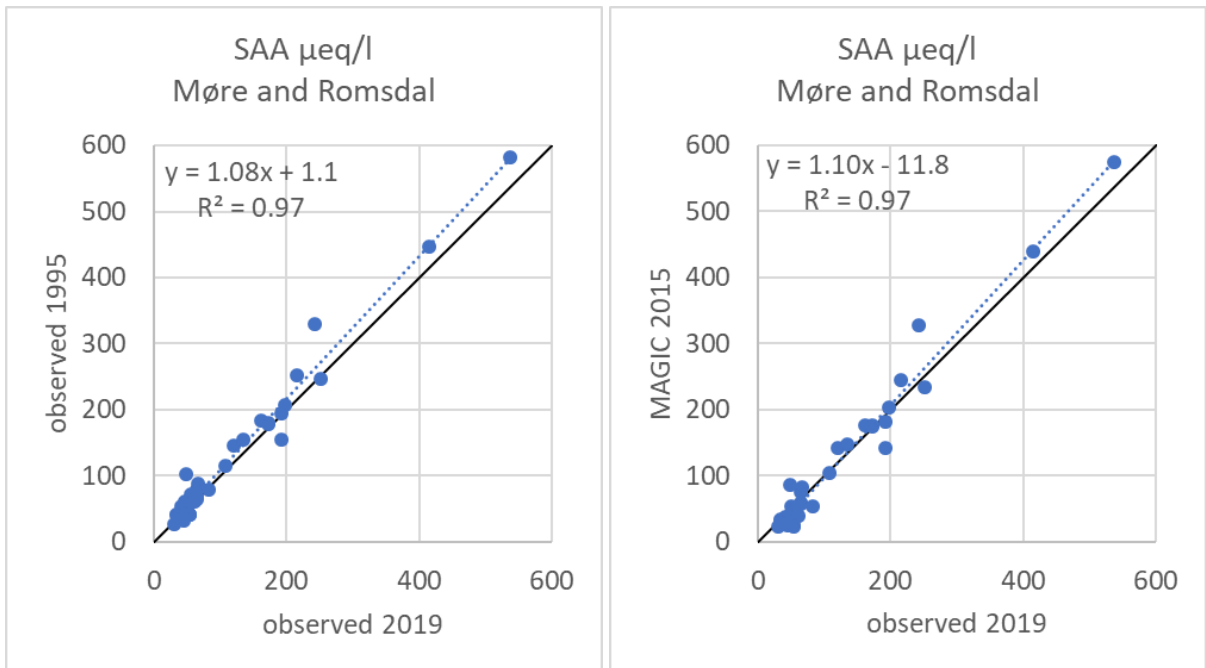
Nitrate concentrations in 1995 and 2019 were very low and did not contribute significantly to the sum of strong acid anions (SAA). SAA concentrations in the lakes were thus only slightly changed in 2019 relative to 1995, and the simulated concentrations in 2019 do not greatly differ from the observed (**Figure 17**).

Concentrations of base cations (SBC) increased slightly in the lakes between 1995 and 2019 (**Figure 18**). Most of the change was due to increased Ca concentrations in some of the lakes (**Figure 19**). The *increase* in Ca cannot be explained by changes in acid deposition, as SO<sub>4</sub> and SAA *decreased* (and only slightly) between 1995 and 2019. It is not due to changes in seasalt deposition as Cl concentrations in the lakes were unchanged in 2019 relative to 1995. And the change is probably not due to differences in amount of precipitation between the years 1995 and 2019, as the concentrations of Mg and Na, the two other dominant base cations, did not change significantly between 1995 and 2019. The increase in Ca concentrations must therefore be due to another factor, such as climate.

ANC concentrations were higher in 2019 than 1995 by about 23 µeq/l on average (**Figure 20**). The increase in ANC is due in part to the increase in SBC (mostly Ca) and in part to the decrease in SAA (SO<sub>4</sub> in some lakes). The simulated ANC for 2015 by MAGIC was lower than the observed in 2019, mostly due to the fact that the observed increase in Ca concentrations from 1995 to 2019 cannot be explained by changes in acid deposition. From the linear regressions it appears that the MAGIC simulations gave on average a slight increase in ANC of about 5 µeq/l (the difference in the intercepts of the two equations). The remaining 18 µeq/l increase in ANC cannot be explained.

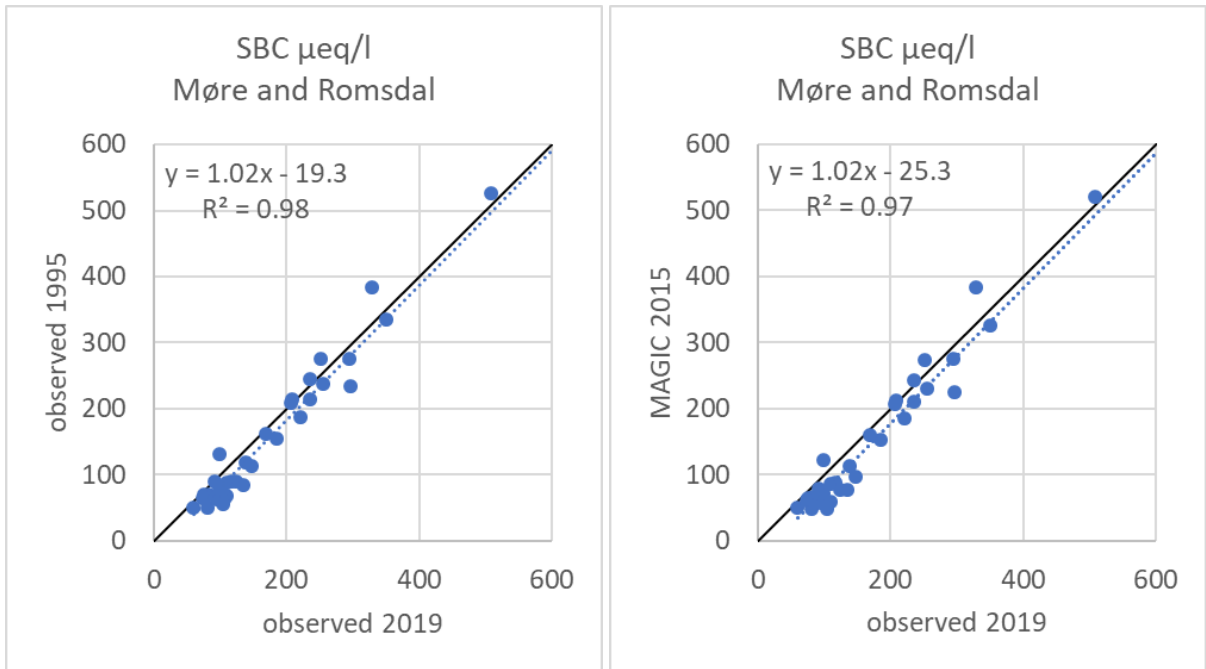


**Figure 16.** Cl concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

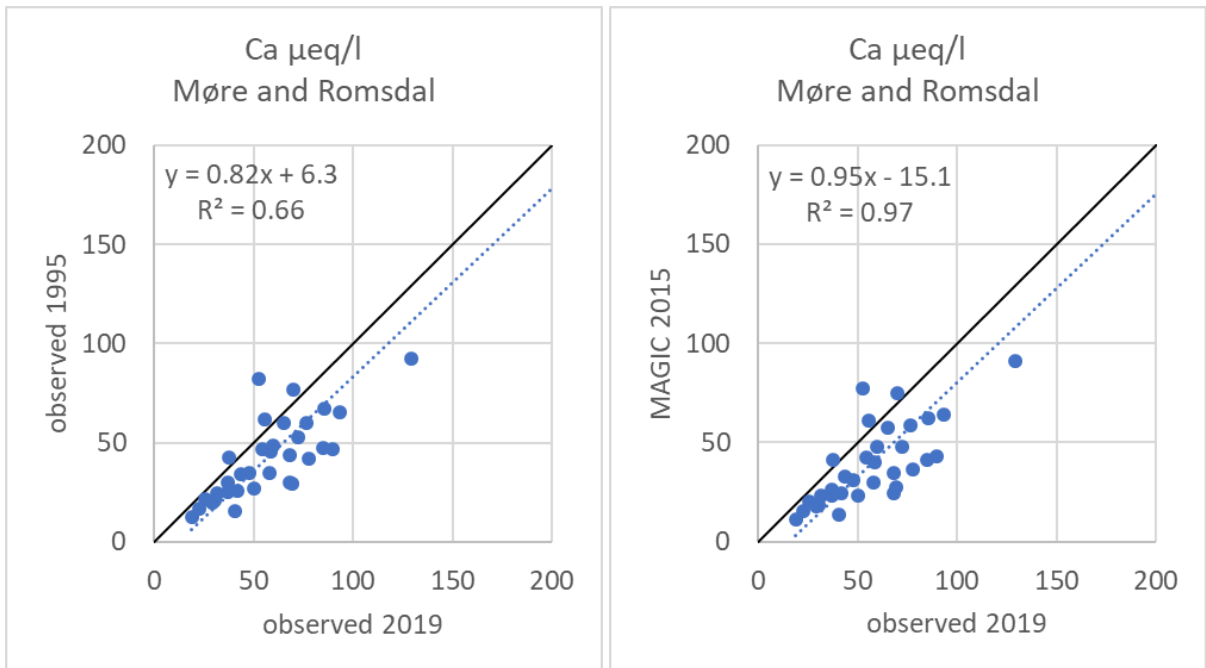


**Figure 17.** SAA concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

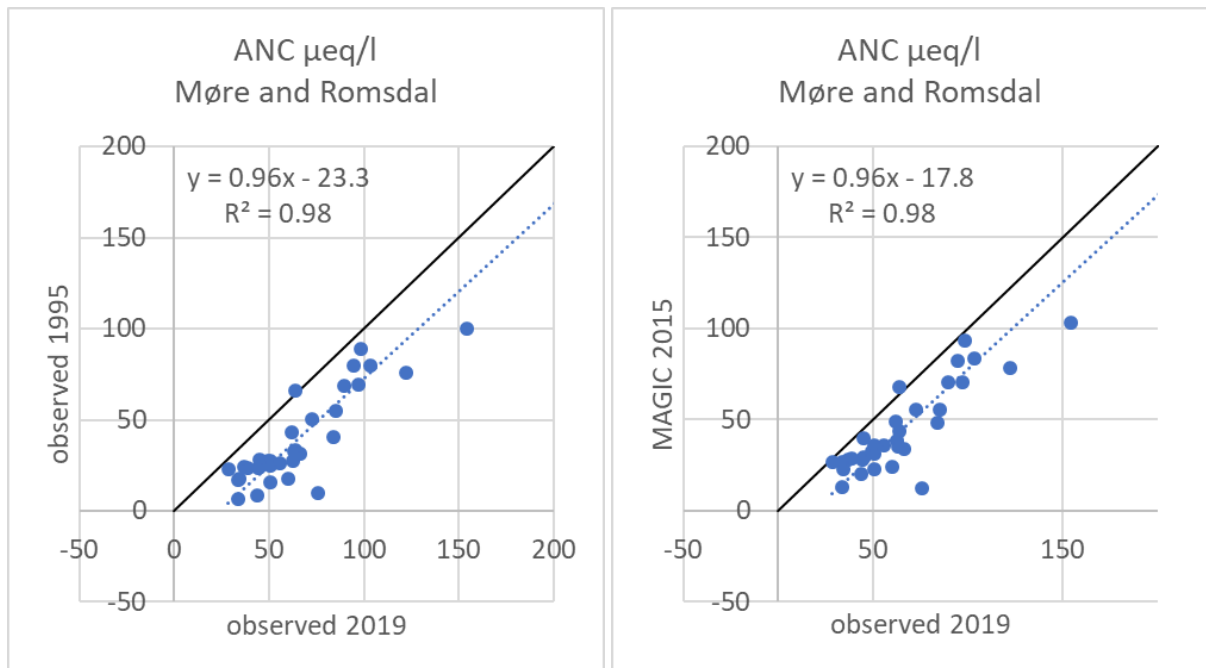




**Figure 18.** SBC concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.



**Figure 19.** Ca concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.



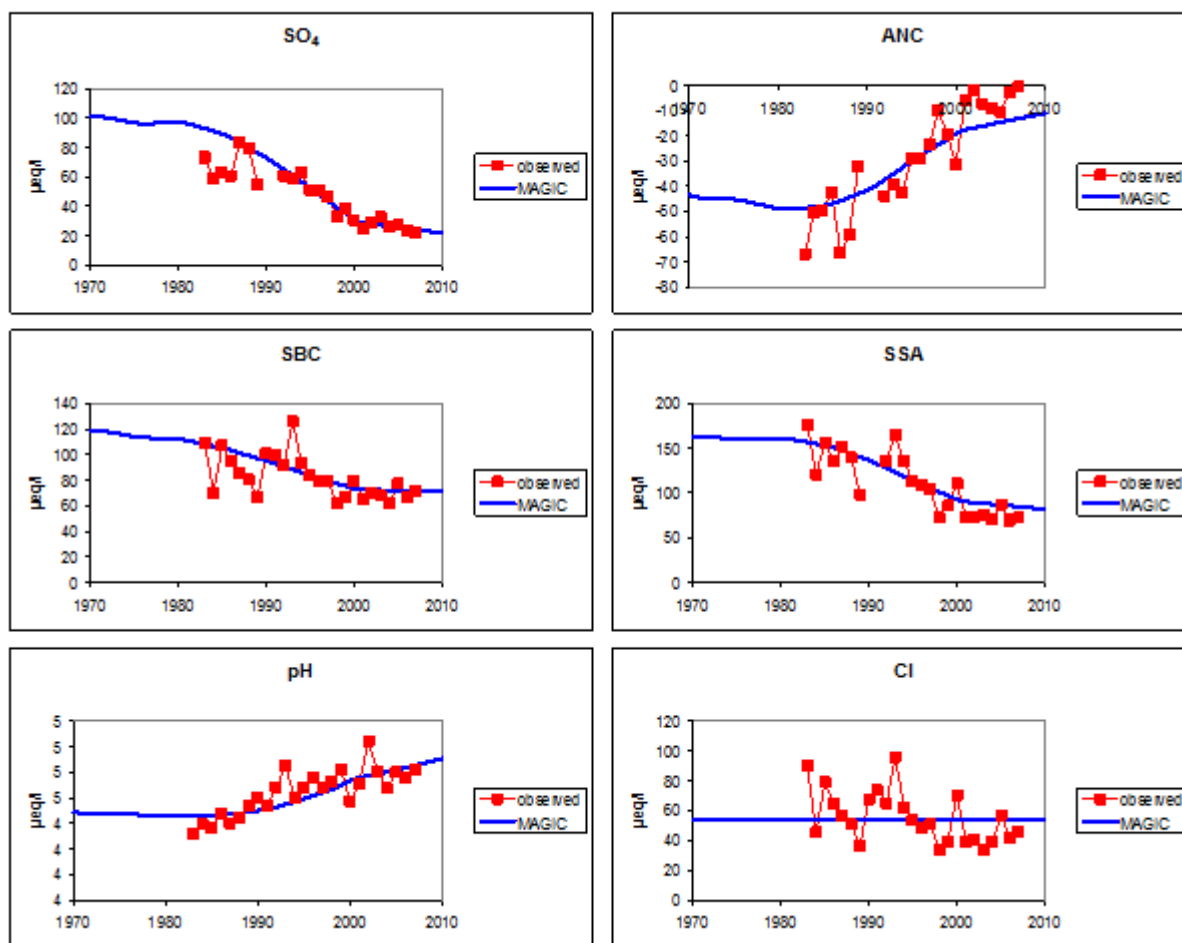
**Figure 20.** ANC concentrations in 34 lakes in Møre and Romsdal measured in 1995 (left-hand panel) and simulated by MAGIC for the year 2015 (right-hand panel) relative to the observed values in 2019.

## 2.4 Discussion

For the acid-sensitive lakes in southern/middle Norway the MAGIC simulations based on the 1995 1000-lake survey data gave an acceptable prognosis for the lakewater chemistry measured in the 2019 resurvey. The forecast decrease in S deposition between 1995 and 2019 turns out to be only somewhat more optimistic than the actual measured S deposition at the selected NILU stations. The observed and simulated changes in  $\text{SO}_4$  concentrations in the lakes can thus be ascribed to decreased S deposition. But both the observed and simulated lakewater chemistry in 2019 has clearly been affected by other factors as well, such as the changed seasalt deposition in the lakes in Sørlandet.

Using data from Lille Hovvatn, one of the 1000 lakes and also one of the lakes sampled annually as part of the Norwegian national monitoring programme (Våge et al., 2020) illustrate how the inclusion of year-to-year variations in seasalt and S deposition greatly improves the precision of the MAGIC simulations (**Figure 21**, **Figure 22**). The MAGIC simulated trends in water chemistry at Lille Hovvatn agree reasonably well with measurements. Year-to-year variations, of course, are not captured by the MAGIC simulations as the model was driven by the smooth deposition trends supplied by EMEP and did not take into account wet and dry years, for example. The observed trend in ANC showed a somewhat greater increase as compared to the simulated. This was apparently mostly due to the strong-acid anion Cl which has been present at lower concentrations since the calibration year 1995. High concentrations of Cl lead to lower ANC (as simulated), whereas lower Cl concentrations lead to higher ANC (as observed).

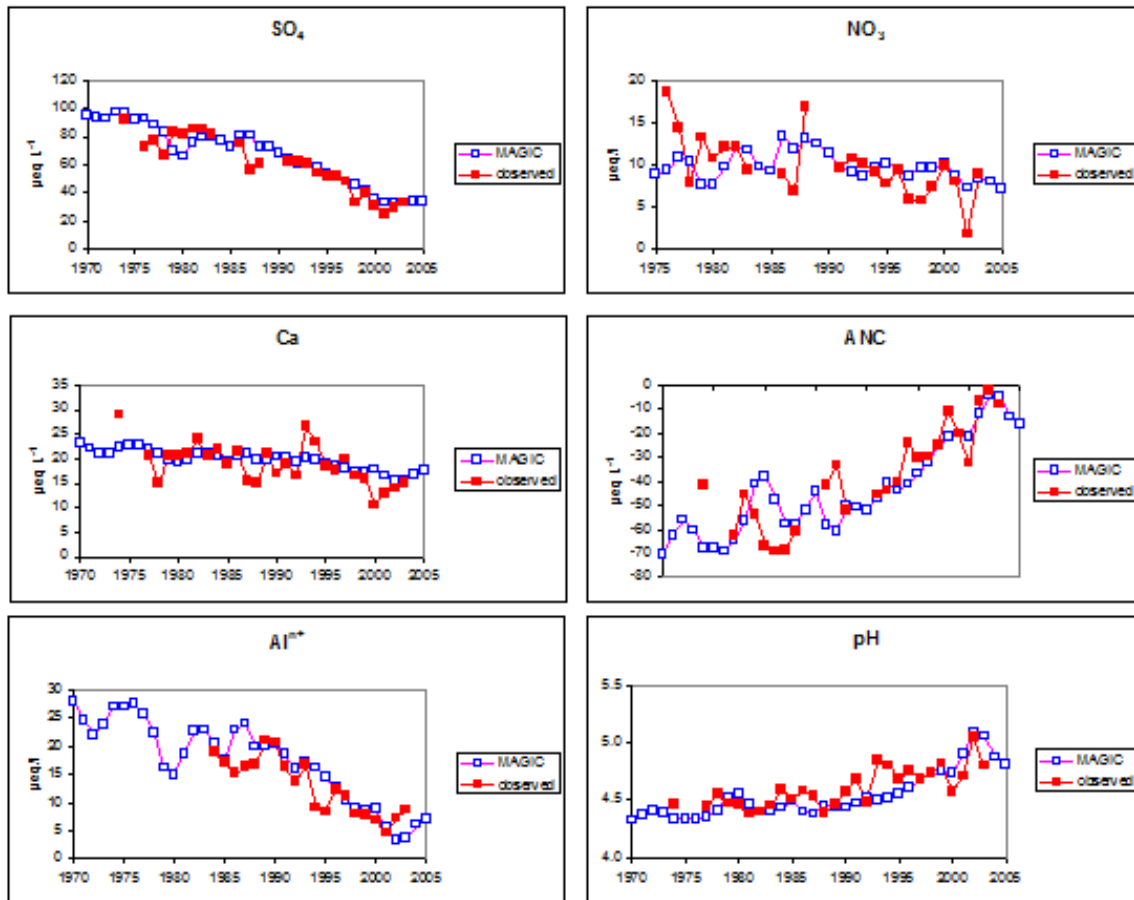
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**Figure 21.** Observed and simulated water chemistry at Lille Hovvatn, southernmost Norway. MAGIC simulations from (Larsen et al., 2008a) using relative changes in S and N deposition from EMEP. Observed data from Hindar and Wright (2005). SSA=sum strong acid anions; SBC=sum base cations.

For the lakes in both Sørlandet and Møre and Romsdal the increase in Ca concentrations in the lakes cannot be explained by changes in S or seasalt deposition. Some other factor such as year-to-year variations in temperature or precipitation must be behind the changes observed.

Our analysis of MAGIC simulated versus observed lake chemistry reached similar conclusions as Helliwell et al. (2014). They looked at MAGIC prognoses forward to the year 2010 based on 1995 data for several groups of sensitive lakes in Europe and then compared these prognoses with the actual measured 2010 lake chemistry. They found that given that the forecast S deposition 1995 to 2010 actually occurred, then the forecast lake chemistry agreed satisfactorily with the observed.



**Figure 22.** Observed and simulated water chemistry at Lille Hovvatn, southernmost Norway. Here the MAGIC simulations included year-to-year measured variations in atmospheric deposition. From Hindar and Wright (2005).

## 2.5 Improving the input data and calibration procedures for a new calibration of MAGIC to the 1000-lake data

### 2.5.1 Why re-calibrate?

The most recent calibration of MAGIC of the 1000-lakes data in Norway was based on the data from the 1995 survey (Larssen et al., 2008a; Austnes et al., 2016). The MAGIC forecasts for 2019 were thus made for 24 years into the future. The 2019 data from the re-survey of the 1000 lakes are the “ground truth” with which the MAGIC forecasts can be compared. The 2019 data document the present condition of Norwegian lakes with respect to acidification and recovery.

Re-calibration to these new data from 2019 would give a new starting point for making forecasts for the future state of water chemistry in Norwegian lakes. Re-calibration could use the actual S and N deposition in the recent past, rather than the estimates provided by EMEP. Moreover, re-calibration could consider changes in seasalt deposition and long-term changes in specific runoff.

Re-calibration could make use of the most recent developments and improvements in the MAGIC model. The latest version 8 dated 1 October 2010 (Oulehle et al., 2012) has new formulations of C and N processes that could be applied to the 1000-lake dataset. Ongoing work at NIVA is adapting MAGIC to the new MOBIUS modelling platform, and a new application to the 1000-lake dataset can make use of current and future modules on the MOBIUS platform. The MOBIUS framework (Norling et al., in review) is an open-source model-building framework designed for rapid model prototyping and has a focus on computational performance. It has been used to develop catchment biogeochemical models (<https://github.com/NIVANorge/Mobius.git> and <ftp://mobiserver.niva.no>).

## 2.5.2 Improving the calibration

The standard calibration procedure for MAGIC entails a stepwise procedure in which assumptions are made first for the sources and fates of the three strong acid anions  $\text{SO}_4$ ,  $\text{Cl}$ , and  $\text{NO}_3$ , second for the parameters controlling the concentrations of inorganic and organic C anions, third for the parameters controlling the solubility of inorganic Al, and finally for the parameters controlling the concentrations of base cations. The various parameters are adjusted either manually or in the case of the base cations with the help of an automated optimisation routine such that the simulated water and soil chemistry in the target year matches the observed. For the regional MAGIC application to the 1000-lake survey the target year (termed “reference year” in the MAGIC model) was 1995, the year for which measured data were available.

Before re-calibrating, it would be appropriate to revisit all the assumptions and approximations made in the MAGIC calibrations to the 1995 1000-lake dataset.

### **Deposition: historical and future scenarios**

The S and N deposition scenarios used in the MAGIC calibration to the 1995 dataset should be revised prior to a new application of MAGIC to the 2019 dataset. The EMEP deposition is normalised to the deposition estimated for each lake based on the lake water chemistry, but the changes in deposition are entirely based on the EMEP scenarios. In a new application, the actual changes in deposition amounts during the period 1995-2020 should be used, such as those observed at NILU stations. In addition, new estimates for future S and N deposition will be required. The current EMEP scenarios run only until the year 2030.

### **Hydrology**

Runoff (m/yr) is one of the fixed parameters in MAGIC. In the calibration to the 1995 lake data the runoff for each lake was taken from the NVE map of specific runoff for Norway for the standard period 1961-1990. For a re-calibration data for the new standard period 1991-2020 can be used. In the cases where the specific runoff has changed due to climate change or other factors, the re-calibration could use the 1961-1990 standard period for years prior to 1990 and the new standard period for years post 1990. Or time trends could be introduced into the input data for runoff.

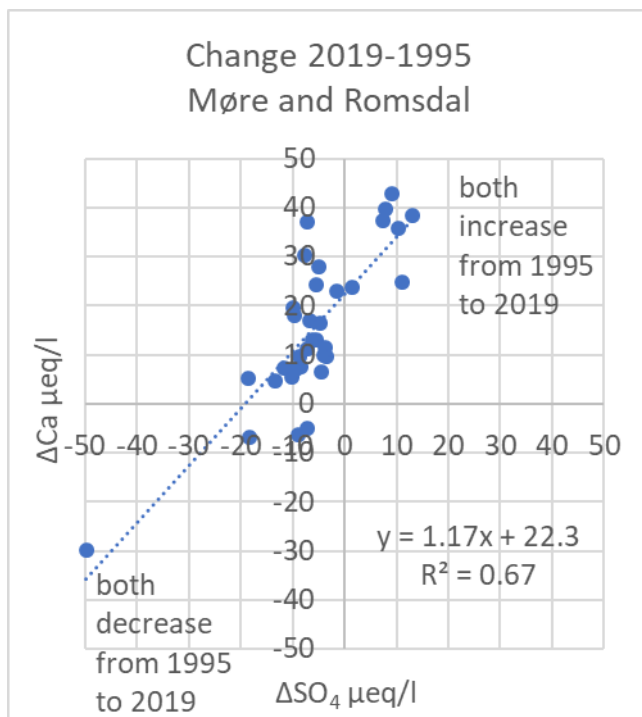
### **Strong acid anions**

The strong acid anions  $\text{SO}_4$ ,  $\text{Cl}$  and  $\text{NO}_3$  (SAA) are assumed in MAGIC to be independent of one another. In the calibrations to the 1000-lake dataset in 1995 they were assumed to be independent of soil properties.

### SO<sub>4</sub> concentrations and S deposition

One of the challenges in calibration of MAGIC to the 1000 lakes is to specify the sources of SO<sub>4</sub> measured in each of the lakes in the reference year. The procedure used by (Larssen et al., 2008a) and (Austnes et al., 2016) in the calibration of MAGIC to the 1995 lake data entailed estimating the amount of S from acid deposition based on the SO<sub>4</sub> flux from each lake in 1995. The flux was calculated from the measured SO<sub>4</sub> concentration and the estimated runoff amount for each lake in 1995. The sources for SO<sub>4</sub> in lakewater were assumed to be comprised of (1) deposition of seasalts, (2) natural background (pre-industrial) deposition of non-marine SO<sub>4</sub>, (3) weathering of soil minerals, and (4) anthropogenic SO<sub>4</sub>. Seasalt SO<sub>4</sub> was assumed to equal 0.103 times the Cl flux in each lake (0.103 is the ratio of these ions in seawater). The natural background was provided in the EMEP data. Excess SO<sub>4</sub> deposition is defined as the amount of SO<sub>4</sub> in excess of the marine contribution. SO<sub>4</sub> from weathering was assumed zero in all cases in which excess SO<sub>4</sub> deposition calculated from the water chemistry and discharge was below 100 meq/m<sup>2</sup>/yr (in 1995). In cases where the calculated excess SO<sub>4</sub> deposition was above 100 meq/m<sup>2</sup>/yr, the weathering component was assumed to account for the remainder of the excess SO<sub>4</sub>. Sulphate adsorption in the soil was assumed to be negligible given the timestep in MAGIC of one year.

The evaluation of the MAGIC forecasts with the observed data for 2019 indicate that this procedure for estimating of excess S deposition in 1995 could be improved. The observed chemistry for lakes in Møre and Romsdal showed that SO<sub>4</sub> concentrations had *increased* in many of the lakes from 1995 to 2019 despite *decreased* S deposition (**Figure 23**). The increased SO<sub>4</sub> concentrations were accompanied by increased concentrations of Ca. As the SO<sub>4</sub> cannot be ascribed to S deposition, it must be due to release from the catchment soils, perhaps as increased weathering. A two-point calibration (cf. section 2.5.3) would improve this method of estimating the role of S deposition on SO<sub>4</sub> concentrations in the lakes.



**Figure 23.** Changes in SO<sub>4</sub> and Ca concentrations in 34 lakes in Møre and Romsdal measured in 1995 and again in 2019.

**Cl concentrations and seasalt deposition**

Estimates of seasalt deposition are based on the assumption that the Cl flux from each lake is entirely due to deposition of seasalt Cl. With lake data from only one point in time (1995), the MAGIC simulations assumed that the seasalt deposition was constant at that level for all years in the past and future. For the lakes in Sørlandet, however, it is clear that seasalt deposition was lower in 2019 than in 1995, and thus the ANC in 2019 was higher than expected. Re-calibration based on lake data from more than one point in time could take into account changes in seasalt deposition over time.

**NO<sub>3</sub> concentrations and N retention**

The flux of inorganic N (oxidised plus reduced forms) in deposition to most lakes was much larger than the flux of inorganic N (almost entirely as NO<sub>3</sub>) from the lakes. In the MAGIC calibrations to the 1995 data it was assumed that the fraction of incoming N retained in the catchment-lake systems observed in 1995 was constant in the past and into the future. The 2019 data provide a second estimate of the % retention. Re-calibration could take into account uncertainty in the estimate of N retention over time, and systematic trends over time in % retention. Concentrations of NO<sub>3</sub> in the lakes were generally low in both 1995 and 2019. NO<sub>3</sub> thus contributed little to the sum of strong acid anions (SAA).

The most recent version of MAGIC (version 8) has three alternative approaches to C and N processes in ecosystems. The simplest assumes constant % retention of N deposition, usually set to the observed value for the calibration year. This was used in the calibrations to the 1995 lake data. The second approach assumes that the % N retention is linked to the C/N ratio in the soil, with an increasing fraction of N leached as the soil becomes more N rich. The third approach (in MAGIC version 8) uses a coupling of C and N in soil via microbial processes. A new calibration of MAGIC to the 2019 lake data might make use of any or all of these three approaches to N retention.

**Base cations**

Simulation of base cation concentrations by MAGIC requires specification of a number of soil parameters (**Table 1**). These are fixed and assumed not to change over time. The calibration for each lake is conducted such that the simulated matches the observed in the reference year (in this case 1995) for each of four water variables (concentrations Ca, Mg, Na, and K with units µeq/l) and four soil variables (exchangeable base cations Ca, Mg, Na, and K with units meq/kg). MAGIC has a trial-and-error optimisation routine to determine the weathering rates of the four base cations and the initial (pre-acidification) amounts of each of the four base cations as a fraction of the soil cation exchange capacity.

The MAGIC calibrations on the 1995 data assumed that the base cation weathering rates were constant over time. Actual weathering rates might change in response to changes in precipitation amount or soil temperature. Similarly, the MAGIC calibrations assumed that base cation uptake by the forest was constant over time. Changes in forest growth rates or silvicultural practices could affect the base cation uptake. Such factors might explain the higher Ca concentrations in the lakes in Sørlandet and Møre and Romsdal in 2019 relative to 1995. Further research is needed to understand the observed increase in Ca concentration. This could serve as a basis for modification of the processes in the model. A two-point calibration (cf. section 2.5.3) would help identifying changes in Ca concentration not related to changes in deposition.

**Table 1.** Soil parameters used in calibration of MAGIC to the 1000-lake dataset (from Larssen et al. (2008a)).

Parameter	Unit
Soil depth	m
Porosity	%
Bulk density	kg/m <sup>3</sup>
Cation exchange capacity (CEC)	meq/kg
SO <sub>4</sub> maximum adsorption capacity	meq/kg
SO <sub>4</sub> half-saturation coefficient	meq/m <sup>3</sup>
Dissociation constant for aluminium hydroxide solid phase (KAl)	log 10
Temperature (annual average)	°C
pCO <sub>2</sub>	%
Organic acids	mmol C/m <sup>3</sup>
Dissociation constants for organic acids (pK)	
Nitrification	% of input

### Fixed soil parameters

The 1995 calibration of MAGIC to the 1000-lake survey necessitated estimates of fixed soil parameters for each of the 1000 lakes (Larssen et al., 2008a) (**Table 1**). Of these soil depth, porosity, bulk density and CEC are usually available or can be estimated from measured soil samples. SO<sub>4</sub> adsorption was assumed to be negligible for the Norwegian lakes given the geologically young soils and the one-year timestep in the MAGIC simulations. Annual average soil temperature is of minor importance as it was used by MAGIC only to calculate the equilibrium points in chemical reactions in the soil. pCO<sub>2</sub> is rarely measured in soil, and the simulations assumed a constant value based on data from the literature. Similarly, the dissociation constants for organic acids in soil were taken from studies in Scandinavian soil solution given by (Hruška et al., 2003). In the calibrations to the 1995 data, nitrification in both the soil and lake was assumed to be 100% inasmuch ammonium (NH<sub>4</sub>) concentrations were assumed to be negligible. NH<sub>4</sub> was not measured in the lakes in 1995. Nitrification in the catchment-lake system as a whole can now be quantified using the measured NH<sub>4</sub> concentrations in the lakes in 2019.

This leaves two key soil parameters to be specified: the dissociation constant for aluminium hydroxide solid phase, KAl (log10) and the concentration of organic acids (mmol C/m<sup>3</sup>) in soil solution. Trials with MAGIC have shown that the simulated water chemistry is sensitive to both these parameters and that they are interdependent. Higher KAl and/or concentration of organic acid results in flatter responses in SBC and ANC to changes in S deposition. Neither of these parameters is available in the soil data for Norway. A two-point calibration of MAGIC (cf. section 2.5.3) could provide an empirical way to obtain more robust estimates of these two sensitive parameters for each of the lakes.

Concentrations of dissolved organic carbon (DOC or alternatively total organic C, TOC) have increased significantly in Norwegian surface waters over the past 30 years most probably due to the decrease in S deposition (de Wit et al., 2007; Monteith et al., 2007; de Wit et al., 2016). The increase in TOC in surface waters can reflect a similar increase in organic C concentrations in soil solution (Hruška et al., 2009), but there are few data available to document such changes in soil solution. Re-calibration could take into account observed changes in TOC in the lakes.



### **New soil data**

For the calibrations to the 1995 1000-lake survey measured data for soils in Norway were available for forest soils (present in about 1/3 of the lakes) but were very sparse for unforested, upland areas as well as peatlands. There were fewer than 50 soil sites that were used for the remaining 2/3 of the lakes.

Evaluation of the MAGIC forecasts with the observed data from 2019 indicates that for acid-sensitive lakes in Sørlandet the forecasts are remarkably good despite the paucity of soils data for the lake catchments. Perhaps the natural spatial variations in soil properties are insufficiently large to greatly affect the MAGIC forecasts. A re-calibration of MAGIC to the 2019 data should, of course, make use of any new soil data that has become available since the previous calibrations, but new soil data are not a prerequisite for re-calibration.

### **Uptake of base cations and N by forests**

For lake catchments that lie in forested areas the uptake of three base cations (Ca, Mg, K) and N was assumed to be constant over time in the MAGIC calibrations to the 1995 1000-lake dataset (Larssen et al., 2008a). Re-calibration could take into account measured changes in forest growth over the period 1995-2020. Such changes may be caused by climate change, forest fertilisation or other silvicultural practices.

## **2.5.3 Two-point calibration**

The most obvious way to improve the calibration of MAGIC is to use data from more than one point in time. The 1000-lake survey in 2019 provides a second set of observed data that could potentially be used to refine the calibrations based only on the 1995 data. Some of the 1000 lakes were also previously sampled in lakes surveys in 1986 and 1974/75. As shown by Larssen et al. (2004) using more than one data set in time could greatly reduce the uncertainty of the MAGIC forecasts for future water chemistry (see **Appendix B**).

Larssen et al. (2004) focused on one site only, Birkenes. They used a Monte-Carlo routine to prepare 10000 sets of input parameters and calibrated these to one point in time (1990). They then checked if the simulated water and soil chemistry agreed with measurements from two other points in time (1980 and 2000). They found that 8891 of the 10000 data sets were successfully calibrated, but only 2234 passed all three 5-year windows.

In the case of a new calibration of MAGIC to the 1000-lake data, the new MOBIUS framework could be used. A Monte-Carlo routine with several thousand parameter sets could be setup and run for each of the 1000 lakes. The procedure could use a protocol similar to that of Larssen et al. (2004) or a more formal Bayesian Markov-chain Monte-Carlo (MCMC) method such as Emcee, which has already been coupled to other Mobius models (Norling et al., in review). Adapting these to MAGIC would allow for substantial code reuse. The resulting two-point calibrations would greatly increase the confidence in the use of MAGIC to forecast lake water chemistry given future changes in acid deposition.

Coupling the Monte-Carlo routine to the model would require a moderate amount of programming to set up the model success evaluation criteria (likelihood function) and the prior distributions (or prior ranges) of the parameters. Once set up running the MCMC routine for MAGIC on 1000 different locations can easily be automated. While different Monte-Carlo algorithms have different strengths,

they all provide the parameter set that gives the best fit of the model to the observed data and also provide a probability distribution (so-called posterior distribution) of the parameters. The posterior distributions facilitate quantification of the sensitivity of the system to perturbation in the parameters and inputs, and also provide estimates of probability distributions of future outcomes of the system, in this case the water chemistry of the lakes.

#### **2.5.4 The way forward**

- Re-calibration of MAGIC to the 1000-lake dataset should start with the 2019 data, as any scenarios of future water chemistry condition in Norwegian lakes will have 2019 as the best estimate of the current conditions.
- New applications of MAGIC can make use of the MOBIUS modelling platform. MOBIUS has other relevant modules available for coupling with MAGIC, such as a hydrology module. MOBIUS offers off-the-shelf procedures for estimating uncertainty and parameter sensitivity in models.
- Prior to re-calibration the processes in MAGIC should be re-visited and updated with recent understanding, especially of soil processes.
- Re-calibration of MAGIC should make use of both the 1995 and the 2019 data in a two-point calibration. Special attention should be given to changes in Ca and SO<sub>4</sub> concentration that cannot be explained by changes in deposition.
- The most recent version of MAGIC (version 8) has three alternative approaches to C and N processes in ecosystems. A new calibration of MAGIC to the 2019 lake data might make use of any or all of these three approaches to N retention.
- Organic C in soils and waters has changed over time, and these changes should be taken into account in new calibrations of MAGIC to the 2019 data.
- Re-calibration of the 2019 dataset can be used to evaluate scenarios of changed acid deposition in concert with other environmental drivers, such as climate change and altered forestry practices.
- The future role of N deposition is of special interest because N deposition plays an increasingly important role as S deposition has decreased substantially over the past 30 years.

A strategy for re-calibration could take two stages. First the re-calibration could use updated input data and a two-point calibration technique with the focus on acidification due to S and N deposition. A second stage could further include modification of processes in MAGIC, such as those influenced by climate changes (Wright et al., 2006) and forestry practices. This might require starting with MAGIC applications to individual sites and lakes.

## 3 Critical loads for surface waters

### 3.1 Background

The Steady-State Water Chemistry (SSWC) model and the First-order Acidity Balance (FAB) model (Henriksen and Posch, 2001; CLRTAP, 2017) are the most commonly used critical loads models for surface waters. These are also the models used in the most recent national application in Norway (Austnes et al., 2018), but only the FAB-generated critical loads are used under the LRTAP Convention. The SSWC model is based on the principle that the acid deposition should not exceed the sum of the non-marine, non-anthropogenic base cation flux from the catchment, i.e. the pre-industrial base cation flux ( $BC_0^*$ )<sup>2</sup>, minus a buffer to protect selected biota ( $ANC_{limit}$ ). The pre-industrial base cation flux is the sum of the non-marine, non-anthropogenic base cation deposition and the base cation flux from weathering minus the long-term removal of base cations by harvest of the forest.

The FAB model is a mass-balance model and takes into account the sources and sinks of base cations, sulphur and nitrogen. For sulphur sources or sinks are assumed to be negligible in the terrestrial part of the catchment at steady state. For nitrogen immobilisation, denitrification and harvest removal are sinks that are taken into account. Both sulphur and nitrogen can be retained in the lake. The base cation part is modelled with SSWC.

The main difference between the models becomes apparent when calculating the exceedance (CLRTAP, 2015a). With the SSWC model, it is assumed that the sulphate ion leaching equals the deposition while nitrogen is retained at present-day levels. The exceedance is thus calculated as the sum of sulphur deposition and nitrate leaching minus the critical load and represents present-day exceedance. Calculation of exceedance in the future assumes that the nitrate leaching remains constant at present-day levels. The FAB model, on the other hand, produces separate critical loads for sulphur and nitrogen. For nitrogen it is assumed that the steady-state retention is lower than the current retention under elevated deposition. Given constant elevated deposition nitrogen will consequently contribute more to acidification in the future as a new steady state is reached. Thus, under FAB the current exceedance will be larger than with SSWC.

### 3.2 The national critical loads application

The methodology for calculating and mapping critical loads for acidification of surface waters in Norway was developed by Henriksen (1998) and later updated by Larssen et al. (2008b) and Austnes et al. (2018). The critical loads are calculated for each grid cell in a  $0.25^\circ \times 0.125^\circ$  longitude-latitude grid covering all of Norway (BLR: "breddegrad, lengdegrad, rute"). The surface water chemistry for the grid cells used in the calculations was set by comparing available water chemistry data for lakes and rivers within each cell. The water chemistry data were primarily results from the national lake survey conducted in 1986 (Lien et al., 1987). The chemistry of the lake or river that was judged to be the most typical was chosen to represent the grid cell. If there were wide variations within a grid cell, the most sensitive area covering at least 25% of the grid cell was selected. Sensitivity was based on

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<sup>2</sup> The asterisk denotes the non-marine fraction (calculated from the chloride concentration and the BC/Cl ratio in seawater)

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water chemistry, topography, and bedrock geology. Geology was determined from the geological map of Norway (1:1 million) prepared by the Norwegian Geological Survey (NGU).

The  $ANC_{limit}$  was originally set to a constant, 20  $\mu\text{eq/l}$ , based on surveys on fish in Norwegian lakes (Lien et al., 1996). This  $ANC_{limit}$  gives a 95% probability of no damage to fish populations. Later, the variable  $ANC_{limit}$  was introduced. Less sensitive systems should have a higher  $ANC_{limit}$  since they will generally have a higher biological diversity, which requires a higher  $ANC_{limit}$  to be held intact (Henriksen and Posch, 2001). An additional adjustment to the  $ANC_{limit}$  was introduced to take into account the effect of naturally occurring organic acids (Lydersen et al., 2004). Many Norwegian lakes are humic, and part of the organic acids act as strong acid anions. An adjusted ANC ( $ANC_{limit, oaa}$ ) which takes this contribution into account gave a slightly better fit with fish status. Results are labelled “*oaa*” (organic acid adjusted) to signify that this adjustment has been applied.

The pre-industrial base cation concentration ( $[BC]_0^*$ ) was originally calculated by the F-factor approach, using the sine function of Brakke et al. (1990). Since Larssen et al. (2008b) calculations of national critical loads have used the MAGIC model to estimate  $[BC]_0^*$ . MAGIC (Model of Acidification of Groundwater In Catchments) is a dynamic process-oriented model used to simulate past and future surface water chemistry in response to scenarios of S and N deposition (Cosby et al., 1985a; Cosby et al., 1985b; Cosby et al., 2001). One of the outputs of MAGIC is an estimate of surface water chemistry prior to the onset of acid deposition. MAGIC was calibrated by (Larssen et al., 2005) to 131 sensitive lakes in southern Norway, of which 83 lakes were acidified ( $ANC < ANC_{limit}$ ). In the current critical loads calculations, a linear regression of MAGIC modelled  $[BC]_0^*$  ( $[BC]_{1860}^*$ ) on  $[BC]_{1986}^*$  for these 83 lakes is used to estimate  $[BC]_0^*$  for each grid cell using the water chemistry data assigned to each cell.

For the FAB model nitrogen removal in harvested forest biomass was estimated by Frogner et al. (1992) and mapped for all of Norway based on forest cover and productivity. Nitrogen immobilisation in the catchments was assumed constant at 0.5 kg N  $\text{ha}^{-1} \text{yr}^{-1}$  and the denitrification factor in the catchments was set to 0.1 (CLRTAP, 2017). In the lakes the mass transfer coefficients for N and S were held constant at 5 m  $\text{yr}^{-1}$  and 0.5 m  $\text{yr}^{-1}$ , respectively; these are the mid-values of the ranges proposed by Dillon and Molot (1990) and Baker and Brezonik (1988), respectively. The lake to catchment area was set constant to 5%. Mean annual runoff data were taken from runoff maps prepared by the Norwegian Water Resources and Energy Directorate (NVE) based on the 1961-1990 normal.

### 3.3 Comparing critical load exceedance with the 2019 lake data

Critical load exceedance indicates that the deposition is sufficiently high to give acidification of surface waters. In theory it should therefore be possible to compare critical load exceedance with measured ANC, to check if the exceedance results are reasonable. However, this type of comparison should be made with some care. First, the exceedance represents a condition of steady state. This means that surface waters may still be acidified even if there is no exceedance, as the steady state is not yet reached, i.e. there is a delay in recovery. For the FAB model it may also be the other way round, as it assumes that nitrogen retention has returned to natural rates. With the current higher nitrogen retention, the given nitrogen deposition may not give acidification even if there is exceedance according to the FAB model. Second, the critical loads and exceedances reflect average conditions in time and space, while measured values represent a single catchment and one point in

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time. Nevertheless, on a large geographical basis one would expect some degree of correspondence between exceedance and acidification levels.

Here we compare the results from the 2019 national 1000-lake survey (Hindar et al., 2020) with the national critical loads and exceedance calculations. Exceedance was calculated with the average deposition for 2012-2016, as in the latest exceedance reporting (Austnes et al., 2018). This is the most recent period for which gridded deposition data at the national scale are available.

Whether there is acidification or not can be decided based on the system developed for classification under the Water Framework Directive in Norway (DirektoratgruppenVanndirektivet, 2018). Here class boundaries are set for different water body types for different physico-chemical and biological quality elements (parameters), targeted at different types of pressures. For comparison with the critical loads output, the class boundaries for acidification status based on ANC were used (**Table 2**), as ANC is also the chemical indicator used in the critical loads calculation. The boundary between good and moderate status defines whether the water body is acidified or not. Based on the WFD classification, the ecological quality ratio (EQR) can also be calculated, indicating the distance from the reference level. When normalised (nEQR) the values can be compared across water body types. nEQR values range between 0 and 1, with higher nEQR representing better status. The five status classes have a range of 0.2 on the scale. The good/moderate boundary is thus at 0.6.

**Table 2.** Type factor boundaries and ANC class boundaries used in the WFD classification. The uppermost boundaries of the type factors were ignored in the classification.

Type	Type factor boundaries (mg/l)		ANC class boundaries ( $\mu\text{eq/l}$ )					
	Ca	TOC	Reference	High	Good	Moderate	Poor	Bad
1a	0-0.25	0-2	10	>5	0-5	-10-0	-20--10	>-20
1b	0.25-0.5	0-2	25	>15	5-15	-5-5	-15--5	>-15
1c	0.5-0.75	0-2	40	>25	10-25	0-10	-10-0	>-10
1d	0.75-1	0-2	55	>40	20-40	5-20	-5-5	>-5
2a	0-0.25	2-5	15	>10	5-10	-5-5	-10--5	>-15
2b	0.25-0.5	2-5	30	>15	10-15	0-10	-10-0	>-10
2c	0.5-0.75	2-5	45	>30	15-30	5-15	-5-5	>-5
2d	0.75-1	2-5	60	>40	25-40	10-25	0-10	>0
3a	0-0.25	5-15	20	>15	10-15	0-10	-10-0	>-10
3b	0.25-0.5	5-15	35	>20	15-20	5-15	-5-5	>-5
3c	0.5-0.75	5-15	50	>30	20-30	10-20	0-10	>0
3d	0.75-1	5-15	65	>40	30-40	15-30	5-15	>5
4	1-4	0-2	90	>60	20-60	10-20	0-10	>0
5	1-4	2-5	125	>70	30-70	15-30	5-15	>5
6	1-4	5-15	125	>70	30-70	15-30	5-15	>5

The comparison of lake acidification status and exceedance of critical loads is summarised in **Table 3**. Only 10 of the lakes were classed as acidified (nEQR  $\leq$  0.6). The vast majority of lakes were neither acidified nor in an area with exceedance (true negatives). This is to be expected, as acid deposition has only affected the southern regions of Norway, and reduced S deposition over the past 25 years has led to recovery in many lakes that were previously acidified. A small number of lakes (4 with

SSWC and 8 with FAB) were acidified ( $nEQR \leq 0.6$ ) and were also in areas with exceedance (true positives). The acidified lakes in non-exceedance areas (6 with SSWC and 2 with FAB; false positives) can in many cases be explained by delayed recovery or local seasalt effects. Nearly all these lakes were close to the exceeded areas and/or the coast. Many also had a low negative exceedance as calculated for SSWC, indicating that the change from exceeded to non-exceeded could be fairly recent.

**Table 3.** Summary of acidified ( $nEQR \leq 0.6$ ) or non-acidified lakes in areas with exceedance or non-exceedance of critical loads according to the FAB or SSWC model. The highlighted values are lakes that were correctly predicted, i.e. true positives and true negatives.

Acidification status	SSWC <sub>0aa</sub>		FAB <sub>0aa</sub>	
	Exceeded	Not exceeded	Exceeded	Not exceeded
$nEQR > 0.6$	174	816	339	651
$nEQR \leq 0.6$	4	6	8	2

The major discrepancy from the expected was the relatively large number of lakes in exceeded areas that were not classified as acidified (174 with SSWC and 339 with FAB) (false negatives). The critical load models predict that these lakes should be acidified given the estimated S and N deposition in 2012-2016. However, the predictions are for steady-state conditions. The higher number of lakes in this category for FAB exceedance can be attributed to the low nitrogen retention assumed in this model, as the current nitrogen retention under non-steady-state conditions is likely to be higher.

One explanation for the discrepancy is that the critical loads are set to protect the most sensitive lakes within the BLRs. The sampled lakes are randomly chosen and may not be among the most sensitive. There will also be bigger lakes and lowland lakes in this selection, which are less sensitive and thus less prone to be acidified. Another explanation can be the unusually high calcium concentrations observed in the 2019 survey (Hindar et al., 2020). This was also shown in section 2.3 above. Higher calcium concentration would give higher ANC and thus less acidification.

In general, there were surprisingly few lakes in the 1000-lake survey that could be classified as acidified. For comparison, in the set of 78 lakes that are sampled on an annual basis as part of the acid rain monitoring (Garmo and Skancke, 2020), in 2019 only two of the lakes were classified as acidified by the WFD ANC criteria. Data from previous years showed more acidified lakes (4 in 2018, 8 in 2017 and 6 in 2016). Such year-to-year variations could affect a significant number of lakes in the much larger 1000-lake survey dataset.

There are two other major factors that may explain the relatively large number of non-acidified lakes in areas with exceedance. The acidification criteria may not be sufficiently strict, or the exceedance is overestimated. An evaluation in 2015 (Moldan et al., 2015) showed that the critical limits resulting from the Norwegian critical loads method were generally lower (less strict) than those of the Swedish method. This resulted in higher critical loads with the Norwegian method for the majority of the studied lakes. In an ongoing Swedish-Norwegian project, the WFD classification systems for acidification for physico-chemical quality elements are compared, and suggestions for more harmonised systems are being developed (Fölster et al., in prep). Results so far indicate that more water bodies are classified as less than good status with the Swedish than the Norwegian system. Although these comparisons with Sweden identify differences, they do not draw conclusions on one system being more correct than the other. A comparison of the WFD good/moderate boundary and

the  $ANC_{limit,0aa}$  (Austnes and Lund, 2014) showed a fairly good correspondence when used on the same dataset, except mainly at high TOC (>5 mg/l) where the WFD boundary was less strict. This can indicate that the WFD system itself is not the main reason for the observed mismatch between exceedance and acidification status. On the other hand, the extremely low number of acidified lakes in 2019 indicates that there should hardly be any exceedance at all.

The exceedance would be overestimated if the S and N deposition in 2019 was lower than the deposition in 2012-2016 used in the calculations. S and N deposition measured at NILU monitoring stations show declines in both S and N deposition between 2012-2016 and 2019 (**Figure 2**; **Figure 4**; **Table 4**). The most relevant stations are those in the higher deposition areas, i.e. Birkenes and Vikedal, and to a lesser extent Treungen, Brekkebygda and Hurdal for both parameters and Nausta and Løken for N deposition. At the latter five stations the difference was small, with slightly higher deposition in 2019 at Treungen, slightly lower at Brekkebygda, Hurdal and Nausta and mixed at Løken. At Birkenes and Vikedal deposition was clearly lower in 2019. This means that the exceedance would have been lower if calculated using 2019 deposition data, but to a varying extent depending on the region.

**Table 4.** Change in wet deposition from 2012-2016 to 2019 at NILU monitoring stations in southern/middle Norway. Data from Aas et al. (2020). The location of the first six stations is shown in **Figure 1**. Hurdal is in the eastern part of southern Norway, Vikedal and Nausta in the western part.

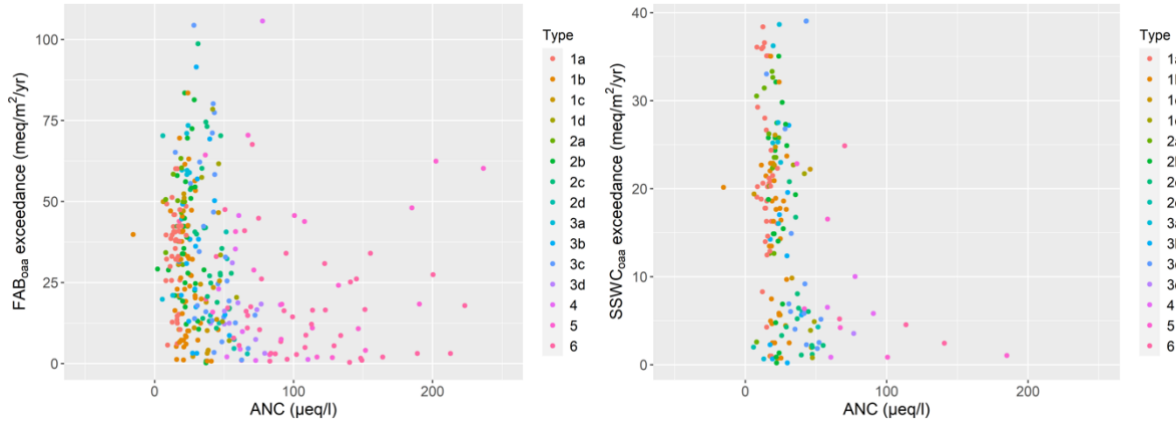
NILU station	2019 vs average 2012-2016 (%)	
	SO <sub>4</sub> -S	NO <sub>3</sub> -N + NH <sub>4</sub> -N
Birkenes	-26	-19
Vatnedalen	11	-10
Treungen	2	5
Løken	-6	2
Brekkebygda (Gulsvik)	-8	-4
Kårvatn	-26	-7
Hurdal	-2	-9
Vikedal	-15	-42
Nausta	-8	-9

The SSWC exceedance is not affected by the nitrogen deposition, but by the nitrate leaching. However, this is also likely to be overestimated, given that it is based on the 1986 nitrate concentrations in the critical loads database. There has been a decline in nitrate concentrations in the past decades (Hindar et al., 2020). The effect of changing the nitrate leaching is further explored in section 3.4. Finally, the exceedance would also be overestimated if the critical loads are too low. Section 3.4 also explores some parameters that could cause this.

Looking at the results in more detail (**Figure 24**) shows that a number of lakes with Ca type > 1 mg/l (type 4-6) have critical load exceedance with the FAB model, but not with the SSWC model. These are lakes that are not likely to be acidified given the current nitrogen retention, as shown by the high ANC. Again, this shows that for comparison with present lake water chemistry, it is most reasonable to use the SSWC exceedance.

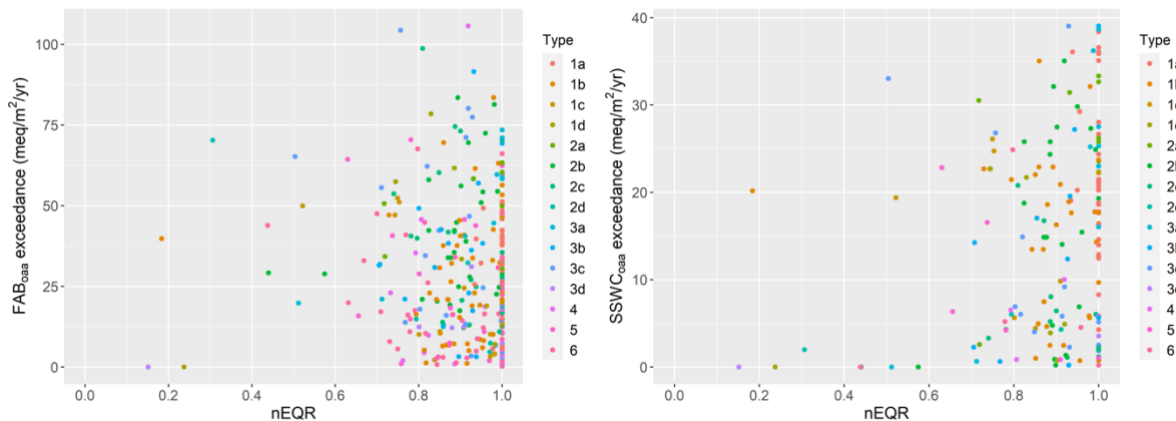
Many of the lakes with exceedance did not only have nEQR > 0.6, but they have nEQR > 0.8, i.e. high status (**Figure 25**), indicating that they were far from being acidified. However, the majority of lakes

with SSWC exceedance were of type 1a, 1b and 2b. Along with type 2a and 3a they were clearly overrepresented compared to the complete dataset. These lake types have particularly small ANC ranges in the WFD classification, so only small year-to-year variations could move them from good to moderate, or even from high to moderate status.



**Figure 24.** Lake  $FAB_{0aa}$  (left) and  $SSWC_{0aa}$  (right) exceedance against ANC for lakes with exceedance  $>0$  meq/m<sup>2</sup>/yr, coloured by lake type. Lakes with ANC  $> 250$  µeq/l are omitted from the plot ( $FAB_{0aa}$  plot: 6 lakes, type 5 or 6;  $SSWC_{0aa}$ : 1 lake, type 6).

The SSWC exceedance is expected to be larger for lakes with lower nEQR, but no such effect was seen (**Figure 25**). This lack of relationship can be indicative of the different natures of these data, as explained at the beginning of this section. A certain randomness is to be expected in this type of comparison. Still, the far higher number of non-acidified than acidified lakes within the exceeded area points to a systematic mismatch. It is difficult to disentangle which factors are most responsible for this. The combined effects of not all lakes being sensitive, particularly high ANC in 2019 and somewhat lower actual deposition than the one used in the exceedance calculations could explain much. However, the results indicate that it is worth revisiting both the WFD classification system and the inputs and fixed parameters used in the critical load exceedance calculations.



**Figure 25.** Lake  $FAB_{0aa}$  (left) and  $SSWC_{0aa}$  (right) exceedance against nEQR, coloured by lake type. Non-acidified (nEQR  $> 0.6$ ) lakes with no exceedance ( $\leq 0$  meq/m<sup>2</sup>/yr) are omitted.



### 3.4 Effects of varying inputs and parameters on the critical load exceedance

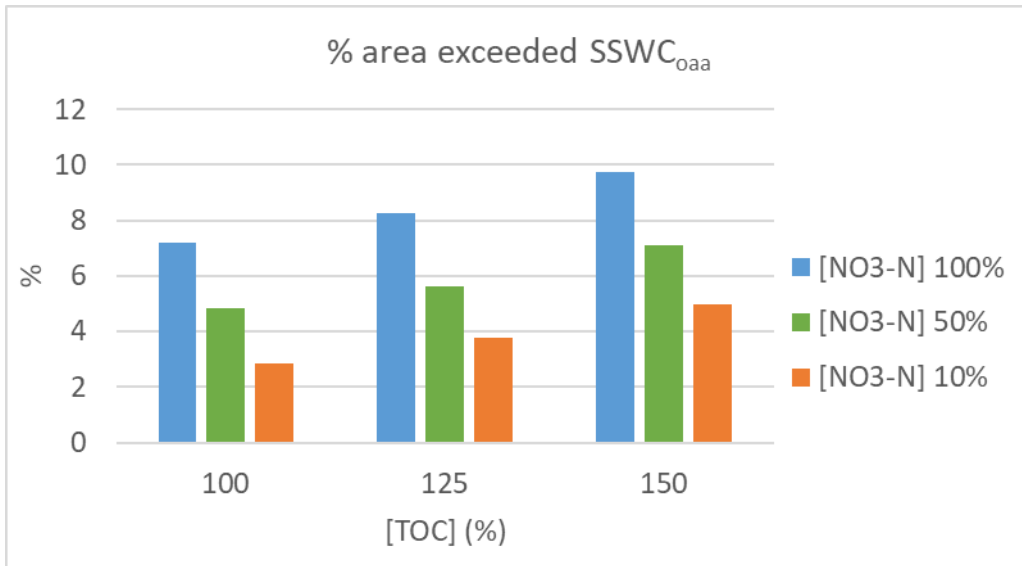
The various inputs and parameters used in the critical loads models and the exceedance calculations affect the exceedance in several ways and to varied extent. Here we vary selected inputs and parameters within reasonable ranges and evaluate the effect on the exceedance. We used the average deposition for 2012-2016. Different approaches to  $[BC]_0^*$  estimation are covered separately in section 3.5. The critical limit will not be explored further here, as any changes should be in line with potential updates of the WFD classification system (Fölster et al., in prep).

Given climate change, the current discharge is likely to deviate from the 1961-1990 normal. According to projections for Norway (Hanssen-Bauer et al., 2015), the discharge is expected to increase, although with regional and seasonal differences, and not very much from 1961-1990 to the new normal period 1991-2020. To test the effect of discharge, it was set to increase by 3 or 5% for all BLRs, with 3% probably being the most realistic. Increasing the discharge generally gives decreased exceedance, but the effect of an increase at this level was marginal. Independent of critical loads model or the combination with other changes the effect on exceeded area was always less than 1%. Though year-to-year variations in discharge may have larger effects on exceedance, the effect of changing the normal period for discharge was minor, so the discharge was kept at 1961-1990 level when exploring other parameter combinations.

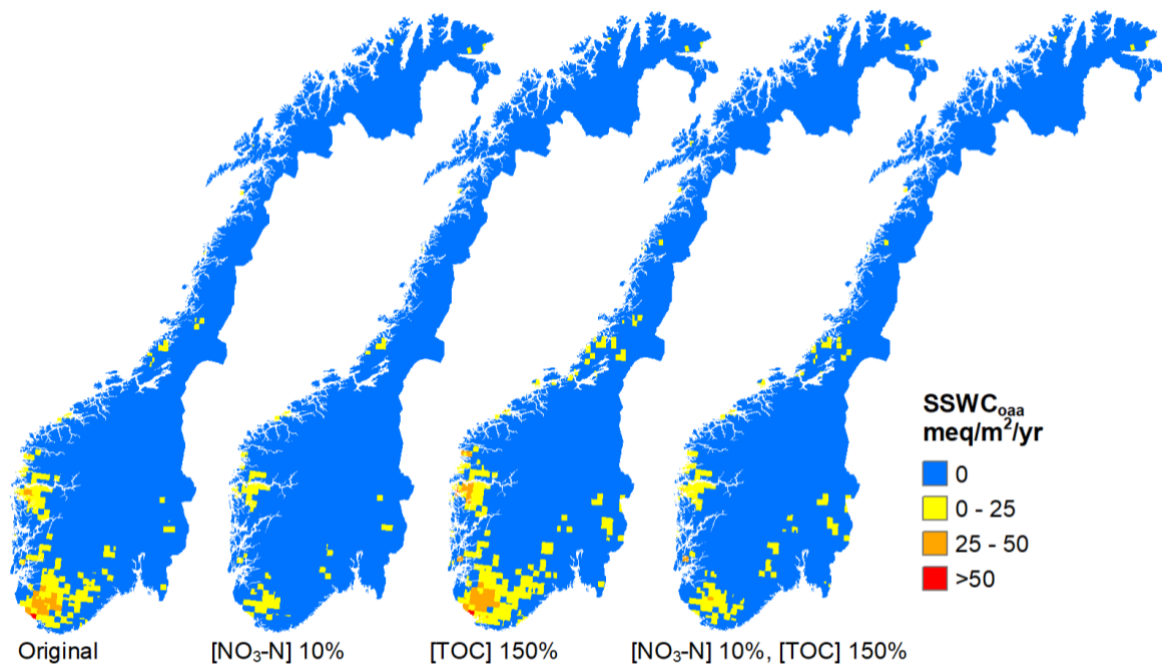
TOC concentration affects the critical loads through the organic acid adjustment. In general, the critical load is lower when TOC is higher. The TOC concentration data in the critical loads database are primarily from the 1995 national lake survey. Given the increase in TOC concentration since then (de Wit et al., 2016), it may be reasonable to update the TOC concentration used. Based on the comparison of results from the 2019 and 1995 lake surveys (Hindar et al., 2020) a general increase in TOC concentration to 125 and 150% of the original was applied.

The nitrate leaching affects the exceedance calculation with the SSWC model. The current nitrate leaching is calculated from the nitrate concentration associated with the grid cells, i.e. data from around 1986. Due to decreased deposition, the nitrate concentrations have decreased since then many places. The decline from 1995 to 2019 in the region Sørlandet was about 10% (**Figure 9**), but larger declines were observed further north in southern Norway (Hindar et al., 2020). To take this and the presumably larger change since 1986 into account, nitrate concentration was reduced to 10% and 50% of the original in the test.

The combined effects of increased TOC and decreased nitrate concentration on the SSWC exceeded area are given in **Figure 26**. **Figure 27** illustrates how both the extent and size of the exceedance are affected. The two types of changes have opposite effects, so the combined effect depends on the actual changes in a given area. A 50% increase in TOC concentration and a 50% decrease in nitrate concentration resulted in about the same % area exceeded on a national scale, while a large change in only one of the parameters had quite a big impact. Large relative changes are observed for both parameters in southern and in particular western parts of southern Norway, but not in exactly the same areas. So locally exceedance may increase or decrease if these two parameters are updated, while the effect on the national level is difficult to predict.



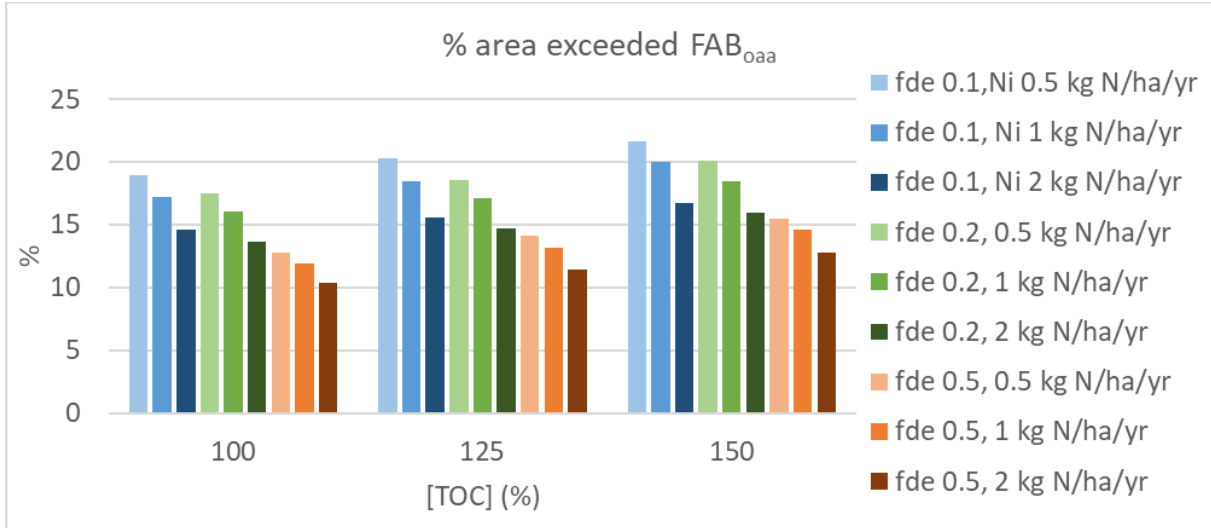
**Figure 26.** Effects of changing the nitrate and TOC concentration on the % of the area of Norway with critical load exceedance, according to the SSWC model and average deposition for 2012-2016. 100% is the currently used TOC and nitrate concentration.



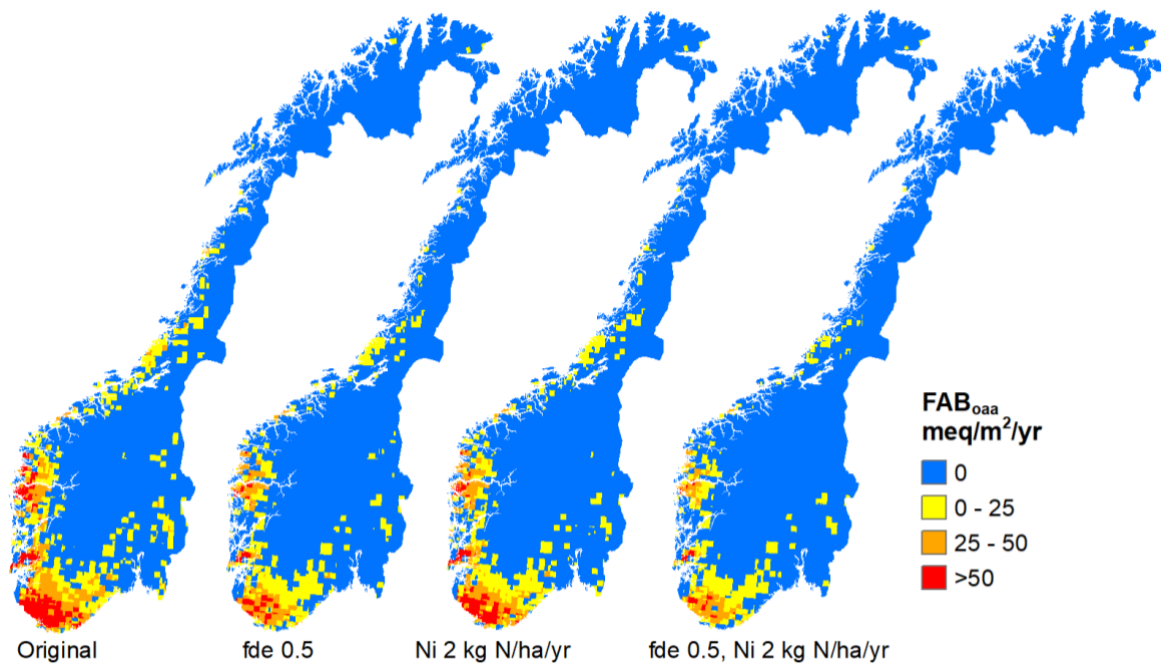
**Figure 27.** Critical load exceedance (SSWC<sub>0aa</sub>, average deposition 2012-2016) for selected combinations of changing TOC and nitrate concentrations compared to the currently used levels (100%).

As explained in section 3.1, the reason for the far higher exceedance with FAB critical loads than SSWC critical loads is the different treatment of nitrogen. Increasing the nitrogen retention would make the exceedance estimates more similar. **Figure 28** and **Figure 29** show the effects of increasing the nitrogen immobilisation and denitrification. Small changes in both only had minor effects, but the combined effect could be substantial. However, TOC concentration also affects FAB exceedance, and

increasing this cancelled out some of the effect of the increased nitrogen retention. Again, the combined effect would depend on the combined changes. If the TOC concentration is increased by 50% it would take a substantial change in at least one of the other parameters to give reduced exceeded area on the national scale.



**Figure 28.** Effects of changing the denitrification fraction (fde), the nitrogen immobilisation (Ni) and the TOC concentration on the % of the area of Norway with critical load exceedance, according to the FAB model and average deposition for 2012-2016. 100% is the currently used TOC concentration.



**Figure 29.** Critical load exceedance (FAB<sub>0aa</sub>, average deposition 2012-2016) for selected combinations of changing the denitrification fraction (fde) and the nitrogen immobilisation (Ni) compared to the currently used constants. The TOC concentration is kept at original levels.

The question is whether it is reasonable to increase the nitrogen retention. The nitrogen immobilisation rate should represent the long-term net accumulation of nitrogen in the root-zone of soils, i.e. an accumulation that does not affect the C/N ratio (CLRTAP, 2017). This immobilisation is commonly estimated based on the nitrogen accumulation in soils since the last glaciation. The value used in the national application (0.5 kg N/ha/yr) is in the range of values found in studies of long-term nitrogen immobilisation (Rosen et al. (1992): 0.2-0.5 kg N/ha/yr; Johnson and Turner (2014): 0.5-1.0 kg N/ha/yr; Höhle and Wellbrock (2017): 0.2-0.8 kg N/ha/yr). However, in Sweden the nitrogen immobilisation is set to 2 kg N/ha/yr for forest soils (Moldan et al., 2017). This was also formerly used in Norway (Henriksen et al., 1996), with reference to (Downing et al., 1993). It may be worth revisiting the literature, identifying the studies that would be most relevant for Norwegian systems. It is also possible to use a differentiated nitrogen immobilisation based on land cover, with higher immobilisation in forest than for other vegetation types. According to the mapping manual (CLRTAP, 2017), nitrogen deposition to bare rock enters the surface water unchanged. This is not currently taken into account in the national application. Given that southern and particularly western parts of southern Norway generally have relatively low forest cover and large areas with barren rock, differentiated nitrogen immobilisation is more likely to result in higher exceedance.

As reviewed by Höhle and Wellbrock (2017), the current nitrogen immobilisation can be much higher (1.8-42 kg N/ha/yr, but also negative rates are found). The steady-state mass balance models used to calculate critical loads by definition take the long-term view and thus the nitrogen immobilisation used in the FAB model should reflect the natural levels. Otherwise there would not be sufficient nitrogen to support the nitrogen immobilisation when N deposition approaches background levels. Prolonged N deposition with higher immobilisation could eventually lead to nitrogen saturation with increased leaching of NO<sub>3</sub> (Aber et al., 1989; Stoddard, 1994). Non-exceedance of FAB critical loads would prevent this from happening and should consequently be the target.

Although signs of N saturation have been observed (Kopáček et al., 2005), is it uncertain whether N saturation will occur in the foreseeable future in areas receiving moderate to low levels of N deposition. Results from long-term N additions to an N-limited forest ecosystem in Sweden indicate no immediate risk of N saturation given current deposition levels (Moldan et al., 2018). The declining nitrate concentrations observed in the Norwegian national lake surveys in 1995 and 2019 do not point to increasing N saturation (Hindar et al., 2020). Moreover, decreased N deposition was shown to give reduced nitrate leaching at three out of four more intensively monitored sites in southern Norway (Kaste et al., 2020). If the immobilisation varies with the deposition level so that most of the nitrogen is retained at any given deposition level, the critical loads estimated by the SSWC model should be sufficient to protect aquatic ecosystems. On the other hand, given the uncertainties regarding the fate of the recently accumulated nitrogen in the soils, the FAB critical loads provide a worst-case scenario, in line with the precautionary principle.

Denitrification (loss of N from soil and water to the atmosphere) is not set as a fixed flux in the critical loads calculations, but as a fraction of the input. This rests on the assumption that denitrification is a slower process than immobilisation (CLRTAP, 2017). Denitrification is thus more in line with current levels at any point in time than the nitrogen immobilisation. The fraction used in the national application (0.1) comes from estimates for well-drained soils (Reinds et al., 2001). The levels used in the test above correspond to moderate and imperfect to poor drainage. A more locally adapted denitrification fraction would require better knowledge of soil conditions. There would, however, be large uncertainties, both in the mapping and in the denitrification fractions used for the different soil types. A simpler approach would be to set the denitrification fraction to 0 for barren

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rock and 0.8 for peatland, in line with the mapping manual, while keeping it at 0.1 for all other terrestrial areas. In the southern and western parts of southern Norway the fraction of barren rock is far higher than that of peatlands, so the combined effect of these changes is likely to be decreased nitrogen retention and increased exceedance.

The remaining sinks in the FAB model have not been tested here. The nitrogen uptake in forests is based on harvest data from the mid-1990s. The harvest volume has increased since then<sup>3</sup>, but the effect of this on the exceedance may be limited, given the relatively low amount of productive forest in southern and in particular western parts of southern Norway. Increasing the lake retention would also increase the natural nitrogen sink and give higher critical loads. Increasing the lake retention of sulphur would also increase the critical load. Adjusting the constants involved in these calculations, however, would only give minor effects, especially for sulphur. Using the actual rather than a fixed lake/catchment ratio would give more realistic values, but on a national scale the effect may be minor.

### 3.5 Comparing two approaches to estimating original base cation concentration

The pre-industrial base cation concentration ( $[BC]_0^*$ ) is an essential parameter in the critical load models. The  $[BC]_0^*$  currently used in the national critical loads calculation is based on results from MAGIC calibrations to selected acidified lakes.  $[BC]_0^*$  was estimated from a linear regression of MAGIC modelled  $[BC]_0^*$  on  $[BC]_{1986}^*$  (see section 3.2). This implies that the ratio between current and pre-industrial base cation concentrations is constant across the country. Given that both the deposition level and the soil base cation saturation varies, this is probably not the case. But since the regression is based on acidified lakes, it can serve as a worst-case approach.

An alternative would be to use the  $[BC]_0^*$  from MAGIC directly. This is not possible for the BLR grid cells, as they are not true catchments and MAGIC has not been calibrated specifically for the BLR grid cells. Austnes et al. (2016) used a matching routine to assign the most recent MAGIC model output for the lakes in the national survey (based on 1995 data) to each of the BLRs. Here the  $[BC]_0^*$  has been extracted from this assigned MAGIC output and set as  $[BC]_0^*$  for the BLRs. The pre-industrial steady state was assumed to be the year 1860, the same as in the regression approach. Critical load exceedance was calculated using this grid-matched  $[BC]_0^*$  and compared to the exceedance resulting from using the linear regression  $[BC]_0^*$ . The exceedance was calculated using average deposition for 2012-2016.

**Table 5** and **Figure 30** show the effect of using the grid-matched  $[BC]_0^*$  on exceedances. Overall the exceeded area was slightly higher than given by the linear regression approach. This means that the grid-matched  $[BC]_0^*$  was lower and thus gave lower critical loads. The maps also show a slightly different distribution of exceedance, with more exceedance in the eastern part of southern Norway and less exceedance in the mid-western part, as well as the northern and south-eastern parts of southernmost Norway.

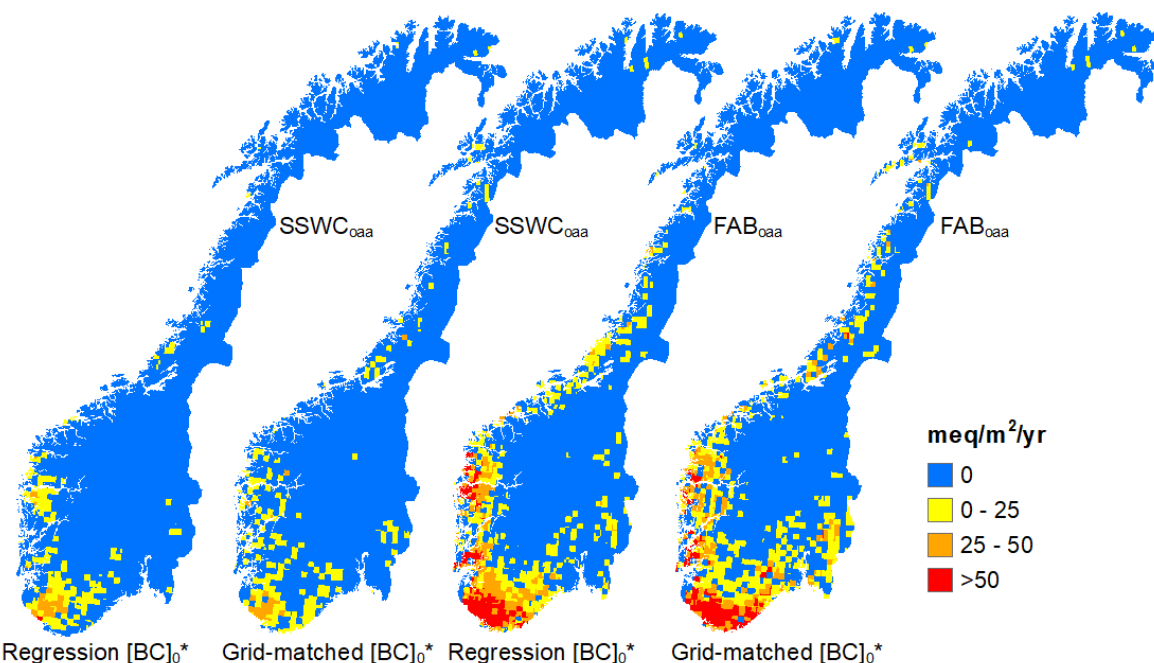
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<sup>3</sup> <https://www.ssb.no/jord-skog-jakt-og-fiskeri/artikler-og-publikasjoner/rekordhoy-tommerhogst>

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**Table 5.** Percentage of the area of Norway with critical load exceedance when using the original linear regression or the grid-matched  $[BC]_0^*$  (average deposition 2012-2016).

$[BC]_0^*$ approach	% exceeded area	
	SSWC <sub>oaa</sub>	FAB <sub>oaa</sub>
Regression $[BC]_0^*$	7	19
Grid-matched $[BC]_0^*$	9	22



**Figure 30.** Critical load exceedance when using the original regression or the grid-matched  $[BC]_0^*$  for SSWC<sub>oaa</sub> (left) and FAB<sub>oaa</sub> (right) (average deposition 2012-2016).

These results are somewhat surprising, as the regression approach was thought to be a worst-case approach. However, as the lakes are matched to BLRs and the BLR water chemistry is biased towards sensitive sites, sensitive lakes are selected also with the grid-matched  $[BC]_0^*$  approach. It is difficult to assess exactly what causes the difference between the results. With two different applications it is likely that there are some differences between the MAGIC model outputs. The input data to the MAGIC modelling behind the regression approach (Larssen et al., 2005) can be assumed to be more accurate, as it included fewer lakes and these were part of the regular national monitoring, so more information was available. On the other hand, the national lake survey calibration from (Austnes et al., 2016) used updated deposition scenarios (including hindcast) and the deposition had finer resolution. The major differences probably result from the matching routine. This is seen also in the shift in geographical distribution of the exceedance.

The matching routine in the grid-matched  $[BC]_0^*$  approach introduces extra uncertainty. The matching gave twelve BLRs with negative critical loads (SSWC), indicating that the  $[BC]_0^*$  was underestimated. Also, the geographical distribution of the exceedance deviates somewhat from where acidification is most commonly observed. Hence, in the choice between these two methods, it is recommended to keep the original approach. If MAGIC output can be used directly and not via a matching routine, this could be a better approach than the linear regression method.

### 3.6 Updating the critical loads – possible approaches

The above discussion has shown that there are reasons to revisit the critical loads calculations. The 2019 national lake survey provides an updated water chemistry dataset that could be used as basis for an update of the critical loads. Two different approaches could be suggested:

1) Using the lakes in the 1000-lake survey as basis for the critical loads:

Critical loads could be calculated for the lakes in the national survey and then extrapolated to the whole country using spatial modelling. This is the approach used in Sweden (Moldan et al., 2017). Then the MAGIC modelled  $[BC]_0^*$  for each lake could be used directly. A re-calibration of the model with updated deposition scenarios and two-point calibration, as described in section 2.5, is expected to give improved estimates of  $[BC]_0^*$ . Using the 2019 lake data would also provide updated TOC and nitrate concentration data.

If this approach is used, it would be reasonable to use the  $0.10^\circ \times 0.05^\circ$  longitude-latitude grid of the LRTAP Convention. In the Swedish application the lake critical loads are transferred to the national grid through inverse distance weighting interpolation, giving most weight to the nearest lakes within a certain radius. This approach could be extended to give more weight to lakes that are more similar to the grid cell in terms of e.g. land cover and geology. Acid sensitive lakes or rivers from other monitoring programmes could also be used. Preferably they would be used directly, but this requires MAGIC to be calibrated. Alternatively, it could be possible to include similarity to these lakes or streams in the spatial modelling.

2) Updating some of the inputs using data from the national lake survey:

A simpler approach not involving re-calibration of MAGIC would be to keep the BLR grid and the  $[BC]_0^*$  associated with them but update some of the other inputs. Since the nitrate and TOC concentration is not used in the estimation of the  $[BC]_0^*$ , it is possible to update this part of the water chemistry associated with the BLRs while leaving the base cation concentrations as they are. This would give critical loads (TOC) more in line with current conditions, and more reasonable SSWC exceedance estimates (nitrate).

The TOC and nitrate concentrations could be assigned to the BLRs using spatial modelling as described above. Here it would probably be sufficient to include distance and land cover as weights. Alternatively, multiple regression models could be set up using the national lake survey dataset with catchment data (e.g. land cover, climate, coordinates, deposition) as explanatory variables.

Independent of choosing 1) or 2), the following should be considered:

- It is advisable to continue using both the SSWC and the FAB models, with SSWC probably being more realistic for current conditions, while FAB serves as a worst-case scenario. However, some changes to the FAB parameters are suggested:
  - The literature on nitrogen immobilisation could be revisited to see if other rates would be more realistic in a Norwegian context. Differentiated immobilisation rates could be considered.
  - Denitrification could be differentiated based on land cover.
  - An evaluation of the change in harvest in areas likely to be exceeded could be made, and if the effect is considered significant, the nitrogen uptake data could be updated. If

approach 1) above is chosen, the nitrogen uptake would have to be revisited in any case, as it has to be assigned to the lake for which the critical load is calculated. Preferably it should be based on the harvest in the lake catchment, but it can also be taken from the BLR to which the lake belongs.

- The land cover distribution could be updated to the actual land cover, either for the lake catchment (1) or the BLR (2). The land cover categories should be lake/water, forest, peatland, grassland/heathland and barren rock/mountain/glacier. The map basis for assigning land cover categories is now much improved and updating this is a prerequisite for some of the suggested changes to the FAB inputs above.
- Even if the effect is not necessarily very big, the discharge data should be updated when the new 1991-2020 normal is available.
- Some lakes should be removed from the national lake survey dataset, based on certain criteria, e.g. very big lakes, lakes influenced by agriculture or urban areas and obvious outliers (cf. section 2.3.1).



## 4 Conclusion

The critical loads methodology used at the national scale in Norway and the MAGIC model as applied to the lakes in the national 1000-lake survey have been evaluated, making use of the data from the 2019 national lake survey. The intent was to assess which parts of the methodologies are most important to improve and which parts *can* be improved in terms of new knowledge and data.

For the acid-sensitive lakes in southern/middle Norway the MAGIC simulations based on data from the 1995 survey gave an acceptable prognosis for the lakewater chemistry measured in the 2019 resurvey. For the lakes in both Sørlandet and Møre and Romsdal the increase in Ca concentrations in the lakes cannot be explained by changes in sulphur or seasalt deposition. Some other factor such as year-to-year variations in temperature or precipitation must be behind the changes observed. Further research is needed to understand the observed increase in Ca concentration.

MAGIC can be re-calibrated to the 2019 lake data and thus produce a new platform for forecasting future changes in acidification of lakes in Norway. Several potential improvements are suggested for a re-calibration of MAGIC using the 2019 data, including two-point calibration. This can be used to evaluate scenarios of changed acid deposition in concert with other environmental drivers, such as climate change and altered forestry practices. The future role of nitrogen deposition is of special interest because nitrogen deposition plays an increasingly important role as sulphur deposition has decreased substantially over the past 30 years.

A comparison of the acidification status of the 1000 lakes with the critical load exceedance showed that most lakes in the areas with critical load exceedance were not acidified. Several possible explanations for this were given. Testing various alternative values for inputs and parameters in the critical loads and exceedance calculations showed that certain combinations of changes could have marked effects on the critical load exceedance.

Two approaches to updating the critical loads are suggested: 1) To calculate critical loads for the 1000 lakes and transfer the results to the national grid. This would require a re-calibration of the MAGIC model to these lakes, but it would give a totally updated basis for the critical loads. 2) Keeping the current setup but updating some of the inputs. Independent of choosing 1) or 2), the FAB parameters should be re-visited, in particular nitrogen immobilisation and denitrification.

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## Appendix A. Outlier lakes Sørlandet

**Table A1.** Outlier lakes in the 2019 national lake survey in the region Sørlandet excluded from the analysis. Highlighted values exceeded the criteria.

	Station ID	NVE nr	Name	Ca	Mg	Na	K	SO <sub>4</sub>	Cl	NO <sub>3</sub>	ANC
Unit				µeq/l	µeq/l	µeq/l	µeq/l	µeq/l	µeq/l	µeq/l	µeq/l
Criterion				>200	>100	>500	>100	>100	>400	>50	>200
<b>Data 2019</b>											
	3205	7954	Grummestadvatnet	161	55	157	10	107	150	17	109
	3389	2508	Skjelbreidtjørna	183	87	254	26	44	264	52	191
	3448	22880	Nordre Vadvatnet	309	88	631	31	125	694	3	236
<b>Data 1995</b>											
	3275	9042	Dalsvatnet	140	119	374	18	135	406	24	87
	3335	21957	HOH 14	158	118	504	18	150	544	15	92
	3337	21881	Engelsvatnet	41	90	448	9	100	496	5	-4
	3389	2508	Skjelbreidtjørna	144	94	220	30	81	257	52	99
	3403	21052	Lauvåsvatnet	61	114	379	17	106	496	13	-44
	3448	22880	Nordre Vadvatnet	110	110	439	22	129	508	5	41
	3450	22892	Steinsvatnet	203	106	352	27	144	460	5	81

## Appendix B. Multiple-point calibration of MAGIC

Larssen et al. (2004) used the long-term data (1974-2000) from Birkenes, one of the calibrated catchments in the Norwegian acid deposition monitoring programme. They calibrated MAGIC to the 5-year observed average chemistry around 1990 and then used the data from 5-year periods around 1980 and 2000 to refine the calibration. They tried to take into account the uncertainties in all the model inputs as well as the uncertainty in the observed stream water chemistry. Each of the model inputs is uncertain. For example, the average soil depth for the Birkenes catchment estimated to 0.4 m might have uncertainty  $\pm 0.1$  m. Six soil parameters and two surface water (stream) parameters were given uncertainty ranges (**Table B1**). They then used a Monte-Carlo procedure to choose values for each of the input parameters from the specified ranges. This was repeated 10000 times to give 10000 independent sets of input parameters. Each of the parameter sets was then used in the automatic optimisation routine for the base cations (**Table B2**). The resulting simulated stream and soil chemistry for the reference year 1990 was compared to the observed. An uncertainty range was calculated for each of the seven water chemistry (Ca, Mg, K, SO<sub>4</sub>, NO<sub>3</sub>, Cl) and five soil chemistry parameters (ECa, EMg, ENa, EK, pH) observed in the 5-year window (**Table B3**). They found that 8891 of the 10000 data sets were successfully calibrated, but only 2234 passed all three 5-year windows. Larssen et al. (2004) found that two-point calibration (5-year periods 1980 and 1990) significantly narrowed the band of forecast water chemistry. Adding the third point (5-year period 2000) did not further improve the predictions. In other words, nearly all the sets of input parameters that passed the 1980 and 1990 windows also passed the 2000 window (**Figure B1**).

**Table B1.** Ranges for soil and stream parameters used as model inputs by Larssen et al. (2004) in the MAGIC application to the Birkenes catchment. For all parameters in the table a rectangular shape was used for the distributions.

	Unit	Min	Max
<b>Soil parameters</b>			
Depth	m	0.30	0.50
Porosity	%	40	60
Bulk density	kg/m <sup>3</sup>	695	850
Cation exchange capacity	meq/kg	95	117
CO <sub>2</sub> -pressure	% of atm.	0.50	2.00
Solution organic charge	μmol/l	0	250
<b>Stream parameters</b>			
CO <sub>2</sub> -pressure	% of atm.	0.05	0.20
Solution organic charge	μmol/l	0	25

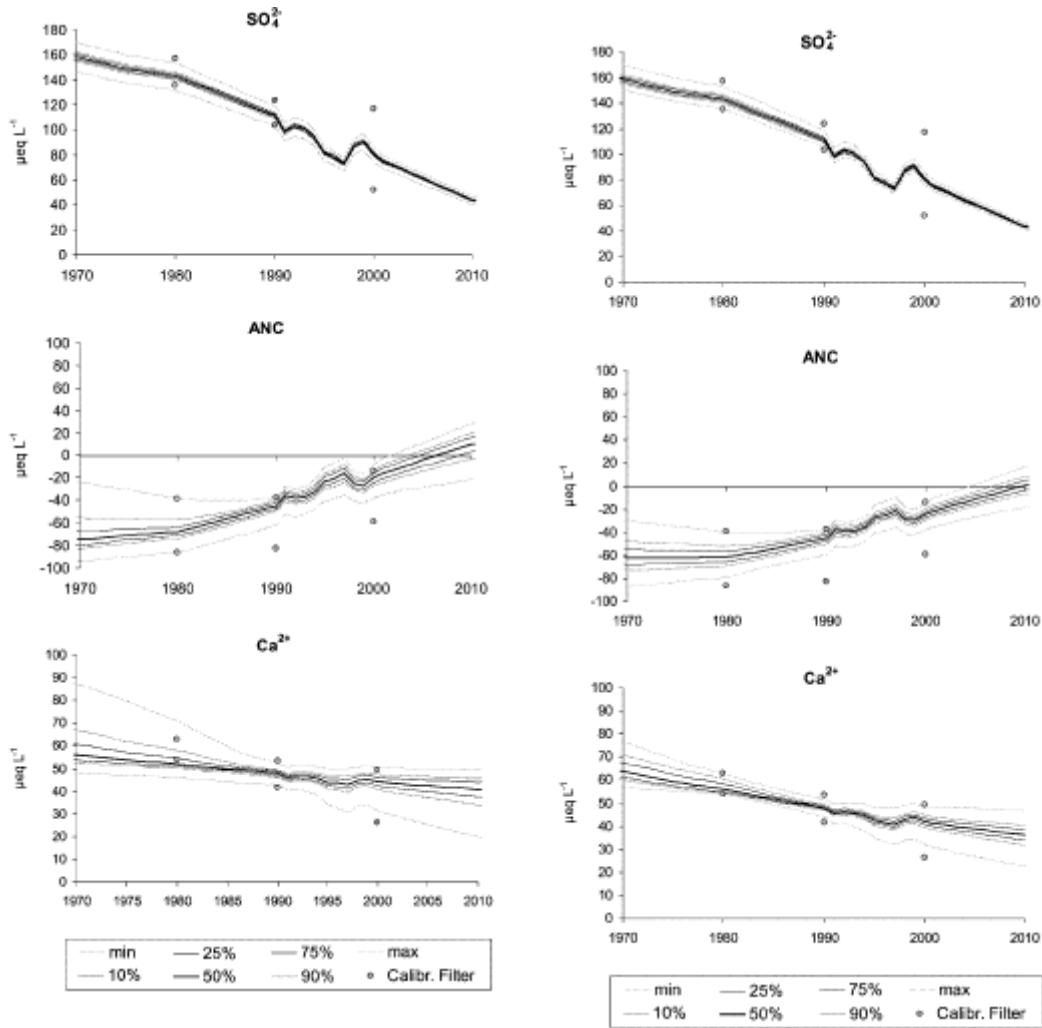
**Table B2.** Initial ranges for the calibrated parameters used by Larssen et al. (2004).

	<b>Unit</b>	<b>Min</b>	<b>Max</b>
<b>Soil weathering rates</b>			
Ca	meq/m <sup>2</sup> /yr	0	100
Mg	meq/m <sup>2</sup> /yr	0	100
Na	meq/m <sup>2</sup> /yr	0	100
K	meq/m <sup>2</sup> /yr	0	100
<b>Aluminium dissolution constants</b>			
Soil	log 10	6	11
Stream	log 10	6	11
<b>Initial soil base cation saturation</b>			
Ca	%	0.1	50
Mg	%	0.1	50
Na	%	0.1	50
K	%	0.1	50
NO <sub>3</sub> uptake	%	0	100

**Table B3.** Limits for surface water chemistry used by Larssen et al. (2004) in the calibration refinement procedure. Units: µeq/l. The limits reflect the observed annual means for each of the 5-year periods.

	<b>Min</b>	<b>Max</b>
<b>1980</b>		
Ca	54	63
SO <sub>4</sub>	135	157
ANC	-86	-39
<b>1990</b>		
Ca	44	51
SO <sub>4</sub>	106	122
ANC	-83	-40
<b>2000</b>		
Ca	26	49
SO <sub>4</sub>	52	117
ANC	-59	-14





**Figure B1.** Larssen et al. (2004) results of the MAGIC application to Birkenes. Shown are the calibration, post-calibration filtering and modelled distributions of concentrations of  $SO_4$ , Ca and ANC in stream water using only one time window (1990 – left-panel) and with all three windows (right-panel).

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