Re-calibration of the MAGIC model with data from the National Lake Survey 2019.

Norwegian Institute for Water Research

REPORT

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Summary

The MAGIC model (Model for Acidification of Groundwater In Catchments) was calibrated to the Norwegian 1000-lake survey data from both 1995 and 2019. The re-survey in 2019 provided a new point in time for generation of new forecasts for future lake chemistry. The acidification pressure on Norwegian lakes has decreased dramatically since the 1970s. For the majority of the lakes in the 1000-lake survey, the large reductions in S deposition since the peak years in the 1970s have resulted in decreased SO_4 concentrations and increased ANC levels. Other driving factors that were previously masked by acid deposition may now become more apparent, as e.g., the effects of climate change. For MAGIC to fully simulate the effects of climate change, additional processes must first be included in the model.

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Re-calibration of the MAGIC model with data from the National Lake Survey 2019.

Preface

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Summary

The MAGIC model (Model for Acidification of Groundwater In Catchments) was calibrated to the Norwegian 1000-lake survey data from both 1995 and 2019. MAGIC is widely used to simulate the effects of acid deposition on the chemistry of surface waters, and to estimate the expected changes given future rates of acid deposition. The 1000-lake re-survey in 2019 provided a new point in time for generation of new forecasts for future lake chemistry using MAGIC. There were 987 lakes sampled in both 1995 and 2019 that included sufficient data for the MAGIC calibrations. We used a version of MAGIC that was recoded in Mobius, NIVA's new model platform https://github.com/NIVANorge/Mobius). The results were the same as from the MAGIC version 777 in Fortran.

Estimates for deposition of sulphur and nitrogen compounds for the period 1990-2020 were obtained from the European Monitoring and Evaluation Programme (EMEP, at met.no). Estimated deposition for years prior to 1990 were scaled and coupled to the EMEP data. The EMEP data released in September 2022 and used in this report gave higher deposition in the peak years compared to the estimates used in the 2008 and 2016 calibrations that were done using the 1995 1000-lake survey data. This affected the calibration of MAGIC parameters such as weathering rates of the base cations, and thus the simulated historical and future acid neutralising capacity (ANC) concentrations in the lakes.

The MAGIC model requires a wide range of input data and parameter values. We reviewed and updated site characteristics such as lake and catchment area, and lake mean depth. Estimates of specific discharge for the new standard period 1991-2020 were obtained from NVE. There were no new data available for soil or forests. Deposition of sea salts was estimated from the observed concentrations of chloride in the lakes. New estimates for weathering of sulphate (SO_4) were made based on EMEP deposition and observed SO_4 concentrations in the lakes. Retention of nitrogen (N) in the catchments and lakes (as % of N deposition) was assumed constant over time.

We did three calibrations: (1) using the 1995-survey data, (2) using the 2019-survey data, and (3) simultaneously using both the 1995- and 2019-survey data (2-point calibration). The calibrations were conducted stepwise with optimisation first of chloride (Cl), SO_4 and nitrate (NO_3) in that order, and then on the four base cations (calcium (Ca), magnesium (Mg), potassium (K), sodium (Na)) simultaneously. Prime focus was on the simulated vs observed values in both 1995 and 2019 for ANC and the eight components that comprise ANC.

The MAGIC calibrations using the 1995-survey data simulated ANC in 2019 to lower values than were observed. This was particularly apparent for the acid-sensitive lakes, i.e. those that had ANC less than 200 µeq/l. The discrepancy in ANC was mostly because simulated Ca concentrations were about 10% lower than measured, and secondly because simulated SO_4 concentrations were lower than observed. The discrepancy in Ca was about three times larger than the discrepancy in SO_4. The MAGIC calibrations using the 2019-survey data gave similar results as for using the 1995-survey data, but with opposite sign. And the 2-point calibration simulated a trend in-between.

The increase in Ca concentrations from 1995 to 2019 is not captured by MAGIC. An increase in Ca has also been seen in data from the 78 "trend" lakes in Norway sampled annually since 1995. That the simulated SO_4 concentrations in 2019 were lower than observed may be because the actual sulphur (S) deposition between 1995 and 2019 was higher than the EMEP estimates. Whether this is indeed

the case can be assessed by comparing S deposition estimated from measurements at NILUs precipitation stations with the EMEP modelled estimates.

Proper estimates of S deposition emerge as a key step in the setting of the input data to MAGIC. This holds both for the absolute S deposition estimated at each lake for the two sampling times, 1995 and 2019, but also to the deposition history from 1800 to 2019. The EMEP values for 1995 and 2019 clearly overestimate S deposition at some lakes and underestimate S deposition at other lakes. The trends in EMEP S deposition directly affect the ability of MAGIC to simulate the observed changes in SO_4 concentrations in the lakes, and as a result also the concentrations of base cations. The long-term depletion of soil base cation pools is directly influenced by the total cumulative S deposition since the start of the industrial revolution.

N deposition had little influence on the ANC concentrations in the lakes, as most of the incoming N was retained in the catchment-lake system. Interannual variations in sea salt deposition caused "noise" in the ANC results.

The acidification pressure on Norwegian lakes has decreased dramatically since the 1970s. For the majority of the lakes in the 1000 lake survey, the large reductions in S deposition since the peak years in the 1970s have resulted in decreased SO_4 concentrations and increased ANC levels. Other driving factors that were previously masked by acid deposition may now become more apparent. Climate change is one of these factors. Increasing temperature, changes in precipitation, and frequency and severity of storms all can affect biological and chemical processes in catchments and the lakes themselves. For MAGIC to fully simulate the effects of climate change additional processes active in the catchment and lake must first be included in the model.

Sammendrag

MAGIC-modellen (Model for Acidification of Groundwater In Catchments) er kalibrert til den norske 1000-sjøers undersøkelsen med data fra både 1995 og 2019. MAGIC er mye brukt for å simulere effekter av sur nedbør på vannkjemi i elver og innsjøer, og for å estimere forventede endringer basert på scenarier for fremtidig avsetning av sure forbindelser. Den nye 1000-sjøers undersøkelsen i 2019 gav et nytt referansepunkt for å bruke MAGIC til å lage nye og bedre prognoser for framtidig vannkjemi. I alt 987 av de 1000 innsjøene hadde tilstrekkelige inngangsdata til å kalibrere MAGIC både for 1995 og 2019. Modelleringen ble gjort med en ny versjon av MAGIC som er omprogrammert for å kunne legges på NIVAs nye modellplattform Mobius (https://github.com/NIVANorge/Mobius). Resultatene basert på den nye versjonen ble funnet å være identiske med output fra den opprinnelige MAGIC versjon 777 som er programmert i Fortran.

Estimater for avsetning av svovel- og nitrogenforbindelser for perioden 1990-2020 er hentet fra EMEP (European Monitoring and Evaluation Programme, ved met.no). Estimert avsetning for årene før 1990 ble skalert og koblet til EMEP-dataene. EMEP-dataene som ble frigitt i september 2022 og brukt i denne rapporten ga høyere avsetning i toppårene sammenlignet med estimatene som ble brukt i 2008- og 2016-kalibreringene til 1995-dataene for 1000-sjøene. Dette påvirket kalibreringen av MAGIC-parametere som blant annet forvitringsrater for basekationer, noe som i sin tur påvirket de simulerte historiske og fremtidige nivåer av syrenøytraliserende kapasitet (ANC) i innsjøene.

MAGIC-modellen krever et bredt spekter av inngangsdata og parameterverdier. I forkant av kalibreringen ble data på innsjøarealer, nedbørfeltarealer og estimater for middeldyp i innsjøene evaluert og oppdatert. Vannføringsdataene ble oppdatert i forhold til den nye avrenningsnormalen 1991-2020 fra Norges Vassdrags- og Energidirektorat (NVE). Det var ingen nye data tilgjengelig for jord eller skog. Avsetning av sjøsalter ble estimert ut fra de observerte konsentrasjonene av klorid (Cl) i innsjøene. Nye estimater for forvitring av sulfat (SO$_4$) ble basert på EMEP-modellert avsetning og observerte konsentrasjoner av SO$_4$ i innsjøene. Retensjon av nitrogen (N) i nedbørfelter og innsjøer (som % av N avsetning) ble antatt å ha holdt seg konstant over tid.

Vi kalibrerte modellen tre ganger: (1) til 1995-dataene, (2) til 2019-dataene og (3) samtidig til både 1995- og 2019-dataene (2-punkts kalibrering). Kalibreringene ble utført trinnvis med optimalisering av komponentene som utgjør ANC, dvs. Cl, SO$_4$ og nitrat (NO$_3$, i nevnt rekkefølge) og deretter med hensyn til de fire basekationene samtidig.

MAGIC-kalibreringene til 1995-dataene simulerte lavere ANC-verdier i 2019 enn det som var observert. Dette var spesielt tydelig for de forsuringsfølsomme innsjøene, dvs. de som hadde ANC lavere enn 200 µeq/l. Avviket i ANC skyldtes hovedsakelig at simulerte konsentrasjoner av kalsium (Ca) var omtrent 10 % lavere enn de målte, og sekundært fordi simulerte SO$_4$-konsentrasjoner var lavere enn de som var observert. Avviket i Ca var omtrent tre ganger større enn avviket i SO$_4$. MAGIC-kalibreringene til 2019-dataene ga lignende resultater for 1995, men med motsatt fortegn. To-punkts kalibreringen (til både 1995 og 2019) simulerte en trend som lå mellom de to første kalibreringene.

Økningen i Ca-konsentrasjoner i innsjøene fra 1995 til 2019 (som også er sett i de årlige prøvene fra de 78 norske «trend-innsjøene») fanges ikke opp av MAGIC. At de simulerte SO_4-konsentrasjonene i 2019 var lavere enn observert kan skyldes at den faktiske S-avsetningen i perioden 1995-2019 var høyere enn EMEP-estimatene. Hvorvidt dette faktisk er tilfelle, kan vurderes ved å sammenligne S-avsetning estimert fra målinger ved NILUs nedbørstasjoner med EMEP-modellerte estimater.

Realistiske estimater for S-avsetning er en nøkkelfaktor i inputdataene til MAGIC. Dette gjelder både den absolutte S-avsetningen estimert for hver innsjø i 1995 og 2019, men også den historiske avsetningen fra 1800 til 2019. De EMEP-modellerte dataene for 1995 og 2019 overestimerer S-avsetningen for noen innsjøer, mens de underestimerer avsetningen for andre. Trendene i EMEP-modellert S-avsetning påvirker MAGIC-modellens evne til å simulere observerte endringer i innsjøenes SO_4-konsentrasjon, og som et resultat av dette, også konsentrasjonene av basekationer. Reduksjonen av jordas basemetning over tid er direkte påvirket av den totale kumulative S-avsetningen siden starten av den industrielle revolusjon mot slutten av 1800-tallet.

N-avsetningen hadde liten innflytelse på ANC-konsentrasjonene i innsjøene, da mesteparten av det innkommende N blir holdt tilbake i innsjøenes nedbørfelt. Mellomårlige variasjoner i sjøsaltavsetning bidrar imidlertid til "støy" i ANC-resultatene.

Forsuringstrykket på norske innsjøer har avtatt betydelig siden 1970-tallet. For flertallet av innsjøene i 1000-sjøers undersøkelsen har de store reduksjonene i S-avsetning siden toppårene på 1970-tallet resultert i reduserte SO_4-konsentrasjoner og økte ANC-nivåer. Andre påvirkningsfaktorer som tidligere ble maskert av den sure nedbøren kan nå bli mer tydelige. Klimaendringer er en av disse faktorene. Økende temperatur, endringer i nedbørmengde og økt hyppighet og intensitet av stormer er alle faktorer som kan påvirke biologiske og kjemiske prosesser i nedbørfeltene og i selve innsjøene. For at MAGIC skal kunne simulere effektene av klimaendringer, må ytterligere biogeokjemiske prosesser i jord og vann først inkluderes i modellen.

1 Introduction

1.1 Background

The MAGIC model (Model for Acidification of Groundwater In Catchments) (Cosby et al. 1985a, Cosby et al. 1985b, Cosby et al. 2001) is widely used to simulate the effects of acid deposition on the chemistry of surface waters, and to estimate the expected changes given future rates of acid deposition. MAGIC has been applied to data from the Norwegian 1000 lake survey from 1995 (Skjelkvåle et al. 1996), in order to make predictions for the future development of water quality in the lakes (Larssen et al. 2008, Austnes et al. 2016). The re-survey of the 1000 lakes in 2019 (Hindar et al. 2020, de Wit et al. In review) provides the opportunity to evaluate the ability of the model calibrated to the 1995 data to satisfactorily simulate the observed lake chemistry data measured in 2019. The re-survey in 2019 also provides a new point in time with which to generate new forecasts for future lake chemistry using MAGIC.

MAGIC has recently been recoded and transferred to NIVA's new model platform Mobius (Norling et al. 2021). This provides more flexibility and simpler routines for auto-calibration and sensitivity analysis, and for calibration to data from more than one point in time. Here we use the recoded version of MAGIC to re-calibrate the model based on the 1000 lakes data from both 1995 and 2019. We expect that a single calibration using two points in time, rather than one, provides a more robust basis for forecasting changes in lake chemistry given future changes in drivers such as acid deposition, climate change and land-use change.

To test this, we calibrate the model in three ways (once to the 1995 survey data, once to the 2019 survey data, and once to both datasets together) and compare the model simulations with observed data from the surveys. We discuss the model performance in the context of uncertainties in input data, calibration parameters, and representation of processes in the model.
 The aim is to provide the best possible basis for forecasting changes in lake chemistry given future changes in drivers such as acid deposition, climate change and land-use change.

2 Methods

2.1 New developments of MAGIC on the Mobius platform

Mobius is a new modelling platform developed at NIVA (Norling et al. 2021). Since its inception in 1984 MAGIC has been programmed in FORTRAN. The model has now been transferred to Mobius, a modelling platform programmed in C++ and open source. MAGIC can be used on individual catchments (with and without a lake) and for running in "batch mode". The latter means that it can be calibrated simultaneously for a larger number of lakes, such as, for example, the 1000-lake data set.

2.1.1 Routines for transferring parameter files from old to new formats

A script has been created that reads and transfers data from the old parameter files to the new format. Input data from both years (1995 and 2019) has been updated and read in.

2.1.2 Programming of auto-calibration and two-point calibration

A python script was created to run auto-calibration of MAGIC in the same way that MAGICOPT did in MAGIC version 777 (FORTRAN). The Python script is open source and is easier to adapt to new runs, for example to allow calibration against multiple measurement points per lake (cf. the two-point calibration here for the 1000 lakes). The calibration now takes place against a sum of squares of the errors in the measured against the modelled values and can later easily be used for calibration against many more observed data points in time.

2.1.3 Programming uncertainty analysis (Monte Carlo)

In the uncertainty analysis, a random selection of parameter sets is made from the parameter space consisting of a selection of the uncertain parameters such as soil depth, cation exchange capacity etc. and a separate calibration is run for each parameter set. This provides a distribution of possible parameter sets and model results for future years. This can provide confidence intervals for future development of the lake water chemistry. The uncertainty analysis is also implemented in Python and is easy to configure. The uncertainty analysis is most useful in applications to one site. It was not used here in the calibrations to the 1000-lake data set.

2.2 Selection of lakes

The 1995 survey comprised 1006 lakes. Of these 990 were successfully calibrated by Larssen et al. (2008) and Austnes et al. (2016). The remaining 16 had incomplete or questionable data or could not be calibrated given the optimisation criteria. The survey in 2019 (1001 lakes) included 983 of the lakes sampled in the 1995 survey (de Wit et al. In review). Five lakes that are strongly impacted by point nutrient sources (i.e., over 50% agriculture and/or PO_4 concentrations > 30 µg L^{-1} in 2019) were removed, leaving 978 lakes for further analysis. There were thus 978 lakes common to both 1995 and 2019 and with complete data available for the MAGIC calibrations.

2.3   Update of deposition data

Modelled estimates for deposition of sulphur and nitrogen compounds for the period 1990-2020 were obtained from The Norwegian Meteorological Institute through the Meteorological Synthesizing Centre - West (MSC-W) of EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe). These data were available online at https://emep.int/mscw/mscw_moddata.html

EMEP now gives estimates for each year. Previously estimates were given for every 5 years. The EMEP data are now on a 0.1 x 0.1 degree (latitude x longitude) grid. We located each lake within a respective EMEP grid cell and used BILINEAR interpolation to obtain the local deposition (see Annex A). We used the EMEP data for grid-average wet plus dry deposition of oxidized sulphur (WDEP_SOX + DDEP_SOX_m2Grid), oxidized nitrogen (WDEP_OXN + DDEP_OXN_m2Grid), and reduced nitrogen (WDEP_RDN + DDEP_RDN_m2Grid).

MAGIC requires estimates of historical S and N deposition at each of the 1000 lakes for the years prior to 1990. These are based on Schöpp et al. (2003) and have not been revised. The historical data are scaled and coupled to the EMEP data in the year 1990 such that the Schöpp et al. (2003) estimate for 1990 agrees with the EMEP estimate for 1990 in every grid cell (Annex A, Figure A1). This work was done by Max Posch at IIASA.

Scenarios for future deposition of S and N in Norway were not released by EMEP in time for this report.

Over the past 25 years EMEP has several times revised the estimates for S and N deposition in Europe. EMEP explains these changes as due to updates in the EMEP MSC-W model and to modifications of the input data (e.g. revised emission data and meteorology). These revisions are made for deposition in the period since 1990. Previous MAGIC calibrations to the 1000-lake data conducted by Larssen et al. (2008) and Austnes et al. (2016) used the EMEP data then available. The EMEP data released in September 2022 and used in this report are again different from the estimates used in the 2008 and 2016 calibrations to the 1995 1000-lake data (Figure 1). For the example shown, lake 36 Lille Hovvatn, the 2022 estimates of S deposition in the peak years are significantly larger than those used previously. Changing estimates of S and N deposition affects the calibration of MAGIC parameters such as weathering rates of the base cations, and thus simulated historical and future ANC concentrations in the lakes.
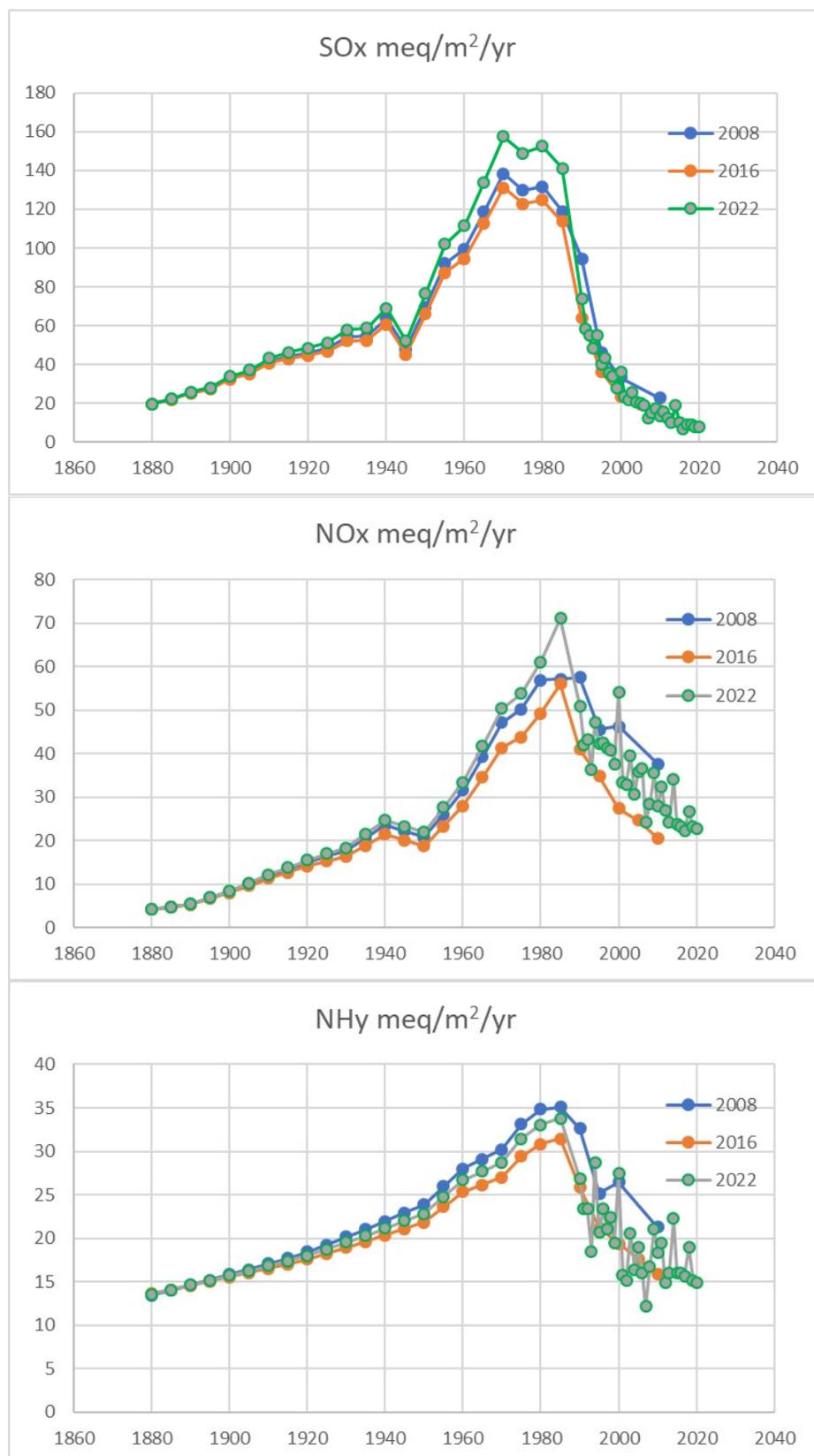
Figure 1. Estimates of S and N deposition from EMEP used to calibrate MAGIC by Larssen et al. (2008), Austnes et al. (2016) and this study (2022). Shown are estimates for lake 36 (Lille Hovvatn located near Evje, Aust-Adger county, Latitude 58.607, Longitude 8.029) of the 1000-lake survey.

2.4   Update of input data and other parameters

The MAGIC model requires a wide range of input data and parameter values. These are listed by Larssen et al. (2008) and must be specified for each lake. We have conducted a review and update of the input data and assumptions prior to re-calibration of MAGIC to the 1995 and 2019 data.

2.4.1 Catchment and lake area

Previous calibration

For the 1995 survey the catchment and lake areas were obtained from NVE's lake database, and for small lakes not included in the database the area was manually estimated from maps by using a planimeter.

Selected approach for new calibration

Catchment areas have been revised based on digital tools from NVE and NIVA. Catchment boundaries were delineated based on a 10x10m digital elevation model (DEM) available at https://www.geonorge.no. The coordinates of the sampling location were used to select lake polygons from NVE's lake database. The location of the outlet was set by intersecting the polygons with NVEs digital river network maps (https://nve.no/map-services/). The DEM was then processed with the TauDEM package version 5 (Tarboton 1997) to delineate the catchment of each lake outlet point. The resulting catchment polygons were also used to extract other catchment properties such as elevation and land cover (https://www.kartverket.no/en; AR50 data).

In cases where the new catchment area estimates differed significantly from the previous estimates, additional checks were made with NVE's catchment delineation tool (https://nevina.nve.no/). In most cases the new estimated proved to be better. The only exceptions were lakes with catchments stretching into Sweden or Finland. Here, the new catchment areas are correspondingly underestimated as areas outside Norway are not included in Nevina. Given that it only concerns a small number of lakes, the new catchment area estimates were applied on the entire data set and used for all the recalibrations.

Lake area data were left unchanged from Larssen et al. (2008), i.e. they were obtained from NVE's lake database or for the smallest lakes manually estimated from maps.

2.4.2 Discharge

To calculate the fluxes of the various components and the water retention time in the lakes, MAGIC needs an estimate of the annual water discharge for each of the 1000 lakes.

Previous calibration

In 1995, the water flow was taken from NVE's national discharge map, which is based on measured discharge in the "normal" period 1961-1990. It was assumed that the annual discharge was constant in all years in the simulation period 1850-2030.

Selected approach for new calibration

NVE has produced national discharge data for the new normal period 1991-2020. These can be used along with the 2019 lake data. Discharge may have changed since the previous 30-year normal due to climate change. NVE also has digitized discharge data.

Both 1995 and 2019 lie within the "new" standard NVE period of 1991-2020. We chose to use these for all years in the simulation period 1850-2030.

2.4.3 Water residence time in the lakes

The residence time of the water is used in MAGIC for two purposes: (1) to calculate denitrification in the lakes and (2) to "smooth out" year-to-year variations in the runoff fluxes from land. Water residence time is calculated from the catchment area, the lake area, the mean depth of the lake and the annual discharge from the outlet. Units are yr.

Previous calibration

In the previous calibration, the lake's mean depth (and thus water residence time) was estimated based on the area of the lake (Table 1).

Table 1. Estimated lake water residence times based on lake area.

Lake area, km^2	> 100	10 - 100	1 - 10	< 1
Water retention time (yr)	5	2	1	0.5

Selected approach for new calibration

In most cases, there are no measured data for mean depth or water residence time for the 1000 lakes. However, in connection with recent developments of the TEOTIL model, Sample et al. (2022) developed a statistical model for estimating lake mean depth based on lake area and additional landscape-related covariates, such as mean slope and height. The water residence times in the 1000 lakes was re-calculated based on these new estimates by means of the new statistical model (Sample et al. 2022). The new estimates were used in the MAGIC calibrations.

2.4.4 Soil data

Previous calibration

- Based on data from NIJOS (now NIBIO); national forest mapping (9x9 km grid)
- The soil data was aggregated up to the critical loads grid (12x12 km)
- 345 lakes were in grid cells with forest data available, while 658 did not.
- Soil data for the lakes in non-forested catchments were obtained from calibrated catchments or from relevant research projects. An overview of how soil data is assigned to the 658 "non-forested" lakes is given in Larssen et al. (2008).

Selected approach for new calibration

We checked NIBIO's forest soil database whether data has been added in later years and could expand the nationwide data set we already have from NIJOS. As little new soil data has been collected from non-forested areas since the previous calibration in 2008, we use the same data as in the 1995 calibration given by Larssen et al. (2008).

2.4.5 Forest uptake data

Previous calibration

- Based on data from NIJOS (now NIBIO); national forest mapping (9x9 km routes)
- 345 lakes were in grid cells with forest data available, while 658 lakes did not.
- Base cation uptake (actually annual net accumulation in biomass) was assumed to be zero for non-forested areas. For forests, the rates were assumed to be Ca 21 meq/m^2/yr, Mg 4 meq/m^2/yr, and K 4.5 meq/m^2/yr.

Selected approach for new calibration

As there are no new forest uptake data available, we use the same data as in the 1995 calibration given by Larssen et al. (2008).

2.4.6 Deposition of sea salts

Previous calibration

In the MAGIC calibrations based on the 1995 data, there were no data from EMEP for the deposition of sea salt ions. Thus, it was assumed for each lake that Cl deposition was equal to Cl flux from the lake (i.e., concentration multiplied by annual water flow). It was assumed that there were no other sources of Cl, such from pollution or weathering of soil minerals. Deposition of Ca, Mg, Na, K and SO$_4$ from sea salts was then calculated from Cl deposition and the ratios for the individual compounds in seawater (Table 2).

Table 2. Sea-salt contribution of base cations and sulphate calculated in relation to chloride deposition (μeq/μeq).

	Ca	**Mg**	**Na**	**K**	**SO$_4$**
Ratio to Cl in seawater (μeq/μeq)	0.0376	0.192	0.858	0.0188	0.104

Selected approach for new calibration

Cl is among the anions included in the calculation of ANC. The main source of Cl is deposition of sea salts. Cl is only weakly retained in the soil. Deposited Cl therefore flows into the lake within a short time (in most cases less than a year). Cl deposition varies from year-to-year, depending on the frequency and intensity of storms. Figure 2 shows year-to-year variations in sea salt deposition at 6 of NILU's rainfall stations. Mg, which is also an important component of sea salt, is used as a surrogate for Cl.
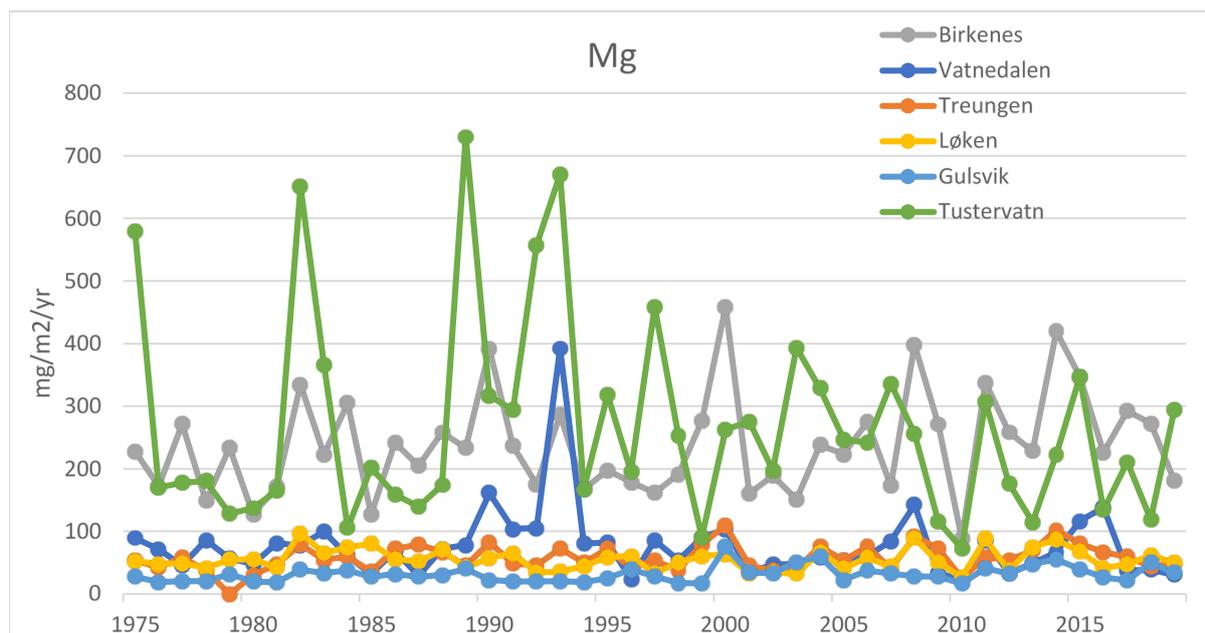
Figure 2. Mg deposition measured at 6 NILU stations in Norway. NILU uses Mg as a surrogate for Cl. Data from (Aas et al. 2020). Figure from Austnes et al. (2020).

For 2-point calibration (1995 and 2019), we assumed as before that Cl dep = Cl flux out of the lakes. We then assume that for all other years the mean of the values in 1995 and 2019 provides the best estimate of Cl dep (Figure 3). Deposition of Ca, Mg, Na, K and SO_4 in sea salt is calculated, as before, from Cl deposition and the ratios in Table 2.

Figure 3. Schematic example of input values for Cl deposition (meq/m^2/yr) for a lake with much lower Cl-flux in 2019 relative to 1995.

2.4.7 S deposition and S weathering

Previous calibration

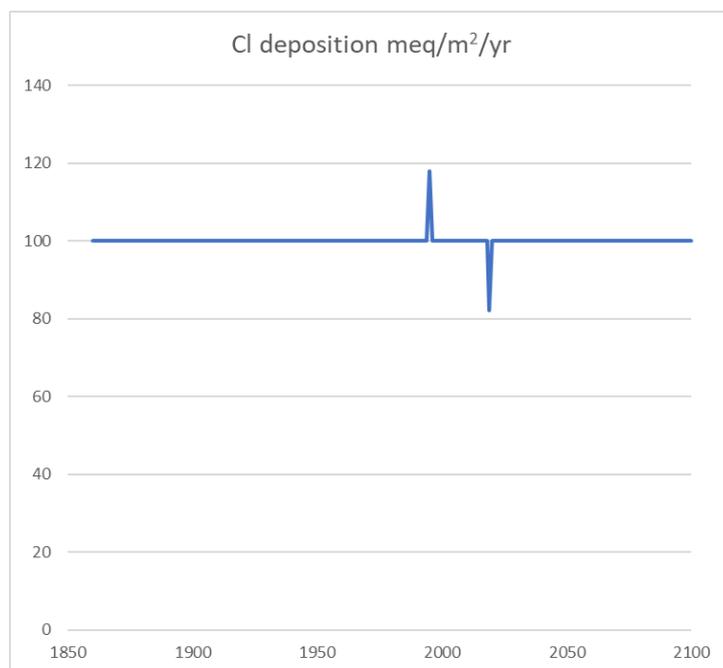The flux of SO_4 from the lakes in 1995 was assumed to be equal to the sum of contributions from sea salts, non-marine SO_4 deposition (the contribution from air pollutants) and weathering. Non-marine SO_4 is also often termed excess-SO_4 and is generally denoted as SO_4^*.

It is generally difficult to use measured deposition data directly in the modelling, because it will always deviate somewhat from the local deposition in the catchment. This is due to local variations in weather, topography and vegetation cover and holds both in cases when the deposition is modelled based on emission data (EMEP) or when it is based on a combination of measured and modelled data (NILU). This is illustrated by the weak correlation between EMEP-modelled non-marine S (S^*) deposition and flux of SO_4^* flux out of the lakes in 1995 (Figure 4). If it is assumed that the catchment-lake system is in equilibrium (steady-state), two problems arise:
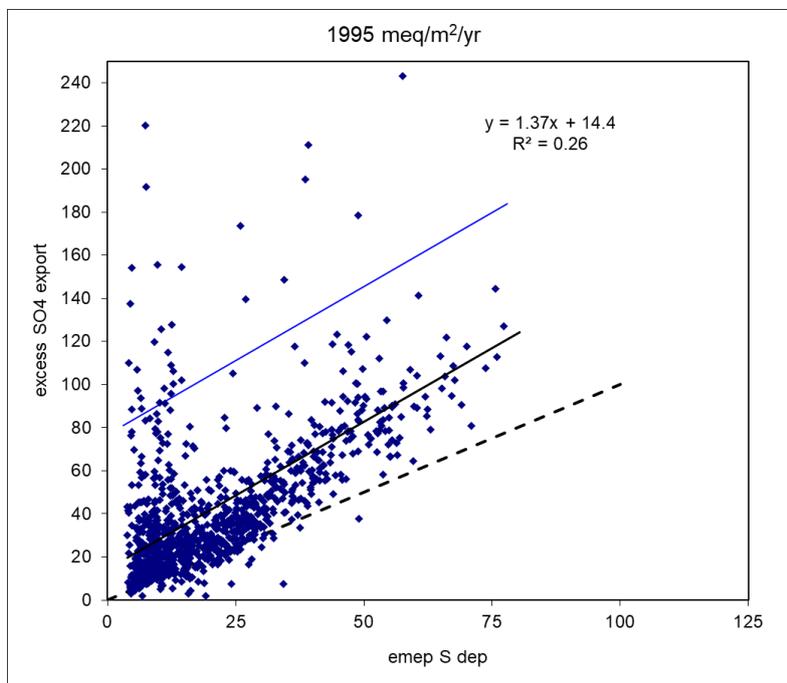

Figure 4. Relationship between EMEP S^* deposition and flux of SO_4^* out of the lakes in 1995. EMEP data are from the Coordination Center for Effects (CCE. Units: meq/m^2/year). Black line: linear regression; blue line: regression plus 60; dotted black line: 1:1.

1. There are lakes where EMEP deposition is greater than the flux of SO_4 out of the lake. These lie below the 1:1 line in the figure. The question then is: where does the surplus of SO_4 end up? In these cases, EMEP's modelled S^* deposition is probably overestimated. It is then better to use flux in the outlet as an estimate for deposition of SO_4^* in these lakes. This means that weathering of SO_4 is set to zero.

2. There are many lakes where the flux SO_4^* out of the lakes is greater than EMEP's modelled S^* deposition. These lakes lie above the 1:1 line in the figure. Then either EMEP's deposition is underestimated and/or a significant contribution from weathering of SO_4 is needed to achieve the balance. Larssen et al. (2008) assumed that all lakes that had SO_4^* flux greater than 60 meq/m2 above the regression line in Figure 4 probably had SO_4 from weathering. The number 60 was chosen because this was the EMEP's maximum S^* deposition in 1995 for the 1000 lakes. With the new EMEP

data displayed in Figure 4 the maximum was 75, but for consistency reasons we decided to use the same number as Larssen et al. (2008). For the 36 lakes above the blue line, all SO_4^* above the regression line (black) was assumed to come from weathering. For the lakes between the regression line and the blue line, it was assumed that the EMEP values underestimated the actual S^* deposition and that there was no S^* weathering. Weathering and deposition of sea salts were assumed not to change over time throughout the simulation period.

Selected approach for new calibration

There are other possible sources and sinks for SO_4 in catchments that can contribute to the fact that the measured flux of SO_4 out of the lakes does not agree with EMEP's S deposition. Adsorption and desorption of S in the soil and redox processes can cause deviations from the assumption of equilibrium (steady state). Data from calibrated catchments and trend lakes show that there is a rapid response to SO_4 concentration in surface water from changes in S deposition. The RAIN experiment at Risdalsheia also showed that reduced S deposition results in a reduced concentration of SO_4 in the runoff water within 0-2 years (Wright et al. 1993). It is therefore unlikely that adsorption or redox processes can explain the large spread around the 1:1 and regression lines in Figure 4. The conclusion is therefore that EMEP's modelled S deposition only gives a rough estimate of the input of SO_4^* in a catchment area/lake -system. Measured flux of SO_4 out of the lakes ("ground truth") therefore seems to be the best way to narrow the uncertainty in the estimates for SO_4^*deposition.

We use the same reasoning for estimating SO_4^* weathering in 2019. EMEP estimated deposition of S^* in 2019 is lower than in 1995 – the maximum deposition is now only 20 meq/m^2/yr, and the mean SO4* flux out of the lakes is about 20 meq/m^2/yr (Figure 5).
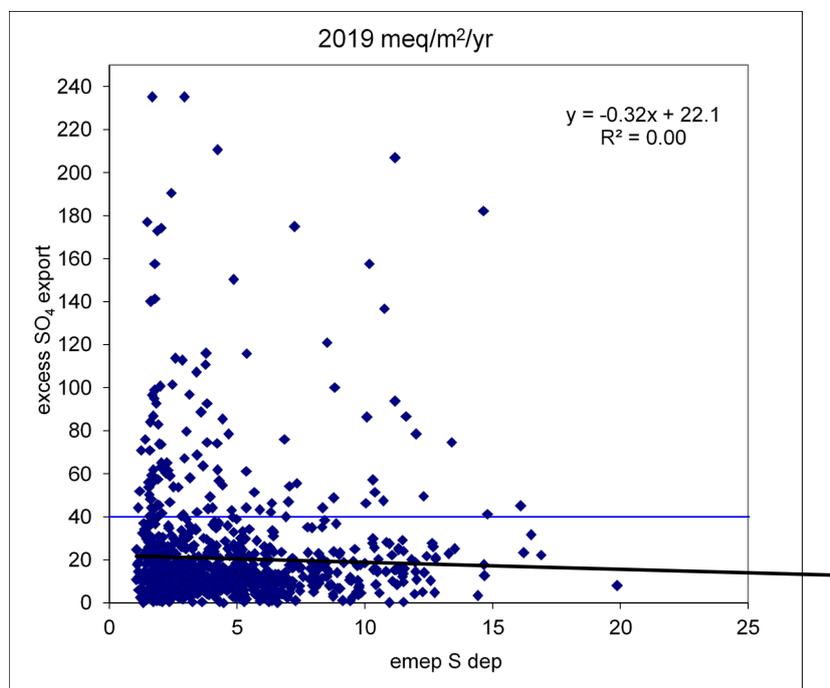

Figure 5. Relationship between EMEP S^* deposition and flux of SO_4^* out of the lakes in 2019. EMEP data were downloaded September 2022 from *https://emep.int/mscw/mscw_moddata.html*. Units: meq/m^2/year). Black line: linear regression; blue line: export = 40. Lakes above the blue line were assumed to have a significant source of SO_4 from weathering.

The regression is no longer statistically significant. We now assume that lakes with SO_4* flux above 40 meq/m^2/yr (above the blue line) have weathering inputs of SO_4*; in these lakes all SO_4* flux greater than 20 meq/m^2/yr was assumed to come from weathering. Given these assumptions and using the new EMEP S* deposition data, there were 36 lakes in 1995 and 116 lakes in 2019 that required SO_4* weathering. 33 lakes had weathering in both years (Figure 6).
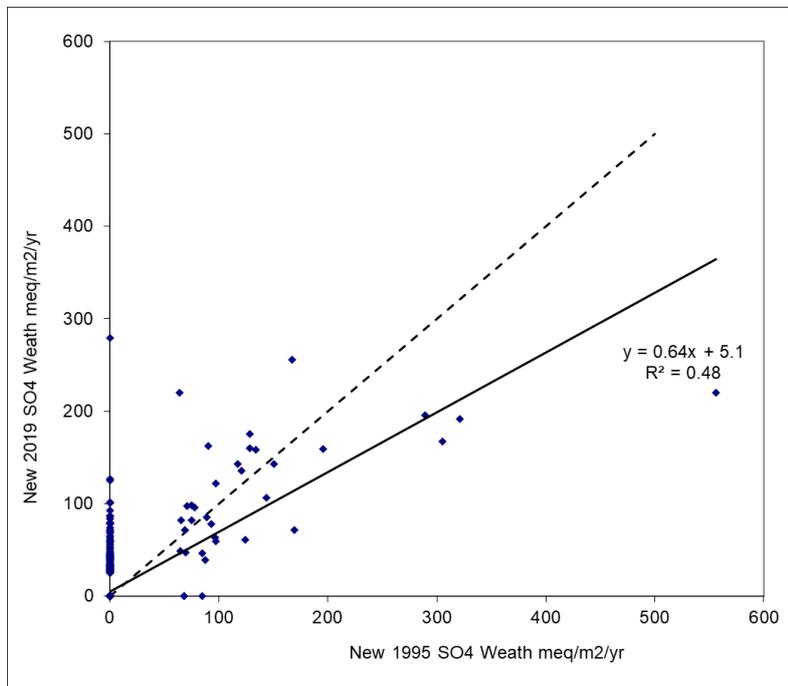

Figure 6. Estimated weathering of SO_4 in 2019 vs 1995. In 1995 there were 36 lakes with SO_4 weathering; in 2019 there were 116 lakes. Black line: linear regression; dotted line: 1:1.

2.4.8 Nitrogen retention

Previous calibration

- N retention: Assumed that the % of atmospheric N that was retained in the catchment and lake in 1995 was the same both backwards and forwards in time.
- Soil: Nitrification set to 100% and denitrification to 0%
- Water: Nitrification set to 100% (reason: NH_4 very low, or not measured), while denitrification is assumed to be a function of theoretical residence time (Tw) and an empirical rate constant.
- UptNH4lake and UptNO3lake set to 0

Selected approach for new calibration

The 2019 survey provides a basis for a new estimate of % N retention. The data show a large degree of scatter and do not indicate a systematic change in the % N retention from 1995 (Figure 7).

The latest version of MAGIC (version 8) has three alternative ways of calculating % N retention: (1) constant % N retention of N deposition, based on input-output in the calibration year, (2) % N retention linked to the C/N ratio in the soil, and (3) % N retention linked to microbial processes for turnover of C and N. The first approach was used on the 1995 data.

The soil data are so sparse that there will be little to gain from calculating % N retention from soil C/N ratio or based on the microbial C and N module in MAGIC. We therefore choose the same approach as in 1995, i.e. that a constant % N retention in the soil is used for all years in each simulation. The 2-point calibrations used the average of % N retention in 1995 and 2019 for each lake. Thus, changes in N deposition drive observed changes in NH_4 and NO_3 concentrations in the lakes.

As NH_4 was measured in all lakes in 2019, % nitrification in the soil and lakes will be based on input and output of NH_4. The model was adapted to implement this (see Appendix B 1. N sinks. Order of calculation. UptNH4lake and UptNO3lake (algae intake) are still set to 0.
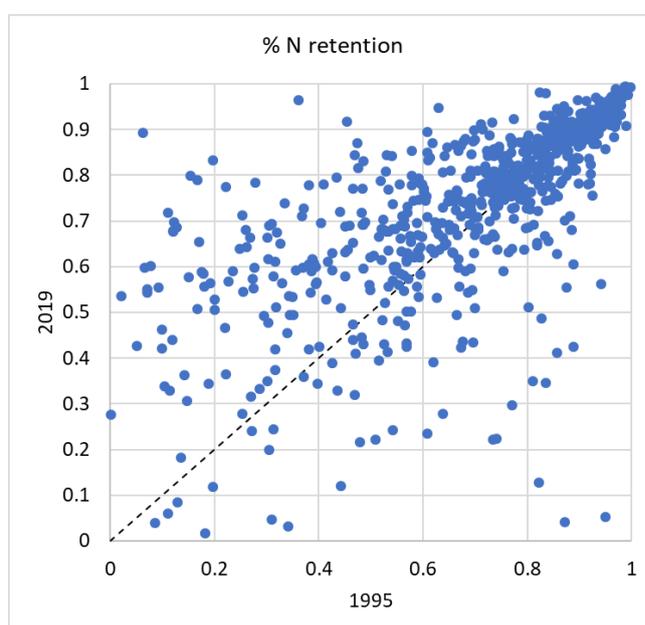
Figure 7. % N retention in 2019 vs 1995. N retention is defined as 1 minus the sum of flux of NH_4 and NO_3 out of the lake divided by the sum of NH_y and NO_x deposition. Most of the retention is in the catchment.

2.4.9 TOC and organic acids in soil and water

Previous calibration

MAGIC requires estimates of the concentrations of organic acids in soil water for simulation of DOC and organic acids in lake water. For surface water, measured values for DOC in 1995 were used. For soil water, DOC concentrations were estimated from observed lake chemistry and soil chemistry (see Appendix B). For both soil water and surface water, DOC was assumed constant over time during the simulation period. Furthermore, it was assumed that the acid portion of DOC consists of tri-protic acids with dissociation constants given by (Driscoll et al. 1994), pK1=2.64, pK2=5.66, pK3=5.94. These were assumed to apply to both groundwater and surface water and were kept unchanged over time throughout the entire simulation period.

Selected approach for new calibration

For surface water, TOC was measured in 1995 and 2019. TOC in Norwegian lakes has increased since the 1980s, and the increase is primarily attributed to the reduction of SO_4 in soil solution (de Wit et al. 2007). The mechanism is that reduced SO_4 leads to reduced ionic strength, which in turn leads to increased solubility of TOC from the soil (Hruška et al. 2009).

In a recalibration of MAGIC to the 1000-lake data, TOC changes over time could be linked to the concentration of SO_4, given that simulated and measured concentrations of TOC and SO_4 agree in 1995. The question is whether TOC in soil water has changed to the same degree as the changes in TOC in surface water. It is in principle possible to link TOC to SO_4 also for groundwater, but there are few observed data available on which to base such a link. This is an issue that is currently being worked on in the Czech-Norwegian EEA project CatchCaN.

We thus will focus only on ANC and its components. ANC is defined as the equivalent sum of the base cations minus the sum of strong acid anions in units of µeq/l.

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

We have not calibrated the other water chemistry components such as pH, labile aluminum (Al), TOC, organic anions and bicarbonate (HCO_3).

2.4.10 Weathering of base cations

Previous calibration

In MAGIC, the calculation of base cation weathering assumes that the system was in equilibrium (steady state) prior to the onset of industrial S pollution (e.g. 1850). For example, for Ca the balance is:

Ca [weathering] + Ca [deposition] + Ca [from the soil's ion exchange complex] = Ca [net uptake in vegetation] + Ca [flux into the water]

Uptake in vegetation is linked to forest growth and net uptake represents Ca loss when biomass is removed during felling.

In 1850, steady state is assumed, i.e. that the stock of Ca in the soil is constant. The calibration routine in MAGIC has an optimization algorithm that calculates the weathering flux as well as the original base saturation for the four base cations in the year 1850. The measured concentration in the lake and the measured base saturation in the soil in 1995 were then simulated using a specified S+N deposition sequence from 1850 to 1995. The weathering rates were assumed unchanged throughout the simulation period 1850-2030.

Selected approach for new calibration

A preliminary assumption is that the weathering rate is constant over time for the four base cations. With two-point calibration, we can more easily assess the assumptions in the model, especially considering whether the observed increase in Ca concentration from 1995 to 2019 can be linked to changes in weathering rate.

2.5   Calibration procedures

We calibrated three times:
– once to the 1995 data and examined the simulated vs observed lake chemistry in 2019
--once to the 2019 data and examined the simulated vs observed lake chemistry in 1995
--once to both the 1995 and 2019 data (2-point calibration) and examined the simulated vs observed lake chemistry in both 1995 and 2019.

The calibrations were conducted stepwise with optimisation first of Cl, SO_4 and NO_3 in that order, and then on the four base cations simultaneously. Prime focus was on the simulated vs observed values for ANC. Details on the calibration procedures can be found in (Larssen et al. 2008).

3 Results

3.1 Single-point calibration to the 1995 data

As expected, the MAGIC calibrations to the 1995 data resulted in complete agreement of the simulated ANC with the observed ANC in 1995 (Figure 8). We thus conclude that the recoding of MAGIC and including it in the Mobius platform was done correctly. The ANC values simulated for 2019, however, were significantly lower than the observed. This is particularly apparent for the acid-sensitive lakes, i.e. those that have ANC less than 200 µeq/l (Figure 9).
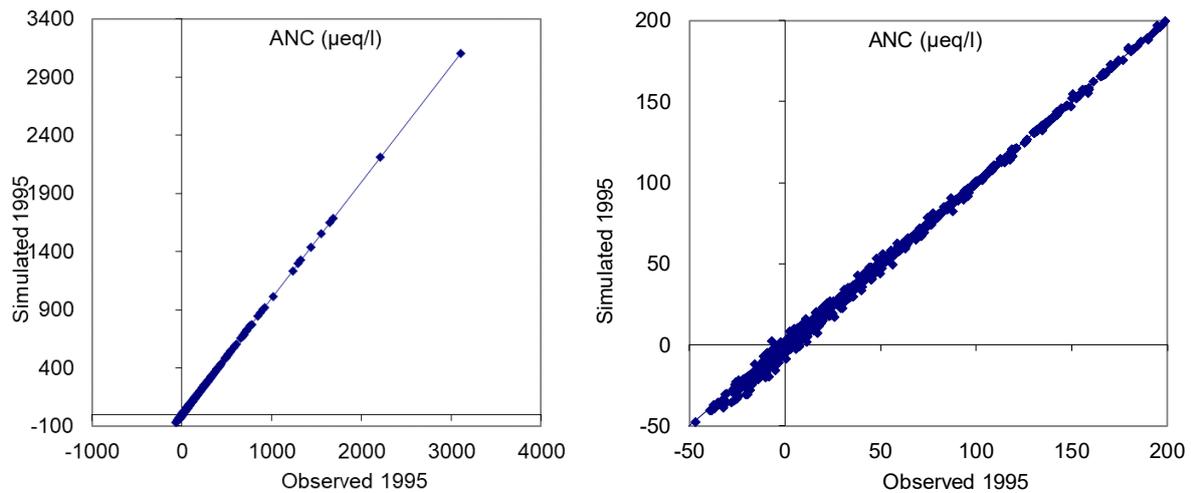
Figure 8. Results from single-point MAGIC calibrations of 969 lakes on the 1995 data. Shown are ANC concentrations (µeq/l) in the lakes simulated vs observed. Left panel: all lakes. Right panel: acid sensitive lakes (those with ANC less than 200 µeq/l). Solid blue line (1:1).
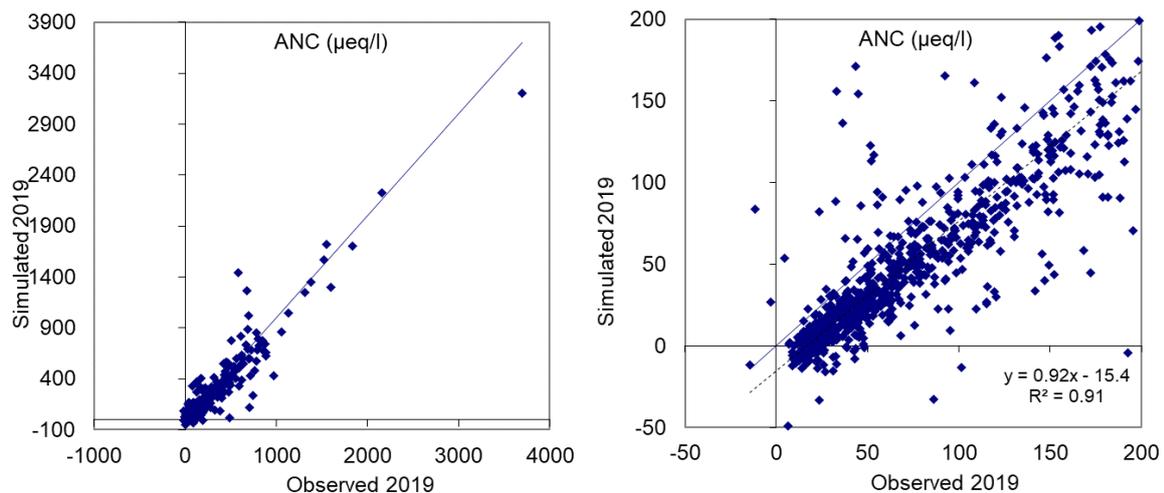
Figure 9. Simulated and observed ANC concentrations in 2019 for the 969 lakes with successful single-point calibrations by MAGIC to the 1995 observed data. Left panel: all lakes. Right panel: lakes with ANC less than 200 µeq/l. Solid blue lines (1:1); dotted black line (least squares linear regression).

ANC is comprised of the equivalent sums of the concentrations of the four base cations (Ca, Mg, Na, K) and NH_4 minus the sum of the three strong acid anions (Cl, SO_4, NO_3). The explanation for the discrepancy between the simulated and observed ANC concentrations in 2019 in the acid-sensitive lakes must be due to discrepancy in one or more of these eight components. The results point to Ca. Simulated Ca concentrations were on average about 10% lower than the observed. Although the simulated SO_4 concentrations were also lower than observed, the discrepancy in Ca was about three times larger than the discrepancy in SO_4 (Figure 10). Simulated Mg concentrations agreed with observed. Simulated concentrations of both Na and Cl were lower than observed but by about the same amounts, thus together they balance. In most lakes the concentrations of K, NH_4 and NO_3 were much lower than other components and had little influence on ANC.

The recalibrated model does not capture the increase in Ca, that is found in the comparison of the 1995 and 2019 data as well as in the Norwegian "trend lakes", a set of 78 acid-sensitive lakes sampled annually since 1986 (de Wit et al. In review). The modelling result is also similar to that obtained by Austnes et al. (2020) in a comparison of the observed 2019 lake data with the MAGIC simulations in the 2016 calibration to the 1995 data (Austnes et al. 2016).

The simulated SO_4 concentrations were lower than the observed in 2019 in the lakes perhaps because the EMEP estimates give a steeper decline in S deposition 1995-2019 than actually occurred. Whether this is indeed the case can be assessed by comparing S deposition estimated from measurements at NILUs precipitation stations with the EMEP modelled estimates. This is beyond the scope of this report.

Figure 10. Simulated and observed concentrations in 2019 of 7 components that comprise ANC (NH_4 concentrations were not measured in 1995) for the 969 lakes with successful single-point calibrations by MAGIC to the 1995 observed data. Shown are data for the acid-sensitive lakes, i.e. those with ANC below 200 µeq/l. Solid blue lines (1:1); dotted black line (least-squares linear regression).

3.2 Single-point calibration to the 2019 data

The MAGIC calibrations to the 2019 data also resulted in complete agreement of the simulated ANC with the observed ANC in 2019 (Figure 11). The ANC values simulated for 1995, however, were on average about 16 µeq/l higher than the observed (Figure 12). This is particularly apparent for the acid-sensitive lakes, i.e. those that have ANC less than 200 µeq/l.
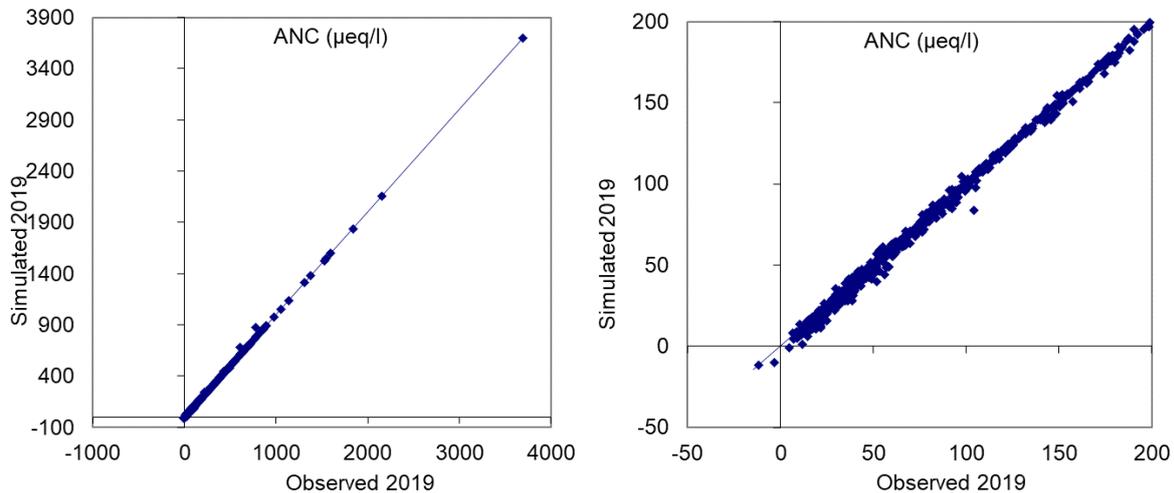
Figure 11. Results from single-point MAGIC calibrations of 847 lakes on the 2019 data. Shown are ANC concentrations (µeq/l) in the lakes simulated vs observed. Left panel: all lakes. Right panel: acid sensitive lakes (those with ANC less than 200 µeq/l). Solid blue line (1:1).
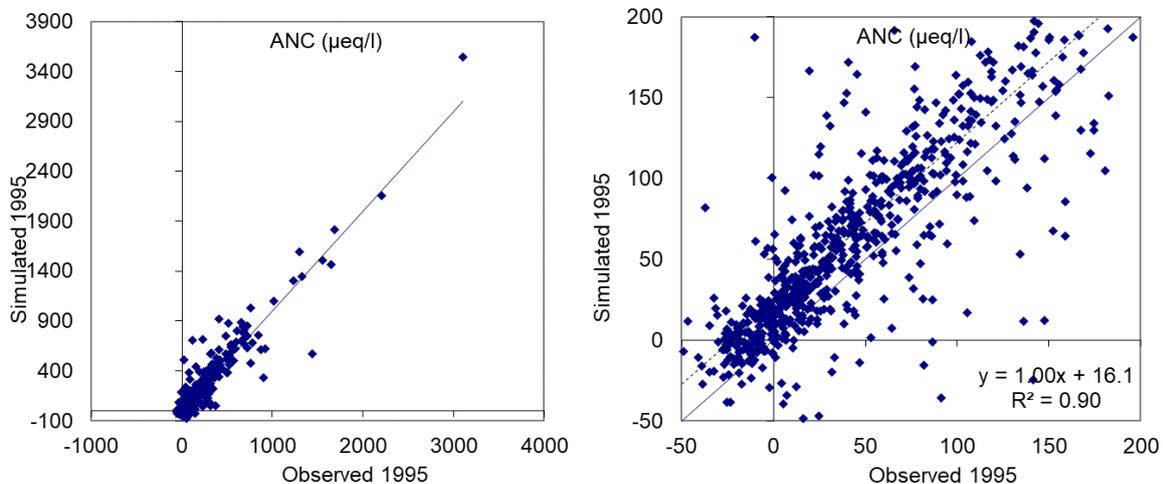
Figure 12. Simulated and observed ANC concentrations in 1995 for the 847 lakes with successful single-point calibrations by MAGIC to the 2019 observed data. Left panel: all lakes. Right panel: lakes with ANC less than 200 µeq/l. Solid blue lines (1:1); dotted black line (least squares linear regression).

Again, the results indicate that Ca and SO_4 account for most of the discrepancy in simulated ANC in 1995 as compared with observed (Figure 13). Simulated concentrations of both Ca and SO_4 were higher in 1995 than the observed. The calibrations to the 2019 data gave higher weathering rates for Ca as compared to the calibrations to the 1995 data (Figure 14).
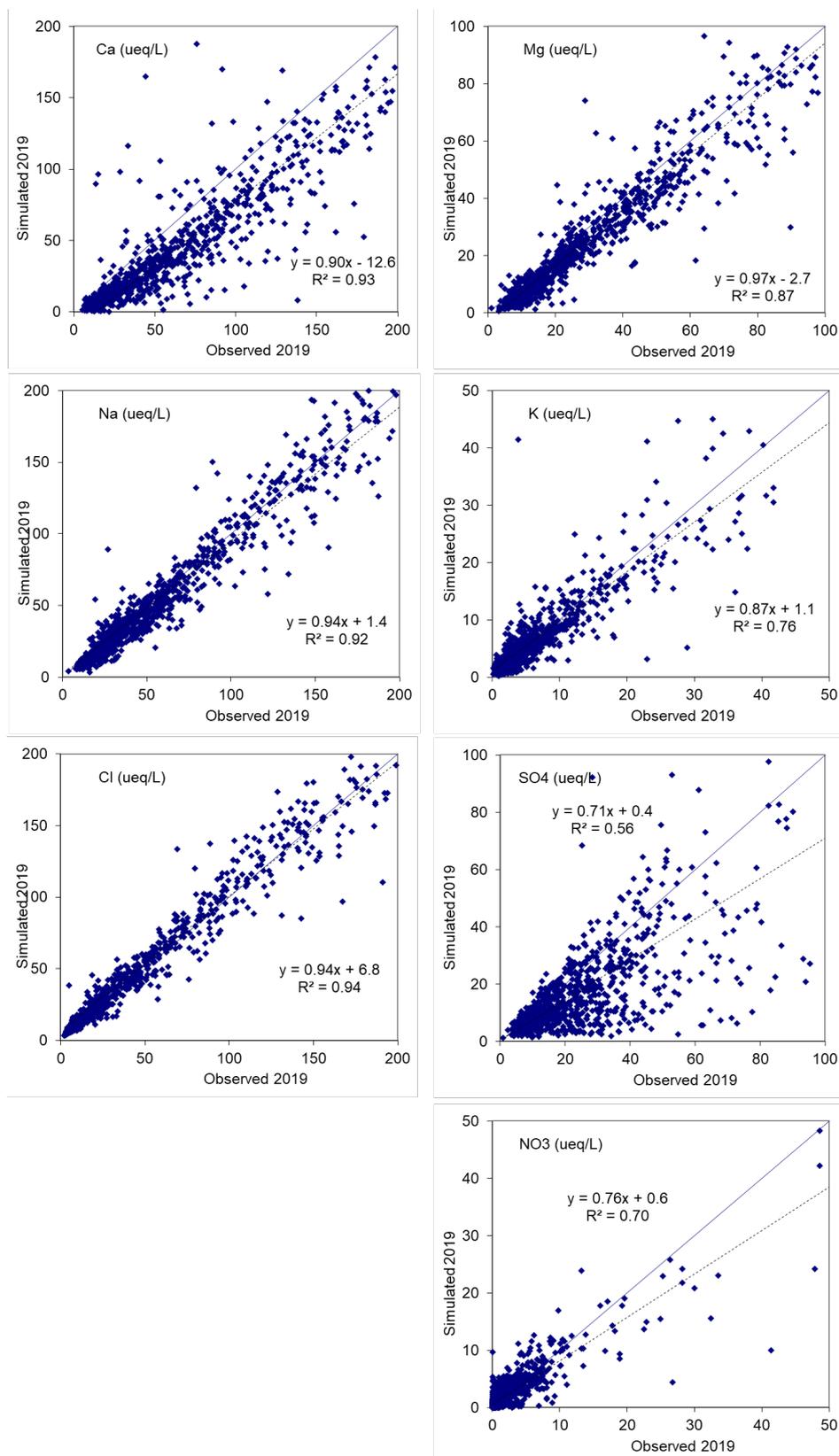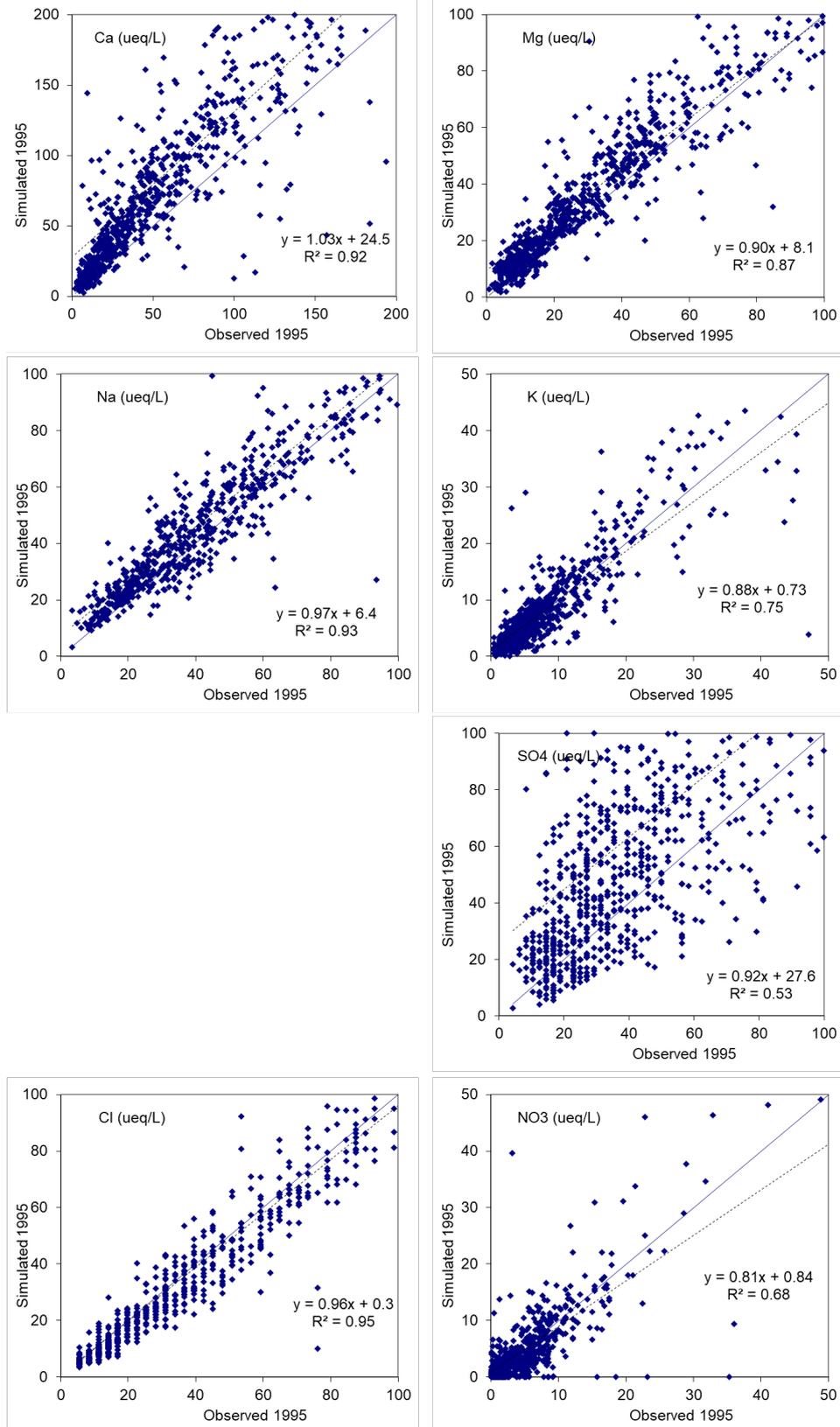
Figure 13. Simulated and observed concentrations in 1995 of 7 components that comprise ANC (NH$_4$ concentrations were not measured in 1995) for the 847 lakes with successful single-point calibrations by MAGIC to the 2019 observed data. Shown are data for the acid-sensitive lakes, i.e. those with ANC below 200 µeq/l. Solid blue lines (1:1); dotted black line (least-squares linear regression).
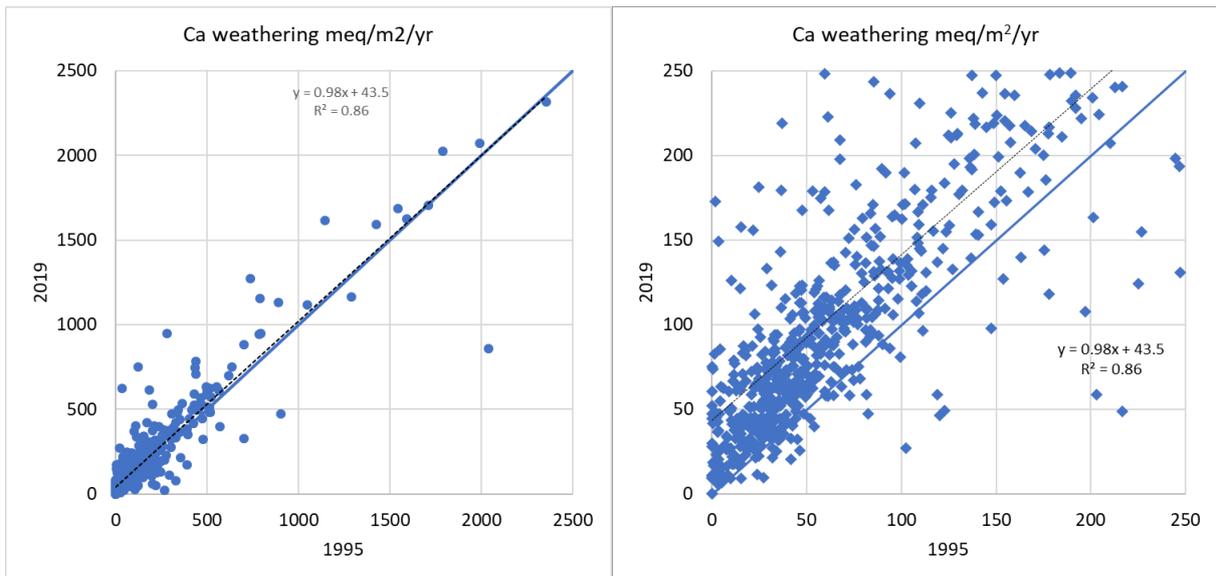
Figure 14. Weathering rates for Ca (meq/m^2/yr) obtained by MAGIC single-point calibrations to the 1995 and 2019 data. Shown are values for the 834 lakes that were successfully calibrated both times. Left panel: all lakes; right panel: lakes with Ca weathering less than 250 meq/m^2/yr.

3.3   Two-point calibration to the 1995 and 2019 data

The 2-point calibration gave simulated ANC values that did not exactly fit either the 1995 nor the 2019 observed ANC (Figure 15). This was expected as the 2-point calibration sought to minimise the difference between the simulated and observed for both the 1995 and 2019 data simultaneously. Thus, the simulated ANC values (and the values for all the eight components contributing to ANC) were a "compromise" between the observed 1995 values and the observed 2019 values. The calibrations appear to give a closer fit to the 1995 data as compared to the fit to the 2019 data.

The values for the eight optimised base cation parameters in the soil also indicate that the two-point calibration is closer to the 1995 one-point than to the 2019 one-point calibration (Table 3).
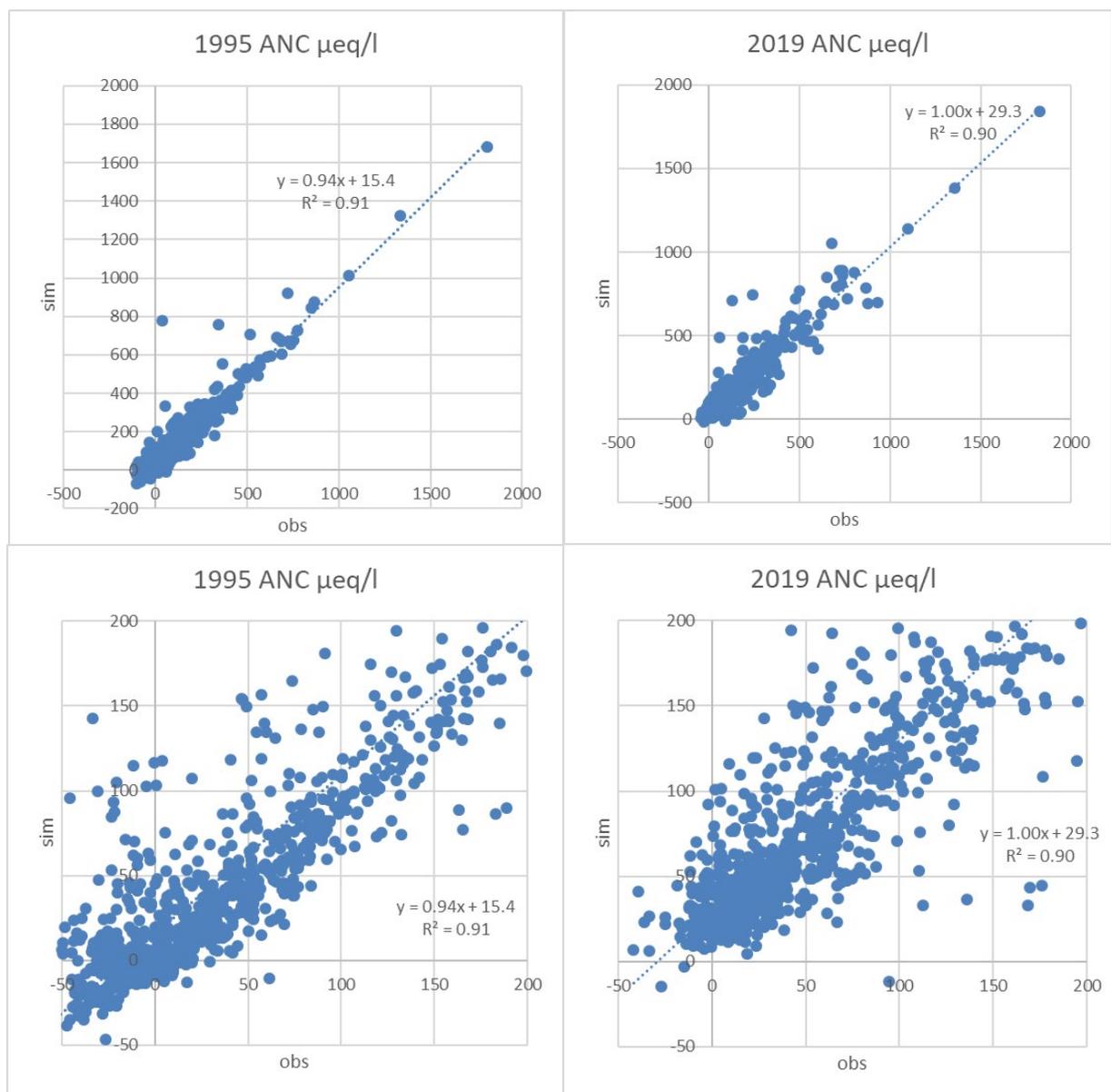
Figure 15. Calibration results from the 2-point calibration to 1995 and 2019. Shown are ANC simulated vs observed for all 933 lakes calibrated. Upper panels -- full ANC range; lower panels – acid-sensitive lakes only (ANC<200µeq/l).

Table 3. Soil parameters optimised in the three separate calibration trials – single-point to the 1995 lake data, single-point to the 2019 lake data and two-point to both data sets. Shown are the median values for the 838 lakes that were successfully calibrated in all three trials.

	Weathering rate (meq/m²/yr)				Initial base saturation (% of CEC)			
	WeCa	WeMg	WeNa	WeK	%Ca	%Mg	%Na	%K
1995	54.8	13.8	15.8	4.7	24.0	8.3	3.8	3.1
2019	88.9	23.3	22.9	3.8	28.3	10.2	4.1	3.3
2-point	60.8	13.1	12.5	3.7	25.0	8.6	3.9	3.2

4 Discussion

4.1 Drivers

4.1.1 Deposition of S

Proper estimates of S deposition emerge as a key step in the setting of the input data to MAGIC. This holds both for the absolute S deposition estimated at each lake for the two sampling dates, 1995 and 2019, but also to the deposition history from 1800 to 2019. The EMEP values for 1995 and 2019 cannot be simply used directly – they clearly overestimate S deposition at some lakes and underestimate S deposition at other lakes.

The trends in EMEP S deposition over the period 1995 to 2019 directly affect the ability of MAGIC to simulate the observed changes in SO_4 concentrations in the lakes, and as a result also the concentrations of base cations. The long-term depletion of soil base cation pools is directly influenced by the total cumulative S deposition since the start of the industrial revolution.

EMEP has revised the estimates of S deposition amounts and trends for the period 1990-2019 several times. The latest estimates (September 2022) indicate larger decreases in S deposition as compared to the observed decline in SO_4 concentrations in the lakes. The effect of changed estimates of S* deposition is illustrated by the MAGIC calibrations to Lake Lille Hovvatn, one of the 1000 lakes (see Appendix C).

4.1.2 Deposition of N and N retention

Our estimates of N deposition at each of the lakes are based on estimated S deposition and are thus subject to additional uncertainty. NO_x deposition is assumed to be equal to the estimated S* deposition times the ratio of NO_x/SO_x given by EMEP for the appropriate grid square. The NH_y deposition is calculated in a similar fashion. Uncertainty in N deposition, however, has little influence on the ANC concentrations in the lakes, as most of the incoming N is retained in the catchment-lake system, and thus with few exceptions NO_3 concentrations in the lakes are low. Some of the outliers have high NO_3 concentrations due to agricultural activities in the catchment.

de Wit et al. (In review) found that nitrate concentrations in the 1000 lakes were lower in 2019 relative to 1995 and occurred independent of N deposition. They suggest that there has been increased catchment N retention, possibly from increased terrestrial productivity.

4.1.3 Sea salt deposition

Deposition of sea salts is a significant driver of episodic acidification, the so-called "sea salt effect" (Hindar et al. 1995). Although sea salt itself has positive ANC, the differential mobility of Cl and Na+Mg in soils can cause episodes of acidic runoff – the Cl leaches fast and mobilises H^+ and labile Al. This is especially important for lakes located near the coast.

Cl concentrations in the 1000 lakes were generally lower in 2019 than 1995, indicating that sea salt deposition was lower prior to the sampling in 2019 as compared to 1995. The interannual variations in Cl deposition are larger than the difference between 2019 and 1995 (Figure 2). Data from NILUs precipitation stations do not indicate a systematic long-term trend in sea salt deposition (Aas et al. 2022).

4.2   Role of other factors

The acidification pressure on Norwegian lakes has decreased dramatically since the 1970s. For the majority of lakes in the 1000 survey the large reductions in S* deposition since the peak years in the 1970s have resulted in decreased SO_4 concentrations and increased ANC levels. Other driving factors that were previously overwhelmed by acid deposition may now become more apparent.

Climate change is one of these factors. Increasing temperature, changes in precipitation, and frequency and severity of storms all can affect biological and chemical processes in catchments and the lakes themselves. de Wit et al. (In review) suggest several of these possible effects in their analysis of the 1000-lake data set. Longer growing seasons could result in greater uptake of N in forests and other vegetation, and thus decreased leaching of N to lakes. Increased temperature might directly or indirectly enhance weathering of soil minerals and result in increased Ca concentrations in lakewater.

The MAGIC model was designed to simulate changes in soil and surface water chemistry due to changes in inputs of acid deposition. For MAGIC to fully simulate the effects of climate change, in particular the effect of higher primary productivity, additional processes must first be included in the model. Some of these changes are currently being explored as part of the Czech-Norwegian-Swedish project CatchCaN. Modifications and extensions include a hydrologic module to allow for simulation of changes in precipitation and a forest growth module to allow for uptake and loss of nutrients in biomass. CatchCaN will also link a soil carbon model to MAGIC. These developments will be made on the new MOBIUS modelling platform at NIVA.

4.3   Single vs two-point calibration

The differences between the one- and two-point calibrations are perhaps best seen in the results from single lakes. We chose Lille Hovvatn (MAGIC ID code 36). See Appendix C.

The choice of which of the three calibrations to use in future studies depends on the application. If the aim is to give the best estimate of pre-industrial "reference" state of the lakes, then perhaps the one-point calibration to the 1995 data is best, as this gives a picture least affected by climate change or other recent changes in drivers. If, on the other hand, the aim is to project future changes in lake water chemistry, then perhaps the one-point calibration to the 2019 is best. The two-point calibration, however, gives perhaps the best estimate of the "average" response of the lakes to changes in deposition of pollutants and sea salts.

4.4   Possible additional studies

1.   The calibrations could be extended to include pH, TOC, labile Al, and HCO_3.

For this report we have focussed on ANC and the eight chemical components that comprise ANC. it is of interest to also simulate changes in pH and labile Al, as these components are critical for fish and other biological organisms.

TOC changes over time could be linked to the concentration of SO_4. The question is whether TOC in soil water has changed in line with the changes in TOC in surface water. In MAGIC this is important as an estimate of the concentration of organic anions in the pre-pollution steady state (e.g. 1850) is

needed to balance the concentrations of base cations from weathering and deposition. In principle it is possible to link TOC to SO_4 also for soil water, but there are few observed data available on which to base such a link.

2. Regional differences could be investigated (for instance, the increase in Ca concentrations).

de Wit et al. (In review) identified significant regional variations in the changes observed in lake chemistry from the 1995 and 2019 1000-lake surveys as well as in the "trend" lake data set. The MAGIC calibrations here could be broken down into regions and the simulated changes compared with the observed data.

3. The new calibrations could be used to run scenarios for future S+N deposition.
EMEP has not yet (as of November 2022) released the new scenarios for projected S and N deposition for the years 2030 and 2050. When these are released, they can be used to project lake chemistry for future years based on the new MAGIC calibrations presented here.

4. The new calibrations provide input data for a MAGIC "library" for Norway.

Moldan et al. (2020) developed a tool, the MAGIC library, which provides an acidification assessment for any surface water in Sweden based on ten parameters describing lake geographical position, surface area, annual discharge and observed lake water chemistry. The MAGIC library consists of two key components: a library of MAGIC model simulations for Swedish 2438 lakes and an analogue matching routine that selects the library lake which is most similar to the evaluation lake described by the ten parameters. The MAGIC library is used by environmental managers in Sweden to assess the pre-industrial "reference state" of lakes for e.g. use in acidification status classification under the Water Framework Directive and for considering the need for liming. Now with the new MAGIC calibrations to the 1000-lake survey, a similar MAGIC library could be constructed for Norway, or perhaps combined with the existing Swedish library.

5. The calibrations could be augmented by including the few lakes that did not have complete and updated site data.

The one- and two-point calibrations were completed on 839 of the 987 lakes that had data common to the 1995 and 2019 data sets. Many of the remaining 148 lakes could be included if the site characteristics and other fixed parameters such as specific discharge were confirmed. This was not possible within the time frame of the current project.

6. The deposition history for S and N could be reviewed.

The MAGIC calibrations presented here use new updated estimates from EMEP for 1990-2020. The deposition estimates prior to 1990, however, have not been re-visited and updated since Schöpp et al. (2003), and are based on older emission estimates and transfer matrices. It is unclear as to whether new estimates of historical S and N deposition would significantly affect the MAGIC calibrations and future forecasts. This could be evaluated by means of a simple sensitivity analysis using MAGIC calibrations and simulations on a few lakes with several alternative historical deposition sequences.

7. The calibrations could be repeated using deposition data from NILU.

NILUs precipitation stations have measurements from the mid-1970s to present. NILU prepares 5-year means of these data, and estimates deposition for all of Norway using a kriging scheme. These data probably give a better picture of S and N deposition in the peak period of the 1970s and 1980s, as well as the steepness of the decline in the period 1995 to 2019.

8.   MAGIC could be used to evaluate the observed trends in Ca concentrations.

In 2019 many of the 1000 lakes had unexpected increased concentrations of Ca, and there were regional differences (de Wit et al. In review). The cause could be an increase in weathering of Ca from soil minerals. Ca weathering is one of the parameters that is optimised in the MAGIC calibration procedure. For 839 of the 987 lakes we now have three separate estimates of Ca weathering, one based on the 1995 calibration, one based on the 2019 calibration and one based on the two-point calibration. These provide the basis for assessing to what extent MAGIC satisfactorily simulates the observed Ca trends or if additional processes are required. The weathering rates can be varied over time in MAGIC, but there is currently no process in MAGIC that is linked to the weathering rate.

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Appendix A: S and N deposition at each lake

MAGIC requires estimates of historical S and N deposition at each of the 1000 lakes for the years prior to 1990. These are based on Schöpp et al. (2003) and have not been revised. The historical data are scaled and coupled to the EMEP data in the year 1990 such that the Schöpp et al. (2003) estimate for 1990 agrees with the EMEP estimate for 1990 in every grid cell (Figure A1). This work was done by Max Posch at IIASA.
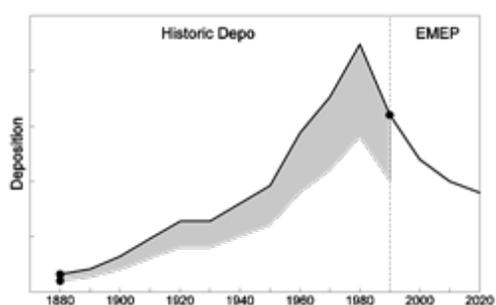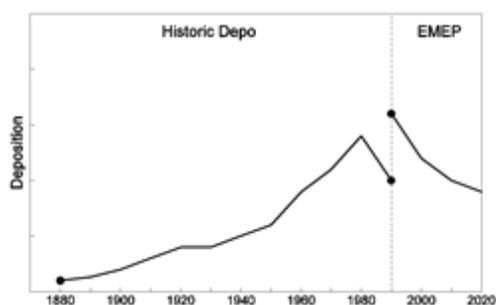

Figure A1. Schematic example of the coupling of historical to EMEP deposition estimates. A) the 'real world' situation, B) the classic method to fit (in 1990) with a constant factor = Dep90EMEP/Dep90hist, giving a big relative change in 1880, C) as done for deposition in the earlier MAGIC modelling (Larssen et al. 2008) and also for this new MAGIC re-calibration, which leaves deposition in 1880 unchanged, D) The red area showing the difference between B) and C) (Figure courtesy of Max Posch, IIASA).
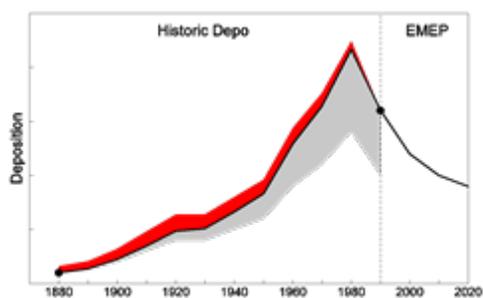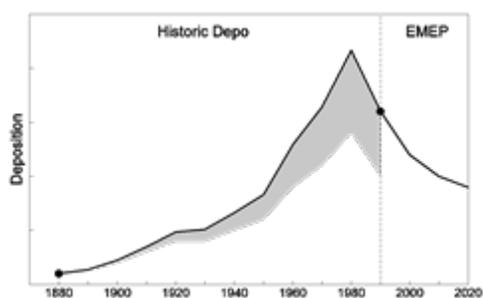
Bilinear Interpolation (from Max Posch, IIASA)

To obtain the value of the function (= deposition field) at an arbitrary point (x,y), when it is known at the four grid points (x_1,y_1), (x_2,y_1), (x_2,y_2) and (x_2,y_1) (see Figure A2), we first interpolate linearly twice (i.e. for fixed y_1 resp. y_2) in the x-direction:

(1a) $\quad f(x,y_1) \approx (1-\lambda)f(x_1,y_1) + \lambda f(x_2,y_1)$

and

(1b) $\quad f(x,y_2) \approx (1-\lambda)f(x_1,y_2) + \lambda f(x_2,y_2) \quad$ with $\quad \lambda = \frac{x-x_1}{x_2-x_1}$

Then we interpolate for a given x linearly between these two values in the y-direction to obtain the desired estimate:

(2a) $\quad f(x,y) \approx (1-\mu)f(x,y_1) + \mu f(x,y_2) \quad$ with $\quad \mu = \frac{y-y_1}{y_2-y_1}$

Inserting eqs.1a,b this results in the bilinear interpolation formula:

(3) $\quad f(x,y) \approx (1-\lambda)(1-\mu)f(x_1,y_1) + \lambda(1-\mu)f(x_2,y_1) + (1-\lambda)\mu f(x_1,y_2) + \lambda\mu f(x_2,y_2)$

Note that the same result is obtained if the interpolation is first done along the y-direction and then along the x-direction. For further details see http://en.wikipedia.org/wiki/Bilinear_interpolation.
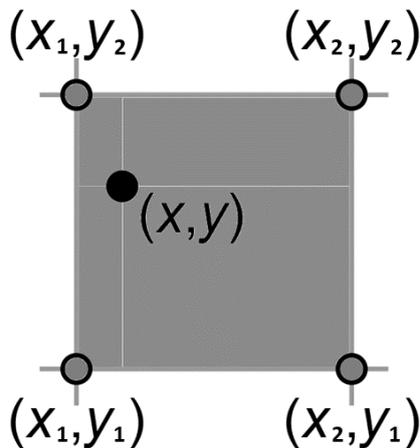
Figure A2: Geometry for interpolating a 2-dimensional function (e.g. a deposition field) at arbitrary (x,y) when it is known at the four points (x_1,y_1), (x_2,y_1), (x_1,y_2) and (x_2,y_2).

Appendix B: Details of calibration procedures in MAGIC

B 1. N sinks. Order of calculation.

Routines to optimize nitrification and NO_3 uptake are completed before the optimization of base cations begins.

NH_4. Set to zero if not measured, set to detection limit if measured but below detection limit.

Nitrification is be optimized BEFORE the optimization of NO_3 uptake because all NH_4 that is nitrified must be added to the NO_3 inputs for each compartment.

1. Optimize nitrification to match observed NH_4 in the lake – use the approach that specifies nitrification as a % of inputs to the compartment (soil or lake)
1) set soil and lake nitrification both to 100%
2) decrease lake % nitrification until simulated NH_4 matches observed value
3) if lake % nitrification reaches 0% and further reductions in simulated NH_4 are still needed to match observed, start reducing soil % nitrification until the simulated and observed NH_4 agree.

2. Optimize NO_3 uptake to match observed NO_3 – use the approach that specifies NO_3 uptake as a % of inputs to the compartment (soil or lake)
1) set soil and lake NO_3 uptake both to 100%
2) decrease lake % NO_3 uptake until simulated NO_3 matches observed value
3) If lake % NO_3 uptake reaches 0% and further reductions in simulated NO_3 are still needed to match observed, start reducing soil % NO_3 uptake until the simulated and observed NO_3 agree.

In MAGIC777 these calculations are done dynamically in the optimizer by running repeated full simulations each time the parameters are adjusted - so that any effects of monthly patterns of nitrification or uptake are included, and any effects of lake retention time effects are also included in the final optimized values of %nitrification and %uptake.

B 2. Estimation of KAl in soil.

KAl, the solubility of aluminium hydroxide, must be specified for the soil compartment in MAGIC. There are few measured data for this parameter in the 1000-lake data set. An estimate can be made using the 1995 lake chemistry (Ca and Mg concentrations) and the soil chemistry (pH, %Ca and %Mg), given the assumption that the concentrations of C and Mg in the soil are the same as those measured in the lake. The soil data are largely from samples collected in the period 1984-1993, thus the 1995 lake chemistry data are the closest in time to the soils data. Assuming selectivity coefficients of SCaAl=0 and SMgAl=0, the concentration of Al can be calculated and then with pH the KAl. The selectivity coefficients of 0 have often been obtained from calibrations of MAGIC to sites with soils similar to those in Norway. The estimated value for KAl is then used for all years.

In the next step the selectivity coefficients are calibrated for each lake at the steady-state condition (year 1800) given the KAl determined above.

B 3. DOC concentration in soil solution.

The DOC concentration (mmol/m3) (organic acid fractions, i.e. sum of dissociated and undissociated components) must be specified for the soil compartment in MAGIC. There are few measured data for this parameter in the 1000-lake data set. As a first approximation 1995 lake data and soil data can be used to estimate DOC in soil solution.
- Assume a pCO_2 in soil solution (common choice is 2%).
- Assume that the concentrations (μeq/l) of the ANC components in the soil solution are the same as in the lake.
- From the soil pH and the KAl (see 0 above) calculate the concentration of labile Al (μeq/l).
- from the ionic balance calculate the concentration of organic anions (μeq/l)
- from soil pH and the organic acid dissociation constants (pK_1=2.64, pK_2=5.66, pK_3=5.94; (Driscoll et al. 1994) calculate the DOC concentration

This gives the organic acid fraction. The estimated value DOC (acid fraction) is then used for all years. From the site density (μeq/mgC) (typical value 10; (Hruška et al. 2003)) the total dissolved organic C (mgC/l) can be calculated.

Appendix C: Lake 36 (Lille Hovvatn) as an example

Lille Hovvatn (MAGIC-1000 ID: 36; NIVA RESA ID: 928-2-20) is a small acidified lake, barren of fish, located near Evje, Aust-Adger county (latitude 58.607, longitude 8.029). The adjacent Store Hovvatn lost its brown trout population in the 1940s, due to acidification. The lake is typical of the thousands of lakes in southernmost Norway that suffered loss or damage to fish populations due to acid deposition.

By chance one of the 1000 lakes is Lille Hovvatn, a lake first sampled in the 1974 regional lake survey (Wright and Henriksen 1978) and the 1986 1000-lake survey (Henriksen et al. 1987), then chosen as a unlimed reference site for the Norwegian liming project (Hindar and Wright 2005), and since 1995 one of the 78 "trend" lakes (de Wit et al. In review). We thus have nearly 50 years of water chemistry data from this lake.

MAGIC (version7) was calibrated to Lille Hovvatn in 2004 (Hindar and Wright 2005). They used the 30-year record of water chemistry from the lake and included 1974–1979 data from Lake Store Hovvatn as surrogate for the period for which no data from Lake Lille Hovvatn were available.

Deposition data were estimated based on measured precipitation chemistry from the NILU (Norwegian Institute for Air Research) station at Treungen (Aas et al. 2004). Treungen is located about 50 km NE of Lille Hovvatn. The precipitation amount is lower and the deposition of both sea salts and pollutant components are lower at Treungen owing to the greater distance from the coast. The Treungen deposition data were scaled up using the procedure given in Wright and Cosby (2003).

Details:
Runoff factor: Use 1973-2002 NILU annual volume and long-term specific discharge (standard period 1961-1990 from NVE map). Scale the yearly runoff variation to the measured precipitation to generate a time series for annual discharge, Qs.
Seasalts: assume that the mean 1995 runoff flux of Cl is correct, calculate a seasalt factor = Cl_{out} /Cl_{in}. Multiply Treungen Cl deposition flux by this factor for all years.
SO_4*: assume that the S* dep in 1995 equals the 1995 runoff flux of SO_4* (no S weathering), calculate a SO_4* factor = SO_4*$_{out}$/measured SO4*$_{in}$. Multiply Treungen S*deposition flux by this factor for all years.
NH_4 and NO_3: multiply Treungen deposition flux by the SO_4* correction factor for all years

NILU now has deposition data for the Treungen up to 2021 (Aas et al. 2022). These were adjusted to estimate deposition at Lille Hovvatn using the same factors as before.

Estimates of deposition of S and N are also available from EMEP (Co-operative Programme for Monitoring and Evaluation of the Long-range Transmission of Air Pollutants in Europe). For the period 1990-2020 these data were obtained from The Norwegian Meteorological Institute through the Meteorological Synthesizing Centre - West (MSC-W).
https://emep.int/mscw/mscw_moddata.html

We used the EMEP data for grid-average wet plus dry deposition of oxidized sulphur (WDEP_SOX + DDEP_SOX_m2Grid), oxidized nitrogen (WDEP_OXN + DDEP_OXN_m2Grid), and reduced nitrogen

(WDEP_RDN + DDEP_RDN_m2Grid). Over the past 25 years EMEP has several times revised the estimates for S and N deposition in Europe. EMEP explains these changes as due to updates in the EMEP MSC-W model and to modifications of the input data (e.g. revised emission data and meteorology). These revisions are made for deposition in the period since 1990. Previous MAGIC calibrations to the 1000-lake data conducted by Larssen et al. (2008) and Austnes et al. (2016) used the EMEP data then available. The EMEP data released in September 2022 and used in this report are again different from the estimates used in the 2008 and 2016 calibrations to the 1995 1000-lake data (Figure 16). For the example shown, lake 36 Lille Hovvatn, the 2022 estimates of historical S deposition are significantly larger than those used previously. Changing estimates of S and N deposition affect the calibration of MAGIC parameters such as weathering rates of the base cations, and thus simulated historical and future ANC concentrations in the lakes.

MAGIC requires estimates of historical S and N deposition at each of the 1000 lakes for the years prior to 1990. These are based on Schöpp et al. (2003) and have not been revised. The historical data are scaled and coupled to the EMEP data in the year 1990 such that the Schöpp et al. (2003) estimate for 1990 agrees with the EMEP estimate for 1990 in every grid cell.

The NILU deposition estimates are significantly different from the EMEP estimates (Figure 16, Table 4). Both estimates are scaled to the year 1995 (or the average of 1994-1996), such that the both the SO_4^* flux and the Cl flux in deposition equals the respective fluxes in runoff. But the trends in SO_4^* deposition over time differ. The EMEP estimates indicate much higher S deposition in the 1970s and 1980s than the NILU measurements. And the EMEP estimates indicate a much steeper decline in S deposition in the period 1990-2020. This problem arises because it is necessary to couple the EMEP estimates for years post-1990 to the Schöpp et al. (2003) estimates for deposition prior to 1990. As the EMEP estimates for the year 1990 appear to be too high, the resulting coupling gives much too high estimates for deposition in the peak years of the 1970s and 1980s.

Because the EMEP estimates for S deposition are too high for the years 1970-1995 and the decline from 1995 to 2020 is too steep, the MAGIC simulations run with the EMEP estimates fail to give satisfactory fits to the measured lake chemistry data 1974-2019. All of the three new MAGIC calibrations to the 1000-lake dataset give much too high concentrations of SO_4 in the years prior to 1995 at Lake 36 Lille Hovvatn (Figure 17). Simulated Ca concentrations are also much too high, as is ANC. The problem does not lie with MAGIC – the manual calibration conducted by Hindar and Wright (2005) in 2004 using NILU deposition data for 1974-2002 gives very good fit to the observed data, and now running this same MAGIC calibrated parameter file onwards with NILU data for subsequent years to 2019 also gives good agreement with the observed data.

The deposition estimates from EMEP for Lille Hovvatn result in much too high flux of SO_4 through the catchment soils, much of which must be compensated by leaching of Ca and other base cations. In the first instance the Ca comes from the soil cation exchange complex, which in turn is replenished by weathering of soil minerals. These processes central to MAGIC are optimised in the calibration procedure, as neither the weathering rate nor the initial (prior to onset of S pollution) stores of Ca in the soil is known. The values for these eight parameters obtained from the calibrations show that the three calibrations on the 1000-lake dataset using the EMEP deposition estimates all need higher base cation weathering rates and higher amounts of exchangeable base cations in the soil to simulate the observed soil and lake chemistry in 1995 or 2019 as compared to the manual calibration based on NILU deposition data (Table 5). The three EMEP based simulations give initial % base saturation of nearly 100%, clearly much too high and totally unrealistic for the thin, poorly-developed podzolic and peaty soils at Lille Hovvatn.
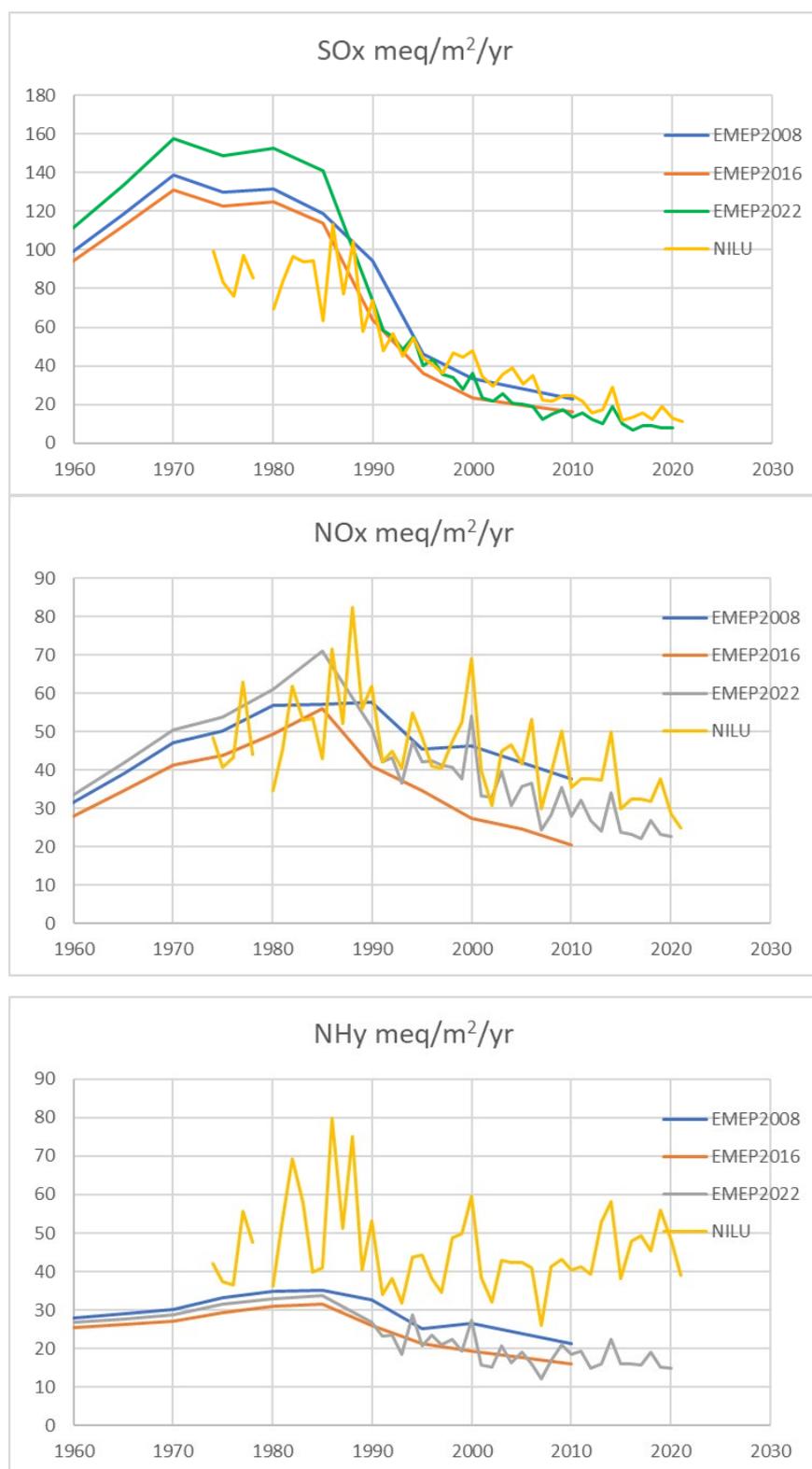
Figure 16. Deposition of non-marine S (SO$_x$), oxidized N (NO$_x$) and reduced N (NH$_y$) at Lille Hovvatn estimated by EMEP and from measured precipitation at Treungen (NILU) (Aas et al. 2022).
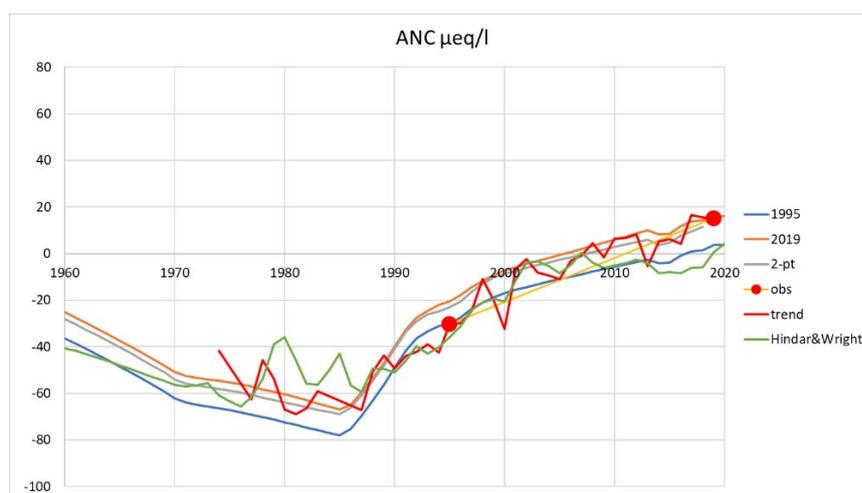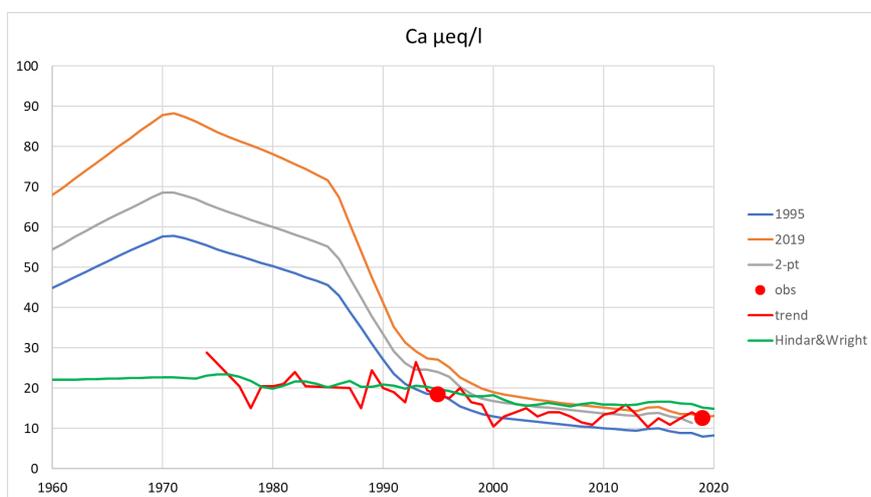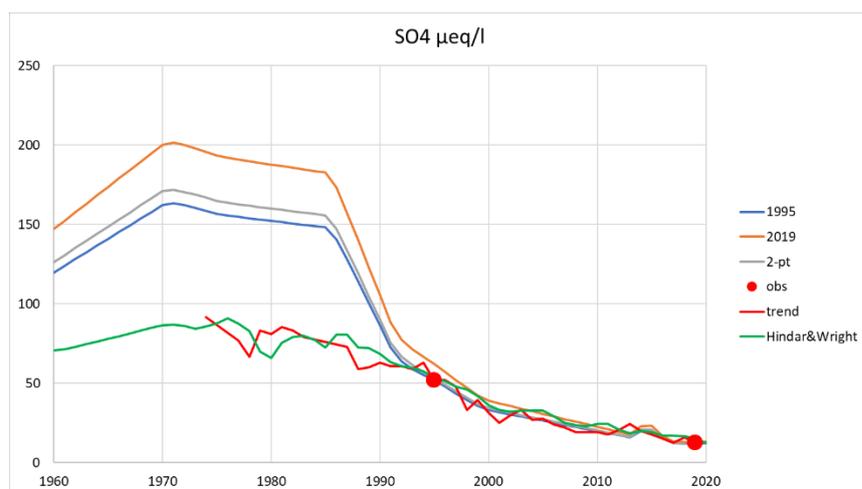
Figure 17. Simulated and observed concentrations of SO_4, Ca and ANC at Lille Hovvatn (lake 36). Shown are the results for Lille Hovvatn from the three MAGIC calibrations to the 1000-lake datasets based on EMEP data with the observed concentrations for the two calibration years 1995 and 2019 (red dots), the observed lake chemistry data (trend), and the MAGIC calibration of Hindar and Wright (2005) based on NILU data and extended here to the year 2020.

Table 4. Non-marine S deposition at Lille Hovvatn estimated by EMEP and from measured precipitation at Treungen (NILU) (Aas et al. 2022). The EMEP estimates are for those used in the MAGIC calibrations to the Norwegian 1000-lake surveys made by Larssen et al. (2008), Austnes et al. (2016), and this report (2022). The EMEP2008 and EMEP2016 values for the year 2020 (italics and red) refer to the CLE (current legislation) scenario.

Year	EMEP2008	EMEP2016	EMEP2022	NILU
1980	131.9	124.8	152.6	69.4
1990	94.5	63.7	74.2	74.1
1995	46.3	36.3	40.1	44.0
2020	*21.9*	*8.9*	7.8	12.8

Table 5. Soil parameters optimised for Lille Hovvatn (lake 36) in the three separate calibration trials – single-point to the 1995 lake data, single-point to the 2019 lake data and two-point to both datasets and in the manual calibration of Hindar and Wright (2005).

	Weathering rate (meq/m^2/yr)				Initial base saturation (% of CEC)			
	WeCa	WeMg	WeNa	WeK	%Ca	%Mg	%Na	%K
1995	28	14	18	2	44	24	7	5
2019	52	25	26	1	54	25	7	4
2-point	40	19	22	2	49	24	7	4
Hindar&Wright	27	6	6	4	27	14	5	4

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