

Modeling Past and Future Acidification of Swedish Lakes

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Abstract Decades of acid deposition have caused acidification of lakes in Sweden. Here we use data for 3000 lakes to run the acidification model MAGIC and estimate historical and future acidification. The results indicate that beginning in about 1920 a progressively larger number of lakes in Sweden fell into the category of “not naturally acidified” ($\Delta\text{pH} > 0.4$). The peak in acidification was reached about 1985; since then many lakes have recovered in response to lower levels of acid deposition. Further recovery from acidification will occur by the year 2030 given implementation of agreed legislation for emissions of sulphur (S) and nitrogen (N) in Europe. But the number of catchments with soils being depleted in base cations will increase slightly. MAGIC-reconstructed history of acidification of lakes in Sweden agrees well with information on fish populations. Future acidification of Swedish lakes can be influenced by climate change as well as changes in forest harvest practices.

Keywords Sweden · Lake · Acidification · Model

INTRODUCTION

Decades of acid deposition have caused acidification of lakes and streams in large areas of Sweden (Almer et al. 1974). Ecological effects include impairment and loss of thousands of fish populations (Tammi et al. 2003). Since the mid-1980s international agreements conducted as part of the United Nations Economic Commission for Europe (UN-ECE) Convention on Long-Range Transboundary Air Pollution (CLRTAP) have achieved substantial reductions in the emissions of sulphur (S) and nitrogen (N) compounds to the atmosphere (UNECE 2012). In response, lakes in Sweden (Wilander and Fölster 2007) and

elsewhere in Fennoscandia have begun to recover from acidification (Skjelkvåle et al. 2001).

The most recent protocol to the Convention, the Gothenburg protocol, entailed nearly 80 % reduction in S emissions from peak years in the late-1970s, and has been largely implemented by the target year 2010. Currently negotiations are underway in the CLRTAP to achieve yet greater reductions in emissions to be implemented by the year 2020.

In 1999, Sweden adopted 15 Environmental Quality Goals as part of an environmental policy for a sustainable Sweden (<http://www.sweden.gov.se/sb/d/5775>). These are to be achieved by the year 2020. One of these is “Natural Acidification Only”. Determination of natural acidification of lakes necessitates assessment of the chemical status in pre-industrial times. The official Swedish criterion of anthropogenic acidification is that the decrease in lake pH relative to pre-industrial status be less than 0.4 pH units (Fölster et al. 2007). This criterion is based primarily on analyses of lake water chemistry and biological status of two organism groups; fish, and littoral invertebrate fauna (Fölster et al. 2007).

During the past 20 years, Sweden’s environmental authorities have conducted several large-scale inventories of lake chemistry. Data are available for ca. 3000 lakes covering the whole country. National data are also available for deposition chemistry, soil chemistry, and forest productivity. Here we use these data with the acidification model Model for Acidification of groundwater in Catchments (MAGIC) (Cosby et al. 1985a, b, 2001) to estimate the pre-industrial condition and historical development of acidification in these lakes. These model simulations are then used to project the extent and degree of anthropogenic acidification in the future given implementation of new reductions in emissions of S and N.

MATERIALS AND METHODS

Sweden has a long coastline on the eastern side and the Scandinavian mountain chain on the western border. Sweden covers 450 000 km², and is a glacially formed landscape with nearly 100 000 lakes. The country lies on the Fennoscandian shield. Bedrock is predominately highly siliceous granitic and gneiss with overburden of largely glacial origin. Much of the country is heavily forested, with 53 % of the surface area being forest and woodland, while farmland constitutes only 8 % of land use. Southern Sweden is predominantly agricultural, with increasing forest coverage northward. Mountains and hills are dominant in the west. The country has large north-to-south gradients in climate, and also coastal to inland gradients in marine influence from the Baltic Sea to the east and the North Sea to the southwest. Nearly all Swedish forests are managed for timber, pulp, and more recently biofuels. Acid deposition is highest in south-western and southern Sweden.

MAGIC

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on soils and surface water chemistry (Cosby et al. 1985a, b, 2001).

The data required to run MAGIC must be spatially and temporally aggregated to represent the whole catchment and lake and the time step of the model. Soil physiochemical parameters are aggregated vertically by the soil profile and spatially to account for the areas of the different soil types within a catchment. The model requires input data for various parameters of lake chemistry, soil chemistry, deposition chemistry, and hydrology (Cosby et al. 2001).

The changes in soil and lake chemistry over time during the calibration period (here from the assumed pre-industrial steady-state condition in 1860 to the calibration period of 2005–2010) are assumed driven by changes in deposition of major ions from the atmosphere and changes in net uptake of base cations and nitrogen by the forest. The time sequence of change in these two fluxes must thus be specified at an annual time step for the calibration period.

Output of MAGIC includes estimation of soil and water acid neutralizing capacity (ANC) at each time step. ANC is a widely used measure of lake acidification status. ANC is defined as the equivalent sum of base cations (Ca, Mg, Na, K) less the equivalent sum of strong acid anions (Cl, SO₄, NO₃). In lakes with high content of humic substances, the contribution of dissociated organic acids may be included in ANC (ANC_{oaa}) (“oaa” organic acid adjusted) (Lydersen et al. 2004). $ANC_{oaa} = ANC - (10.2/3) \times DOC$, where

DOC is the concentration of dissolved organic carbon (mgC L⁻¹), and ANC is in $\mu\text{eq L}^{-1}$.

The MAGIC model has been extensively applied and tested over the past 20 years at many sites and in many regions around the world (Cosby et al. 2001). Overall, the model has proven to be robust, reliable, and useful in a variety of scientific and environmental management activities. MAGIC has been used to assess the recovery of European surface waters from acidification given a S and N deposition scenario of full implementation of the Gothenburg protocol (Wright et al. 2005).

Lakes

A total of 2985 unique lakes were used in this study. Three separate datasets were used: 170 “trend” lakes, sampled four times annually since the mid-1980s; 1526 “synoptic” lakes, sampled in nationwide surveys conducted in 1995, 2000, and 2005; and 1625 “liming reference” lakes sampled in 2007–2008. Since 2005 one-fifth of the synoptic lakes have been sampled each year. 336 of the lakes were included in more than one of these three datasets; duplicates and triplicates were excluded from further analysis. We used the most recent lake chemistry data from the period 2005–2010 for each lake. Water chemistry data for these lakes are held at the Swedish University of Agricultural Sciences Department of Aquatic Sciences and Assessment (<http://info1.ma.slu.se>). Lake annual discharge and lake catchment land use characteristics were obtained from SLU (Jens Fölster pers. comm.).

Soil

Forest soil data for 1993–2002 came from the Swedish Survey on Forest Soils and Vegetation (SK). SK is a regularly repeated survey of forest vegetation and soil chemical and physical qualities, at the permanent plots of the Swedish National Forest Inventory. Data are downloadable from <http://www-markinfo.slu.se>. These were aggregated up (arithmetic averages across soil horizons at each sampling point weighted by soil mass) for each lake catchment. For each lake catchment an average soil properties from the nearest 1–20 soil samples were lumped together to represent catchment soil. The distance of relevant samples from the lake increased from south to north and was estimated for each lake according to procedure described by Moldan et al. (2004). In the absence of a national soil inventory of soils in non-forested land we were relegated to assigning from data available from various research projects. This problem was most obvious at lakes with zero percentage forest cover and therefore no forest soil sampling points in their catchments. For those catchments we assigned an

average soil properties determined by soil sampling at two areas in Norrbottens County (Svarén 1996).

Uptake by Forests

Base cation uptake (actually annual net accumulation in biomass) was assumed to be zero for non-forested areas. For forests we used region and forest type specific uptake rates as calculated by the Swedish ASTA program (International and national abatement strategies for transboundary air pollution) (Akselsson et al. 2006) weighted by the percent forest cover at each catchment.

We assume that forest harvesting is patchy such that the forest in any lake catchment will be composed of patches of young, medium-age, and mature trees. Thus the uptake fluxes used in MAGIC were set equal to the long-term average annual removal of nutrients in biomass (tree harvesting).

In the hindcast we assumed that the net annual uptake of nutrients in the forest was equal to the amount removed from the ecosystem at harvest divided by the mean rotation time. We estimated the biomass (and thus nutrient) removal from forest land based on historic data on forest area and information on harvesting of stems, branches, tops, and stumps in Sweden: 1860–1980 stems only were harvested; 1980–2000 stems plus a percentage of branches and foliage increasing from 0 in 1980 to 16 % in 2000; 2000–2010 further increase in removal of branches and foliage to 28 % in 2010. Beginning in 2000 a fraction of the nutrients removed was compensated by recycling of ash from biomass combustion. Ash recycling was introduced gradually from 2000 and reached 9 % of area in 2010. Thus, the net increase in nutrient removal from 2000 to 2010 was only from 16 to 19 %. For year 2010 and beyond no further changes in forestry practices were assumed.

Deposition

Deposition of S and N for the period 1880–2010 was provided by Coordination Centre for Effects (CCE) and came from estimates of European emissions of S and N (Schöpp et al. 2003) and the EMEP modeled deposition for each of the 50 × 50 km European Monitoring and Evaluation Programme (EMEP) squares (Fig. 1). EMEP is part of the UN-ECE LTRAP; EMEP measures and models European regional air quality. The Coordination Centre for Effects (CCE) (<http://www.rivm.nl/cce>) is also part of LTRAP and is responsible for the development of modeling and mapping methodologies for the integrated assessment of European air pollution effects. Each lake was assigned to an EMEP grid square.

The EMEP deposition values were scaled to each lake/catchment based on measured lake chemistry for most recent year 2005–2010 using the following procedure:

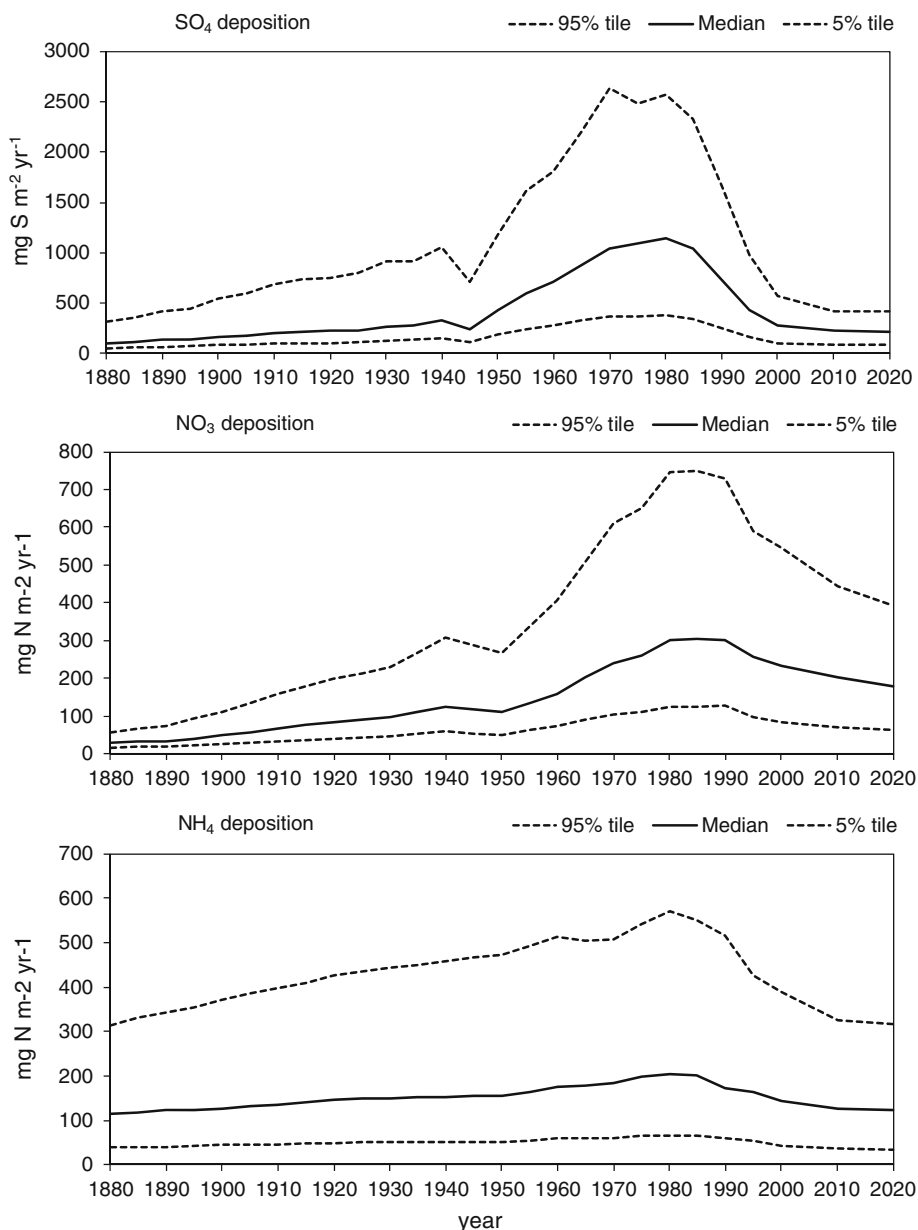
First SO_4 deposition was calculated. We started by assuming that all SO_4 in lake water came from atmospheric deposition. SO_4 deposition was thus set equal to the SO_4 output flux at each lake (SO_4 output flux = SO_4 concentration × Q , where Q is the annual runoff volume). Deposition of sulfate is composed of the marine seasalt fraction ($\text{SO}_{4\text{ss}}$) and the non-marine anthropogenic fraction (SO_4^*). $\text{SO}_{4\text{ss}}$ was calculated at each lake from the chloride (Cl) output flux (Cl output flux = Cl concentration × Q), assuming that all Cl is derived from seasalt deposition and the SO_4/Cl ratio is that of seawater. SO_4^* was then set to total $\text{SO}_4 - \text{SO}_{4\text{ss}}$.

An independent estimate of SO_4^* deposition is available from the EMEP model for all of Sweden, and these were used to check for lakes at which the contribution of SO_4 from internal sources such as weathering of soil minerals in the catchment and lake sediments might be significant. The EMEP model provides estimates of SO_4^* for each 50 × 50 km grid in the model domain. Within each grid, however, local variations topography and surface vegetation can cause large differences in actual deposition of SO_4 to the individual lake catchments. The estimates of SO_4^* deposition derived for each lake were regressed against the EMEP estimates of SO_4^* deposition from the grid containing each lake to provide an estimate of the average enhancement of SO_4^* deposition above the EMEP estimate caused by local landscape conditions (SO_4^* dep local = $1.8 \times \text{SO}_4^*$ dep EMEP + 26, units $\text{meq m}^{-2} \text{ year}^{-1}$). At 81 of the 2903 sites the lake output flux of SO_4 exceeded two times the regression relationship between local and EMEP deposition. For these outliers we assumed that the excess was result of weathering of SO_4 in the soils or lake sediments and reduced the estimated local SO_4^* deposition to the average regression response for all lakes.

Deposition of NO_x and NH_y were calculated from the SO_4^* deposition and the molar ratios in the EMEP modeled deposition (NO_x/SO_4 , NH_y/SO_4) provided by the CCE. Deposition of Cl was assumed equal to the Cl export flux at each lake. The deposition of Na, Mg, Ca, K, and marine SO_4 were calculated from the deposition of Cl and the ionic ratios of these ions in seawater. Deposition of Na, Mg, Ca, and K other than from seasalt was calculated at each lake (together with mineral weathering of these cations) during the calibration procedure. Deposition of base cations and Cl were assumed constant throughout the entire simulation period 1860–2030 except for years 1983–2009, when sea salt deposition was varied from year to year based on observed Cl concentrations in lakes. Deposition sequences for the period 1880–2010 for the three components S, NO_x , and NH_y were specified by the CCE for each EMEP grid square. These were normalized to the deposition values calculated for the year 2005–2010.

We used the scenario for the year 2020 “cost optimized baseline” (COB) for S and N deposition supplied in 2010 by the CCE. COB is based on full implementation of

Fig. 1 Deposition (grid square average) of S, NO_3^- , and NH_4^+ for three EMEP grid squares in Sweden 1860–2030. Shown are the values for the three EMEP squares representing the 95th, 50th and 5th percentiles of the distribution of S deposition in the year 2000. Data for 1880–2010 provided by the Coordination Centre for Effects (CCE), Bilthoven. Values for 1860 were assumed equal to “background” (natural deposition only) and ramped from 1860 to 1880. Values for 2020 assume “cost optimized baseline” (COB) supplied by CCE, and ramped from 2010 to 2020 then held constant to 2030. Units: $\text{mgS m}^{-2} \text{ year}^{-1}$ and $\text{mgN m}^{-2} \text{ year}^{-1}$



existing European and national legislation for S and N emissions by the year 2020. For the forecast we assumed that no further changes in S and N deposition occur after the year 2020. The MAGIC simulations were run out to the year 2030 to fully allow the catchment-lake systems to adjust to the new deposition levels projected for 2020.

Calibration

MAGIC (version 777) was calibrated to each lake using an automated optimization routine. The optimization routine sought to match both the simulated with the observed lake chemistry (most recent year in the period 2005–2010) and

the simulated with the observed 1995 soil chemistry (year for which soil chemistry was available). The windows for the automatic calibration were set to $\pm 2 \mu\text{eq L}^{-1}$ for all major ions and ± 0.2 units for % saturation of the four base cations. These calibrations assumed a constant percent retention of N in the catchment/lake system for each site.

We calculated lake water equivalent concentration of dissociated dissolved organic carbon (DOC) with MAGIC from the observed DOC and pH in the lakes assuming a triprotic organic acid with pKa values of 2.64, 5.66, and 5.94 (Hruška et al. 2003). Further we assumed that DOC did not change in either soil solution or lake water over the simulation period.

Acidification Measures

ANC or ANC_{OAA} are typically used as measures of lake acidification status (Henriksen and Posch 2001). ANC is a good explanatory variable for biological effects of acidification, such as population status of fish in Swedish lakes (Holmgren and Buffam 2005) and brown trout in Norwegian lakes (Bulger et al. 1993; Lien et al. 1996; Lydersen et al. 2004). Larssen et al. (2010) grouped ANC_{OAA} levels into several classes, representing the probability of water quality suitable for brown trout populations in Norway. These are: >75 % probability of damaged population ($\text{ANC}_{\text{OAA}} < -37 \mu\text{eq L}^{-1}$); 25–75% probability of damaged population (-37 to $-20 \mu\text{eq L}^{-1}$); 2.5–25% probability of damaged population (-20 to $0 \mu\text{eq L}^{-1}$); 0.5–2.5% probability of damaged population (0 to $12 \mu\text{eq L}^{-1}$); and <0.5% probability of damaged population ($>12 \mu\text{eq L}^{-1}$).

In Sweden the official measure of acidification for lakes is defined as lake pH more than 0.4 pH units below the pre-industrial reference pH level (Fölster et al. 2007). This measure is based on adverse effects on two organism groups: fish and littoral fauna. The procedure specified for obtaining the pre-industrial pH value for a given lake entails first estimation of pre-industrial lake ANC and then calculation of pre-industrial pH. The method specifies that pre-industrial ANC be calculated by MAGIC. Pre-industrial pH is then calculated from ANC using the equation:

$$\text{ANC} = ([\text{CO}_3^{2-}] + [\text{HCO}_3^-] + [\text{org}^-]) - ([\text{H}^+] + [\text{Al}^{n+}]),$$

where org^- is the sum of organic anions, Al^{n+} is the sum of cationic aluminum species, and units are $\mu\text{eq L}^{-1}$. Org^- is calculated from present-day DOC concentration assuming no change over time and the triprotic model of Hruška et al. (2003). Concentrations of bicarbonate and carbonate are calculated assuming CO_2 content is four times atmospheric (0.156 % by volume). Concentrations of cationic Al are assumed negligible (Fölster et al. 2007). ΔpH gives a measure of the cumulative change since the onset of acid deposition.

MAGIC also simulates changes in the soil store of base cations. Base cations are added to the soil by atmospheric deposition and weathering of soil minerals, and removed from the soil by leaching and by uptake in growing vegetation. The change in store of base cations provides a measure of whether the soil is deteriorating or recuperating. We use the annual percent change in store of exchangeable base cations in the soil as a measure of terrestrial ecosystem acidification.

RESULTS

Calibration was successful for 2903 lakes; simulated concentrations of major ions and ANC in lake water and for

the four base cations in soil agreed with the observed values to within the precision specified by the calibration windows. The main reason for those that did not calibrate was noise in the data when modeling Na and Mg at sites with large seasalt inputs. At present-day (2005–2010) acid lakes are found all over Sweden, but predominately in south-western, south-central, and north-eastern parts of the country (Fig. 2).

The results indicate that beginning in about 1920 a progressively larger number of lakes in Sweden fell into the category of “not naturally acidified” according to the ΔpH acidification measure (Fig. 3). By this definition, of course, there were no acidified lakes prior to the onset of acid deposition. The simulations indicate that the peak in acidification (1273 of the 2903 lakes) was reached in about 1985, and since 1985 a substantial number of lakes have recovered. By the year 2010, 455 of the 1273 acidified lakes had recovered ($\Delta\text{pH} < 0.4$). The decline in S deposition (Fig. 1) has been faster than the recovery of the lakes; the lag time is due to several factors including the slow replenishment of base cations in the soil, and the recent increases in removal of base cations in biomass.

Today (2005–2010) most of the acidified lakes under the criterion of ΔpH greater than 0.4 are located in south-western and south-central Sweden (Fig. 2).

The ANC_{OAA} measure shows a similar history. Reconstructed values of ANC_{OAA} indicate that a small number of lakes had $\text{ANC}_{\text{OAA}} < 12 \mu\text{eq L}^{-1}$ in pre-industrial times (Fig. 4). The number of lakes with low ANC_{OAA} increased slowly in the first half of the 1900s, and then accelerated steeply from about 1950. As for ΔpH , the peak was reached in about 1985. Since then a progressively larger number of lakes have recovered to $\text{ANC}_{\text{OAA}} > 12 \mu\text{eq L}^{-1}$. By the year 2010 recovery had progressed such that water quality was approximately the same as in the 1960s.

The simulated changes in soil pools of base cations in the catchments of the lakes indicate that at peak acidification in 1985 a majority of the catchment soils were being depleted by more than 0.2 % per year (Fig. 5). By the year 2010 this number had decreased substantially.

The model simulations for the next 20 years indicate that further recovery from acidification will occur given implementation of the COB scenario for emissions of S and N in Europe. COB entails existing national and international legislation for emissions of S and N, and is intended to be fully implemented by the year 2020. By the year 2030 less than 10 % of the lakes will remain acidified by the measure $\text{ANC}_{\text{OAA}} < 12 \mu\text{eq L}^{-1}$. By the measure $\Delta\text{pH} > 0.4$ the fraction of the 2903 lakes that are “not naturally acidified” will decrease to 15 %. But the fraction of catchments with soils being depleted in base cations will increase slightly.

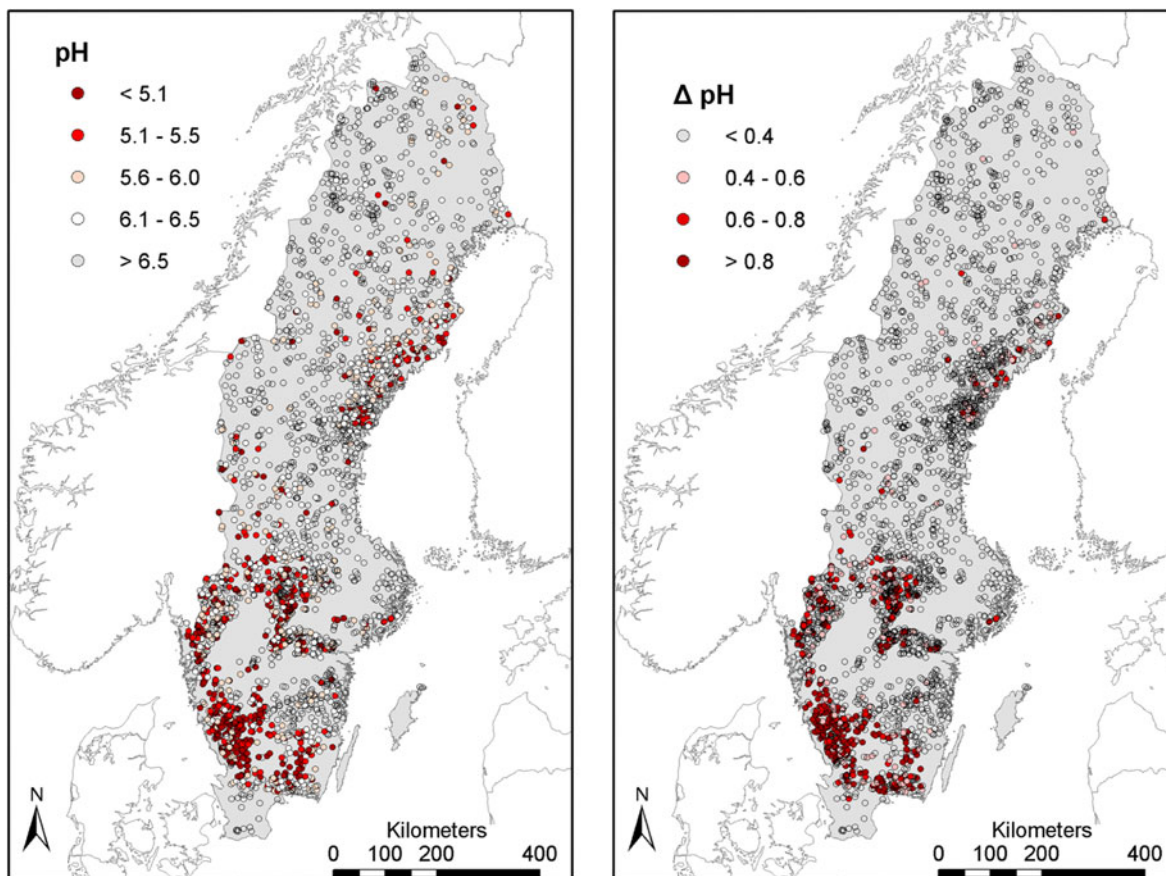


Fig. 2 Maps of the 2903 lakes in Sweden calibrated in this study. *Left panel* pH levels for the calibration year 2005–2010. *Right panel* acidification status in 2005–2010 by the criterion of change in pH since pre-industrial time

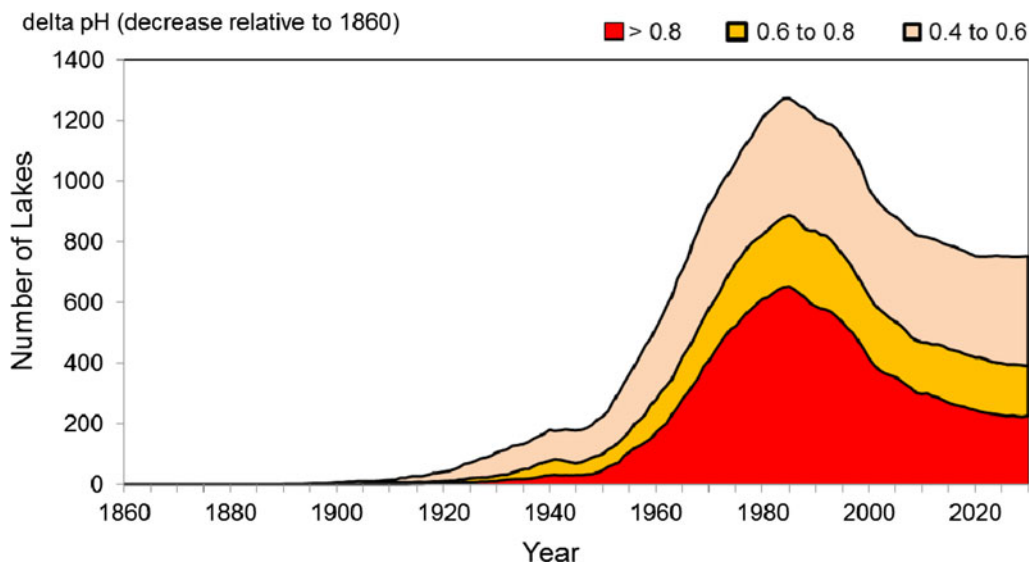


Fig. 3 Number of lakes (of total 2903) falling into various ΔpH classes. ΔpH is defined as the decrease in pH relative to pH in 1860

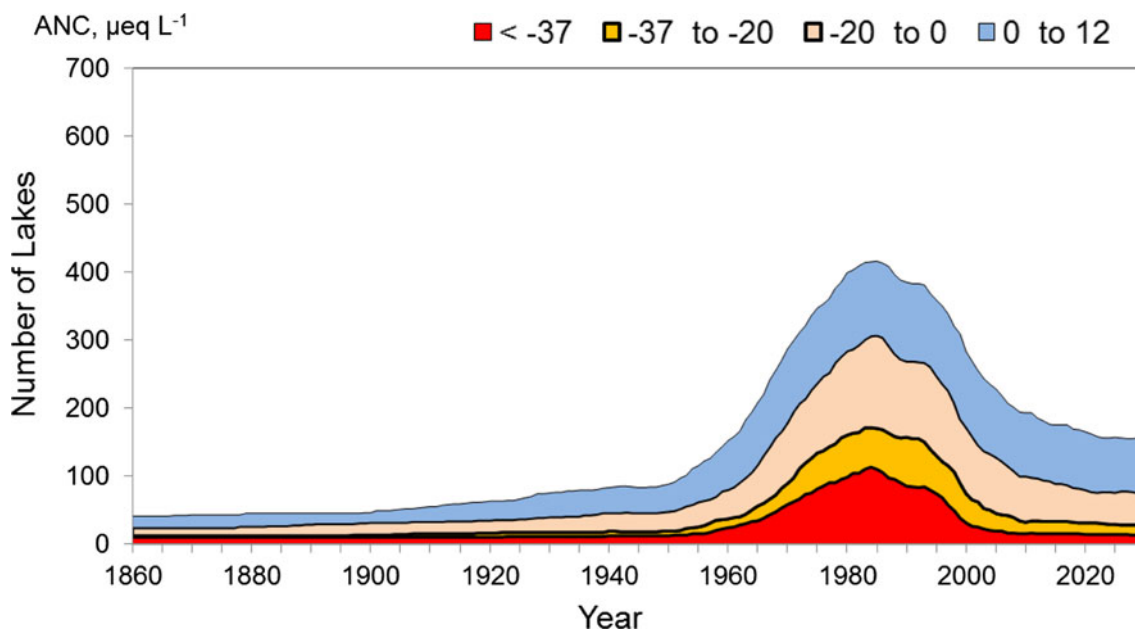


Fig. 4 Number of lakes falling into various classes of ANC_{oaa} (ANC organic acid adjusted, units $\mu\text{eq L}^{-1}$). The classes represent ANC_{oaa} levels for which water quality is suitable for brown trout populations in Norway (Larsen et al. 2010). *Red* >75 % probability of damaged population (ANC_{oaa} < -37 $\mu\text{eq L}^{-1}$); *orange* 25–75 % probability of

damaged population (-37 to -20 $\mu\text{eq L}^{-1}$); *pink* 2.5–25 % probability of damaged population (-20 to 0 $\mu\text{eq L}^{-1}$); *blue* 0.5–2.5 % probability of damaged population (0 to 12 $\mu\text{eq L}^{-1}$); *white* <0.5 % probability of damaged population (>12 $\mu\text{eq L}^{-1}$)

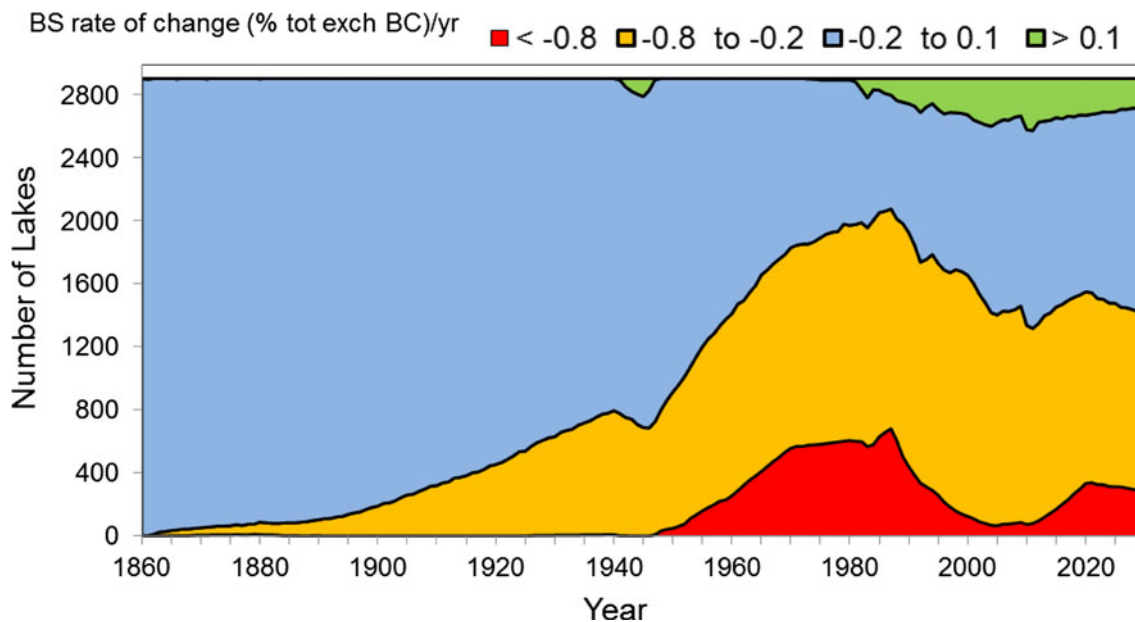


Fig. 5 Number of lakes falling into various classes of rate of change of soil pool of exchangeable base cations. *Red* soils are strongly acidifying; *orange* soils are weakly acidifying; *blue* no large change; *green* soil base cation pools are recovering

DISCUSSION

The two maps of lakes in Sweden show the present-day (2005–2010) acidity of the lakes (pH map) on the one hand, and the acidification status of the lakes ($\Delta\text{pH} > 0.4$) on the

other hand (Fig. 2). Acid lakes with $\text{pH} < 5.1$ are found in south-western, south-central, and north-eastern Sweden as well as scattered throughout the rest of the country. But acidified lakes ($\Delta\text{pH} > 0.4$) are found predominately in south-western and south-central Sweden, regions receiving

the highest deposition of S and N. Many of the acid lakes in northern Sweden are thus naturally acidic, predominately due to high concentrations of natural organic acids. Acidic lakes (i.e., those with low pH) are not necessarily acidified (i.e., decrease in pH).

There are well-established empirical relationships between ANC, pH, and DOC in natural waters. Lakes with low ANC have low pH. The relationship is not linear; at low ANC levels ($<100 \mu\text{eq L}^{-1}$) small changes in ANC give large changes in pH, while at high ANC ($>200 \mu\text{eq L}^{-1}$) large changes in ANC give small changes in pH. The natural organic acids in DOC give a lower pH for a given ANC. The triprotic model of Hruška et al. (2003) implies that 1/3 of the organic acids are nearly 100 % dissociated at pH above 4, while the other 2/3 dissociate and thereby buffer changes in pH in range pH 5–7. The ANC–pH relationship is thus such that for a given ANC, the pH will be lower at higher DOC level.

The patterns in recovery of modeled ANC_{OAA} for the 2903 lakes over the period 1990–2005 agree roughly with those observed in 73 “trend lakes” (Wilander and Fölster 2007). The trend lakes are sampled four times yearly. The data from the trend lakes unfortunately begin too late to assess whether the modeled peak in acidification did in fact occur around 1985. The modeled recovery patterns also agree with those of the repeated synoptic surveys of Swedish lakes conducted at 5-year intervals 1990–2005 (Wilander and Fölster 2007). The summary of Wilander and Fölster (2007) entails the scaling up from the sampled synoptic lakes to all lakes in Sweden based on the statistical design of the lake selection for the synoptic surveys. The recovery patterns in the trend lakes, the upscaled synoptic lakes, and the 2903 modeled lakes here are all similar with the steepest recovery in the early 1990s and with lower rates in subsequent years.

Loss and damage to fish populations have long been the major concern with regard to acid deposition and acidification of surface waters. Brown trout is one of the more sensitive fish species, and is the principal fish species in most lakes in Sweden and the rest of Fennoscandia (Tammi et al. 2003). Decades of research have established that chronic levels of acid deposition result in decreased ANC and pH and increased concentrations of labile inorganic aluminum (Al^{n+}) in surface waters (Schindler 1988). Al^{n+} is the toxic agent for many life stages of brown trout (Baker and Schofield 1980; Hesthagen et al. 2008). Organic anions combine with both Al^{n+} and H^+ ; high DOC waters are thus generally less toxic to fish. This is the basis for the organic acid correction ANC_{OAA} , which gives a statistically better relationship to brown trout status in Norwegian lakes than ANC (Lydersen et al. 2004; Hesthagen et al. 2008).

The relationship between lake water chemical parameters and fish communities has also been evaluated for

Swedish data (Holmgren and Buffam 2005). They tested nine different indices based on pH, H^+ , alkalinity, ANC, Al^{n+} , and Ca on a dataset of about 60 lakes in southern Sweden, and produced logistic regression models of nearly equally good fit ($r^2 = 0.8\text{--}0.9$). Most likely the ANC_{OAA} parameter would also give a good fit, but their study was made before the ANC_{OAA} index was proposed.

MAGIC reconstructed history of number of low ANC_{OAA} lakes in Sweden agrees well with information on the status of fish populations (Tammi et al. 2003). Tammi et al. (2003) present fish status data collected in 1995–1997 for 3821 lakes in Norway, Sweden, and Finland, and relate the observed fish status to acidification. For the 2290 lakes in Sweden with fish data, extrapolated up to all lakes >4 ha, about 6 % of the brown trout populations were lost and an additional 4 % potentially affected by acidification. The latter was based on a critical limit of ANC $20 \mu\text{eq L}^{-1}$ (Lien et al. 1996). Together this amounts to about 10 % of the brown trout populations lost or affected in 1995, very similar to percent of the lakes with MAGIC reconstructed $\text{ANC}_{\text{OAA}} < 12 \mu\text{eq L}^{-1}$ in 1995 (Fig. 4). Tammi et al. (2003) also report that a small number of lakes never supported brown trout populations; many of these may be naturally low pH, low ANC_{OAA} , and high DOC lakes as indicated by the MAGIC reconstructions for the pre-industrial (1860) state (Fig. 4).

The MAGIC simulations indicate that acid deposition during the 1900s led to progressive depletion of the pools of exchangeable base cations in soil (Fig. 5). At the peak in 1986 base cation pools in nearly 2000 of the 2903 catchments were being depleted at the rate of $>0.2 \text{ % year}^{-1}$. With the decrease in acid deposition since the mid-1980s, this trend has reversed, and the rate of depletion of base cation pools decreased at many sites and at some sites replenishment began. This turn-around agrees with the results from Swedish national soil surveys conducted since 1983 (Stendahl 2007). These surveys show a decrease in the number of soils falling into soil acidity classes “acid” and “very acid”. The change was largest for sites in southwestern Sweden.

Exchangeable base cation pools in soil are replenished by deposition of base cations from the atmosphere and weathering of soil minerals, and depleted by leaching and by removal of base cations in biomass. Leaching is enhanced by the flux of sulfate and nitrate through the soil. More intensive forest harvesting practices increase the removal of base cations in biomass. An assessment of the relative contributions of these four fluxes suggests that in year 2000 acid deposition and stem-only forest harvest resulted in net depletion of base cation pools in southern and central Swedish forest soils (Akselsson et al. 2006, 2007). More intensive harvest entailing removal of tops and branches would nearly double the rate of soil

acidification in southern Sweden. This is reflected in the MAGIC simulations (Fig. 5), which assume increased removal of biomass in the next 20 years; soils begin to lose base cations once again despite the projected lower S deposition under the COB scenario. Lower base cation pools in soil could result in re-acidification of sensitive lakes in the future.

Increases in the fraction of incoming N leached from the lake catchments could greatly affect future acidification in the lakes. At present more than 90 % of N deposition is retained in catchment soil and vegetation; average NO₃ concentration in the 2903 lakes was less than 4 µeq L⁻¹. The strong acid anion NO₃ thus contributed very little to ANC levels in the lakes. But clearly catchment soil cannot continue as a large sink for incoming N forever. Chronic deposition of N leads to a risk of nitrogen saturation, commonly described as the situation in which an increasing fraction of incoming N is leached to runoff (Aber et al. 1989). Swedish coniferous forests are typically N-limited (Tamm 1991), and long-term experiments with N addition indicate that development of N saturation requires many decades (Wright and van Breemen 1995; Moldan and Wright 2011). The experimental evidence suggests that NO₃ leaching in the future probably does not threaten the recovery of surface waters in Sweden.

Harvest of forest biomass alleviates this threat somewhat by removing N from the catchment, but this is more than offset by the removal of base cations in biomass (Akselsson et al. 2006, 2007).

Future acidification of Swedish lakes can be influenced by climate change as well as changes in forest harvest practices (Moldan et al. 2009). Climate change can influence the rates of many processes in catchment and lakes, including base cation weathering, growth of forest and uptake of base cations and N, increased leaching of N, and changes in concentrations of DOC (Wright et al. 2006). Modeling studies suggest that the combination of chronic N deposition, climate change, and increased forest harvest pose a risk for increased leaching of NO₃ to lakes, with the resultant increase in acidification (Moldan et al. 2009). This conclusion agrees with similar modeling studies in Finland (Aherne et al. 2008, 2011).

The MAGIC model has evolved from a pure research device to a robust and versatile tool with procedures and software that allow handling of large datasets and thousands of lakes and catchments. In Sweden MAGIC has been designated as the official mechanism for reconstructing past acidification status of lakes. MAGIC has been recently used for national-scale scenario analyses for combinations of future acid deposition, climate change, and forest practices in Finland (Aherne et al. 2011; Forsius et al., in preparation). MAGIC has been applied to national lake datasets in several countries to derive critical and

target loads for S + N deposition, and to evaluate the potential environmental improvement given alternative future scenarios for S and N emissions to the atmosphere. These outputs feed into European-wide assessments carried out by the Coordination Centre for Effects and are used for the Convention on Long-range Transboundary Air Pollution.

New protocols under negotiation may result in yet lower deposition relative to COB, and thus give Sweden a yet better chance to meet the environmental objective of “natural acidification only”. But the MAGIC simulations here and other similar scenario analyses indicate that changes in forest practices and climate change could worsen lake acidification in the future, and thus point to the need for greater reductions in sulfur and nitrogen deposition.

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