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Modelling regional response of lakewater chemistry to changes in acidic deposition: the MAGIC model applied to lake surveys in southernmost Norway 1974–1986–1995

Bernard J. Cosby[1] and Richard F. Wright[2]

[1] Department of Environmental Sciences, University of Virginia, Charlottesville, VA 22903 USA.
[2] Norwegian Institute for Water Research, Box 173 Kjelsås, N-0411 Oslo, Norway.

Abstract

Two methods for modelling regional responses of lake water quality to changes in acidic deposition in southernmost Norway were examined. Both methods are based upon the MAGIC model but differ in mode of regional application; one uses site-specific while the other uses Monte-Carlo methods for model calibration. The simulations of regional responses from both methods were compared with observed responses based on data from three lake surveys in southernmost Norway conducted in 1974, 1986 and 1995.

The regional responses of the two modelling approaches were quite similar and agreed well with the observed regional distributions of lakewater chemistry variables. From 1974 to 1986 the observed data indicated that despite a decline of approximately 10% in sulphate (SO_4) deposition, the mean acid neutralizing capacity (ANC) of lakes in southernmost Norway declined by approximately 6 μeq l^{-1}. Both modelling approaches simulated no change or a very small decline in mean ANC for that period. From 1986 to 1995 the observed data indicated that, in response to an approximate 40% decline in SO_4 deposition, the mean ANC of lakes in southernmost Norway increased by 11–16 μeq l^{-1}. The modelling approaches simulated increases of 9–10 μeq l^{-1} in mean ANC for the same period. Both simulations and observations indicate that >65% of lakes in southernmost Norway were acidic in 1974 and 1995. Both simulation methods predict that >65% of the lakes in southernmost Norway will have positive ANC values within 10 years of reductions of SO_4 deposition to 20% of 1974 levels.

Of the two regionalization methods the site-specific method appears preferable, because whereas the Monte-Carlo method gives results for a region as a whole, the site-specific method also reveals patterns within the region. The maintenance of a one-to-one correspondence between simulated and observed systems means that simulation results can be mapped for a geographically explicit presentation of model results. The ability to examine geographic patterns of response is becoming increasingly important in regional assessments.

Introduction

Assessment of the effects of acidic deposition on aquatic resources (lakes and streams and the biological communities therein) requires a regional perspective. It is not enough to understand and predict the responses of one or a few intensively studied systems. Informed policy decisions should be based on assessment of the response of all of the resources of concern. Yet, even when this extensive information is available, it is usually necessary to distill the estimates to provide a few measures of the effects of interest. For instance, if synoptic surveys of current water chemistry were available for all aquatic resources within a region (potentially hundreds or thousands of lakes or stream segments), the policy-relevant summary of those extensive data might be simply the number of lakes with acid neutralizing capacity (ANC) less than 20 μeq l^{-1}, or the number of river stretches with pH less than 5.5. The nature of regional analyses, then, requires a large number of observations on the freshwaters of interest in order to provide reliable estimates of a few properties of the entire population. These data are usually derived from synoptic surveys. Surveys are commonly implemented at a single point in time in order to provide estimates of the current properties of the population.

For lakes in a region, if the assessment is concerned not just with the current chemical status, but also with the chemical variation through time of the lake population (e.g., trends in response to changing deposition), there are two approaches to collecting the additional data necessary to estimate and understand the trends. The first approach (termed here 'site-specific method') is to sample the same lakes repetitively through time. The changes in lake variables

between sampling times can then be calculated and examined for each lake sampled, and population estimates for the trends can be derived directly (mean change, etc.). The second approach (termed here 'distributed' or 'Monte-Carlo') is to use independent survey samples taken at different times. In this approach, the same lakes may not have been sampled during each survey with the result that it may not be possible to calculate changes for individual lakes within the sample population. The estimates of trends that can be derived from this approach are limited to changes in population parameters (e.g. changes in means rather than mean changes as derived from the site-specific approach). As sample sizes of each survey are large and are unbiased (i.e. are true sub-samples of the parent population), estimates derived from the two approaches will converge.

If the policy assessment requires estimates of future water chemistry (or of water chemistry prior to the earliest survey data available), mathematical models of water chemistry responses to changing atmospheric deposition are needed to predict the future (or to reconstruct the past) condition of the systems. The models need be applied to many lakes in the region and the simulated results distilled to provide a few measures of the forecast (or hindcast) regional water chemistry. The statistics applied to simulated data from the model could be the same as those applied to the survey data. The model would be expected to reproduce the population properties of the survey data set to which it was calibrated. The model would also be expected to reproduce fairly closely the population properties of surveys taken at other times or the trends in water chemistry variables between sampling times (i.e., 'model validation'). The data needed to calibrate and 'validate' a model for assessment activities can be derived either from independent surveys or from long-term monitoring of the same resources. Just as empirical descriptions of resource properties derived from the two approaches will converge, the two approaches become equivalently useful for calibrating models (and establishing confidence in model applications) as the sample sizes of the survey become large and unbiased.

In practice, long-term monitoring of a large number of lakes (a combination of the two approaches mentioned above) is unusual. Individual surveys of relatively large numbers of lakes have been implemented, but these large synoptic surveys occurred at a single point in time and were not repeated at later times. In other cases, long-term (years-to-decades) monitoring programs have been implemented and have resulted in time series data for individual lakes, but the numbers of lakes monitored in these long-term programmes has usually been small. It would be useful to compare the two sampling approaches: 1) used to estimate regional conditions or regional trends in resources; and 2) used to calibrate and 'validate' a mathematical model of regional response.

Norway has implemented three national lake surveys at approximately ten-year intervals (1974, 1986 and 1995) (Wright and Snekvik, 1978; Henriksen *et al.*, 1988; Skjelkvåle *et al.*, 1997). Each survey sampled hundreds of lakes and was more or less independently designed and implemented. Within these survey data sets, 83 lakes were sampled in all three surveys and so provide a smaller long-term monitoring data set (spanning approximately 20 years). These data thus provide examples of both approaches to regional sampling. These data are used here to compare the two sampling approaches for estimating regional water chemistry characteristics of the Norwegian lakes and their trends over twenty years. The survey data (using each sampling approach) are used to calibrate and evaluate MAGIC (Cosby *et al.*, 1985a,b), a model of water chemistry response to atmospheric deposition. The model is applied to southernmost Norway using the two regionalization methods.

Methods

THE NORWEGIAN LAKE SURVEY DATA

Regional lake surveys were first conducted in southern Norway from autumn 1974 to spring 1975 (Wright and Henriksen, 1977) and in southernmost Norway in autumn 1974 and 1975 (Wright and Snekvik, 1978). Lakes in these surveys were selected with the restriction that they were headwater lakes in pristine watersheds with no local sources of pollution from agriculture, municipal effluent, industry or hydroelectric development. More than a thousand lakes were sampled in the combined surveys.

In 1986 Norway conducted a second regional lake survey—the '1000 Lake Survey' (Henriksen *et al.*, 1988). The lakes in this survey were selected: (1) from areas underlain by bedrock expected to give runoff waters with low buffer capacity, such as granites, gneisses and migmatites; and (2) from areas known to be affected by acidification. The lakes surveyed in 1986 were located in headwater areas with no direct human influence. Most of the lakes sampled were larger than 0.2 km^2.

In 1995, Norway conducted a third regional lake survey, this time with two sets of lake populations. The first population was based on a randomized stratified selection of lakes ('statistical selection') (Skjelkvåle *et al.*, 1997). This survey was a part of the Northern European Lake Survey 1995, where selection of lakes, sampling and analytical methods were standardized across several countries (Henriksen *et al.*, 1998). In Norway, 1008 lakes were included in this survey. The second lake population was a resurvey of the lakes that had been sampled and analyzed in the '1000 Lake Survey' in 1986. Many lakes, however, were limed in Norway after the 1986 survey. When lakes with liming or other disturbances in the catchment were excluded there remained only 485 lakes in the resurvey population.

The data used here were limited to surveyed lakes in the region of southernmost Norway (Fig. 1). This region is

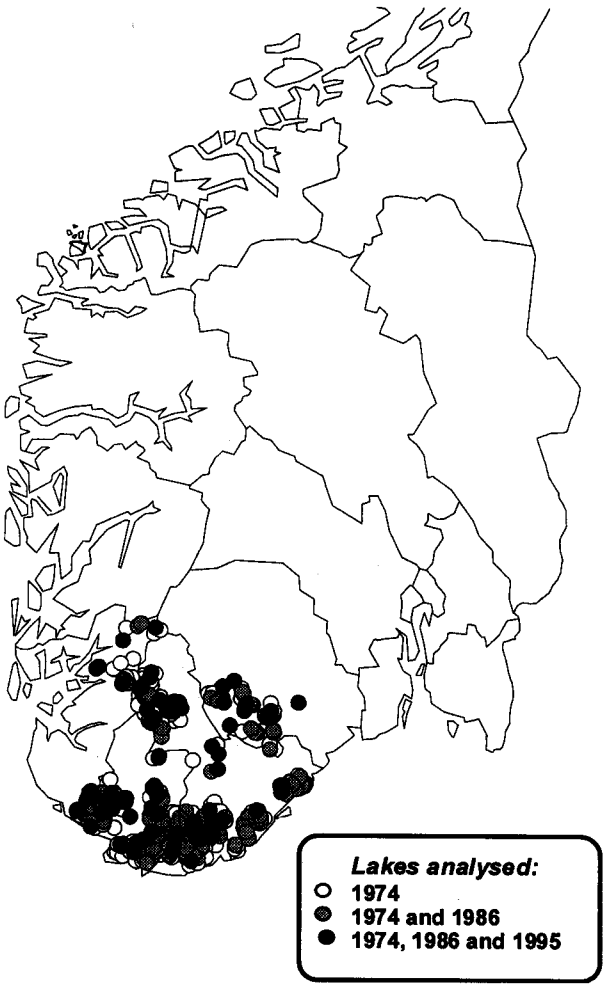

Fig. 1. *Map of southern Norway showing locations of lakes surveyed (data from Skjelkvåle et al., 1997).*

defined by township number, and includes some parts of the county of Telemark, all of East Agder, all of West-Agder and parts of Rogaland. Only lakes sampled in the autumn were selected. In the case of the first surveys in the 1970s, only those lakes sampled in the autumn of 1974 were selected. In the case of the 1995 survey, only those lakes sampled also in 1986 were included in this study.

Table 1. Numbers of lakes sampled in southernmost Norway in each survey year and the number of re-sampled lakes for each survey and for all three surveys.

	Year		
	1974	1986	1995
total lakes sampled in region	463	434	220
number in common with previous sampling		180	220
number in common with previous two samplings			83

After this selection process between 220 and 463 lakes were available in each survey year (Table 1). These survey data sets (one for each year) will be referred to here as the 'lake survey data' and were used for the Monte-Carlo regional application of MAGIC. There were 83 lakes that were sampled in all three survey years (Table 1). These data will be referred to as the 'monitored lakes data' and were used for the site-specific regional application of MAGIC.

The survey data include concentrations of calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+), potassium (K^+), chloride (Cl^-), sulphate (SO_4^{2-}) and nitrate (NO_3^-). Concentrations of ammonium (NH_4^+) are negligibly small. The definition of acid neutralizing capacity (ANC) used here is based on charge balance:

$$ANC = SBC - SAA \quad (1)$$

where SBC (sum of base cation) is the equivalent sum of base cation concentrations (Ca^{2+}, Mg^{2+}, Na^+ and K^+) and SAA (sum of strong acid anions) is the equivalent sum of acid anion concentrations (Cl^-, SO_4^{2-}, NO_3^-).

THE MAGIC MODEL AND REGIONAL APPLICATIONS

Model description

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on surface water chemistry (Cosby *et al.*, 1985a,b; 1989a). The model simulates soil solution chemistry and surface water chemistry to predict annual average concentrations of the major ions. MAGIC consists of: (1) a set of simultaneous reactions involving SO_4 adsorption, cation exchange, dissolution-precipitation- speciation of aluminium (Al) and dissolution-speciation of inorganic carbon (C) and; (2) a mass balance section in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering, net uptake and loss in biomass and losses to runoff. At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from this pool change over time owing to changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. The degree and rate of change of surface water acidity thus depend both on flux factors and the inherent characteristics of the affected soils.

Cation exchange is modelled using equilibrium (Gaines-Thomas) equations with selectivity coefficients for each base cation and Al. Sulphate adsorption is represented by a Langmuir isotherm. Aluminum dissolution and precipitation are assumed to be controlled by equilibrium with a solid phase of Al trihydroxide. Aluminum speciation is calculated by considering hydrolysis reactions as well as complexation with SO_4 and fluoride (F). Effects of carbon

dioxide (CO_2) on pH and on the speciation of inorganic C are computed from equilibrium equations. Organic acids are represented in the model as tri-protic analogues. First-order rates are used for retention (uptake) of NO_3 and NH_4 in the catchment. Weathering rates are assumed to be constant. A set of mass balance equations for base cations and strong acid anions is included. Given a description of the historical deposition at a site, the model equations are solved numerically to give long-term reconstructions of surface water chemistry.

MAGIC has been used extensively to reconstruct the history of acidification and to simulate the future trends at a large number of individual catchments in both North America and Europe (Jenkins *et al.*, 1990a,b; Lepistö *et al.*, 1988; Whitehead *et al.*, 1988; Wright *et al.*, 1994), including tests of the model against manipulation experiments (Cosby *et al.*, 1995, 1996; Wright *et al.*, 1990a).

Regional applications of MAGIC

There are two methods for modelling the regional response of surface waters using MAGIC. The site-specific method relies on the calibration of MAGIC for each catchment in the region included in the surface water survey. This method uses numerical techniques to select parameter values for MAGIC that result in a minimum sum of squared errors between simulated and observed water chemistry variables for each catchment. The site-specific method produces an ensemble of calibrated parameter sets for which there exists a one-to-one correspondence between model simulations and observed catchments. It is possible, therefore, using this method, to calculate changes or trends for each simulated lake and derive statistical descriptions of the simulated trends.

The second method, called the Monte-Carlo method relies on the calibration of MAGIC to simulate the observed statistical distributions of water chemistry variables in the region as a whole. This method uses Monte-Carlo simulation techniques to select joint distributions of input parameters for MAGIC that result in joint distributions of simulated water chemistry variables with the same properties as the joint distributions of observed water chemistry variables. The Monte-Carlo method does not maintain a one-to-one correspondence between simulated and observed catchments. Rather, the Monte-Carlo method produces an ensemble of calibrated parameter sets whose statistical properties are consistent with the observed data. The estimates of trends that can be derived from these simulations are therefore limited to changes in population parameters (e.g. changes in means rather than mean changes as derived from the site-specific method).

The application of MAGIC to a catchment requires data sets that describe atmospheric deposition, hydrology, soils physical and chemical characteristics and surface water chemistry and, if applicable, changes in land use and forestry practices within the catchment. In regional applications of the model using the site-specific method, these data are necessary for each catchment. If any one of the data sets is not available for a site, some means of estimating the required data must be devised. Usually this can be accomplished by spatially extrapolating from appropriate observations at other locations using criteria based on nearest distance, soil type, landscape type or other appropriate similarities. In regional applications using the Monte-Carlo method, the model also requires each type of data for each simulation. However, since there is no one-to-one correspondence between lakes and simulations, there is no need for a one-to-one assignment of soils data or deposition data or land use data, etc. Rather, the regional distributions (i.e. statistical descriptions) of all variables in the required data sets are used as inputs to the model, including any correlations that may be present among and within the data sets.

Here, both the site-specific and Monte-Carlo methods are used in regional applications of MAGIC to southernmost Norway. The model is applied to the regional water quality surveys in southern Norway for 1974, 1986 and 1995, and to the 83 lakes sampled in all three surveys. MAGIC is calibrated for the Monte-Carlo method using the 1974 data from the regional lake survey and for the site-specific method using the 1974 water chemistry values for the 83 'monitored' lakes. Both calibrated regional models are then used to predict the observed regional water chemistry in 1986 and 1995 as a test of the model utility. Both models are also used to forecast the regional response to observed and expected future reductions in acid deposition.

MONTE-CARLO REGIONAL SIMULATIONS

The regional calibration of MAGIC for southernmost Norway using the Monte-Carlo method is based on procedures and simulations of Cosby *et al.* (1989b) and Hornberger *et al.* (1989) in a previous calibration to the 1974 survey data. The ensemble of weighted model simulations derived from the joint distributions of inputs and parameters from this previous calibration is used as the starting point here. The Monte-Carlo parameter sets for MAGIC calibrated to the 1974 Norwegian lake survey and the calibrated regional model are used to simulate water chemistry distributions for 1986, 1995 and 2020 using observed and predicted deposition. These regional predictions are compared to both the observed data (when available) and to the regional predictions generated using the site-specific method.

Specification of regional data

The distributions of annual rainfall and runoff and soil and surface water temperatures for southern Norway were derived from a climate atlas for the region (Holtan, 1986). Data from several soil studies for the region were used to define allowable ranges (input distributions) for soil depth, bulk density, cation exchange capacity, and base saturation

(Lotse and Otabbong, 1985; Seip *et al.*, 1979a,b; Stuanes and Sveistrup, 1979; Frank 1980; Abrahamsen *et al.*, 1976).

The chemistry of precipitation in southern Norway can be depicted as a mixture of marine aerosols (Cl, Na, Mg, and to a lesser extent K, Ca, and SO_4) and pollutant species transported from distant sources (H, SO_4, NO_3, and NH_4) (Wright and Dovland, 1978). Dry deposition and impaction also deposit these components. The concentration of seasalt components in precipitation decreases sharply with distance from the coast, reflecting the rapid deposition of the relatively large marine particulates. The concentration of SO_4 in precipitation displays a gentler gradient reflecting lower deposition rates for gaseous and small particulate components. The input distributions of the deposition parameters used in the model represent this pattern. These input distributions were inferred directly from the lake survey data. Observed distributions of Cl, SO_4, NO_3 and NH_4 concentrations in the surveyed lakes (corrected for evapotranspiration by assuming a water yield of 80%) were used to set atmospheric deposition inputs of these ions to the model. Deposition of the base cations in 1974 was set assuming sea salt ratios (to Cl) of these ions in deposition and using the estimated 1974 Cl deposition. Historical deposition for the regional model was estimated using the procedure of Cosby *et al.* (1985b) which scales the estimated 1974 deposition inputs for the lakes to the temporal pattern of reconstructed emissions for northern Europe (Mylona, 1996). This scaled deposition history (Fig. 2) was used for SO_4, NO_3 and NH_4 ions. Deposition of all other ions was assumed to be constant.

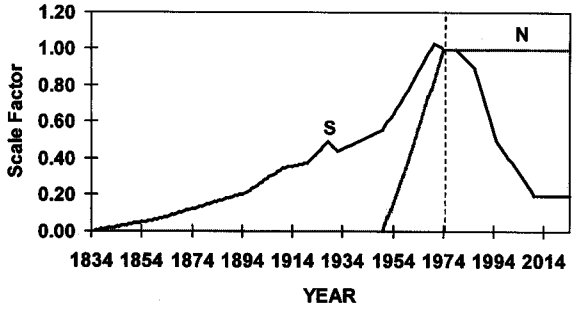

Fig. 2. *Deposition sequences (scaling factors relative to 1974) used in MAGIC calibrations. Hindcast N deposition scale factor was assumed equal to S scale factor for the Monte-Carlo method. See text for details.*

Calibration of the model

The model was implemented using yearly time steps and one soil layer. Soil and deposition parameter values were selected at random from their specified distributions. Simulations were run from 1834 to 1974. For each simulation, the simulated 1974 lake concentrations were compared with the range of concentrations for all major ions observed in the 1974 survey. If the simulated concentrations of all ions fell within their observed 'windows' (defined by the maximum and minimum of the observed data) the simulation was accepted; if one or more did not, the simulation was rejected. This sampling, simulation and evaluation procedure was repeated 3000 times of which 1176 'acceptable' simulations (fit observed windows) were obtained.

Weights for each simulation in the sample of 1176 were then selected such that the simulated joint distribution of alkalinity, Ca, and SO_4 matched the observed joint distribution of those ions (based on the 463 surveyed lakes). ANC, Ca and SO_4 were chosen as criterion variables because (1) they are important indices of soil water and surface water acidification and (2) they embody important correlations in the joint distribution of the total suite of ions measured. Each of the three axes (ANC, Ca^{2+}, and SO_4^{2-}) of the joint distribution of these variables was divided into six class intervals—giving a possible 216 histogram 'bins' in the joint distribution for the region. Not all joint histogram bins contained observed data. The weight for a given simulation was then calculated according to the bin to which it was assigned as

$$w = n_0/n_s \qquad (2)$$

where w is the weight, n_0 is the number of observed lakes assigned to a given bin, and n_s is the number of simulated lakes assigned to that bin. These weights are used in calculating the population characteristics (means, standard deviations, etc.) of the model simulations. Bins with simulations and no observations resulted in a weight of zero.

SITE-SPECIFIC REGIONAL SIMULATIONS

The regional application of MAGIC using the site-specific method involved calibrating MAGIC individually for each of the 83 lakes that were sampled in the three Norwegian Lake Surveys. The calibration method used was similar in most particulars to that used by Wright *et al.* (1992) and Frogner *et al.* (1994) in previous applications of MAGIC for mapping critical loads in Norway. The calibration procedure required soils, land use, lakewater and atmospheric deposition data for each lake. The requisite data was estimated for each lake by: (a) using observed lake chemistry in the calibration year to set atmospheric deposition; and (b) assigning soil properties by associating each catchment with soil properties determined from either forested or moorland soil sampling sites. Once calibrated, the parameter set for each lake was used to simulate water chemistry for that lake in 1986, 1995 and 2020 using the observed and expected time series of deposition inputs as described in the deposition scenario section (below). These regional predictions are compared to both the observed data (when available) and to the regional predictions generated using the Monte-Carlo method.

Specification of catchment data

For lakes with forested catchments (45 of the 83 lakes) soils data derived from the Norwegian Institute for Land Inventory (NIJOS) soils data bases were used. These data are from areas in productive spruce, pine and birch forests (Esser and Nyborg, 1992; Esser, 1994). One soil pit was located at the corner of each 9×9 km cell in a gridded network laid over Norway. The soil pit was dug to at least a 50-cm depth where possible. Soil depth, pH, bulk density, cation exchange capacity, and base saturation were measured for each soil horizon. Each lake was assigned soils data from the grid cell in which the lake was situated. The 38 lakes with heathland catchments used soils data derived from one of two sites: Vikedal on the west coast of Norway, or Storgama in the inland east of Norway. The soils data for Storgama and Vikedal come from the Norwegian Forest Research Institute (NISK) (A. Stuanes, pers. comm.), and were originally collected as part of the Norwegian Pollution Control Authority monitoring program for long-range transported air-pollutants. The data were aggregated by Reuss (1990) and have been previously used for site-specific applications of MAGIC (Wright *et al.*, 1990b).

Observed concentrations of Cl and non-marine SO_4 in the surveyed lakes were first corrected for evapotranspiration and then used to set atmospheric deposition inputs for the model. Lake Cl was assumed to be derived from sea salts and the 1974 observed lake Cl concentrations were used to set the 1974 atmospheric deposition of Cl, Na, Mg and K (using seasalt ratios of those base cations). Lake SO_4 concentrations were assumed to be in steady state with atmospheric inputs (i.e. it was assumed that SO_4 adsorption in soils was negligible). The 1974 observed lake SO_4 concentrations were used to specify 1974 atmospheric deposition of SO_4. The ratios of Ca, NO_3 and NH_4 each to SO_4 in deposition for the period 1988–1992 for seven sites in southernmost Norway (Tørseth and Pedersen, 1994), were used with the estimated 1974 SO_4 deposition at each lake to set 1974 deposition of Ca, NO_3 and NH_4 for each lake.

Historical deposition for the regional model was estimated using the procedure of Cosby *et al.* (1985b) which scaled the estimated 1974 deposition inputs for the lakes to the temporal pattern of reconstructed emissions for northern Europe. Different emissions patterns were used for sulphate (Mylona, 1996) and for NO_3 and NH_4 (Simpson *et al.*, 1997) (Fig. 2). All other ions in deposition were assumed to be constant throughout the simulation period.

Calibration of the model

Site-specific calibration proceeded through a number of steps. First, the values of certain parameters within the model were set using directly measured or estimated values (the 'fixed' parameters). The model was then run using observed or assumed atmospheric and hydrologic inputs. The simulated values of certain state variables (the 'criterion' lakewater and soil chemical variables) were compared to observed values of these variables for the calibration year. If the observed and simulated criterion values differed, the values of another set of parameters in the model (called 'optimized' parameters) were adjusted to improve the fit. After a number of iterations, the simulated-minus-observed values of the criterion variables usually converge to zero (within some specified tolerance). The model was then considered calibrated. If new assumptions (or values) for any of the fixed variables or inputs to the model are subsequently adopted, the model must be re-calibrated by re-adjusting the optimized parameters until the simulated-minus-observed values of the criterion variables again fall within the specified tolerance.

The model was calibrated to each of the 83 lakes using the 1974 lake data as criterion variables. The model was configured with one soil layer if total soil depth in the assigned soil profile was less than 0.5 metres. If the soil depth was greater than 0.5m, two soil layers were used in the model with the upper layer being 0.5 m thick and the remaining depth of the soil occurring in the simulated lower layer. For the site-specific application, a triprotic organic anion was added to soil in order to match observed soil solution pH. No organic acids were simulated in the Monte-Carlo approach. Soil pH data were not available at the time of the Monte-Carlo calibrations and so calibration of organic acids in soils was precluded.

Where forests exist within a given catchment net uptake of Ca, Mg and K from catchment soils was simulated for the site-specific method. The procedure for estimating the magnitudes of these uptake fluxes is given by Wright *et al.* (1992). Uptake of base cations by forests was not included in the Monte-Carlo simulations. Net uptake of base cations was assumed to be zero for heathland catchments.

FORECAST SCENARIOS

The calibrated models were used to predict regional lake water chemistry in 1986 and 1995 when regional lakewater chemistry data were available for comparison. Time sequences of atmospheric deposition were needed to drive the models for these forecasts.

For both regional approaches sea salt deposition for 1986 and 1995 was set using Cl concentrations observed in the lakes in those years. In the Monte-Carlo approach, the ratios of the regional mean Cl concentrations in 1986 and 1995 to the regional mean Cl concentration in 1974 were calculated. These ratios were multiplied with the 1974 deposition inputs of Na, Mg, K and Cl in each of the Monte-Carlo simulations to get deposition of those ions for 1986 and 1995, respectively. In the absence of other information the changes in sea salt deposition were assumed to be linear between observation years. In the site-specific approach, the calculations were done similarly except that the ratios to Cl concentrations in the lakes in

1986 and 1995 were calculated separately for each lake (and applied separately to each lake). For both approaches the deposition of these sea salt ions from 1995 to 2020 was set constant at the 1995 values.

Future SO_4 deposition was set using the average observed pattern of atmospheric deposition of SO_4 for seven stations in southern Norway from 1978 to 1995 (Tørseth, 1996). That sequence of deposition was normalized to the deposition in 1974 to produce a scaled sequence of SO_4 deposition from 1974 to 1995 for use in the model (Fig. 2). The scale factor for each year was multiplied by the 1974 deposition to obtain SO_4 deposition in that year for any simulation. Deposition of SO_4 from 1995 to 2010 was assumed to decline linearly from the 1995 value to 20% of the 1974 value. This simulated SO_4 deposition in 2010 (20% of 1974 deposition) is the expected deposition for southern Norway when the Oslo protocol for SO_2 emission reductions is implemented (Norwegian Meteorological Institute, unpubl.). Deposition of SO_4 from 2010 to 2020 was set constant at the 2010 level. The same scaled sequence for SO_4 deposition was used in both the site-specific and Monte-Carlo applications.

Deposition of NO_3 and NH_4 was assumed to increase by 27% from 1974 to 1980 and to remain constant at the 1980 level until 2020. The increase of 27% and the constant deposition from 1980 to 1995 are consistent with observed N deposition patterns for southern Norway (Simpson *et al.*, 1997). The same scenario of NO_3 and NH_4 deposition was used for all simulations in both regional approaches.

Rates of uptake of both NO_3 and NH_4 in the soils were calibrated in both regional approaches such that simulated agreed with observed concentrations in 1974. For the Monte-Carlo regional approach, it was assumed that both historical and future uptake of N occurred constantly at the 1974 rates. The uptake rates of NO_3 and NH_4 in 1986 and 1995 in the site-specific simulations were adjusted using observed lake NO_3 and NH_4 concentrations. Base cation uptake by forests (included in the appropriate site-specific catchment simulations) was assumed to remain constant for both historical and future periods.

Results and discussion

SIMULATED AND OBSERVED DISTRIBUTIONS FOR 1974

Examination of goodness-of-fit of a calibrated model to observed data is usually based on pairwise comparisons of simulated and observed values of variables for each lake modelled. This type of comparison is not possible for the Monte-Carlo regional approach and not desirable for the site-specific approach when the intent is to reproduce the regional characteristics of water quality. In these regional applications of MAGIC, therefore, an examination of goodness-of-fit of the calibrated models to the observed data is based on comparisons of the *distributions* of the observed and simulated variables. Results presented here will examine four characteristics of the distributions: the mean, standard deviation, maximum and minimum.

The calibration of MAGIC required the simultaneous simulation of the distributions of seven lake water chemistry variables: Ca, Mg, Na, K, Cl, SO_4 and NO_3. Both methods matched closely the observed distributions of each of these ions in the calibration year 1974 (Table 2). The statistics for the simulations generated by the site-specific method are nearly identical with the observed

Table 2. Minimum, mean, standard deviation and maximum of the observed (obs) and simulated (sim) distributions of water quality variables for the calibration year 1974. The variables listed are the criterion variables used in the calibration procedures. All concentrations are $\mu eq\ l^{-1}$.

		site-specific calibration			Monte-Carlo calibration		
		min	mean ± S.D.	max	min	mean ± S.D.	max
Ca	obs	11	40 ± 26	138	6	47 ± 28	144
	sim	11	40 ± 25	129	5	40 ± 25	132
Mg	obs	9	30 ± 17	72	7	38 ± 20	103
	sim	9	30 ± 17	72	5	39 ± 21	113
Na	obs	21	89 ± 61	270	13	107 ± 69	313
	sim	21	89 ± 60	247	20	115 ± 72	309
K	obs	2	6 ± 4	26	1	7 ± 4	27
	sim	2	6 ± 4	25	1	5 ± 3	22
Cl	obs	17	93 ± 69	260	14	122 ± 78	339
	sim	17	93 ± 69	260	11	121 ± 81	336
SO_4	obs	15	71 ± 35	144	10	88 ± 40	202
	sim	15	72 ± 35	144	12	86 ± 39	183
NO_3	obs	1	6 ± 4	19	0	7 ± 4	31
	sim	1	6 ± 4	19	0	9 ± 5	19

statistics for the monitored lakes (Table 2). While the differences between simulated and observed statistics for the Monte-Carlo application to the lake survey data are somewhat larger (Table 2), the simulated distributions are still acceptable. The larger differences arise because of the 'coarser' resolution of the bin-filling routine used in the Monte-Carlo calibration procedure compared to the more-or-less exact fits provided by the site-specific method.

Calibration to individual ions was necessary because of the mass-balance, process-based structure of MAGIC. The primary water chemistry variable of interest in examining lake response to acidic deposition, however, is the ANC of the water. From a regional perspective it is often sufficient to examine the values and trends in ANC, SBC and the SAA in order to gain understanding of responses to acidic deposition.

The use of different historical sequences of deposition of N compounds (Fig. 2) is a potential source of discrepancy between the results from the two methods. The Monte-Carlo procedure was based on the assumption that the historical deposition of NO_3 and NH_4 followed the same pattern as that of sulphate (Cosby *et al.*, 1989b; Hornberger *et al.*, 1989). At that time there was no independent estimate for historical N deposition in Norway. The site-specific simulations, on the other hand, used separate deposition sequences for N compounds, based on recent estimates by Simpson *et al.* (1997). The use of these different historical deposition sequences, however, probably has only a minor effect on the simulated ANC. For most of the lakes NO_3 (and NH_4) concentrations are low, and thus most of the incoming N is retained in the terrestrial catchment or lake itself. Both methods assume that the fraction of N retained in the past (and future) is the same as for the year 1986.

In systems that have significant inputs of seasalts (as do many of these lakes in southernmost Norway), the total cation and anion concentrations are relatively large. The ANC is then the difference between two relatively large numbers, and the effects of changing cations and anions on changes in ANC can be difficult to discern. It is convenient in such cases to examine 'non-marine' base cation (SBC*) and 'non-marine' acid anion (SAA*) concentrations. Non-marine ion concentrations are calculated by subtracting from the measured concentration of each ion in a sample of lakewater the amount of that ion potentially contributed by seasalt. The seasalt contribution of each ion is estimated using the ionic ratio of each ion to Cl in seawater and the measured concentration of Cl in the lakewater sample. There is good agreement among simulated and observed distributions of these defined variables for the calibration year for both methods of regional application of MAGIC (Table 2) (Fig. 3).

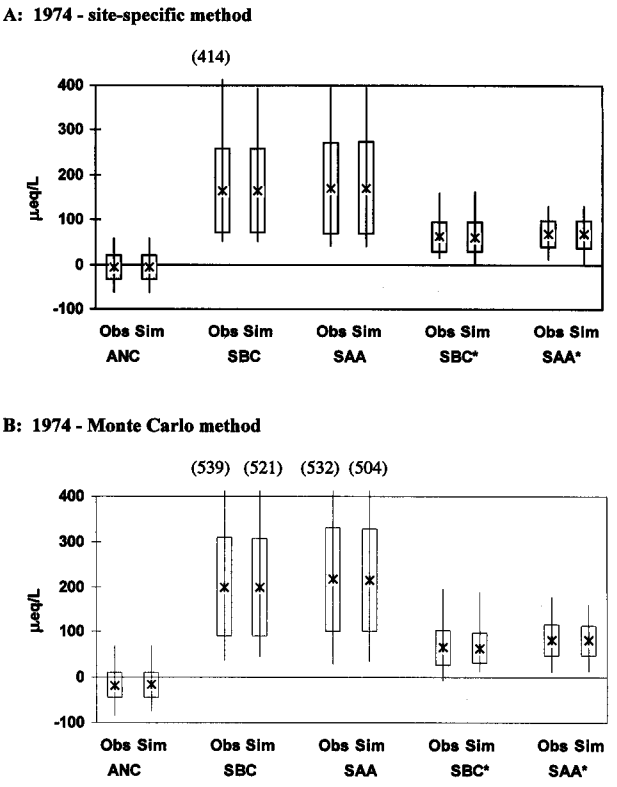

Fig. 3. *Simulated and observed distributions of variables in 1974 (the calibration year) for the two methods of regional application of MAGIC: A) the site-specific method; and B) the Monte-Carlo method. Variables are defined in the text. The x indicates means; boxes indicate one standard deviation, lines indicate maximums and minimums.*

SIMULATED AND OBSERVED DISTRIBUTIONS FOR 1986 AND 1995

The calibrated models were used to forecast the distributions of SBC, SAA and ANC for 1986 and 1995 using observed changes in deposition of S and N from 1974 to 1995. These forecast distributions were compared with the observed distributions for the site-specific method applied to the monitored lakes (Fig. 4) and for the Monte-Carlo method applied to the lake survey data (Fig. 5).

For both regionalisation methods, the simulated SBC and SAA distributions agree reasonably well. The differences between simulated and observed mean values of SBC and SAA are all relatively small compared to the magnitudes of the means. The errors in the simulated means are a small percentage of the mean values (less than 15% for both methods in both 1986 and 1995). Both simulation methods simulate the variances in SBC and SAA approximately correctly (Figs. 4 and 5). The site-specific method also simulates the maximums and minimums relatively precisely, coming within 10% or 5 µeq l^{-1} for both SBC and SAA in both years (Fig. 4). While the minimums simulated by the Monte-Carlo method are relatively pre-

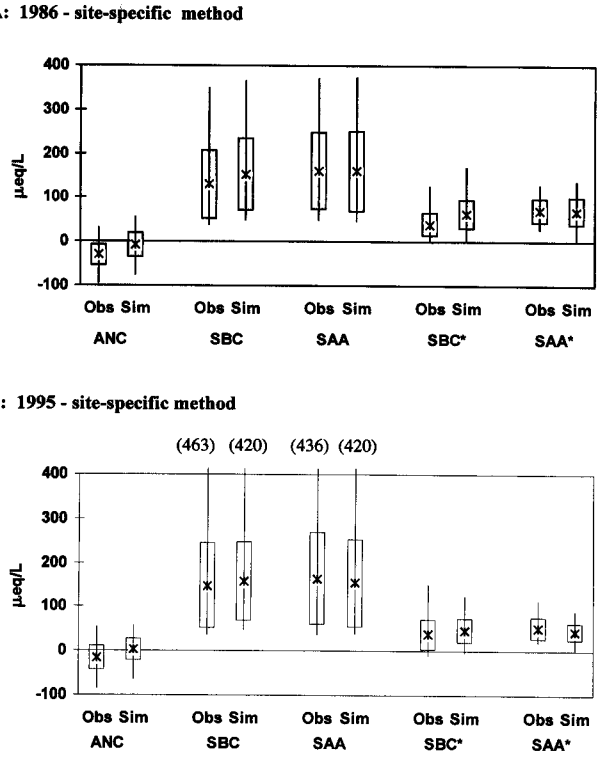

Fig. 4. *Observed distributions of variables from the monitored lakes compared with model forecasts of those distributions made using the site-specific regional application of MAGIC. A) 1986; and B) 1995. Variables are defined in the text. Means are indicated by x; boxes indicate one standard deviation; lines indicate maximums and minimums.*

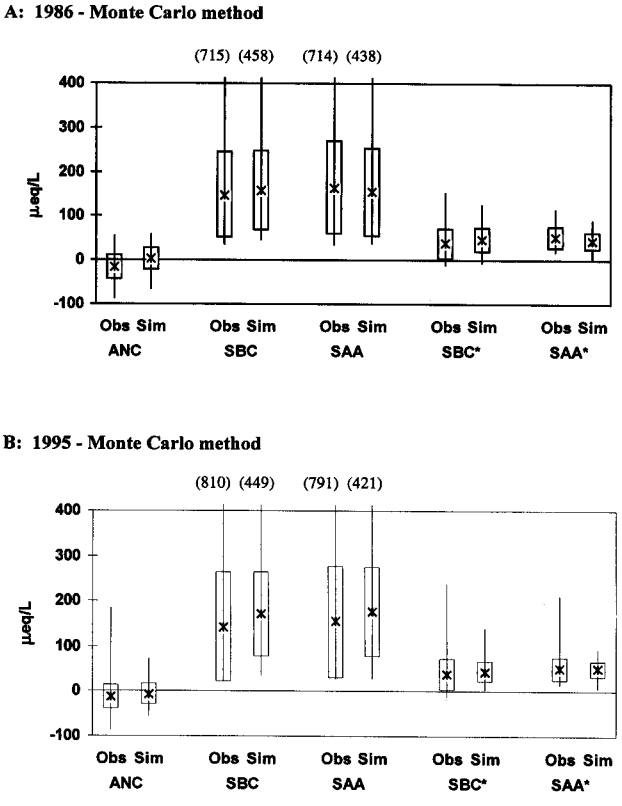

Fig. 5. *Observed distributions of variables from the regional lake surveys compared with model forecasts of those distributions made using the Monte-Carlo regional application of MAGIC. A) 1986; and B) 1995. Variables are defined in the text. Means are indicated by x; boxes indicate one standard deviation; lines indicate maxima and minima.*

cise, the maximums of both SBC and SAA are underestimated in both years (Fig. 5).

For both regional modelling methods there is a tendency to overestimate ANC in both 1986 and 1995 (Figs. 4 and 5). The variances and the ranges of values agree reasonably well with those observed. However, the mean and the entire simulated distribution of ANC are shifted upward relative to the observed in both years. The simulated ranges of ANC from the Monte-Carlo method, however, were smaller than the observed. The upward shifts (errors) in the simulated ANC distributions are larger for the site-specific method than for the Monte-Carlo method.

The differences between the simulated and observed ANC distributions can be separated into errors arising from incorrect simulation of the base cation component of ANC and errors arising from incorrect simulation of the acid anion component. For the site-specific method (Fig. 4) the overestimation of ANC in 1986 (relative to observed) was primarily due to an overestimation of non-marine base cation concentrations. In 1995, however, the site-specific method reproduces the non-marine base cation distribution reasonably well and the overestimation of ANC resulted from an underestimation of acid anion concentrations. For the Monte-Carlo method, the overestimation of ANC in both years appears to be a result of approximately equal amounts of overestimation of SBC* and underestimation of SAA*.

COMPARABILITY OF OBSERVED DATA SETS

Some of the difference between modelled and observed frequency distributions can be ascribed to variations in the observed data sets. The monitored lakes are a subset of the lake surveys in all three years, but were not explicitly chosen to be a statistically representative subset of the survey lakes. The comparisons of the regional models each to its own observed data set are no better than the comparisons of the two observed data sets to each other, provided that the two observed data sets had the same statistical properties in 1974, the year in which the models were calibrated to the data.

The SBC and SAA concentrations of the monitored lakes have smaller variances, a smaller range and less skew (because of much lower maxima) than do SBC and SAA concentrations from all the lakes surveyed in all three years (Fig. 6). In 1986 and 1995 the means of SBC and SAA are lower in the monitored lakes, but in 1995 they are slightly higher than the means for SBC and SAA from the lake surveys. The same patterns are apparent but less

pronounced for the SBC* and SAA* concentrations (Fig. 6) with the exception of SBC* in the monitored lakes in 1974 which is equivalent to that in the lake surveys. The result of these patterns in SBC and SAA produces a shifting pattern of ANC comparisons for the two data sets. In 1974 the monitored lakes are less acidic (higher ANC) than the survey lakes (Fig. 6A). By 1986, this has reversed and the monitored lakes are more acidic than the survey lakes (Fig. 6B). In 1995, the distributions of ANC of the two data sets are nearly identical (except for the differences in skew resulting from a much higher maximum in the lake survey data).

These results (Figs. 3, 4, 5 and 6) indicate that simulated and observed distributions agree more closely (for both modelling methods) than observed distributions agree with each other in 1974. This is not unexpected and indicates that the calibration 'errors' (differences in calibrated and observed distributions) are smaller than the 'noise' in the data (the differences between two observed data sets for the calibration year). The results (Figs. 3, 4, 5 and 6) also indicate that differences between simulated and observed forecast distributions (model success or reliability for forecasts) are of the same order as differences between the two observed distributions. Both modelling methods are comparable in prediction of future distributions, as do the two sets of observations in predicting each other.

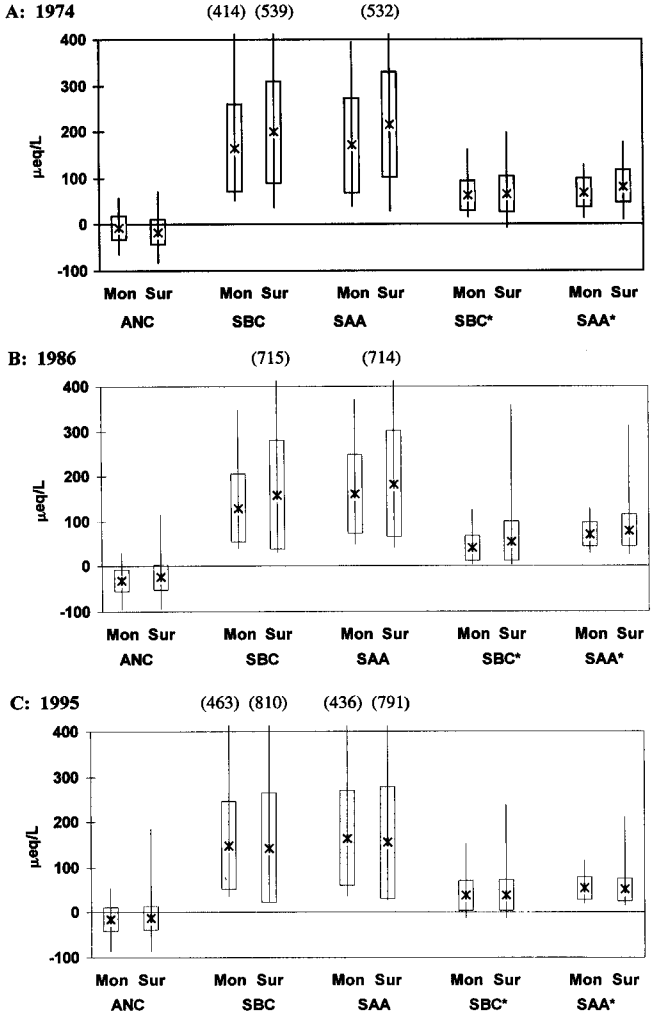

Fig. 6. *Observed distributions of variables for the monitored lakes (Mon) and the lake surveys (Sur). Variables are defined in the text. Means are indicated by x; boxes indicate ± one standard deviation; lines indicate maximums and minimums.*

TRENDS IN ANC FROM 1974 TO 1995

In southernmost Norway there has been a decrease of approximately 50% in deposition of S from 1974 to 1995 (Fig. 2). The effects of this deposition reduction on lake water chemistry has been examined using the forecast simulations as well as the survey data sets to examine trends in water chemistry over the 21 years from 1974 to 1995.

The two modelling approaches produce similar trends in mean ANC over the 21 years (Fig. 7A). The simulated ANC means in 1974 differ because of differences in the data sets to which the models were calibrated (Fig. 7B). The changes in simulated mean ANC from 1974 to 1986 and from 1986 to 1995, however, are essentially the same for each modelling approach (± 2 µeq l^{-1} change from 1974 to 1986; an increase of 9–10 µeq l^{-1} from 1986 to 1995). The simulated response of essentially no change from 1974 to 1986 corresponds to the initial period of only 10% deposition reduction (Fig. 2). The increases in simulated ANC from 1986 to 1995 occurred in response to the additional 40% deposition reduction (Fig. 2).

The two observed data sets show trends that are different from the modelled trends and different from each other in magnitude of response (Fig. 7B). Both data sets indicate that mean ANC of lakes in southern Norway decreased during the period 1974 to 1986 even though acidic deposition also declined slightly during that period. The decrease in mean ANC was greater for the monitored lakes (a loss of 25 µeq l^{-1}) than for the lake surveys (a loss of 6 µeq l^{-1}). For the period from 1986 to 1995 both data sets showed recovery of ANC (Fig. 7B) in response to the 40% reduction in acidic deposition. The mean ANC of the monitored lakes increased by 11 µeq l^{-1} and the mean ANC of the lake surveys rose by 16 µeq l^{-1}.

Both modelling approaches failed to produce the observed downward trend in ANC from 1974 to 1986. The period from 1974 to 1986 was a period of relatively little change in deposition. The models (based on mean hydrological and climatic inputs that do not vary from year-to-year) responded to essentially no change in deposition by simulating essentially no change in lake ANC.

The acidification observed in the lakes may have resulted from a number of causes. Year-to-year variability in acid deposition, sea-salt deposition, rainfall amount and snowmelt dynamics can have pronounced effects on lake

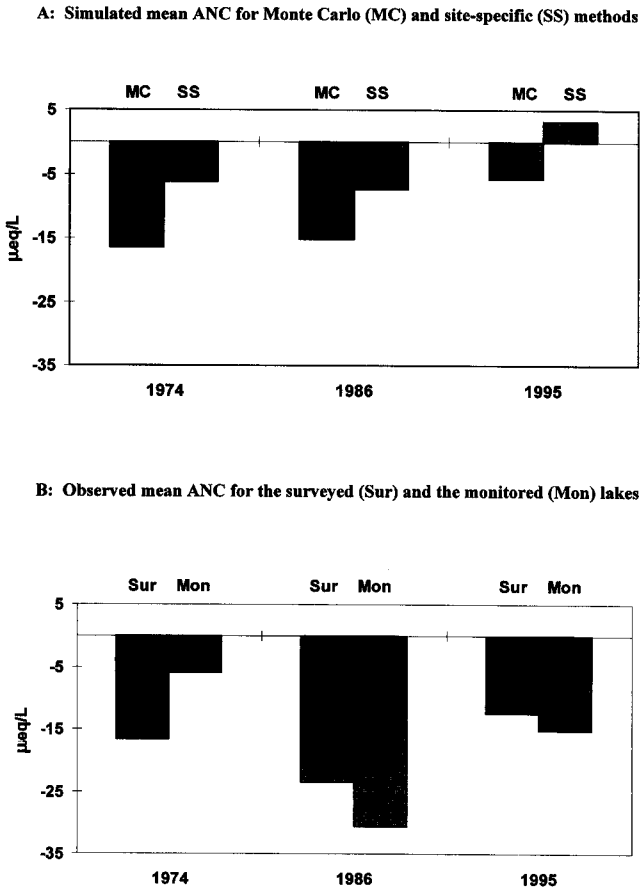

Fig. 7. *Mean ANC values in three years for: A) simulations using the Monte-Carlo and site-specific methods for regional application of MAGIC; and B) observations from the lake surveys and the monitored lakes.*

chemistry in any given year. Either 1974 or 1986 could have been an 'outlier' year, and the trend inferred from the two sampling times may thus have been exaggerated (or subdued). The small but finite turnover time in the lakes could produce a delayed response to changes in deposition and catchment runoff.

Still unexplained is the large decline in ANC in the monitored lakes (25 µeq l^{-1}) relative to the lake surveys (6 µeq l^{-1}) between 1974 and 1986. The mean ANC of the monitored lakes is consistently lower than the mean ANC of the lake surveys in 1986 and 1995, but not in 1974 (Fig. 7B). The 1974 data from the monitored lakes may contain an anomaly. Changes in non-marine Na concentrations in the monitored lakes between 1974 and 1986 can explain a large part of the difference in the change in ANC relative to that inferred from the lake surveys. The changes in mean Ca*, Mg* and K* from 1974 to 1986 are very similar for the monitored lakes and the lake surveys (Fig. 8A). The lake surveys show no change in mean Na* from 1974 to 1986, while the change in mean Na* for the monitored lakes (a loss of 9 µeq l^{-1}) is approximately half of the difference in decrease in ANC inferred from the two data sets.

A possible explanation for the discrepancies in Na* changes could be that measured Na concentrations were too high in 1974 or too low in 1986 in the monitored lakes. The mean value of the Na/Cl ratio for all samples in each data set for each year (Fig. 8B) indicates that the measured Na values in 1974 may have been biased toward high values. In all other years for the monitored lakes and for all years in the lake surveys, the average Na/Cl ratio is near 0.87, the sea salt value. A reduction of 9 µeq l^{-1} in the Na concentration of each sample in the 1974 monitored lakes data set would: a) remove the discrepancy in change in Na* between the monitored lakes and the survey lakes from 1974 to 1986; b) result in a mean Na/Cl ratio for the monitored lakes in 1974 of .87; and c) reduce the apparent decline of mean ANC of the monitored lakes between 1974 and 1986 from 25 µeq l^{-1} to 16 µeq l^{-1}. A bias of 9 µeq l^{-1} in measured Na for 1974 is equal to 10% of the mean Na concentration (Table 2).

The observed data thus indicate that, despite a decline of approximately 10% in acidic deposition from 1974 to

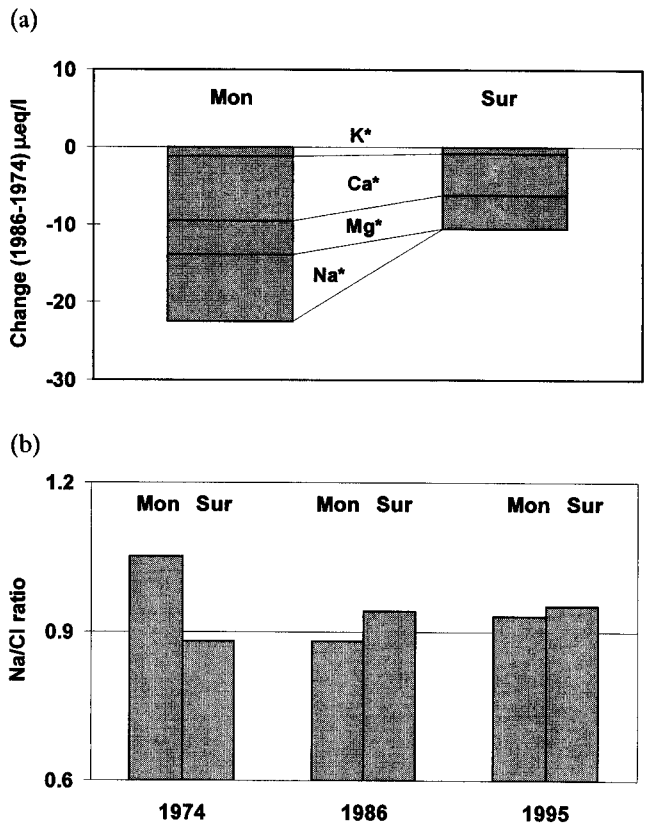

Fig. 8. *(a) Changes in mean values of non-marine base cation concentrations for the monitored lakes (Mon) and the survey lakes (Sur) from 1974 to 1986. (b) The mean of the Na/Cl ratios for samples from the monitored lakes (Mon) and for samples from the survey lakes (Sur) for three years.*

1986, the mean ANC of lakes in southern Norway declined by approximately 6 μeq l^{-1} (with a cautious upper limit for the decline of 16 μeq l^{-1}). Both modelling approaches simulated no change or a very small increase in mean ANC for the period. The observed data indicate that, in response to an approximate 40% decline in deposition from 1986 to 1995, the mean ANC of lakes in southern Norway increased by 11 to 16 μeq l^{-1}. Both modelling approaches simulated an increase of 9–10 μeq l^{-1} for the same period.

FORECASTS BEYOND CURRENT OBSERVATIONS

The two regional calibrations of MAGIC were then used to predict the water chemistry of the lakes in southern Norway in the year 2020. The deposition scenario for this forecast was taken as the expected deposition reduction that will occur in southernmost Norway after 1995 in response to S emission controls implemented in Europe as part of the Oslo protocol. The target date for the complete implementation of controls is 2010. The forecasts here have been run until 2020 to allow some additional time for delayed responses to be simulated.

Relative to the 1974 level deposition of SO_4 was reduced by 10% from 1974 to 1986, by an additional 40% from 1986 to 1995, and by yet another 40% from 1995 to 2010, with constant deposition at the final low level (equal to 20% of 1974 level) from 2010 to 2020. Deposition of both NO_3 and NH_4 were increased by 27% (relative to 1974 levels) from 1974 to 1980 and were maintained constant at the higher level from 1980 to 2020.

The site-specific method simulates a larger recovery from 1995 to 2020 than does the Monte-Carlo method (Fig. 9). The higher distribution of simulated ANC from the site-specific method in 2020 is not simply the result of differences in ANC in the calibration year, 1974 (the site-specific method was calibrated to data that had a mean ANC 9 μeq l^{-1} higher than the data used to calibrate the Monte-Carlo method). The mean ANC simulated in 2020 by the site-specific method is 31 μeq l^{-1} compared with a mean 2020 ANC of 11 μeq l^{-1} simulated by the Monte-Carlo method.

The differences in simulated recovery of ANC produced by the two methods probably reflect the differences in soils data used in the two methods for calibrating the model. The lake water chemistry data used in each method were very similar. The same methods were used to estimate deposition in the calibration year for each method and the same deposition histories were used for both methods. There was, however, a difference in the quality and quantity of soils data used in the two methods. When the original calibrations of the Monte-Carlo method were performed, there were only a few published data on soils in southern Norway for use in model calibration. The site-specific calibration performed for this analysis used soil data bases that were much more extensive. The additional

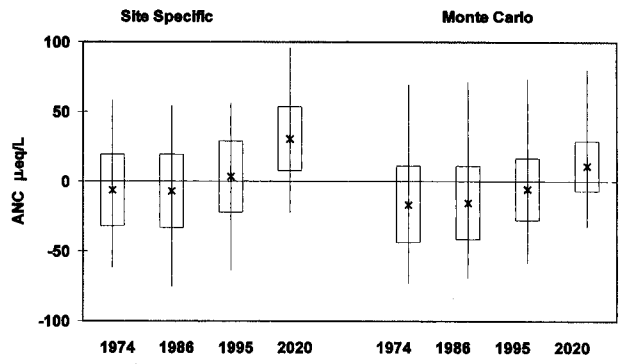

Fig. 9. *Distributions of simulated ANC in lakes in southernmost Norway for the calibration year (1974) and for three forecast years. The distributions are given for both the site-specific and the Monte-Carlo methods used for regional application of MAGIC. The same deposition scenario was used for both methods (see text).*

soils data used for the site-specific application (including the one-to-one mapping of soils to lakes) should have resulted in tighter constraints on model calibration. The use of more and better soil data in the site-specific method probably justifies a greater confidence in the forecast results.

Regardless of differences in magnitude of recovery, however, both methods predict that better than two-thirds of the lakes in southernmost Norway will have positive ANC values within 10 years of the realization of the deposition reductions expected from the Oslo protocol (Fig. 9). This is a marked improvement over the situation in 1974 (or even in 1995) when >65% of lakes in southernmost Norway were acidic.

Concluding remarks

Two methods for modelling regional responses of lake water chemistry to changes in acidic deposition are both based upon the MAGIC model but differ in their modes of regional application. The two approaches agreed closely and were generally consistent with the observed data. Both methods reproduced the statistical characteristics of the calibration data sets satisfactorily. Both methods reliably simulated distributions of water chemistry variables beyond the calibration year with reproducibility at least as good as that inherent in a comparison of the two observed data sets for the same year. The site-specific method performed better at simulating the 'tails' of the distributions, particularly the maxima.

While both methods of application of MAGIC for regional assessment were successful, the site-specific method should be used when data are sufficient. The tighter constraints imposed by site-specific model calibration result in greater fidelity in reproducing the distributions of water chemistry variables. The maintenance of a one-to-one correspondence between simulated and observed data means that simulation results can be mapped

for a geographically explicit presentation of model results. The ability to examine geographic patterns of response is becoming increasingly important in regional assessments.

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