

Modelling the hydro-geochemistry of acid-sensitive catchments in Finland under atmospheric deposition and biomass harvesting scenarios

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Abstract The dynamic hydro-geochemical Model of Acidification of Groundwater in Catchments (MAGIC) was used to predict the response of 163 Finnish lake catchments to historic and future atmospheric deposition (1880–2100) and future tree harvesting practices. Deposition was assumed to follow current legislated European emission reduction policies (CLE) and a scenario based on maximum (technically) feasible reductions (MFR). Future harvesting was assumed to shift from stem-only harvesting (SOH) to whole-tree harvesting (WTH) owing to the potential increased utilisation of biofuels.

Despite the modest changes in atmospheric deposition under CLE (compared to current day), these reductions are predicted to halt the decline in soil base saturation; however, further reductions are required to improve soil and lake water chemistry. The MFR scenario predicted a significant long-term improvement in soil base saturation leading to continued long-term recovery in surface waters (all lakes with ANC > 0 by 2100). However under the WTH scenario, significant long-term impacts (re-acidification) were predicted for soil and surface water chemistry (14 lakes with ANC < 0 by 2100). To some extent the long-term negative impacts were reduced under MFR, indicating that increased utilisation of biofuels will necessitate ‘trading emissions for timber’, or soil amendment, to maintain ecosystem quality and sustainable forest growth. The current practice of SOH is close to the sustainable maximum harvesting under current (legislated) atmospheric deposition in Finland.

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Introduction

In Finland there has been a substantial decrease in atmospheric sulphate (SO_4^{2-}) deposition (30% in northern and up to 60% in southern Finland) since the late 1980s (Kulmala et al. 1998; Vuorenmaa 2004)

owing to large reductions in sulphur (S) emissions in both Finland and other European countries (EMEP 2004). The protocols to the Convention on Long-range Transboundary Air Pollution (LRTAP) of the United Nations Economic Commission for Europe (UNECE) and legislation of the European Union (EU) have been the key international instruments driving this development. Reductions in acidifying depositions (primarily S) have resulted in large-scale recovery of lakes and streams from acidification across Europe and North America (e.g., Stoddard et al. 1999, 2003; Skjelkvåle et al. 2003, 2005). In concert, surface waters in Finland have shown (chemical) recovery from acidification. Mannio (2001) detected a statistically significant decline in SO_4^{2-} concentrations in 60–80% of the lakes in the Finnish Regional Monitoring of Lake Acidification network (RMLA; $n = 163$), depending on region. Forsius et al. (2003) estimated that some 1,400 lakes of size 4–100 ha (27% of Finnish headwaters) have shown an increase in alkalinity during the 1990s. The first signs of a (biological) recovery of perch (*Perca fluviatilis*) populations have also been recorded (Rask et al. 2001).

Further reductions in acidifying emissions are expected to result in continued recovery of acid sensitive ecosystems. It is anticipated that full implementation of the 1999 Gothenburg Protocol to the LRTAP Convention will cut European S emissions by at least 63%, nitrogen (N) oxides by 41% and ammonia by 17% by the year 2010, compared to 1990. However, there are a number of (confounding) factors, such as climate and land-use change, that can influence the extent of chemical recovery from acidification (Beier et al. 2003; Larssen 2005; Aherne et al. 2004, 2006). The role of climate change in the recovery of acidified surface waters has recently received considerable attention (e.g., see special issue in Hydrology and Earth System Sciences 12, 2008, Posch et al. 2008). Concomitantly, in response to environmental concerns, the use of biomass energy has become an important mitigation strategy against climate change, e.g., the EU have set a target of doubling the share of renewables in gross inland energy consumption to 12% by 2010 (European Commission 1996). The use of biomass in Finland has steadily increased over the past 25 years; forest biomass is now an integral part of modern energy systems, e.g., biomass accounts for 7.9% of the fuel

mix in district heating (Ericsson et al. 2004). The utilisation of biofuels in Finland will be further increased over the next few years. The most significant part of this growth will be based on the use of forest fuels, produced from logging residues (Ranta 2005). Accordingly, forest harvesting practices are expected to shift from more traditional stem-only harvesting (SOH) practices to whole-tree harvesting (WTH). However, long-term mass balance studies have shown that WTH may lead to base cation deficits in forest mineral soils in southern, central and north-eastern Finland (Joki-Heiskala et al. 2003).

Dynamic process-oriented models can be employed to evaluate the future (chemical) response of soils and surface waters to changes in deposition and land-use. Several dynamic (hydro-chemical) models, such as Model of Acidification of Groundwater in Catchments (MAGIC: Cosby et al. 1985, 2001), Soil Acidification in Forest Ecosystems (SAFE: Warfvinge et al. 1993) and Simulation Model of Acidification's Regional Trends (SMART: De Vries et al. 1989), have been extensively applied at site-specific and regional scales to predict changes in soil and surface water chemistry (e.g., De Vries et al. 1994; Alveteg et al. 1995; Aherne et al. 2003; Wright et al. 2005). Recently, dynamic modelling has also become an important part of the effects-oriented work under the LRTAP Convention (Hettelingh et al. 2007). In Finland, SMART has been used to model the response of soils and surface waters at site-specific (Iivonen et al. 1993; Kämäri et al. 1995, 1998; Forsius et al. 1997; Holmberg et al. 2000) and regional scales (Iivonen and Kenttämies 1994; Bilaltdin et al. 2001; Posch et al. 2003). However, few modelling studies have included the influence of forest harvesting strategies on soil hydro-geochemistry (the exceptions being Forsius et al. 1997; Holmberg et al. 2000) and none on surface waters. In the 'hydro-geochemical modelling community', the response of surface waters has typically been addressed using MAGIC; however, to-date MAGIC has been applied to very few sites (primarily as part of SMART, SAFE and MAGIC model comparisons) in Finland (Warfvinge et al. 1992; Forsius et al. 1998; Jenkins et al. 2003; Wright et al. 2006).

This paper presents the calibration of MAGIC to the 163 RMLA lake catchments in Finland using extensive data-sets on lake chemistry, catchment characteristics, soil properties, atmospheric deposition and nutrient

uptake by forests. The objective of the study was to evaluate the impacts of future emission reduction policies and tree harvesting practices on soil and surface water chemistry. A secondary objective was to provide a detailed analysis of the model calibration process, the regional model inputs and their required data processing. Future deposition was assumed to follow current European emission reduction policies and a scenario based on maximum technologically feasible reductions (Amann et al. 2005). It is anticipated that during the coming decades the increased demand for biomass for energy production will lead to changes in forest harvesting; forest harvesting practices was assumed to follow current SOH and a maximum removal scenario based on WTH.

Materials and methods

Study sites

The acidification monitoring network (RMLA), maintained by the Finnish Environment Institute (SYKE), consists of 163 lakes located throughout Finland (Fig. 1). The lakes were subjectively chosen by expert judgement for use in national acidification

studies. They were initially surveyed for chemistry during 1987 and have been monitored regularly since 1990. Since its establishment the network has been the focus of several investigations, including studies on trace metals (Mannio et al. 1995; Tarvainen et al. 1997), fish populations (Rask et al. 1995a, b) and chemical trends (Mannio and Vuorenmaa 1995; Forsius et al. 2003; Vuorenmaa and Forsius 2008). The lakes are predominantly small (median area = 10 ha) headwater or seepage lakes (Table 1). Catchment characteristics were determined from topographic maps (1:20,000). The terrestrial land cover is dominated by forests (median = 75%) and peatlands (median = 15%); the catchments are relatively undisturbed (only 7 lakes have catchments with >3% agricultural fields). All the lakes are acid-sensitive with low base cation concentrations, low alkalinity and sometimes elevated labile aluminium (Al^{3+}) concentrations (Mannio 2001; Forsius et al. 2003; Table 1). Given their wide geographic distribution, the long-term climate normals (1961–1990) vary widely with long-term annual precipitation ranging between 0.48 and 0.70 m, temperature between -2.3 and $+4.4^\circ\text{C}$, and hydrologic runoff between 0.24 and 0.42 m for the study catchments (Table 1 and section “Climate and runoff data”).

Fig. 1 Location of the 163 lakes in the Finnish lake acidification monitoring network (RMLA) included in this study (left), stratified according to the 1990–1994 average observed lake ANC, and location of the 441 soil sites used (right) with percent base saturation ranges

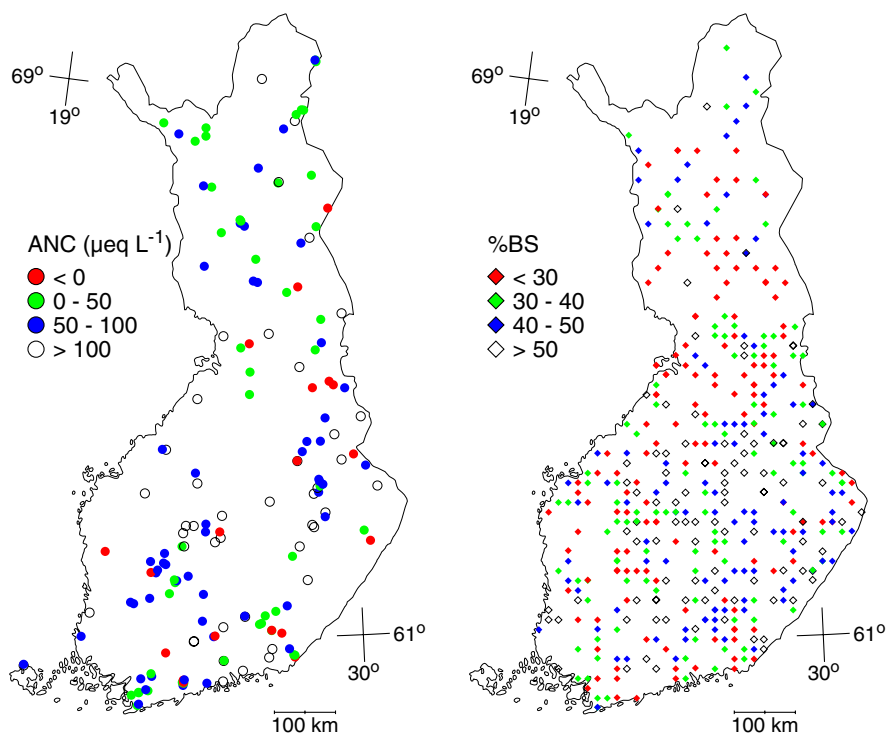


Table 1 Statistical summaries (5th percentile, median and 95th percentile) for catchment characteristics, catchment-average soil properties, long-term net stem-only removal inforest biomass, climate variables and annual average chemistry (2000–2004) for the Finnish lake acidification monitoring network (RMLA) lakes ($n = 163$; see Fig. 1 for locations)

Variable	Unit	5 Percentile	Median	95 Percentile
Catchment area	ha	21.3	108.6	797.2
Forest area	% Terrestrial area	35.1	74.7	97.0
Peatland area	% Terrestrial area	0.4	14.7	60.6
Soil depth ^a	m	0.30	0.54	0.68
Lumped soil bulk density ^a	kg m ⁻³	711.5	1056.4	1317.9
Cation exchange capacity ^b	meq kg ⁻¹	9.00	19.20	41.12
Soil base saturation ^a	% CEC	23.18	37.15	57.50
Exchangeable calcium ^a	% CEC	16.55	25.62	37.33
Exchangeable magnesium ^a	% CEC	4.16	7.46	13.78
Exchangeable sodium ^a	% CEC	0.71	1.24	1.76
Exchangeable potassium ^a	% CEC	1.76	2.83	4.63
Net base cation uptake	meq m ⁻² a ⁻¹	0.89	6.38	14.57
Net nitrogen uptake	meq m ⁻² a ⁻¹	0.51	5.26	11.19
Lake area	ha	2.3	10.0	137.3
Maximum lake depth	m	1.61	5.4	20.35
Retention time	year	0.08	0.88	6.05
Catchment runoff	m	0.243	0.331	0.420
Precipitation	m	0.482	0.608	0.701
Temperature	°C	-2.3	2.2	4.4
Calcium	μeq l ⁻¹	11.00	63.87	143.21
Magnesium	μeq l ⁻¹	8.23	39.49	80.09
Sodium	μeq l ⁻¹	14.94	47.85	106.85
Potassium	μeq l ⁻¹	2.30	8.70	18.71
Ammonium	μeq l ⁻¹	0.20	0.64	3.13
Sulphate	μeq l ⁻¹	13.10	45.82	118.64
Chloride	μeq l ⁻¹	9.03	21.44	81.24
Nitrate	μeq l ⁻¹	0.11	0.61	2.62
Acid neutralising capacity ^c	μeq l ⁻¹	0.84	78.97	181.72
pH	pH units	4.87	5.84	6.77
Labile aluminium	μeq l ⁻¹	0.00	1.00	5.27
Dissolved organic carbon	mg l ⁻¹	2.23	7.33	21.36
Gran alkalinity	μeq l ⁻¹	-8.50	30.80	104.92

^a Catchment soil characteristics are summarised from the closest permanent sampling plots in the 8th national forest inventory^b Effective cation exchange capacity^c Acid neutralising capacity estimated as the sum of base cations minus the sum of strong acid anions (see Eq. 1)

Historic and future soil and surface water chemistry (1880–2100) was simulated for each lake catchment using MAGIC. The required model inputs were obtained from site-specific observations and data interpolated from regional assessments. Considerable effort was placed on generating site-specific model inputs for each study catchment.

Model description

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. The model was first described by Cosby et al. (1985) and developments are reviewed

in Cosby et al. (2001). The model predicts monthly and annual average concentrations of the major ions for soil solution and surface water chemistry. MAGIC has been in use for >20 years and has been applied extensively in North America and Europe (see Appendix I in Cosby et al. 2001). Several refinements or additions to MAGIC have been proposed or implemented through the years as a result of the many applications of the model. These changes address inadequacies in the original structure and incorporate new processes that research has indicated to be of increasing importance in natural systems (Sullivan et al. 1996; Sullivan and Cosby 1998; Wright et al. 1993; Aherne et al. 2004). The latest version of MAGIC (7.77ext; 07 December 2006) was used in the current study.

MAGIC includes short-term processes described by equilibrium reactions that regulate ionic balance, and long-term input-output processes that control the catchment mass balance. These processes include weathering of minerals, adsorption and exchange of base cations and Al^{3+} by soils, anion retention by soils, buffering of soil solution pH by weak organic and inorganic acids and biologically mediated uptake of cations and anions. The equilibrium equations are represented by inorganic Al^{3+} reactions for surface and soil water, cation exchange reactions, and inorganic carbon equations that describe the chemical change that occurs as soil water enters the stream channel. The rates of change for each ion in soil water and surface water are calculated through the mass balance equations. Collectively, the mass balance equations describe the input-output relationships for base cations and strong acid anions. Soil and surface water organic acidity is represented by a triprotic pH model (Driscoll et al. 1994). Additional details describing the model structure are available in Cosby et al. (1985) and Cosby et al. (2001).

MAGIC represents the catchment with aggregated, uniform soil compartments (up to three in the current version), and a surface water compartment that can be either a lake or a stream. Time series inputs to the model include: deposition of ions from the atmosphere (wet plus dry deposition); discharge volume and flow routing within the catchment; biological production (nutrient uptake), removal and transformation of ions; internal sources and sinks of ions from weathering or precipitation reactions; and climate data. Constant parameters in the model include physical and chemical

characteristics of the soils and surface waters, and thermodynamic constants. Soil base cation weathering rates and initial base cation exchangeable fractions are calibrated using observed (or ‘target’) values of surface water and soil chemistry for a specified period. In this instance calibration refers to an automated optimisation procedure that is a component of the MAGIC model suite (MAGICOPT), generally used for regional applications (e.g., see Clair et al. (2007) for regional calibration of 400+ lakes). The minimum required site-measured target variables are: surface water concentrations for the major ions and soil exchangeable fractions for base cations, calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^{+}) and potassium (K^{+}).

In the current study all catchments were represented by one soil compartment receiving deposition and releasing discharge to the lake compartment. The soil compartment represented the aggregated (lumped) horizons of the catchment soils, i.e., mineral and organic soils were not segregated within the modelling structure but instead lumped into one soil compartment. Simulations were carried out using an annual time-step, with a number of simplifying assumptions applied consistently across all study lakes. Discharge volume and flow routing within the catchments were not varied during the calibration (and simulation) period; discharge was described using long-term means (see section “Climate and runoff data”) with 100% routed to the lake. Simulations were carried out between 1880 and 2000 (hindcast) and 2000–2100 (forecast). Detailed process-oriented N dynamics were not modelled; instead a fixed catchment-specific (percent) retention was estimated from input to output observations. Essential data for the regional application are time-series of annual atmospheric deposition (wet plus dry), biological production and removal (forest harvesting), physical and chemical characteristics of the soils and lakes, and observed soil and lake chemistry (calibration targets) for each study lake (see Table 1).

Data sources

Lakes

Chemical data for the RMLA lakes (Fig. 1) have been sampled (at least) annually since 1990. A sample of water from each lake was taken either from

the middle of the lake (1 m depth) or at the outlet each autumn during the thermal overturn phase (from early September in the north to mid November in the south). Samples were analysed for Ca^{2+} , Mg^{2+} , K^+ , Na^+ , chloride (Cl^-), SO_4^{2-} , nitrate (NO_3^-), ammonium (NH_4^+), Al^{3+} , dissolved organic carbon (DOC), pH and Gran alkalinity (Table 1) according to standardised methods (e.g., Forsius et al. 1990; Vuorenmaa and Forsius 2008). Charge balance acid neutralizing capacity (ANC) was calculated as the sum of base cations minus the sum of acid anions:

$$\text{ANC} = (\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+ + \text{Na}^+ + \text{NH}_4^+) - (\text{SO}_4^{2-} + \text{NO}_3^- + \text{Cl}^-) \quad (1)$$

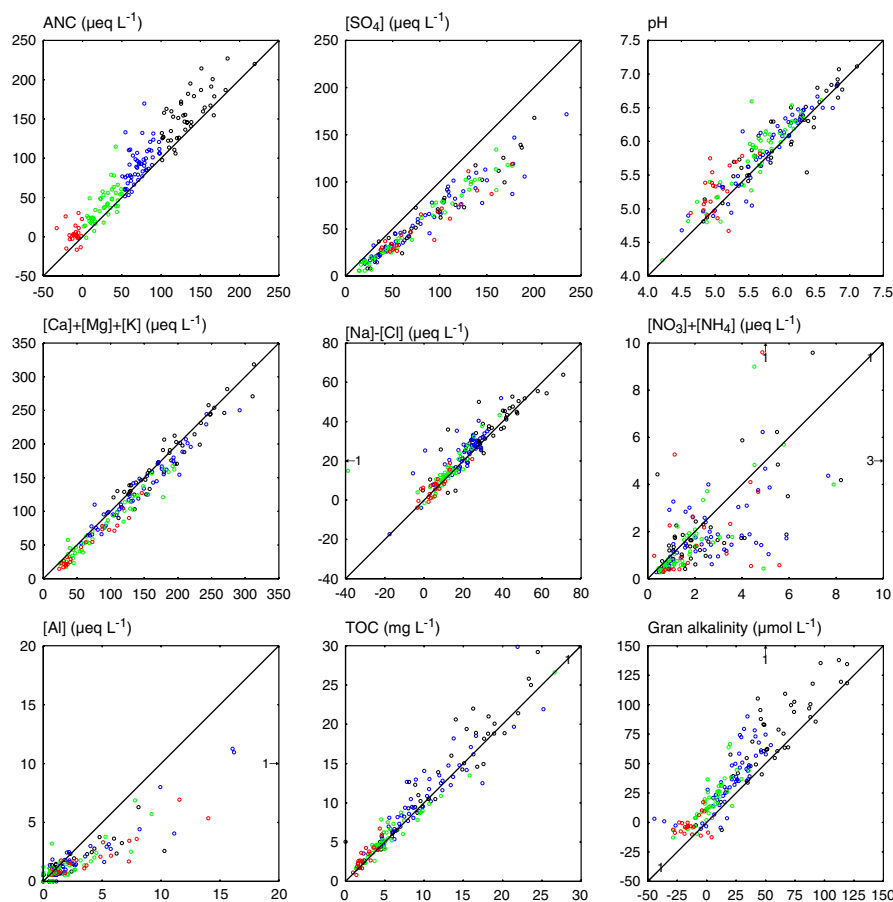
The 5-year period 1990–1994 was used to describe observed annual average lake concentration data (targets) for model calibration (with the calibration year = 1992). A second 5-year period (2000–2004: Table 1) was used to further compare simulated and observed data (a post-calibration check). During this 10 year period lake chemistry has changed

significantly (Fig. 2). The lake concentrations of ANC, pH, alkalinity, and to some extent DOC, have (visually) increased; whereas SO_4^{2-} , base cations and Al^{3+} decreased (Fig. 2). These changes are consistent with trends observed in surface waters across Europe (see special issue in Hydrology and Earth System Sciences 5, 2001) and have mostly been attributed to changes in S emissions. Median pH for the study lakes is 5.8, Al^{3+} is $1.0 \mu\text{eq l}^{-1}$, Gran alkalinity is $30.8 \mu\text{eq l}^{-1}$, DOC is 7.3 mg l^{-1} and base cations and SO_4^{2-} are 162.8 and $45.8 \mu\text{eq l}^{-1}$, respectively, for the 2000–2004 averaging period (Table 1). Lakes with low ANC values are located throughout Finland (Fig. 1), although study sites with negative ANC are restricted to more central and southern regions.

Soils

Soil physico-chemical properties were described using a network of 488 permanent plots located throughout Finland, established as part of the eighth

Fig. 2 Average observed water chemistry variables for the period 1990–1994 (horizontal axes) compared with those for the period 2000–2004 (vertical axes). Colours indicate a stratification of the data according to their 1990–1994 ANC values (see Fig. 1 for locations): ANC < 0 (red), 0–50 (green), 50–100 (blue) and ANC > 100 $\mu\text{eq l}^{-1}$ (black). The colour coding indicates the correlation between ANC with the other lake variables shown



national forest inventory (8NFI). The plots were systematically laid out at the intersections of a 16 km × 16 km grid in the southern part of the country (<65°30' N) and a 24 km × 32 km grid in northern Finland (>65°30' N; see Fig. 1 for sampling density). Samples of the humus layer ($O_f + O_h$) and 0–5, 5–20 and 20–40 cm mineral soil layers were taken systematically from five pits at each plot and composited into a single sample for each layer. From the 60–70 cm layer a single sample was taken from one of the five soil pits (Tamminen and Starr 1990). Soils were analysed for exchangeable base cations, exchangeable acidity, organic matter (OM), carbon (C) and nitrogen content according to standard procedures. Exchangeable cations were determined from a 0.1 M $BaCl_2$ extraction using an inductively coupled emission spectrophotometer. Exchangeable acidity (H^+) was determined from the same 0.1 M $BaCl_2$ extraction by titration with NaOH to pH 7. Effective cation exchange capacity (CEC) was calculated as the sum of base cations (Ca^{2+} , Mg^{2+} , K^+ , Na^+) and exchangeable acidity ($Al^{3+} + H^+$). Base saturation was calculated as the sum of base cations divided by the effective CEC. Further details of sampling and analyses are given in Tamminen and Starr (1990).

Lumped (pit average) soil physico-chemical data for each of the permanent plots were derived by weighting soil properties by depth and density; e.g., for exchangeable fraction E_X (where $X = Ca^{2+}$, Mg^{2+} , K^+ and Na^+):

$$E_X = \frac{\sum_{i=1}^m z_i \rho_i CEC_i E_{X,i}}{\sum_{i=1}^m z_i \rho_i CEC_i} \quad (2)$$

where z_i , ρ_i , CEC_i and $E_{X,i}$ refer to the thickness, bulk density, CEC and exchangeable fraction of soil layer i ($i = 1, \dots, m$), respectively. Bulk densities were estimated from soil OM using empirical relationships (pedotransfer functions) by Van Wallenburg (1988) for the organic soil layer and Harrison and Bockock (1981) for the mineral soil layers. Pedotransfer functions are commonly employed to estimate bulk density, with the best-fit models for mineral soils based on OM (De Vos et al. 2005).

The 488 permanent soil plots do not, in general, lie within the study catchments. Mineral soil properties for each study catchment were defined by interpolating (averaging) the three (geographically) nearest soil

plots. However, prior to this several of the permanent plots were removed on the grounds that their chemistry was not 'characteristic' of acid sensitive soils (exchangeable $Ca^{2+} > 500 \text{ meq kg}^{-1}$), resulting in 441 permanent plots (see Fig. 1 for plot locations in relation to study lakes). Peatlands are present in the majority of the catchments; as such, mineral and organic soil data were spatially-weighted to derive the final catchment average (Table 1, Fig. 3). Soil physico-chemical data for organic soils are not included in the 8NFI, instead generic organic soil data were derived from literature studies; further the hydro-chemically active depth for peat soils was assumed to be 30 cm. Based on Minkinen and Laine (1998), Westman and Laiho (2003) and Sundström et al. (2000), bulk density was set to 80 kg m^{-3} , exchangeable Ca^{2+} and Mg^{2+} were set to 28% and 9%, and effective CEC was set to 325 meq kg^{-1} . The lumped catchment soil characteristics were strongly dependent on the physico-chemical properties of the mineral soils. Peat soils and organic forest soil horizons have low bulk densities and accordingly had limited influence on the final catchment average soils, e.g., the average exchangeable Ca^{2+} pool for mineral soils and the lumped catchment average differed by less than 10%. The median soil depth for the study sites was 0.54 m; median lumped catchment soil bulk density was 1056 kg m^{-3} (Table 1). The median soil base saturation was 37% (Table 1 and Fig. 3). Lower base saturation soils (<30%) occur throughout Finland (Fig. 1) and are strongly associated with low pH soils (Fig. 3). The apparent high base saturation is a function of effective CEC used in the current study, potential CEC results in lower median soil base saturation (27%); the size of the base cation pool (which is independent of effective or potential CEC) ultimately defines catchment sensitivity to acid deposition. The median effective CEC for all study sites was 19 meq kg^{-1} (Table 1). Organic matter is an important source of CEC in the soils as indicated by the positive correlation between carbon (% C) and CEC (not shown). The negative correlation between CEC and soil pH (Fig. 3) has been previously documented by several researchers in the northeastern United States of America and Scandinavia (Skylberg 1994; Ross and Bartlett 1995; Johnson et al. 2000; Johnson 2002), and for the study soils may be related to the correlation between CEC and organic matter. The

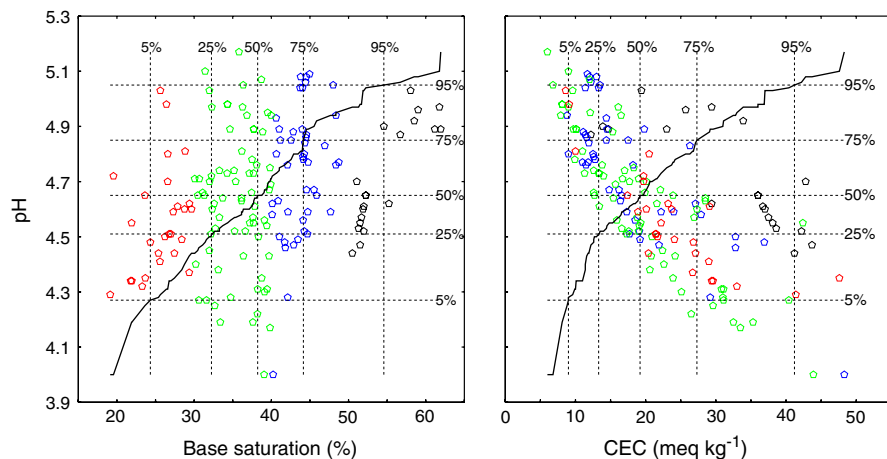


Fig. 3 Average catchment base saturation (%) against soil pH (left) and CEC (meq kg^{-1}) against pH (right) for the 163 study sites. Colours indicate a stratification of the data according to base saturation (see Fig. 1 for locations): %BS < 30 (red), 30–40 (green), 40–50 (blue) and >50% (black). The plots also

show a ‘percentile grid’, i.e. selected percentile values (5%, 25%, median, 75% and 95%) and the ‘percentile-percentile curves’ (thick solid lines) connecting all percentiles of the distributions of the respective two variables

charge development in soil organic matter can be viewed as the dissociation of a weak acid according to the extended form of the Henderson-Hasselbalch equation (Bloom and Grigal 1985; Stevenson 1994; Johnson 2002).

Climate and runoff data

Long-term average annual climate (1961–1990) for each study catchment was described using precipitation and temperature data from the Finnish Meteorological Institute. Further, these data were used as inputs to the Finnish watershed simulation and forecasting system (WSFS, Finnish version of the Swedish HBV-model system) to estimate long-term (1961–1990) catchment runoff (Vehviläinen and Huttunen 2002). The WSFS is used for hydrological forecasting and real-time monitoring in Finland (Vehviläinen 2007). The long-term climate normals for temperature, precipitation and runoff (used in the current study) were generated under the FINESSI project, which provided a common platform for integrating observations of present-day climate and possible impacts of future climate change. Under FINESSI, climate and runoff data were generated for all of Finland on a $10 \text{ km} \times 10 \text{ km}$ grid resolution (Fig. 4). The lowest runoff ($<200 \text{ mm a}^{-1}$) occurred mainly in the eastern and southern regions of Finland (Fig. 4). The long-term normals (precipitation,

temperature and runoff) for each study catchment were described using the values of the grid cell within which each lake was located (see Table 1).

Forest uptake

The long-term average annual (net and gross) uptake of base cations (Ca^{2+} , Mg^{2+} and K^{+}) and N by forests in the study catchments was calculated from the average annual volume growth and the nutrient concentrations in the removed (harvested) biomass (net: stem and bark; gross: all above-ground woody components). The average annual forest growth in Finland was estimated from the national forest inventory for 1970–1976 (Kuusela 1977). The total forest area and total annual volume growth for three tree species (deciduous, spruce, pine) were given separately for 21 forest management districts with their average effective temperature sum (ETS) values. From these data the average annual growth for each tree species at any location in Finland as a function of the local ETS was estimated. Biomass densities of each tree species (Mälkönen 1975; Rosén 1982) were used to convert average annual volume growth to mass increments. Finally, the annual average nutrient uptake was calculated using data on nutrient contents in biomass based on field measurements (Mälkönen 1975; Olsson et al. 1993; Rosén 1982). Grid-based long-term average annual

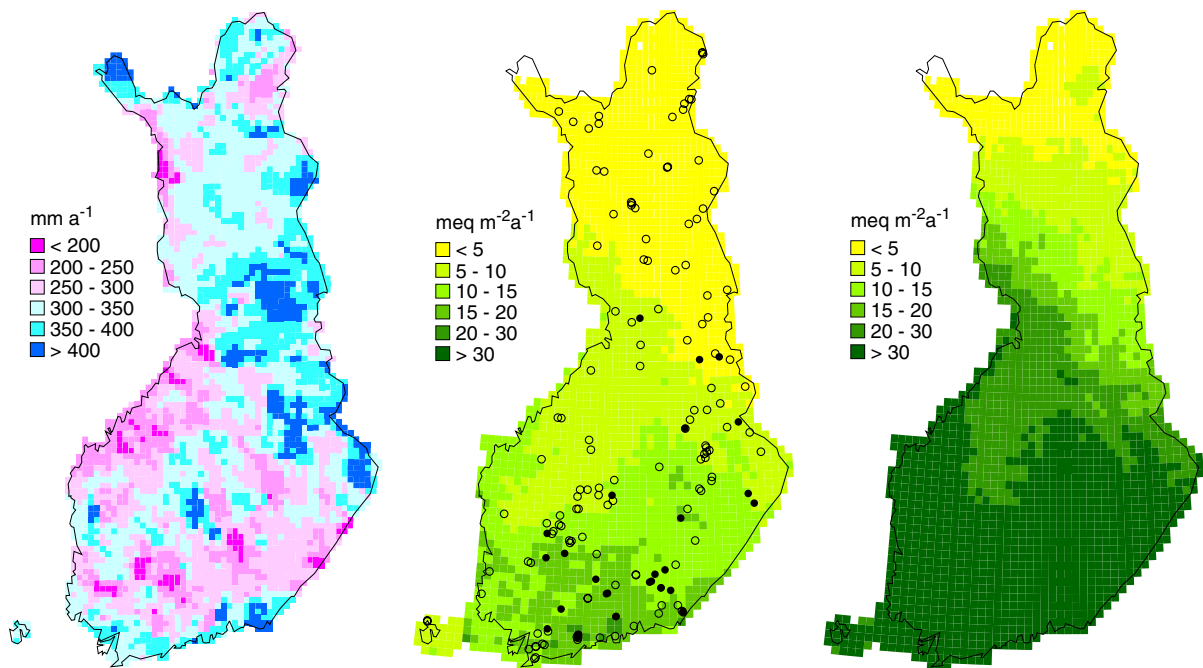


Fig. 4 Left: The 1961–1990 average annual runoff (mm a^{-1}) on a $10 \text{ km} \times 10 \text{ km}$ national grid covering Finland (left) as computed by the FINESSI project (URL: www.finessi.info/finessi). Annual average net base cation ($\text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^{+}$) uptake (removal) in Finnish forests in case

of stem-only harvesting (centre) and whole-tree harvesting (right) on a $0.250^{\circ} \times 0.125^{\circ}$ grid. The black (open and closed) circles in the centre map show the locations of the 163 study sites

(net and gross) uptake values are available for Finland with a spatial longitude–latitude resolution of $0.250^{\circ} \times 0.125^{\circ}$ (Johansson et al. 1990). Both net and gross uptake show a strong north–south gradient, with greater uptake in southern Finland (Fig. 4). Catchment estimates were interpolated (distance averaged) from the four nearest grids and multiplied by the percentage of forest area in each catchment to estimate net and gross uptake for each study site. The median long-term average annual net uptake of base cations and N for the study sites are 6.4 and $5.3 \text{ meq m}^{-2} \text{ a}^{-1}$, respectively (Table 1). In contrast, the median average annual gross uptake of base cations and N are 22.4 and $28.4 \text{ meq m}^{-2} \text{ a}^{-1}$, respectively. Nutrient uptake data were estimated as static long-term average annual values throughout the rotation period of the forest, rather than dynamic time-series of nutrient uptakes following (standard) biomass growth curves, i.e., annual uptake is constant.

Two future (2010–2100) scenarios for forest harvesting were used in the current study: (i) a ‘stem-only harvesting’ (SOH) or ‘base’ scenario,

which assumes a constant net uptake (business as usual), and (ii) a ‘whole-tree harvesting’ (WTH) scenario, which assumes a constant gross uptake. Both harvesting scenarios follow a common sequence between 1880 and 2010, i.e., constant net uptake (SOH), from 2010 the WTH scenarios was phased in linearly until 2020 and assumed constant thereafter. The SOH scenario remained constant throughout (2010–2100).

Deposition

The spatial distribution of S and N deposition to average land-cover on a $50 \text{ km} \times 50 \text{ km}$ grid resolution for the years 1990 and 2000 was taken from the output of the EMEP/MSC-W eulerian dispersion model (Tarrason et al. 2005). The deposition of S and oxidised and reduced N in Finland show the well-known north–south gradient (Fig. 5). The modelled deposition shows reasonable agreement to observed data at 19 bulk deposition monitoring (BDM; see Fig. 5 for their locations) stations operated by SYKE (Fig. 6; Vuorenmaa 2004). The average annual total

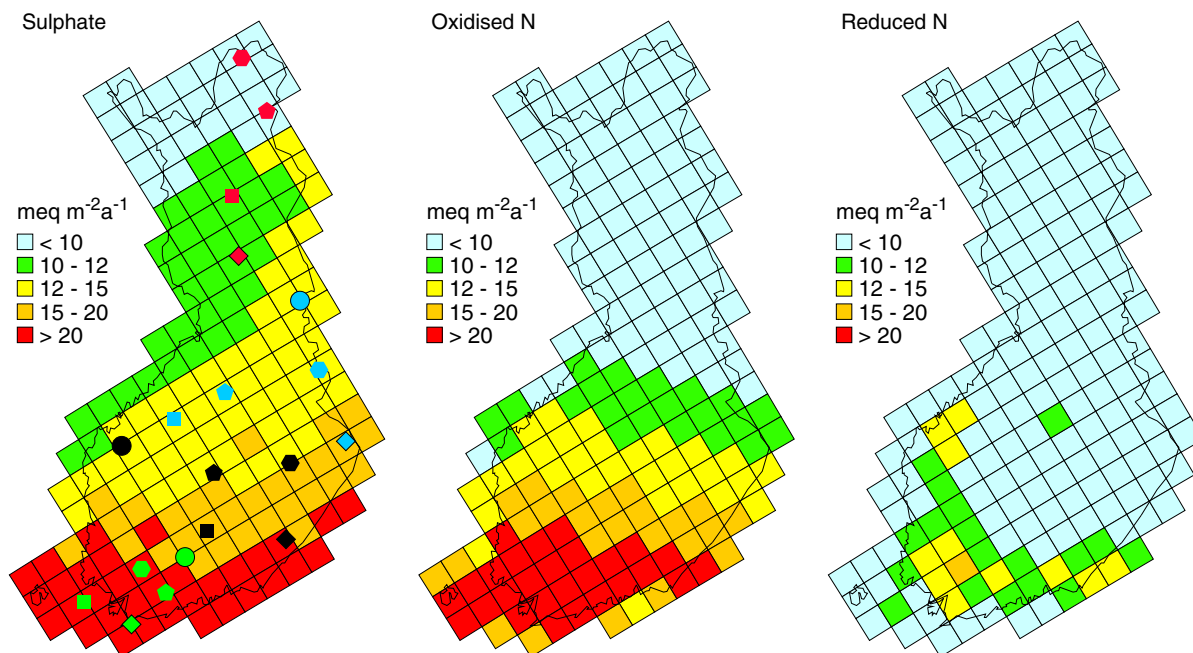


Fig. 5 Average annual deposition of sulphate (left), oxidised nitrogen (centre) and reduced nitrogen (right) in the year 2000 on the EMEP 50 km \times 50 km grid (Note: 1 meq equals 16 mg of S or 14 mg of N). The 19 different symbols in the left map

show the location of the bulk deposition monitoring (BDM) stations operated by the Finnish Environment Institute (SYKE; Vuorenmaa 2004); see also Fig. 6

SO_4^{2-} deposition modelled to average land-cover (as opposed to forest land-cover) compares well with observed data at the 19 BDM stations during the years 1990–1994 (calibration period; see Fig. 6a). In contrast, modelled total nitrogen deposition (sum of NO_3^- and NH_4^+) shows much more scatter, especially at higher deposition loads (Fig. 6b). The deposition of base cations (Ca^{2+} , Mg^{2+} , K^+ and Na^+) and Cl^- have been mapped at a resolution of $0.250^\circ \times 0.125^\circ$ for the years 1991–1995 by interpolating observations from a nation-wide network of approximately 38 stations measuring monthly bulk deposition (Järvinen and Vänni 1990). Base cation and Cl^- deposition are also consistent with observations at the 19 BDM stations (Fig. 6c; only shown for base cations). However, this is to be expected as the 19 BDM stations were part of the original 38 stations used to map deposition to Finland.

Initially, catchment-specific estimates of base cations, Cl^- , S and N deposition during the calibration year (1992) were estimated by interpolating (inverse distance averaging) the surrounding grid values. These data represent bulk deposition or total deposition to an average land-cover; however,

depending on catchment characteristics, such as land-cover, elevation, aspect, etc., total deposition may be much greater. Sulphate and Cl^- inputs were assumed to be conservative with respect to input-output fluxes, i.e., they were not retained in soils or lakes. As such, the total SO_4^{2-} input (in deposition) was set to match the SO_4^{2-} output from the lake catchments. This appears to be a reasonable assumption for the study lakes (Fig. 6d). Any difference in the input-output balance for SO_4^{2-} and Cl^- were attributed to (unmeasured) dry deposition. Imbalances in Cl^- were assumed to be of sea-salt origin, as such, all ions were balanced in proportion to their sea-salt ratios. Imbalances in SO_4^{2-} were attributed to anthropogenic sources and assumed to be balanced by anthropogenic Ca^{2+} deposition.

Sulphur and N deposition history on a 50 km \times 50 km grid resolution for the period 1880–1990 was obtained from Schöpp et al. (2003). The historical pattern is relatively uniform over Finland; therefore, a single relative deposition curve for each pollutant was used at all study sites (Fig. 7). Similarly, a single relative deposition curve for the period 1980–1990 was generated from the BDM stations. The

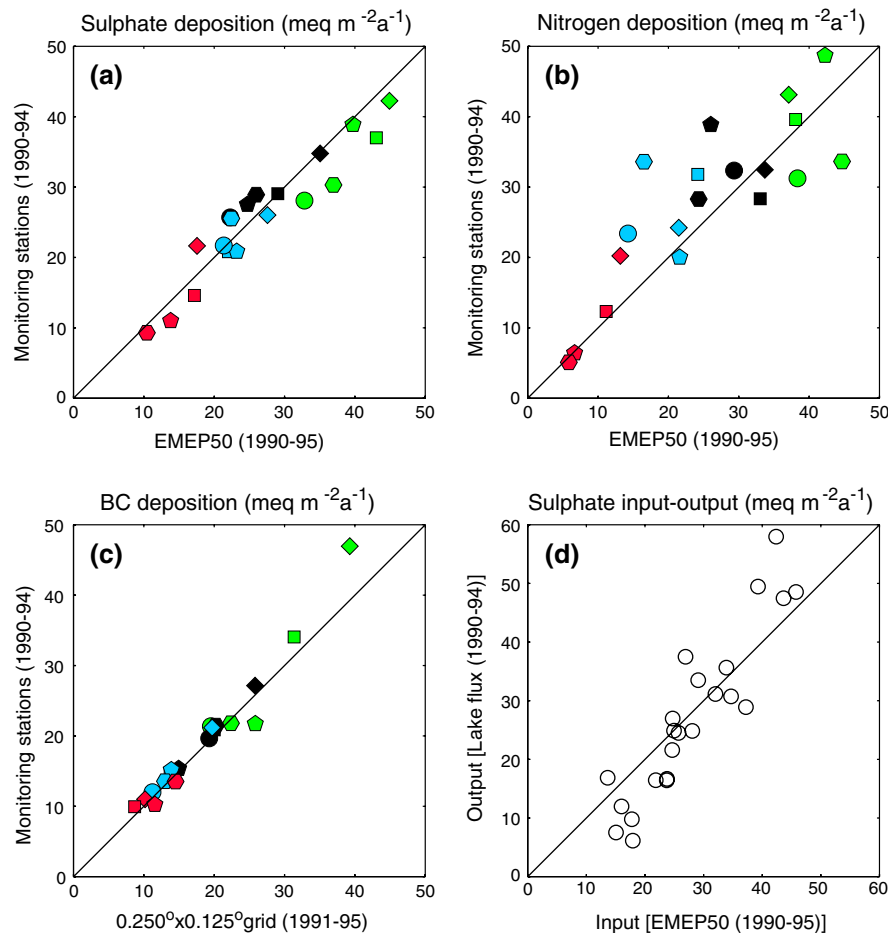


Fig. 6 Comparison of (a) observed sulphate deposition at the 19 bulk deposition monitoring (BDM) stations (1990–1994) against modelled sulphate deposition to average land cover derived from the EMEP 50 km × 50 km (EMEP50) grid by bi-linear interpolation from the four closest grid centres (1990–1995), (b) observed nitrogen (nitrate and ammonium) deposition at the BDM stations (1990–1994) against modelled EMEP50 (1990–1995), (c) observed base cation (Ca^{2+} +

deposition sequences were scaled to unity for the year 2000 (reference year). Sulphur has by far the largest variation in the past, whereas ammonium varied the least (Fig. 7). Combining these curves with the depositions in the year 2000 (Fig. 5) allows the calculation of S and N deposition at every (grid) location in Finland at any time between 1880 and 2000. Calcium and Mg^{2+} deposition were assumed to follow the same historical sequence as S, the remaining base cations and Cl^- were assumed constant throughout the simulation period. Sulphur and N deposition in (especially) southern Finland have been associated with elevated base cation deposition (particularly Ca^{2+}), which has been

Mg^{2+} + K^+ + Na^+) deposition at the BDM stations (1990–1994) against interpolated (mapped) deposition on a grid-resolution of $0.250^\circ \times 0.125^\circ$ (1991–1994) and (d) sulphate input–output budget for the study lakes summarised to the EMEP50 grid (only grids with 3 or more lakes are shown). The 19 different symbols in graphs (a–c) allow the location of each station to be identified in Fig. 5

attributed to atmospheric emissions from oil shale-fired plants in north-eastern Estonia (Lummel and Arkhipov 1996, Sofiev et al. 2003). The emission of fly ash in association with sulphur dioxide from these point sources has (historically) alleviated the impact of acidifying deposition in Finland. Base cation deposition has declined substantially in association with S; however, this decline appeared to level off during the 1990s (Vuorenmaa 2004). As such, Ca^{2+} and Mg^{2+} deposition were assumed constant from the calibration year onwards (1992).

Two future (2010–2100) scenarios for S and N deposition have been used in the current study: (i) a

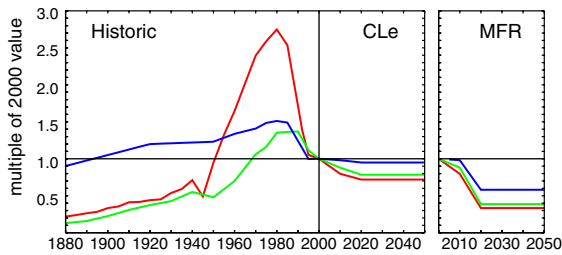


Fig. 7 Historical and future development of sulphate (red), nitrate (green) and ammonium (blue) deposition in Finland for the CLe (left panel) and MFR scenario (right panel). Note that these are scaling curves, having the value of one in 2000. The absolute deposition for any year is obtained by multiplying the deposition at a given location in 2000 (from Fig. 5) with the fraction for the year shown in these graphs

‘Current Legislation’ (CLE) scenario, which assumes the implementation of the 1999 Gothenburg Protocol of the LRTAP Convention as well as the EU National Emissions Ceiling (NEC) Directive, and (ii) a ‘Maximum Feasible Reductions’ (MFR) scenario, which assumes implementation of all technically feasible emission reduction measures by 2020 (Amann et al. 2005). All deposition scenarios follow a common sequence between 1880 and 2010, from 2010 the two scenarios were phased in linearly until 2020 and assumed constant thereafter (Fig. 7). The MFR scenario shows considerably greater reductions; relative to 2000, S deposition in 2020 is reduced by 67%, with CLe showing 28% reductions (Fig. 7).

Model calibration

Site-specific parameter files were prepared for each study site using catchment and soil physico-chemical characteristics based on, or estimated from, field observations (lake area, lake retention, soil bulk density, soil cation exchange capacity, etc.), and considered ‘fixed’ in the model (see Table 1). In addition, a number of default parameter values were uniformly applied across the region, e.g., the partial pressure of carbon dioxide ($p\text{CO}_2$) in the soil was set at ~ 15 times atmospheric. Buffering by organic acids was described using a triprotic analogue model (Driscoll et al. 1994; Cosby et al. 2001); the three dissociation constants (pK_a ’s) required by the model were taken from Hruska et al. (2003) and applied equally to soils and surface waters (see Table 2). Furthermore, a number of model parameters were calibrated to observed data; lake DOC concentrations, $p\text{CO}_2$ and aluminium solubility (or gibbsite equilibrium) constants were calibrated to observed lake pH, bicarbonate (alkalinity) and Al^{3+} concentrations, respectively. The solubility of aluminium is typically described by an equilibrium reaction with aluminium trihydroxide ($\text{Al}(\text{OH})_3$); however, an increasing number of studies have shown that equilibrium with $\text{Al}(\text{OH})_3$ is a poor predictor of Al^{3+} (Holmberg et al. 2001). In the current study, a squared relationship between the activity of Al^{3+} and hydrogen provided a better fit to surface water Al^{3+} concentrations. Soil

Table 2 Input parameters for the soil and water compartments

Parameter	Units	Soil	Lake
Sulphate adsorption half saturation ^a	meq m^{-3}	100	–
Sulphate adsorption maximum capacity ^a	meq kg^{-1}	0.1	–
Aluminium solubility constant	log	9.0	4.5
Aluminium solubility exponent		3.0	2.0
Partial pressure of carbon dioxide	$\text{atm} (\times 100)$	0.51	0.09
Dissolved organic carbon (DOC)	mmol m^{-3}	44.0	18.7
DOC dissociation constants (pK_{a1}) ^a	–log	3.04	3.04
DOC dissociation constants (pK_{a2}) ^a	–log	4.51	4.51
DOC dissociation constants (pK_{a3}) ^a	–log	6.46	6.46
Calcium uptake	$\text{meq m}^{-2} \text{a}^{-1}$	4.11	–
Magnesium uptake	$\text{meq m}^{-2} \text{a}^{-1}$	1.13	–
Potassium uptake	$\text{meq m}^{-2} \text{a}^{-1}$	1.08	–
Nitrogen uptake	$\text{meq m}^{-2} \text{a}^{-1}$	5.26	–
Nitrate retention (soil)	%	98	–
Nitrification ^a	%	100	100

Only the median values are shown for the study sites. Additional inputs are given in Table 1

^a Default values applied to all study sites, i.e., the same value for all sites

solution Al^{3+} concentrations were not available; therefore the default cubic relationship was assumed (Table 2). Soil solution DOC concentrations and aluminium solubility were calibrated against soil solution pH and cation exchange selectivity coefficients. The coefficient for N in-lake retention was set to 5 m a^{-1} for all lakes (Kelly et al. 1987). Soil and surface water nitrification was set to 100%. Nitrogen (NO_3^- and NH_4^+) retention (or uptake) in the terrestrial and lake compartments was set to match observed lake concentrations (on average, 92% of the incoming N is retained in the catchment). Nitrogen transformations were described as a catchment net retention calculated simply as the difference between input and output flux for the calibration year. This percentage was assumed to be constant throughout the simulation.

Base cation weathering rates and initial soil exchangeable fractions were calibrated using MAGICOPT. The procedure uses numerical techniques to select parameter values that result in a minimum sum of squares error between simulated and observed target variables for each catchment during the calibration period (Rosenbrock 1960). To account for uncertainty in a number of the fixed parameters (lake retention, soil depth, soil bulk density, cation exchange capacity and sulphate adsorption half saturation), a ‘fuzzy’ optimisation method was employed. Multiple calibrations were carried out for each study lake; during each simulation the fixed parameters were randomly varied within specified uncertainty bands ($\pm 10\%$). In addition, uncertainty bands (or tolerance levels) were also applied to the target lake water ($\pm 2 \mu\text{eq l}^{-1}$) and soil chemistry ($\pm 0.2\%$) variables. The tolerance levels were somewhat arbitrarily chosen as the numerical optimisation routine efficiently minimises the difference between simulated and observed target variables (hence the narrow bands). For each study lake, 10 calibrations were performed with MAGICOPT; any simulation that reproduced all target variables (see section “Model description”) was considered successful. Model performance was evaluated by comparing simulated and observed data during the calibration year (1992) and 10 years post-calibration (simulated year: 2002; observed data: 2000–2004). Goodness-of-fit for predicted against observed was assessed using model efficiency, which is sensitive to outliers (Alewell and Manderscheid 1998). An efficiency of 100% indicates perfect agreement between predicted and observed data.

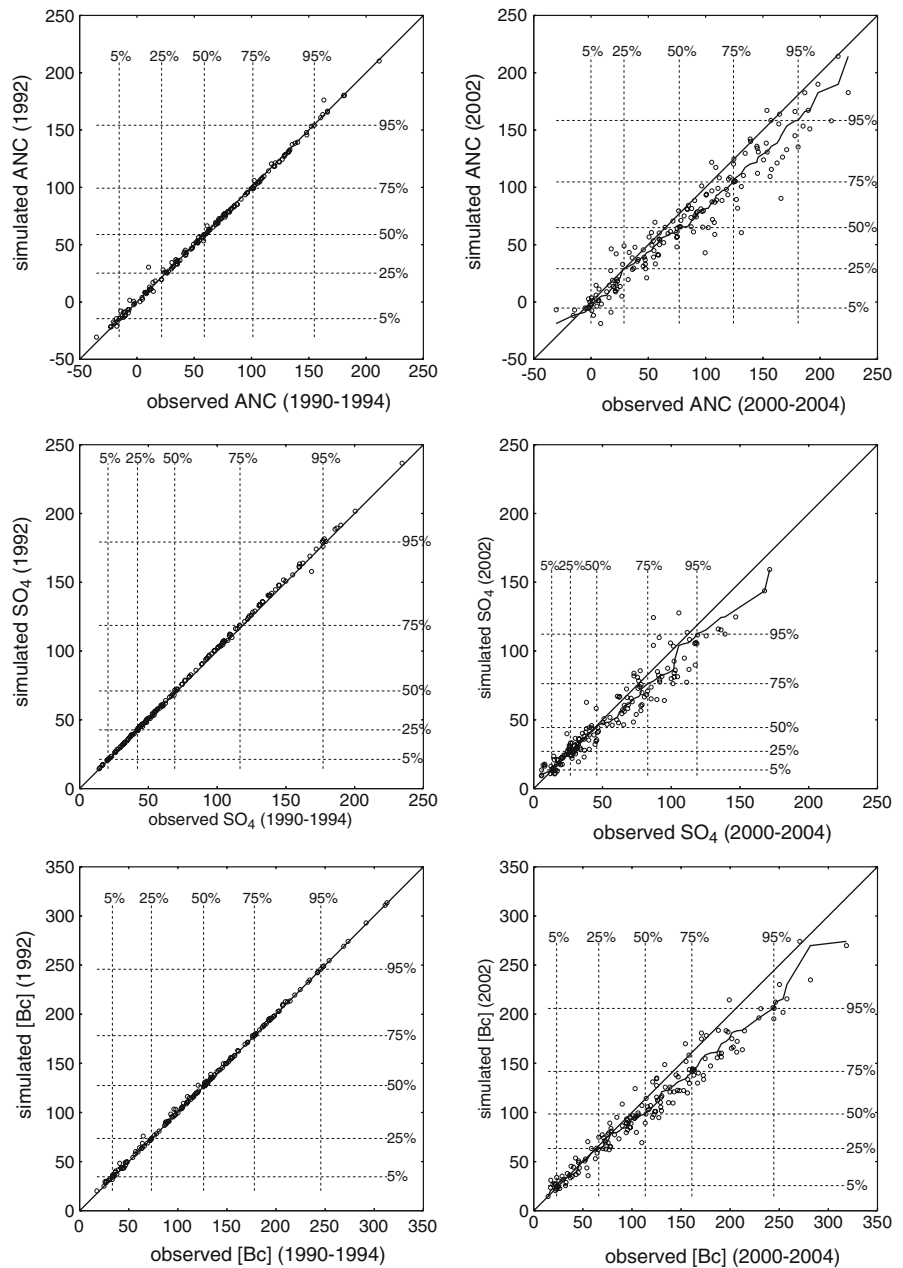
Future soil and surface water chemistry for each lake were simulated for the period 2000–2100 under two S and N deposition scenarios and two forest harvesting scenarios (2×2 scenario matrix). Climate and runoff were assumed to be constant. The reference or ‘base’ scenario was composed of the CLe (deposition) and SOH (forest harvesting) scenarios, which represent the ‘present’ future, based on current legislation and practices. The multiple simulation results for each lake (maximum of 10) were combined using median statistics. Future recovery patterns (until the year 2100) were evaluated using three key indicators: ANC and pH in lake water and base saturation in the catchment soils. Lake ANC and pH are standard indicators used in acidification studies, as both are strongly indicative of biological recovery (Hutchinson et al. 1989; Lien et al. 1996). In addition, ANC is the most widely used chemical criterion in critical load calculations for surface waters (Henriksen et al. 1995). Base saturation is a key indicator of soil sustainability, and has also been used as a chemical criterion in forest soil critical loads (Holmberg et al. 2001).

Results and discussion

Calibration to the Finnish RMLA lake catchments

The fuzzy optimisation procedure attempted 10 calibrations on simulations from 1880 to 2000 (calibration year: 1992) under the historical deposition sequence. The number of calibrations per lake ranged from 3 to 10, with the majority of lakes having >5 successful calibrations (155 lakes; 53 lakes with 10 successful calibrations). There was excellent agreement between simulated and observed ANC, base cations ($\text{Bc} = \text{Ca}^{2+} + \text{Mg}^{2+} + \text{K}^+$) and SO_4^{2-} during the calibration year (model efficiency = 100% for each parameter; Fig. 8 left column). In general, most modelled parameters show ‘perfect’ agreement with observations during the calibration year. This is not surprising as the ‘optimisation procedure’ rejected simulations that fell outside the specified tolerance levels ($\pm 2 \mu\text{eq l}^{-1}$). A more rigorous evaluation of model performance is to compare simulated and observed data outside the calibration period. Overall, modelling efficiency for simulated and observed data ten years beyond the

Fig. 8 Comparison of the five-year mean observed ANC (top), $[\text{SO}_4^{2-}]$ (centre) and base cation concentration $[\text{Bc}] = [\text{Ca}^{2+}] + [\text{Mg}^{2+}] + [\text{K}^+]$ (bottom; all in $\mu\text{eq l}^{-1}$) for the 163 study lakes in 1990–1994 (left column) and 2000–2004 (right column) compared with the respective simulated values for the years 1992 and 2002, respectively. Also shown are the percentile grids and the 1-to-1 lines



calibration year indicate good agreement ($\text{ANC} = 87\%$, $\text{SO}_4^{2-} = 90\%$ and $\text{Bc} = 91\%$; Fig. 8 right column). However, simulated lake ANC, SO_4^{2-} and Bc are in general lower than the observed. Sulphate deposition is the principal driver responsible for the divergence between simulated and observed. Historic and future S deposition (1880–2100) was described using a scaling sequence (normalised to the reference year) that was applied to all study lakes. However, there is considerable

variation in deposition to each study lake; a single scaling sequence did not capture the regional variation in deposition. Secondly, it was assumed that all SO_4^{2-} was entirely from atmospheric sources (section “Model calibration”). However, in a number of regions SO_4^{2-} input to lakes was lower than SO_4^{2-} output (Fig. 6d). These lakes may be influenced by other sources of SO_4^{2-} ; acid SO_4^{2-} soils have been observed in western and south western areas of Finland owing to sediments from the post-glacial

Litorina sea (Mattsson et al. 2007). Furthermore, in the current study SO_4^{2-} adsorption characteristics in soils were parameterised assuming a low regional capacity; however, a number of soils may have much higher potential adsorption (desorption) capacity. Despite this potential discrepancy, the current study still captures the lake dynamics, with the mismatch more evident in lakes with higher SO_4^{2-} concentrations. Moreover, the largest deviations occur for lakes with the highest ANC ($>100 \mu\text{eq l}^{-1}$), which are not important with respect to acidification.

The calibrated base cation weathering rates ranged between 4 and $88 \text{ meq m}^{-2} \text{ a}^{-1}$ (median = $32.5 \text{ meq m}^{-2} \text{ a}^{-1}$; Table 3) and generally show lower weathering in northern Finland (Fig. 9). This is consistent with previous weathering rate studies in Finland, based on soil geochemistry and mapped on a spatial longitude–latitude resolution of $0.250^\circ \times 0.125^\circ$ (Johansson and Tarvainen 1997). However, in the current study the calibrated weathering rates are lower (compared to Johansson and Tarvainen (1997): $18\text{--}162 \text{ meq m}^{-2} \text{ a}^{-1}$, median of $87 \text{ meq m}^{-2} \text{ a}^{-1}$). Johansson and Tarvainen (1997) presented average weathering rates on a grid basis, whereas the current study specifically selected acid sensitive lake catchments. Base cation weathering is dominated by Ca^{2+} release; similarly initial base saturation was dominated by Ca^{2+} (Table 3). Median percent base saturation in 1880 was approximately 6% greater than current (43.0% and 37.2%,

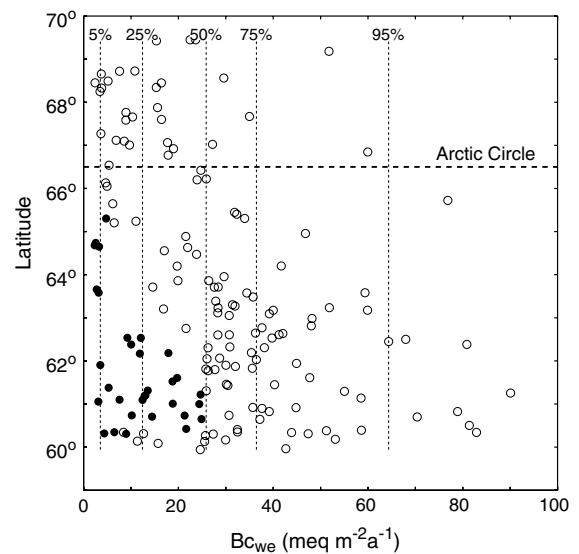


Fig. 9 Calibrated base cation weathering for the 163 catchments versus latitude. The 32 filled circles are those catchments for which base cation uptake exceeds deposition plus weathering under whole-tree harvesting (see Figs. 4 and 11)

respectively; see Tables 1 and 3). The logarithm of the calibrated selectivity coefficients are generally between -2 and $+2$, which is in agreement with estimates for a wide range of soils types in the Netherlands (De Vries and Posch 2003).

Model calibration has often been criticised as a ‘making-it-fit’ exercise; however, calibration is a critical phase in the modelling process. All ecosystem

Table 3 Statistical summaries (5th percentile, median and 95th percentile) for the calibrated soil parameters (weathering rate, initial (1880) cation saturation, and cation exchange selectivity coefficients) for the study sites ($n = 163$; see Fig. 1 for locations)

Calibrated parameter ^a	Units	5 Percentile	Median	95 Percentile
Weathering rate				
Calcium	$\text{meq m}^{-2} \text{ a}^{-1}$	2.3	14.0	39.9
Magnesium	$\text{meq m}^{-2} \text{ a}^{-1}$	1.0	9.9	23.7
Sodium	$\text{meq m}^{-2} \text{ a}^{-1}$	0.9	7.9	19.0
Potassium	$\text{meq m}^{-2} \text{ a}^{-1}$	0.0	0.7	5.0
Initial cation saturation (1880)				
Calcium	% CEC	18.3	27.6	44.8
Magnesium	% CEC	6.0	9.6	18.7
Sodium	% CEC	1.0	1.6	2.1
Potassium	% CEC	2.2	4.2	6.9
Exchange selectivity coefficients				
Aluminium–calcium	log	-2.6	-0.1	2.1
Aluminium–magnesium	log	-1.8	1.0	3.5
Aluminium–sodium	log	-4.4	-2.5	-1.1
Aluminium–potassium	log	-4.7	-3.5	-2.0

^a Calibration refers to an automated optimisation procedure (MAGICOPT) that is a component of the MAGIC model suite

models are based on a simplified description of the system under study, moreover they contain incomplete descriptions of parameters and initial and boundary conditions (Janssen and Heuberger 1995). As such, the calibration of model parameters through fitting output parameters to observational data is fundamental to dynamic, process-oriented models. However, the complex structure of ecosystems and the limited number of observations result in model calibrations that are not unique. One ‘true’ model for a given system does not exist; rather any model may be termed ‘valid’ unless proven invalid. As such, model evaluation is important (Alewell and Manderscheid 1998). At minimum model evaluation requires two elements: a performance criterion and a benchmark (modified from Kirchner et al. 1996). Specifying a benchmark answers the question, ‘good compared to what?’. In the current study, model performance was evaluated using model efficiency. This criterion has been recommended in preference to other criteria such as Normalised Absolute Mean Error (NAME: see Janssen and Heuberger 1995) owing to its sensitivity to outliers (Alewell and Manderscheid 1998). However, an explicit performance criterion for model rejection, such as an efficiency $<50\%$, was not defined in the current study. Rather we have relied on MAGIC’s reputation: MAGIC has been extensively applied and tested during the last 20 years at many sites and regions around the world, and overall has proven to be robust, reliable and useful in a variety of scientific and managerial activities (Cosby et al. 2001). Moreover, a judicious censoring of post-calibration simulations based on observations ten years beyond the calibration period was not required as there is good agreement for the entire lake population (efficiency for dominant ions $>80\%$); and more importantly, the most sensitive lakes (observed ANC $<100 \mu\text{eq l}^{-1}$) show better agreement.

Influence of deposition on recovery

The reduction in S deposition is the main driver of acidification recovery in Finland (Forsius et al. 2003; Posch et al. 2003). The median nitrate concentration of the study lakes is only $0.61 \mu\text{eq l}^{-1}$ (Table 1) and increasing trends in nitrate for the RMLA lakes have not been observed. Future changes in N emissions are not expected to have any major impact on the acidity

status of Finnish lakes (Forsius et al. 2003) and therefore, are given limited attention in the present study. Further, it was assumed that N saturation will not lead to N leaching in the future. The base scenario (CLE and SOH) showed very marginal recovery in lake chemistry between 2000 and 2100 (Fig. 10 and Table 4). The median simulated ANC for the CLE scenario increased by $3.0 \mu\text{eq l}^{-1}$. Simulated median pH increased by 0.1 of a pH unit; median Bc concentration in lakes decreased by $15.0 \mu\text{eq l}^{-1}$ (sulphate decreased by $12.4 \mu\text{eq l}^{-1}$). Median soil base saturation similarly showed limited recovery, with increases of $<1\%$. These marginal recoveries are (initially) somewhat surprising considering that S deposition is reduced by 28% relative to 2000 under the CLE scenario. However, the largest reductions in S depositions, and largest recoveries, occurred during the 1980s and 1990s (Figs. 7 and 10). In general, the base scenario assumed modest reductions between 2000 and 2020 and was held constant thereafter. Nonetheless, the future recovery of surface water chemistry will continue, but more slowly than during the previous decades (Table 4). In addition, future reductions in emissions are likely to stop soil acidification (base cation depletion).

Posch et al. (2003) used the SMART model to predict the possible recovery of 36 acid-sensitive

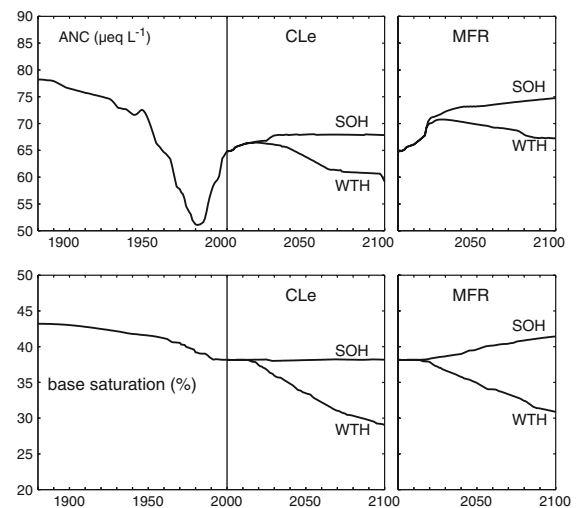


Fig. 10 Temporal development of the median of lake water ANC (top) and the catchment base saturation (bottom) for the 163 study lakes under the two emission scenarios (CLE: left panel and MFR: right panel) and two harvest scenarios (SOH and WTH). Note: Median statistic refers to the median for the 163 study catchments

Table 4 Simulated soil and surface water chemistry (5th percentile, median and 95th percentile) during 2000, 2025, 2075 and 2100 under current legislated (CLE) and maximum

feasible emission reductions (MFR), and stem-only (SOH) and whole-tree harvesting (WTH) scenarios

Parameter	Percentile	Biomass Scenario	CLE					MFR			
			2000	2025	2050	2075	2100	2025	2050	2075	2100
Sulphate	5		13.9	10.1	10.1	10.1	10.1	5.4	5.3	5.3	5.3
	50		45.2	32.8	32.8	32.8	32.8	16.7	16.5	16.5	16.5
	95		116.0	81.2	81.2	81.2	81.2	42.5	41.2	41.2	41.2
Acid neutralising capacity	5	SOH	−5.7	0.4	0.6	0.9	0.9	7.6	9.0	9.4	9.7
	50	SOH	64.8	66.8	68.0	67.9	67.8	71.5	73.2	74.0	74.8
	95	SOH	158.1	166.6	167.3	168.5	167.7	169.9	170.6	171.6	172.9
pH	5	SOH	4.9	4.9	4.9	4.9	4.9	5.0	5.0	5.0	5.0
	50	SOH	5.8	5.9	5.9	5.9	5.9	6.1	6.1	6.1	6.1
	95	SOH	6.8	6.8	6.8	6.8	6.8	6.8	6.8	6.8	6.8
Base cations	5	SOH	26.2	22.7	22.5	22.3	22.3	19.3	19.0	18.8	18.7
	50	SOH	102.2	89.1	88.0	87.5	87.2	78.4	76.5	76.7	77.3
	95	SOH	208.0	185.7	184.1	183.4	183.0	162.1	161.6	162.3	163.1
Base saturation	5	SOH	23.9	23.5	23.7	23.9	24.1	24.2	26.8	27.7	28.4
	50	SOH	38.2	38.2	38.1	38.2	38.2	38.5	39.6	40.7	41.4
	95	SOH	54.9	54.6	54.2	53.2	53.0	54.9	55.5	57.4	59.2
Acid neutralising capacity	5	WTH	−5.7	−0.3	−2.2	−4.1	−5.7	7.0	5.6	4.6	3.3
	50	WTH	64.8	66.2	63.8	61.0	59.2	70.7	69.8	68.7	67.3
	95	WTH	158.1	161.0	155.3	154.0	153.2	169.1	168.4	167.6	166.2
pH	5	WTH	4.9	4.9	4.9	4.9	4.8	5.0	5.0	5.0	4.9
	50	WTH	5.8	5.9	5.8	5.8	5.7	6.0	6.0	6.0	6.0
	95	WTH	6.8	6.8	6.8	6.8	6.8	6.8	6.8	6.8	6.8
Base cations	5	WTH	26.2	22.2	20.4	17.8	17.1	18.4	15.9	14.8	13.5
	50	WTH	102.2	87.6	82.3	78.3	76.3	77.3	71.0	69.6	66.9
	95	WTH	208.0	180.6	169.8	164.7	161.4	159.9	150.1	144.3	140.4
Base saturation	5	WTH	23.9	21.6	17.6	13.5	9.9	22.6	19.3	16.9	14.0
	50	WTH	38.2	36.8	33.5	30.6	29.0	37.2	34.9	32.9	30.9
	95	WTH	54.9	53.2	47.4	44.4	41.9	53.5	50.6	48.0	46.4

Finnish headwater lakes, located mainly in southern parts of the country. The model was used to simulate soil and water chemistry until 2030 under a 'current legislation scenario'. In concert with the current study, Posch et al. (2003) indicated that the positive trends in lake ANC, which have been observed over the past decade, would continue into the future, albeit at a slower pace. Posch et al. (2003) also predicted that during 2010–2030 all lakes will have reached a positive ANC, a pre-requisite for the recovery of fish populations. In the current study, based on a larger and more spatially representative data set, four lakes have negative ANC in 2030, and 28 have $\text{ANC} < 20 \mu\text{eq l}^{-1}$. Wright et al. (2005) applied the

MAGIC and SMART models to twelve acid-sensitive surface water regions in Europe. The model results indicate that even after complete implementation of the Gothenburg protocol and other current legislation, acidification will continue to be a significant problem in southern Norway, southern Sweden, the Pennines in the UK, the Tatra Mountains (Slovakia), and the Italian Alps. In each of these regions more than 5% of the lakes will not meet the ANC criterion to protect sensitive aquatic organisms ($\text{ANC} > 20 \mu\text{eq l}^{-1}$; Lien et al. 1996).

The MFR scenario (under SOH) showed significant recovery in lake (and soil) chemistry between 2000 and 2100 (Fig. 10 and Table 4). The median

simulated ANC under the MFR scenario increased by $9.9 \mu\text{eq l}^{-1}$; approximately 14% of lakes are predicted to have $\text{ANC} < 20 \mu\text{eq l}^{-1}$ by 2100 under MFR (no lakes with $\text{ANC} < 0 \mu\text{eq l}^{-1}$), compared to 23% in 2000. Simulated median pH increased by 0.3 of a pH unit; median Bc concentration in lakes decreased by $24.9 \mu\text{eq l}^{-1}$ driven by the decrease in S deposition (median sulphate in lakes decreased by $28.7 \mu\text{eq l}^{-1}$). The simulated median soil base saturation showed significant recovery under MFR, with a 3% increase by 2100. More importantly, base saturation showed continued recovery throughout the forecast period (and beyond) under MFR. In contrast, CLe showed no improvement beyond 2020 (Table 4). Under MFR, the increase in base saturation drives continued recovery in ANC, pH, etc. The MFR scenario presents a considerably more optimistic future with a 67% reduction in S deposition by 2020 relative to 2000. The MFR scenario may be viewed as an optimistic scenario based on maximum emission reductions currently technically feasible. Nonetheless, the scenario has been used in numerous regional, continental and global studies (e.g., Dentener et al. 2006a, b). In general, it is the only (realistic) scenario that consistently indicates (further) improvement in ecosystem quality, such as reduced impacts of N on natural vegetation (Dentener et al. 2006a) and air quality (Dentener et al. 2006b). In concert, the current study shows considerable long-term improvement in lake (and soil) chemistry under MFR. Despite the obvious benefits of the scenario, and its popularity in the scientific community, it ‘apparently’ is not being considered by the political community. Moreover, despite its ‘optimistic outlook’, it does not consider progressive energy use scenarios, which may have similar (or better) effects on reducing emissions (Dentener et al. 2006a).

Influence of harvesting on recovery

Tree growth has a significant effect on the acid status of forest soils owing to the excess accumulation of base cations compared to anions during tree growth (e.g., see Olsson et al. 1993). During recent years there has been a tendency to increase the use of harvest residues (branches and tops) as biofuels represent an attractive alternative to fossil fuels for many countries in support of climate policies. The present production of forest chips in Finland is

2.6 million $\text{m}^3 \text{a}^{-1}$, with a technically feasible maximum potential of 15 million $\text{m}^3 \text{a}^{-1}$. More than half of this potential is in harvest residues. If all of this potential was utilised for energy production, it would amount to about 8% of the total Finnish energy consumption (Hakkila 2004). There are plans to greatly increase the use of forest chips for energy production in Finland; according to the national forestry plan the aim is 8 million $\text{m}^3 \text{a}^{-1}$ by the year 2015. However, more intensive harvesting (whole-tree) will result in a higher export of nutrients (and alkalinity) from forest sites compared to the current stem-only harvesting practices, resulting in similar effects on the forest ecosystems as acidification owing to air pollutants (Olsson et al. 1993; Sverdrup and Rosén 1998).

Base cation budgets on a regional scale have been estimated for Finland (Joki-Heiskala et al. 2003) and Sweden (e.g., Sverdrup and Rosén 1998; Akselsson et al. 2007). Joki-Heiskala et al. (2003) demonstrated that there would be a depletion of base cations in forest mineral soils in southern, central and north-eastern Finland (40–50% of the mapping grids) under WTH in the long-term. In concert, Akselsson et al. (2007) indicated that the present pools of exchangeable base cations will decrease even when SOH is employed, and that the losses of Ca^{2+} and K^+ will increase substantially under WTH, especially in spruce forests, for large parts of Sweden.

In the current study, two future (2020–2100) biomass scenarios were used to evaluate the impacts of forest harvesting on acidification status: the SOH or ‘base’ scenario assumed a constant net uptake (business as usual) and WTH assumed a constant gross uptake. These scenarios clearly show the impacts of forestry practices on the chemistry of soils and surface waters at acid-sensitive sites. Under CLe, the SOH scenario resulted in a steady-state with only marginal improvement regarding acidification recovery; the more stringent emission reductions under MFR were required for recovery towards pre-acidification values. The WTH scenario indicated a pessimistic future of re-acidification (Fig. 10 and Table 4) under both CLe and MFR. The CLe and WTH scenario predicated a decrease in ANC, pH and a significant decrease in base saturation. More importantly, there was an increase in the percentage of study lakes with $\text{ANC} < 20 \mu\text{eq l}^{-1}$ (3% increase to 22%, 14 lakes with $\text{ANC} < 0$). Clearly WTH will offset any benefits

from emission reductions under CLe (Fig. 10). The future under WTH and MFR is less pessimistic, for example ANC and pH in 2100 are similar under CLe and SOH, and the number of lakes with ANC < 20 will decrease to approximately 17% (one lake with ANC < 0). The simulations suggest that further emission reductions are required to mitigate the impacts under a shift from SOH to WTH; greater biomass removal will dictate further emissions reductions to maintain ecosystem quality. However, even under MFR, the base saturation showed significant impacts indicating that ANC and pH will ultimately decrease into the future (Fig. 10 and Table 4). Regardless of atmospheric deposition scenario, increased biomass removal under the WTH scenario resulted in decreasing ANC and soil base saturation. The current SOH forestry practices are less detrimental and more environmentally sustainable.

The leaching of base cations, mainly driven by S deposition, is a significant component of the base cation catchment mass balance. Looking at the steady-state situation, where the base cation uptake should not exceed the long-term sources of base cations (deposition and weathering), clearly indicates that whole tree harvesting is not sustainable in the long-term for a significant proportion of these acid-sensitive catchments. In 32 out of the 163 study sites, soil amendment (fertilisation) is required to compensate for the net loss under WTH, even without considering the leaching of base cations (Fig. 11). Moreover, the majority of the sites (150 out of 163) would experience decreasing base saturation under WTH when acid deposition is considered (Fig. 10).

The simulations are based on the assumption that the harvesting practices are carried out at the scale of the whole forested catchment area (range: 3–100%,

Table 1), representing the most extreme situation. For the WTH scenario it was also assumed that all harvest residues were removed. In a study for Sweden, Akselsson et al. (2007) assumed a removal of 75% harvest residues. Moreover, as catchment-specific forestry scenarios were not available, a constant base cation uptake sequence was assumed. In general, this will not have a significant influence on the predictions for 2100, as the simulation period covers a forest rotation cycle. However, the dynamic pattern of tree uptake between 2020 and 2100 is not captured, with uptake during the earlier growth years being underestimated. In contrast to the regional steady-state mass balance calculations (Joki-Heiskala et al. 2003), site-specific dynamic uptake scenarios may easily be incorporated into dynamic model frameworks (e.g., see Forsius et al. 1997) thus providing more detailed information on the temporal behaviour of forest ecosystems. Ideally, detailed nutrient uptake sequences should be incorporated when the information becomes available. This is necessary more so for point or plot scale assessments; for regional studies (such as the current) the combination of numerous uptake sequences may result in a more ‘constant’ regional uptake. Nevertheless, both the regional mass-balance calculations and the present dynamic modelling study clearly show that base cation budgets and acidification recovery are important topics to be considered for sustainable forestry management policies in acid-sensitive regions.

Uncertainties

The application of MAGIC to the RMLA lakes presents one of the most comprehensive regional applications of MAGIC. However, it is inevitable that

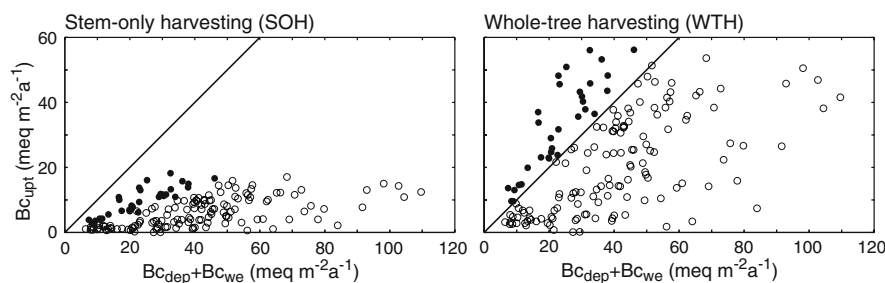


Fig. 11 Base cation ($Bc = Ca^{2+} + Mg^{2+} + K^{+}$) input (deposition + weathering) versus Bc output (net uptake) for stem-only harvesting (left) and whole-tree harvesting (right) after

2020. The 32 points above the 1-to-1 line (filled circles) represent catchments where long-term whole-tree harvesting is not sustainable (see Fig. 4 for their locations)

there are uncertainties associated with any model application. These can be roughly grouped into two categories: (a) uncertainties owing to model structure (conceptualisation), and (b) uncertainties in the model parameters and input data. Although some of the process descriptions used in simple hydro-geochemical models have been called into question (e.g., Neal 1997), MAGIC has shown to be a reliable tool for simulating the impact of atmospheric deposition on soils and waters. A different issue is the impacts of climate change; MAGIC (and similar models) are not explicitly conceptualised to include climate-sensitive processes (Posch et al. 2008). Instead, it is mainly the lack of site-specific input data that limit model applications on a regional scale. It is axiomatic that models should be rigorously tested. However, many ‘modelling studies’ suffer from generous treatment of input data, obvious failings in documentation and rigorous evaluation of model outputs. In the current study, considerable effort was given to the description of the input data, the derivation of site-specific model parameters, and the collation and processing of regional data-sets. Where multiple regional data-sets existed, each was evaluated for consistency. Similarly, site-specific parameters and scenarios were applied where information was available. Nonetheless the model results (predictions) are subject to considerable uncertainty.

Elevated deposition of base cations has been shown to have a significant impact on the acidity of rainfall in Finland. Despite its importance little is known about the historic sequence of base cation deposition. In the current study it was assumed that historical base cation deposition followed S deposition. Similarly, (high-resolution) site-specific S and N deposition sequences were not available; deposition was assumed to follow a single regional sequence. However, the single regional sequence did not completely reproduce observed lake chemistry ten years beyond the calibration period. Furthermore, one lake sample annually was assumed to adequately reflect average lake chemistry. Catchment specific soil data were not available for the study sites; soil physico-chemical properties from the three nearest NFI plots were assumed to adequately reflect the soils at the study sites. In addition, information on organic soils was not available. In practice, soil physico-chemical properties may vary enormously within a forest stand, and more so across a catchment. The lumped parameter

approach assumed that upland mineral soils and peat soils equally contribute to lake chemistry irrespective of their landscape position. It was assumed that the net retention of N within catchments (difference between input and output flux) was constant throughout the simulation. This may be viewed as a ‘best case’ scenario given that potential increased leaching may have a significant impact on future recovery. Nitrogen deposition is highly variable in time and space and is clearly more challenging to model (see Fig. 6); nonetheless, observations to date suggest that future changes in N emissions are not expected to have a major impact on the acidity status of Finnish lakes. Model predictions depend strongly on model calibration, and calibration depends on available observational data. In the current study, calibration was limited to one observation period (calibration year: 1992). However, previous studies have shown that differences between modelled and observed data in general decrease with increasing length of the time series used in calibration. Furthermore, uncertainties in predicted future quantities are lower when using longer time series for calibration (Larssen et al. 2005).

There are numerous perils inherent to regional modelling studies. The issue of data resolution is clearly paramount. Regional modelling studies are generally not afforded the luxuries experienced by intensive monitoring sites, and typically have to ‘make do’ with the available data. Within the current study, considerable effort was given to generating ‘site-specific’ model inputs. Nonetheless, the inputs represent a range of spatial and temporal resolutions: catchment characteristics at a 1:20,000 scale, climate and runoff at 10 km grid resolution, catchment physico-chemical properties at the three nearest soils plots, and lake chemistry sampled annually. The choice of data resolution within the current study was pragmatic; they were the best available data. Despite the practical limitations, data resolution only presents an issue where it influences model predictions. Sulphate deposition is the main driver in MAGIC; accordingly, the lack of site-specific deposition sequences in the current study had the greatest influence on model simulations. The divergence between simulated and observed data ten years beyond the calibration period is principally driven by the ‘regionally optimistic’ SO_4^{2-} deposition sequence. While this has important implications for the comparison of observed and simulated data

(beyond the calibration period), it has less importance in relation to the assessment of the relative impact of proposed (or potential) future emission reduction scenarios.

The assumption that (in the future) all forests will be subject to WTH is overly ambitious. Clearly there are forest regions that (for practical reasons) cannot be completely harvested. Nonetheless, the estimated base cation uptake rates used in the current study are conservative, as such; the predicted influence of harvesting on recovery may be underestimated. The estimated uptake rates were based on data from the national forest inventory during the 1970s. Current forest growth rates are higher (potentially) owing to changes in climate. However, these removal estimates are somewhat skewed as they also include a significant proportion of WTH in conjunction with SOH removals. The current study was designed to be independent of climate effects as long-term (1961–1990) meteorology and hydrology were fixed to baselines prior to significant global climate change. It is the relative difference between the two harvesting scenarios (SOH and WTH) which is important to the current study. Ultimately, future studies should incorporate 'realistic' base cation (nutrient) uptake sequences and more importantly, where the information is available, models should use site-specific deposition sequences and be calibrated to more than one observation period.

Conclusions

The impact of future atmospheric deposition and biomass harvesting scenarios on soil and surface water chemistry was evaluated for 163 lake catchments in Finland using MAGIC. The model calibration utilised extensive national data-sets on lake chemistry, catchment characteristics, soil properties, deposition and nutrient uptake by forests. The study represents one of the most detailed regional-scale hydro-geochemical model assessments for lake catchments. MAGIC predicted limited chemical recovery for soils and lakes under the current legislation scenario. Simulations suggested that only the maximum (technically) feasible emission reduction scenario would result in significant long-term chemical recovery of soils and surface waters, returning soil base saturation and water quality close to pre-acidification values.

The use of forest biomass for energy production in Finland has steadily increased over the past 25 years; to meet this demand future harvesting is proposed to shift from stem-only to whole-tree harvesting. This increased use of forest harvest residues for biofuel production (WTH scenario) was predicted to have a significant negative influence on the base cation budgets causing re-acidification at the study catchments. Sustainable forestry management policies need to consider the combined impact of air pollution and harvesting practices. Clearly there is a need for further emission reductions to mitigate the negative impacts of WTH, if such a policy is implemented. Additionally, increased fertiliser use, such as wood ash applications, may also be required to maintain soil nutrient status and lake water quality in these forested ecosystems. The modelling framework presented in this paper provides a suitable tool for more detailed studies regarding this matter.

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