Acid Rain Research

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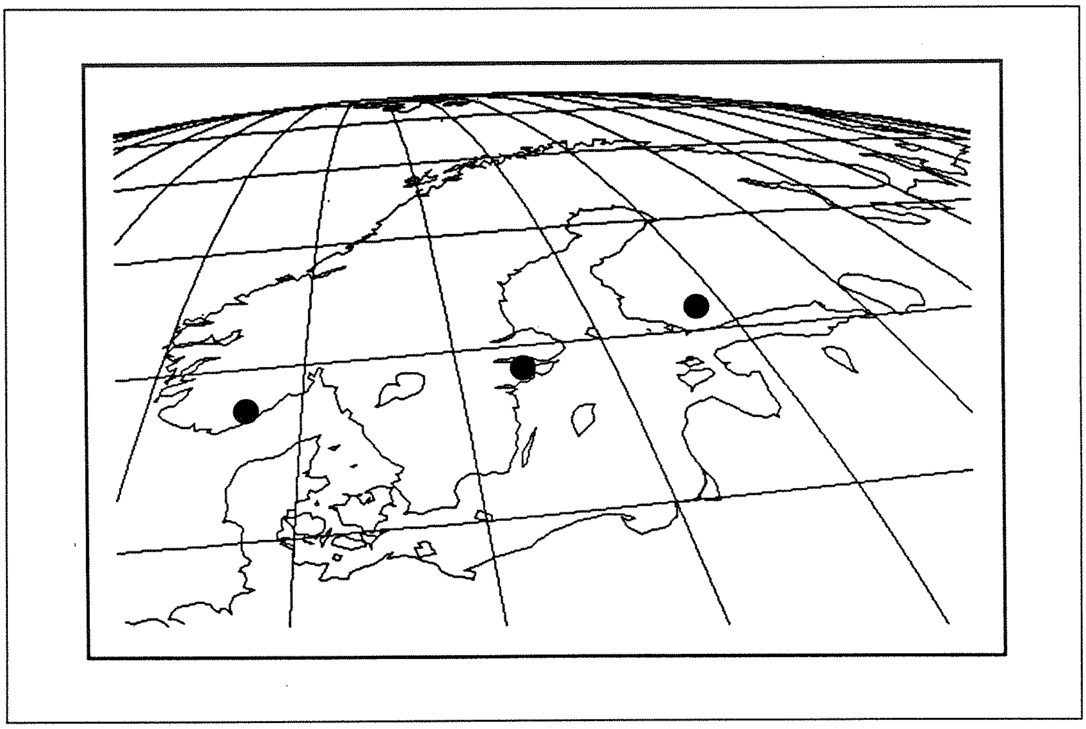
Dynamic models for predicting soil and water acidification: Application to three catchments in Fenno-Scandinavia



Norwegian Institute for Water Research NIVA

NIVA – REPORT

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Dynamic models for predicting soil and water acidification: Application to three catchments in Fenno-Scandia

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Abstract:

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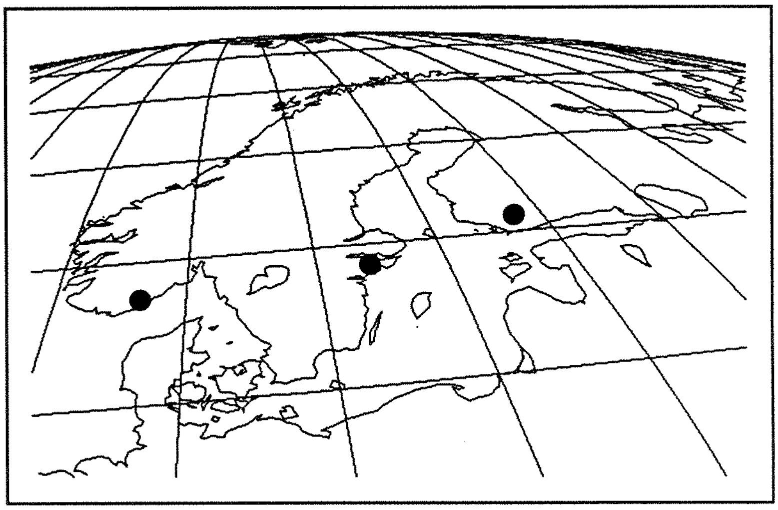

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ABSTRACT

Prediction of future acidification as a result of changes in emissions and deposition necessitates the use of models. We report here the application of 4 dynamic process-oriented models of soil and water acidification to data from three calibrated catchments: Birkenes, an acidified site in southernmost Norway, Stubbetorp, a transitional site in east-central Sweden, and Yli-Knuutila, a well-buffered site in southernmost Finland. We calibrate the models to the observed conditions at present-day, and then use the calibrated models to predict acidification over the next 50 years given 4 different scenarios of future acid deposition. The model predictions are compared and the differences evaluated with respect to the use of these models in determining critical loads.

All models yield similar estimates for pre-acidification soil and water chemistry in 1840. They also produce comparatively similar hindcasts over the period 1840-1988. The models all indicate that (1) at Birkenes >55% reduction in sulfur deposition is required to reverse soil and streamwater acidification; (2) at Stubbetorp, although the stream currently has pH 6.0 and positive alkalinity, major reduction in sulfur loading is required to prevent acidification in the future; (3) at Yli-Knuutila the thick soils with high base saturation can protect against acid deposition at present-day rates for many decades into the future.

Critical loads for sulfur calculated by these dynamic models are roughly similar for Birkenes and Stubbetorp. The level to which deposition must be reduced (target load) depends not only on the criterion used but also on the number of years in the future for which this criterion is to be satisfied. Streamwater (using the criterion of alkalinity) is generally more sensitive than soils (using the criterion of Ca/Al in soil solution). At Yli-Knuutila the critical load for the uppermost soil horizons may be substantially lower than for the soil and catchment as a whole. Here models with multiple soil layers such as SAFE are appropriate.

PREFACE

This report covers work conducted in 1990-91 under the "Scenario project" funded by the Nordic Council of Ministers Air Group. Participating in the Scenario project are: R.F. Wright (Norway, project leader), M.Holmberg (Finland), M. Posch (Finland), and P. Warfvinge (Sweden). Here we report results of model applications to three calibrated catchments in Fenno-Scandia. We have divided the work as follows: M. Holmberg ran the MIDAS model and worked out the uptake sequences for Yli-Knuutila. M. Posch ran the SMART model. P. Warfvinge ran the SAFE model and worked out the uptake and deposition sequences for Stubbetorp. R. F. Wright ran the MAGIC model and edited the report. We thank B. Andersen (COWI-consult, Denmark) for providing the hindcast scenario for deposition at Stubbetorp, P. Grennfelt (IVL, Sweden) for providing the two scenarios for future deposition of sulfur and nitrogen, J. Kämäri (VYL, Finland) for providing unpublished water data from Yli-Knuutila and for assisting in the application of SMART, L. Maxe (KTH, Sweden) for providing unpublished data from Stubbetorp, and A. Nissinen (Univ. Helsinki) for providing unpublished soils data for Yli-Knuutila.

1. INTRODUCTION

Acid deposition, acidification of soils and surface waters, and the consequent biological effects are widespread phenomena in Fenno-Scandia. Efforts to mitigate acidification include various Nordic and European initiatives to reduce the emissions of acidifying compounds SO_2, NO_x and NH_3. These efforts are based on the premise that reduction in emissions and hence acid deposition will result in recovery of acidified ecosystems and the protection of other ecosystems currently threatened by acid deposition.

Prediction of future acidification as a result of changes in emissions and deposition necessitates the use of models. Several acidification models are available. These include simple static empirical models and more complex dynamic process models.

The concept of critical load is now widely used as a criterion for decisions regarding future emissions of acidifying gases. The critical load for surface waters is defined as "A quantitative estimate of the loading of one or more pollutants below which significant harmful effects on specified sensitive elements of the environment are not likely to occur according to present knowledge" (Nilsson and Grennfelt, 1988).

Procedures for determination of critical load for a given ecosystem or region entails the use of models. One category of models to be used in mapping critical loads are dynamic process-oriented models (Sverdrup et al., 1990).

We report here the application of 4 dynamic process-oriented models of soil and water acidification to data from three catchments in Fenno-Scandia: Birkenes (Norway), Stubbetorp (Sweden), and Yli-Knuutila (Finland). These sites were chosen because of the availability of extensive high-quality sets of data for deposition, soils and surface waters, and because they span a range in present-day acidification status. Birkenes is a highly acidified site in southernmost Norway (stream pH 4.5 and alkalinity -55 μeq/l). Stubbetorp in east-central Sweden receives acid deposition but still has pH 6.0 and alkalinity 74 μeq/l. Yli-Knuutila in southernmost Finland is well-buffered with pH 6.0 and alkalinity 189 μeq/l.

We calibrate the models to the observed conditions at present-day, and then use the calibrated models to predict acidification over the next 50 years given 4 different scenarios of future acid deposition. The model predictions are compared and the differences evaluated with respect to the use of these models in determining critical loads. This work was conducted as part of the activities supported by the Nordic Council of Ministers Air Group.

2. MODEL DESCRIPTIONS: MAGIC, MIDAS, SAFE & SMART

Four models, which are available for use in Sweden, Norway and Finland, were chosen for the scenario comparison: MAGIC (Cosby et al., 1985a, 1985b), MIDAS (Holmberg et al., 1989), SAFE (Sverdrup et al., 1987, Warfvinge, 1988) and SMART (de Vries et al., 1989).

All these models are process-oriented dynamic models that attempt to describe the long-term impact of atmospheric deposition, net uptake by vegetation, weathering and cation exchange on the chemical composition of soil and the outflowing water. In all the models solution chemistry is governed by charge and mass balance principles (Table 1).

Table 1. Characteristics of the models.

MODEL	MAGIC	MIDAS	SAFE	SMART
Soil layers	one	one	three	one
Cation exchange	Gaines-Thomas equilibrium	Gaines-Thomas kinetic	Gapon mass-transfer	Gaines-Thomas equilibrium
Aluminum in solution	equilibrium	equilibrium	equilibrium	equilibrium
Sulfate adsorption	Langmuir	-	-	Langmuir
Weathering	input	input	kinetic	input
Nitrification	input	-	kinetic	input
Nitrogen uptake	input	-	input	input

Application of the models at the catchment scale requires lumping chemical and physical properties of the soil profile into one or several homogeneous layers characteristic for the entire catchment. For these applications yearly time-steps were used. Historical atmospheric deposition and net uptake of major ions and the future scenarios are calculated independent of the models.

MAGIC is generally used as a one-box model (Cosby et al., 1985a, 1985b). MAGIC calculates a separate Gaines-Thomas equilibrium for the exchange of each of the cations Al, Ca, Mg, K, and Na. Sulfate adsorption is described by a Langmuir isotherm. MAGIC keeps track of mass budget and chemical equilibria of all major ions including organic acids.

MIDAS (Holmberg et al., 1989) uses a kinetic expression for the ion exchange, described in equilibrium by the Gaines-Thomas equation. The model works with net acidity (trivalent in the cation exchange) and the equivalent sum of calcium and magnesium. Soil solution pH and [Al^{3+}] are calculated using forcing functions to produce equilibrium.

SAFE (Sverdrup et al., 1987, Warfvinge, 1988) uses PROFILE as a subroutine for weathering rates and initial conditions. Weathering rates are calculated from measurements of soil texture, mineralogy and moisture. The model uses a mass-transfer equation for the ion exchange of base cations, which converges upon an equilibrium described by the Gapon equation.

SMART (de Vries et al., 1989) works with divalent base cations, [H^+], [Al^{3+}], sulfate, nitrate, ammonium and bicarbonate in solution. Gaines-Thomas equilibrium equations govern Al/BC and H/BC exchange.

3. SITE DESCRIPTIONS

3.1 Birkenes

The Birkenes catchment is 0.41 km^2, located in southernmost Norway about 20 km inland from Kristiansand (Table 2). The catchment is dominated by 80-year-old Norway spruce (Picea abies L.). A small mire occupies part of the catchment. The soils are mainly podzols and brown earths developed on stony moraine on granitic bedrock. Peaty soils are located along the stream channels.

In 1971 the Birkenes catchment was instrumented for input-output budgets, including a weir and level recorder at the outlet and a complete air and precipitation station to measure inputs. The catchment has been monitored now for nearly 20 years, first as part of the SNSF-project and since 1980 as part of the Norwegian monitoring program for long-range transported pollutants under the auspices of the Norwegian State Pollution Control Authority (SFT 1986). Results of the monitoring activities are published annually in SFT's report series (SFT 1991).

Table 2. Measured soil and catchment characteristics (aggregated).

	unit	Birkenes	Stubbetorp	Yli-Knuutila
Area	km^2	0.41	0.9	0.07
Temperature	^oC	5	7	5
Depth	m	0.4	0.8	1.5
Porosity	%	0.50	0.55	0.45
Bulk density	kg m^{-3}	936	1300	1480
CEC	meq kg^{-1}	46	10	124
%Ca		9.0	28.0	59.0
%Mg		4.5	5.0	36.0
%Na		1.6	3.0	1.0
%K		2.7	1.0	1.9
%BS		17.8	37.0	97.9

Birkenes is located in the zone of maximum acid deposition in Norway. The site receives about 161 meq/m^2/yr SO_4, 96 meq/m^2/yr NO_3, 100 meq/m^2/yr NH_4, and 117 meq/m^2/yr H^+ of which about 3/4 comes as wet and 1/4 as dry deposition (Table 3) (SFT 1991). Streamwater is highly acidified and has a volume-weighted mean pH of about 4.5, high levels of inorganic aluminum and sulfate as the major anion (Table 3) (SFT 1991). The streamwater is toxic to fish; the last reported sighting of brown trout in the stream was in about 1950; the lake downstream is also barren of fish.

Birkenes is one of 20 sites in Norway for intensive-monitoring of forest health. Needle loss in spruce at Birkenes is moderate, but the 3-year data series is too short in duration to indicate whether this needle loss reflects forest damage or merely a natural phenomenon.

Table 3. Deposition (wet + dry) and runoff fluxes of major ions at Birkenes, Stubbetorp and Yli-Knuutila. Units: fluxes meq m^{-2} yr^{-1}; concentration ueq l^{-1}. HCO_3^- is measured as titrated (Gran) alkalinity. SBC = Ca + Mg + Na + K + NH_4. SAA = SO_4 + NO_3 + Cl. Alk = SBC - SAA. Organic anions (A^-) calculated as Alk + H^+ + Al^{n+} - HCO_3^-.

	Birkenes (1973 - 1988)			Stubbetorp (1986-1988)			Yli-Knuutila (1984-1988)		
	Input	Output		Input	Output		Input	Output	
	flux	flux	conc	flux	flux	conc	flux	flux	conc
mm	1471	1190		680	280		613	172	
H^+	117	35	30	51	0	1	42	0	1
(pH)			4.5			5.9			6.0
Ca^{2+}	22	66	56	7	41	148	32	69	400
Mg^{2+}	28	41	35	5	20	73	6	50	290
Na^+	128	135	113	17	29	102	16	29	170
K^+	10	7	6	2	4	16	6	6	37
NH_{4+}	100	1	1	30	0	0	33	1	7
SO_4^{2-}	161	160	134	53	52	184	90	91	530
Cl^-	146	146	123	21	21	75	16	16	94
NO_3^-	96	10	8	38	1	2	29	16	91
HCO_{3-}	0	0	0	0	13	46	0	14	81
A^-	0	41	35	0	7	32	0	18	108
SBC	288	250	210	61	94	339	93	155	904
SAA	403	316	265	112	74	261	135	123	715
Alk.	-115	-66	-55	-51	20	78	-42	32	189

3.2 Stubbetorp

The Stubbetorp catchment (0.9 km^2) is situated in Kolmården; a hilly, forested area on the Swedish east coast, approximately 150 km south of Stockholm (Table 2). It was established as a research basin by the Swedish Meteorological and Hydrological Institute (SMHI) in 1983 when precipitation measurements and water chemistry sampling in the main stream were initiated. In 1985 a weir was constructed for the measurement of runoff.

The catchment is drained by a small stream. The runoff is usually large during snowmelt and the autumn rains. During most years there is low flow even during summer, but in the summers of 1983, 1989 and 1990 the stream was completely dry for several months. The streamwater is normally well-buffered, but during periods of high flow there is a drop in pH and alkalinity. Seasalt ions (Cl, Na, and Mg) are high due to the proximity to the Baltic Sea. Streamwater chemistry is otherwise characterized by sulfate as dominant anion, and calcium and magnesium as major cations (Table 3).

Stubbetorp is dominated by bedrock of gneissic granites with generally thin overburden comprised of stony till. The overburden has been depleted of fine material as a result of wave action during the emergence of the land in the late post-glacial period. Soil cover is patchy on the hilltops. Sand deposits occur in depressions and in the main valley. The soils in the catchment are therefore rather permeable. Part of the water flow in the catchment occurs through fractures in the bedrock. Today the relief in the catchment is from 130 m a.s.l. to 80 m a.s.l. at the weir.

Stubbetorp is 83% covered with productive forest of Scots pine (84%), while Norway spruce (13%) and deciduous tree species (3%) are less important. The remaining area (9%) is impediment and small mires (7%).

The soils are mostly podzolic and brown forest soils. The chemistry of the upper soil (O-, A- and B-horizons) has been surveyed at about 90 points covering the area while samples at greater depths (>0.8 m) have only been taken at 4 sites. Both the physical and chemical properties of the soil vary strongly with depth and also show a large spatial variation. The measured soil properties have been aggregated to mean values for the catchment (Table 3).

Because of the large area with little or no soil cover, the estimated mean soil depth of the catchment is only 0.8 m. the uncertainty in the estimated soil depth may be large (approx. +/- 30%). Due to the large interdependence of the soil parameters with depth this uncertainty will also affect the other parameters.

3.3 Yli-Knuutila

Yli-Knuutila, Vihti, is a 0.07 km^2 forested catchment in southern Finland, about 40 km NW of Helsinki. The site is operated by the Water and Environment Research Institute. Hydrological observations began in 1953 (Seuna 1983). Sampling for streamwater quality has been carried out since 1963, following various routines. Bulk deposition is collected monthly. Throughfall measurements were conducted in 1987 and 1988.

The catchment is forested with spruce and pine of average age 100 years. Current standing stock is 273 m^3/ha. The stands have been managed by thinning. The entire catchment will be clearcut in 1991. Soils at Yli-Knuutila are podzols and brown earths developed on clays and sands of average depth 1.5 m (Table 2).

The area receives a rather high input of sulfur and nitrogen. Total sulfur deposition (wet and dry) is estimated at 90, total NO_3 at 29, and NH_4 at 33 meq/m^2/yr, respectively (Kallio and Kauppi 1990) (Table 3).

Streamwater has high concentrations of most ions, due in part to the large evapotranspiration rate relative to precipitation. Major cations are calcium and magnesium balanced by alkalinity and sulfate (Lepistö et al. 1988). Despite the high alkalinity pH is only 6.0, due to the buffering affect of organic acids. The stream also has relatively high concentrations of nitrate, and in this respect is unusual for the 3 sites studied here (Table 3).

4. DEPOSITION AND NUTRIENT UPTAKE SCENARIOS

Four scenarios for future deposition and uptake of sulfur and nitrogen have been employed in this study (Figure 1). The values of SO_4, NO_3, and NH_4 deposition for the years 1840 to 1987 are derived from historical emission estimates. Three alternatives for future deposition of sulfate and nitrate were used: 1) no reductions 1988-2040 (base case); 2) minimum reductions i.e. linear decrease by 30% for sulfate and 10% for nitrate from 1988 to 2005 and constant thereafter (30/10 case); 3) maximum reductions i.e. linear decrease by 55% for sulfate and 40% for nitrate from 1988 to 2005 and constant thereafter (55/40 case). Future ammonium and base cation deposition was assumed constant in all cases.

Historical rates of net uptake of nitrogen and base cations in biomass are estimated for each site. At Birkenes net uptake of NH_4 and NO_3 are assumed to be 99% and 90% of deposition, respectively. Net uptake of base cations is assumed to be zero except for K, which is set at 7 $meq/m^2/yr$. These values were chosen such that deposition minus uptake equals measured present-day flux in streamwater (Figure 2).

All these three deposition scenarios were used assuming that future uptake of nitrate was unchanged relative to the present-day. In addition the maximum reduction scenario (55/40 case) was combined with a future nitrate uptake scenario in which the leakage of nitrate to streamwater increases exponentially from the present-day until "nitrogen saturation" is reached, at which point nitrate flux in runoff equals nitrate deposition (55/40 + N-leach case) (Figure 2).

At Stubbetorp a more rigorous approach was taken. Here net uptake of base cations is based on growth rates and stand history. The present day uptake rate was calculated on the basis of literature values for percentages of Ca, Mg and K in stems and branches. The historical uptake pattern, based on the age distribution, was scaled to present-day uptake. Net uptake of nitrogen compounds was again set as a percentage of deposition (99% for NH_4 and 98% for NO_3). Future uptake was estimated by this same procedure under the assumption that the forest practices in the future would continue at the same

Deposition at Birkenes

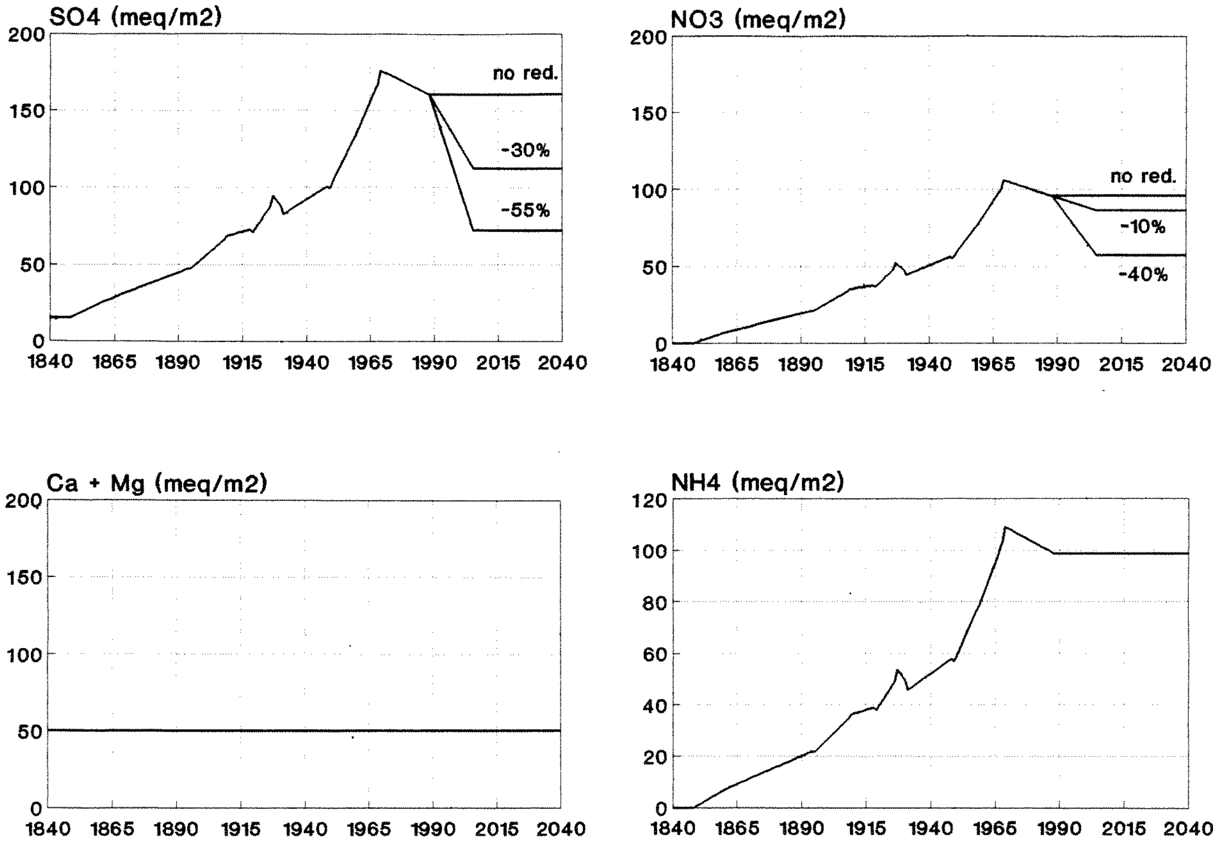


Figure 1. Deposition of SO_4, NO_3, NH_4, and Ca+Mg at Birkenes, Stubbetorp and Yli-Knuutila. Shown are estimated historical deposition patterns estimated from European emissions of SO_2, NO_x, and NH_3 and including both wet and dry deposition. Three scenarios for future deposition for SO_4 and NO_3 are used: no reduction from present-day (no red.), minimum reduction to year 2005 and constant thereafter (min. red.), and maximum reduction to 2005 and constant thereafter (max. red.). Dry deposition rates vary depending on degree and age of forest cover. The deposition patterns for Cl and Na+K (not shown) are similar to that of Ca+Mg. Units: $meq/m^2/yr$.

Deposition at Stubbetorp

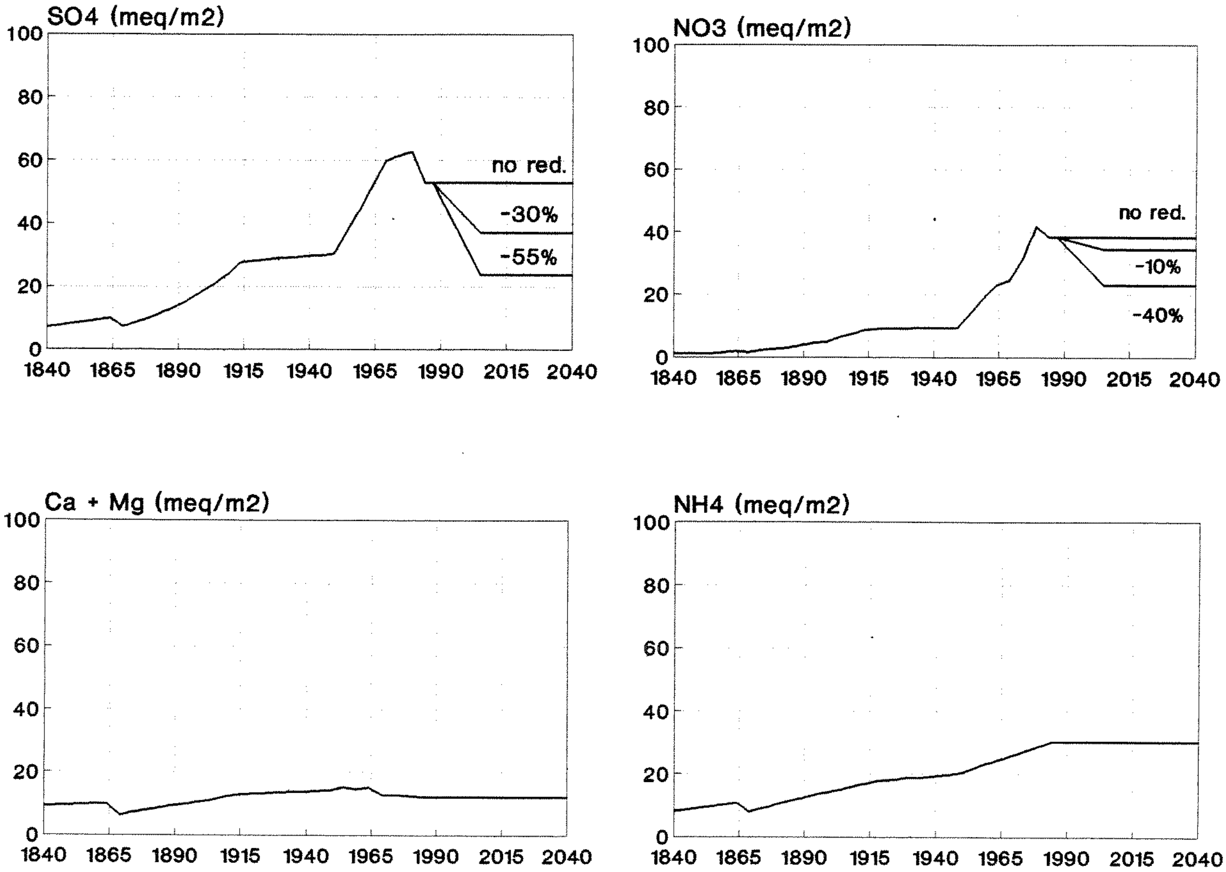
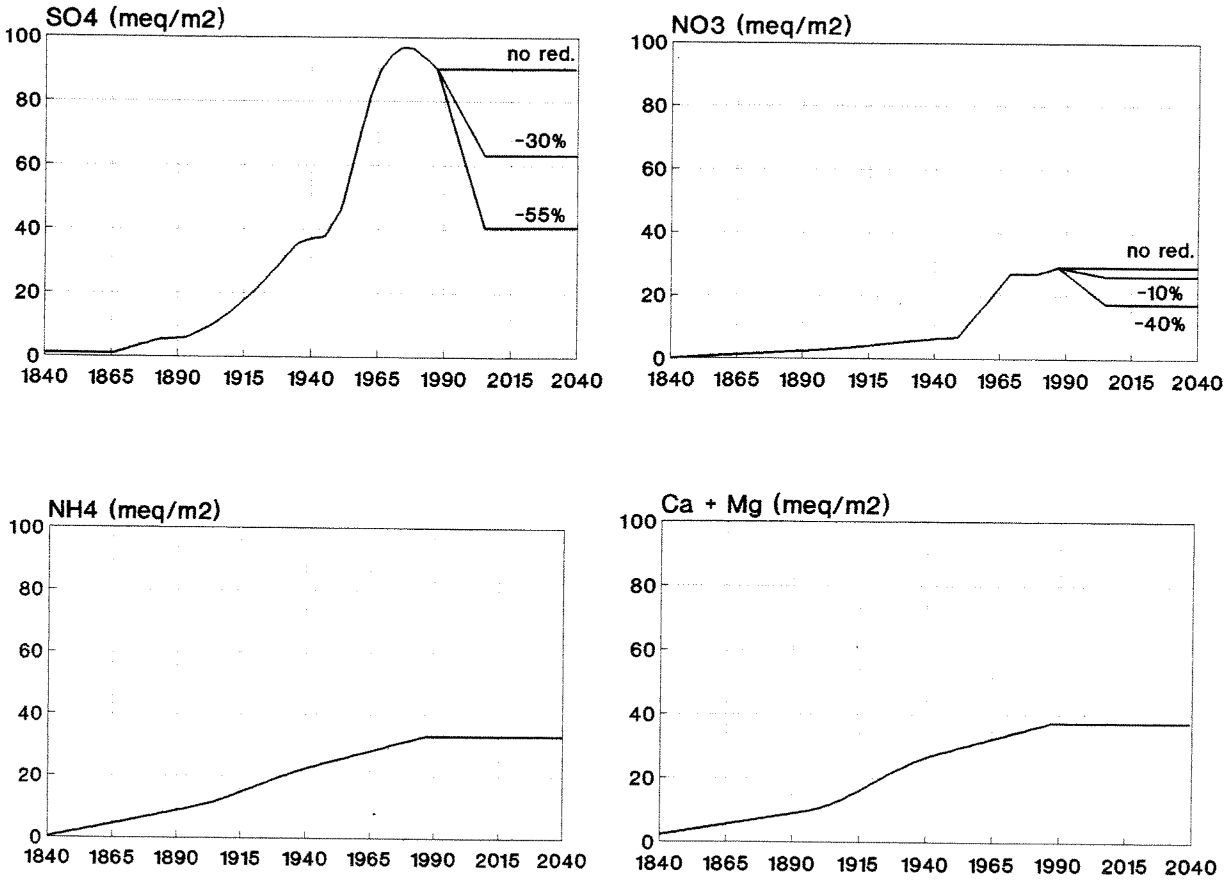


Figure 1 (cont.)

Deposition at Yli-Knuutila



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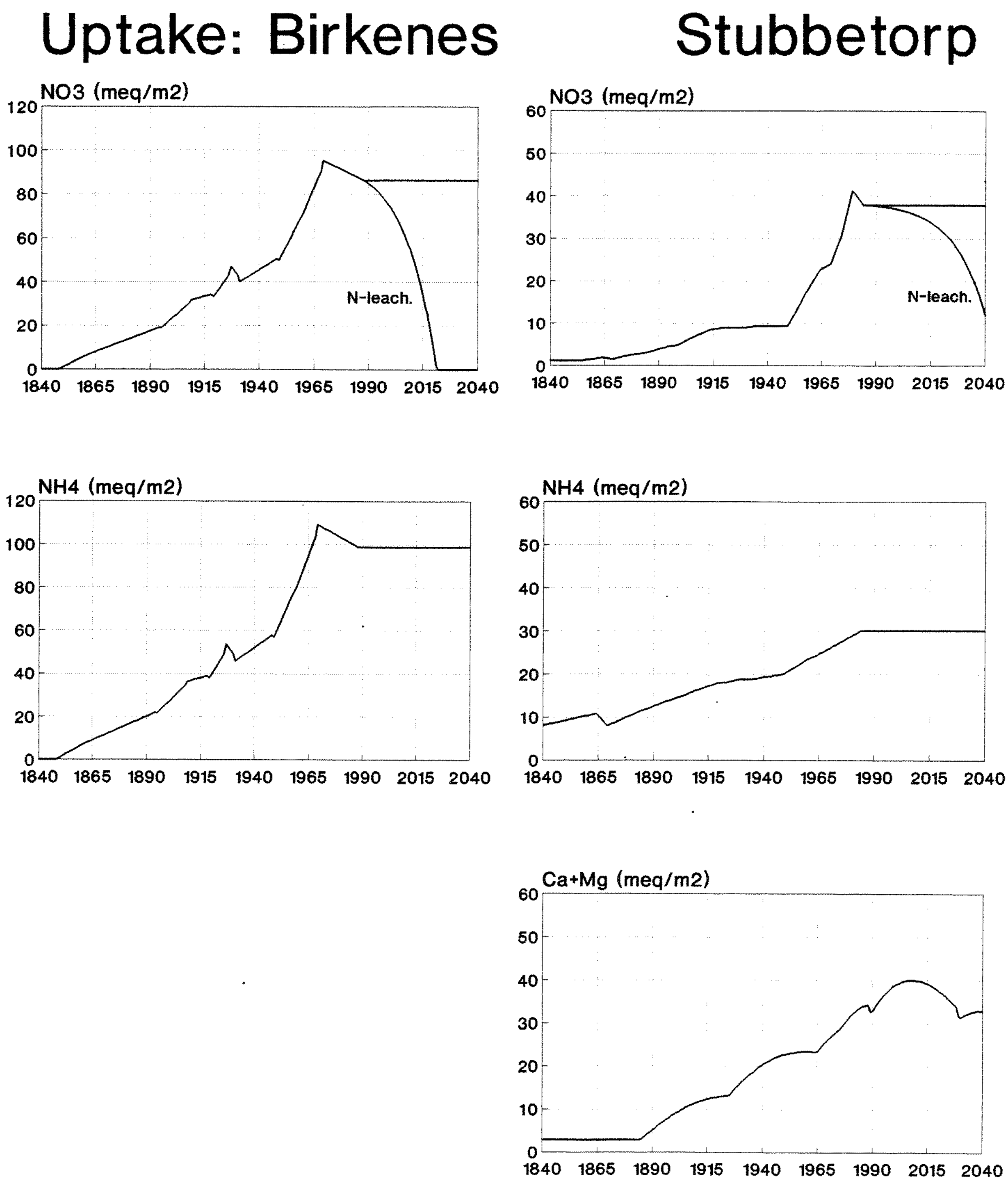

Figure 2. Net uptake of NO_3, NH_4, and Ca+Mg at Birkenes, Stubbetorp and Yli-Knuutila. At Birkenes net uptake of K is similar to that for NH_4. At Stubbetorp net uptake of K and SO_4 is similar to that of Ca+Mg. At Yli-Knuutila uptake of K is similar to that for Ca+Mg. For all other ions at these sites the net uptake is assumed to be zero. Units: meq/m^2/yr.

Uptake at Yli-Knuutila

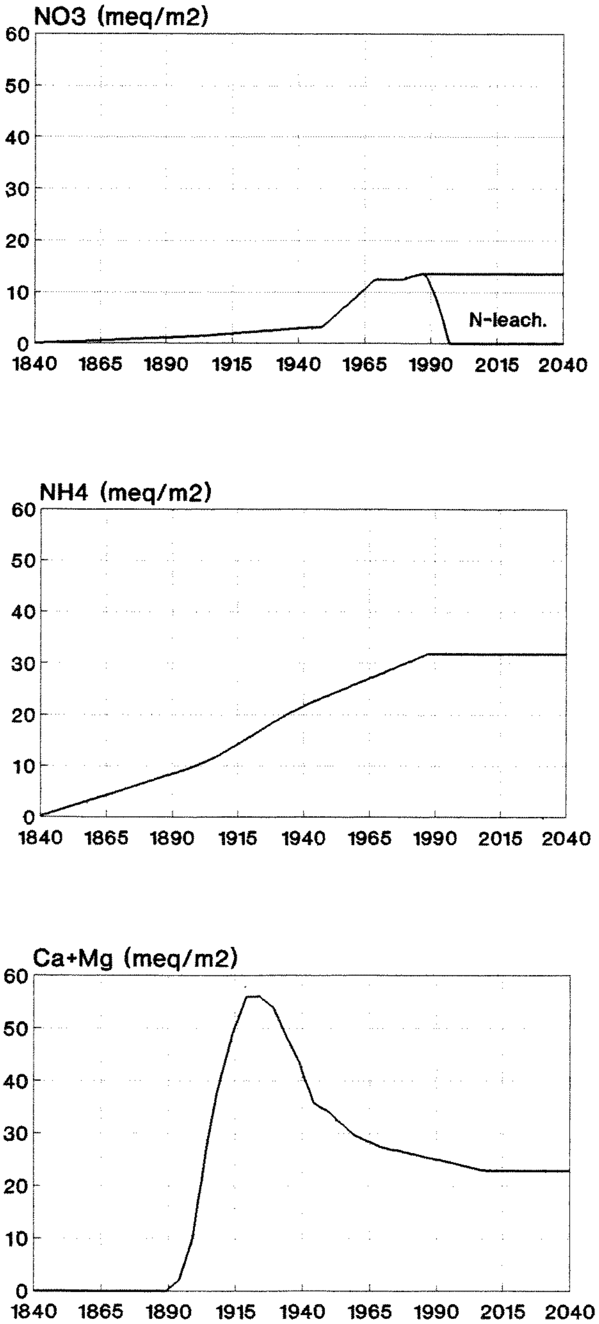

Figure 2 (cont.)

intensity as in the past with trees harvested upon maturity at 80 years and the area replanted.

This rigorous approach was also used at Yli-Knuutila. Again the net uptake of base cations and nitrogen in biomass was calculated from estimates of forest growth and biomass removal at the site. Measurements of net uptake and rates of forest growth were taken from studies at similar sites elsewhere in Finland (Kubin 1984, Vuokila and Väliaho 1980). Future uptake was calculated in the same manner assuming that the forest remains untouched for 50 years into the future (ie. no clearcut in 1991).

5. RESULTS

5.1 Calibration

The models were calibrated to reproduce present-day (1987-88) soil and streamwater chemistry. The historical deposition and uptake patterns were used to drive the models and produce the chemical changes in soil and streamwater over time. The models each require an estimate of weathering rates for base cations, and these are obtained by either trial and error or in the case of SAFE from information on soil texture and mineralogy. The models were calibrated independently of one another but all using a common set of data for each catchment.

5.2 Model comparisons

Several key parameters are used to compare the models. For soils base saturation is the key measure of acidification. To compare the models we use %Ca+%Mg saturation; the models all provide outputs of this parameter (SAFE gives %Ca+%Mg+%K). Typically %K+%Na comprises only a small fraction of base saturation, and does not exhibit major change over time with acidification relative to %Ca+%Mg.

For streamwaters we use pH and alkalinity. These are key parameters with respect to acidification and effects to aquatic organisms and are outputs produced by the models.

We define alkalinity as the sum of base cations minus sum of strong acid anions. This definition is now widely used in critical load work.

For all 3 sites the calibrated models yield similar estimates for pre-acidification soil and water chemistry in 1840 (Figures 3-5). They also produce comparatively similar hindcasts over the period 1840-1988. The models all suggest that the sites have undergone soil and water acidification over the past 140 years, and the models are in agreement as to the magnitude and timing of this acidification.

Weathering rate is a central parameter determining catchment sensitivity and critical load. Weathering rate is notoriously difficult to determine directly. SAFE calculates weathering rates from soil mineralogy, texture and moisture. Rates are 13, 68 and 71 meq m^{-2} yr^{-1} for Birkenes, Stubbetorp, and Yli-Knuutila, respectively. These values are similar to those determined by MAGIC of 20, 39 and 54 meq m^{-2} yr^{-1}.

Of greater interest, of course, is the ability of the models to predict future acidification of soil and water given various scenarios of future acid deposition. For the 3 sites examined here all models predict the lowest base saturation in soil and the lowest pH and alkalinity of streamwater in 2040 for the base case (Figures 3-5). All models predict that Birkenes will continue to acidify unless sulfate deposition is reduced by >55%, that Stubbetorp is threatened by acidification at present-day levels of sulfate deposition, and that Yli-Knuutila can tolerate >50 years of acid deposition at current rates without experiencing adverse effects.

5.3 Scenario comparisons

At each catchment the predicted response of soil and streamwater chemistry varies depending on the scenario for future deposition of sulfur and nitrogen and future nitrogen leaching. At Birkenes, for example, the soil and streamwater continue to acidify under the no reduction, minimum reduction and N leaching scenarios, and only with the maximum reduction does the acidification stabilize in the future (Figure 6). All the models predict this general response to the 4 scenarios at Birkenes.

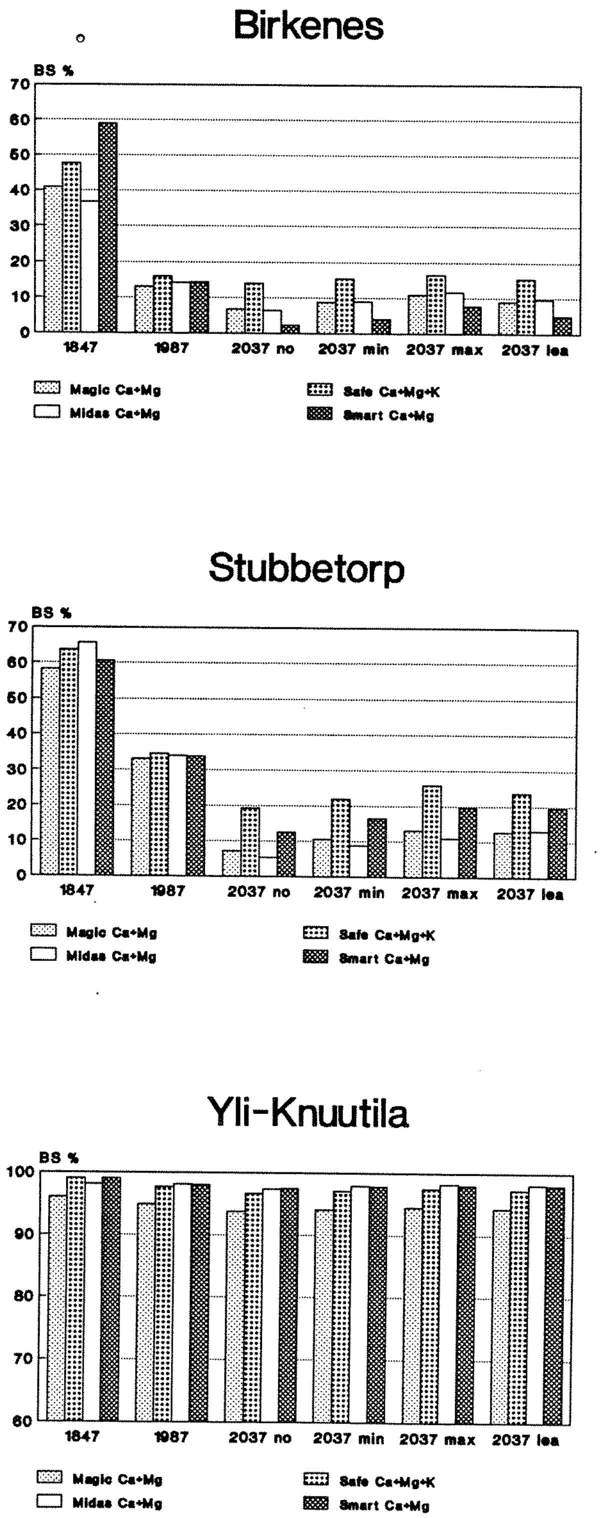

Figure 3. Reconstructed and predicted soil base saturation (%) using 4 acidification models given 4 different scenarios for future deposition and nitrogen leaching.

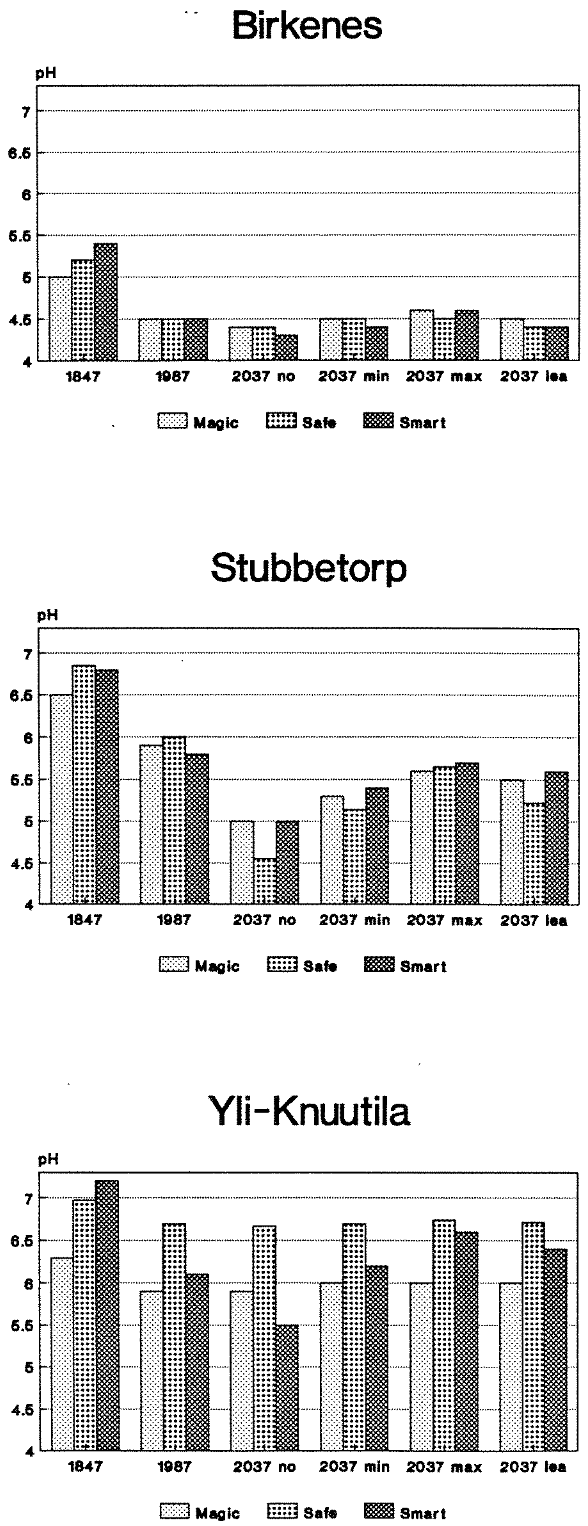

Figure 4. Reconstructed and predicted streamwater pH using 3 acidification models given 4 different scenarios for future deposition and nitrogen leaching.

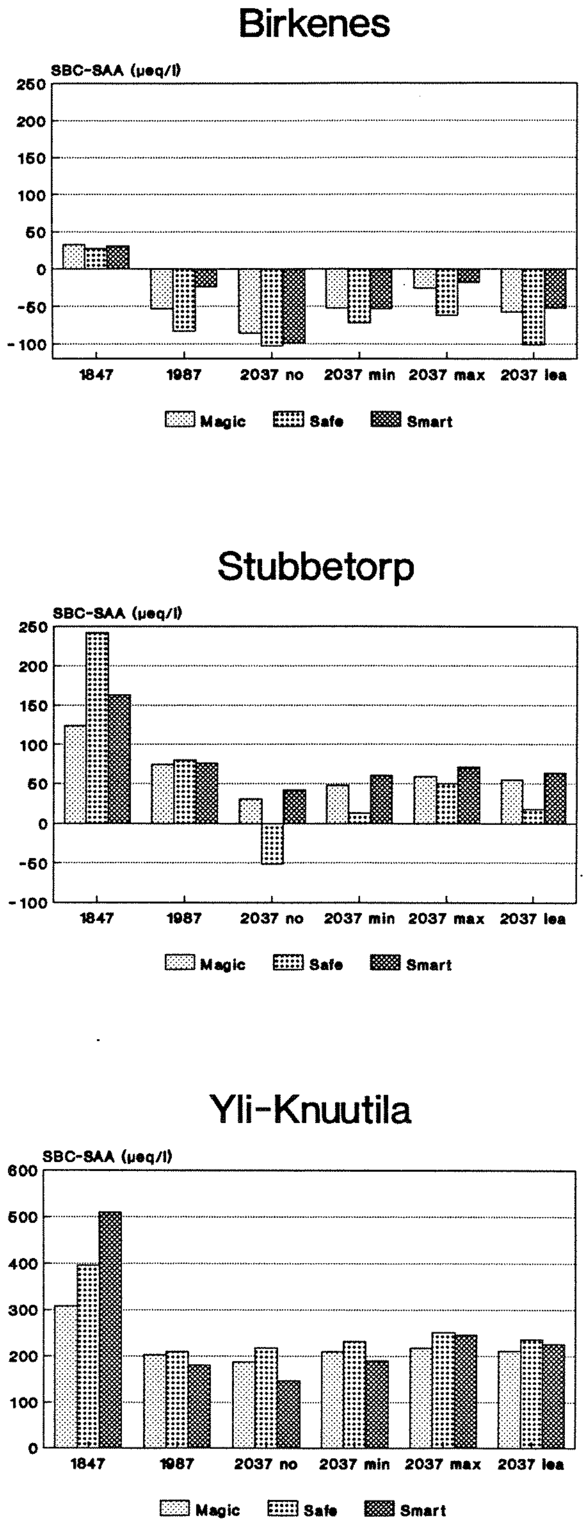

Figure 5. Reconstructed and predicted streamwater alkalinity (defined as SBC-SAA) using 3 acidification models given 4 different scenarios for future deposition and nitrogen leaching.

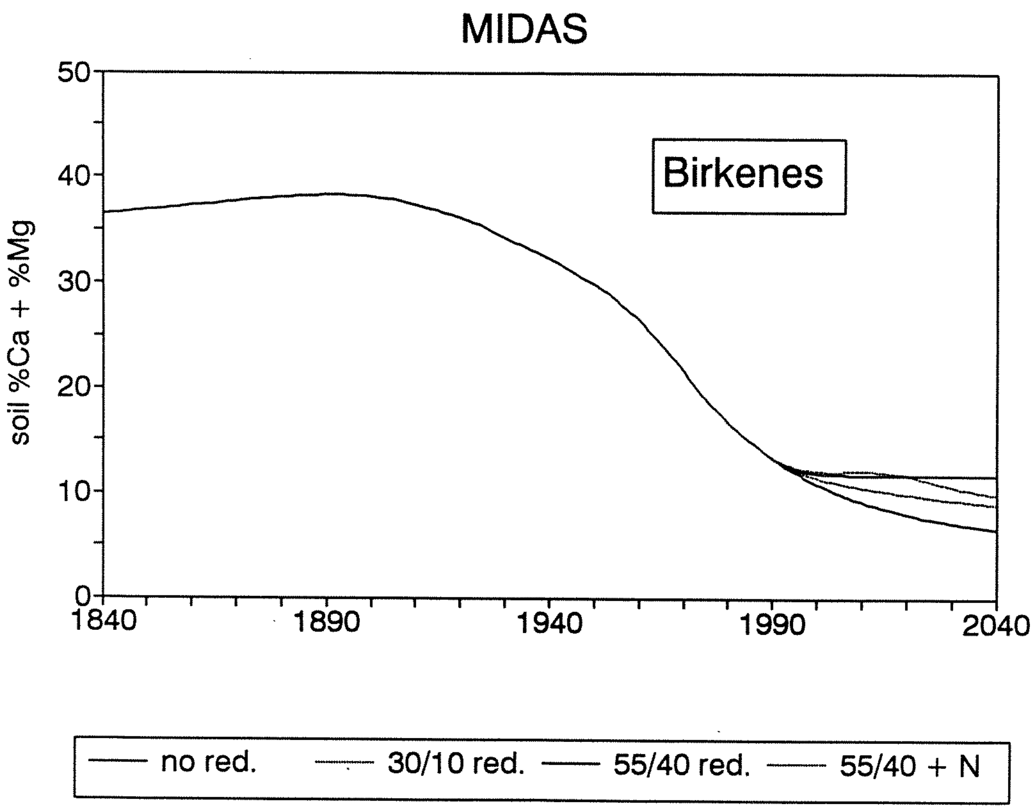

Figure 6. Birkenes. Soil base saturation (%) as calculated by MIDAS under 4 different scenarios for future deposition and nitrogen leaching.

Similarly at Stubbetorp the models all predict continued decrease in base saturation under all 4 scenarios (Figure 7a). A 55% reduction of sulfur deposition is insufficient to halt the ongoing soil acidification. The models also predict that pH and alkalinity of streamwater will continue to decline under all scenarios (Figure 7b). Alkalinity declines from present-day levels of 74 μeq/l but remains above zero for the next 50 years even under the no reduction scenario.

At Yli-Knuutila the soils have very high base saturation (>95%) and thus are well-buffered against acid deposition. No significant change in soil acidification is predicted for the future at Yli-Knuutila even with no reduction in sulfur deposition. Similarly the streamwater is well-buffered with measured present-day alkalinity of about 180 μeq/l.

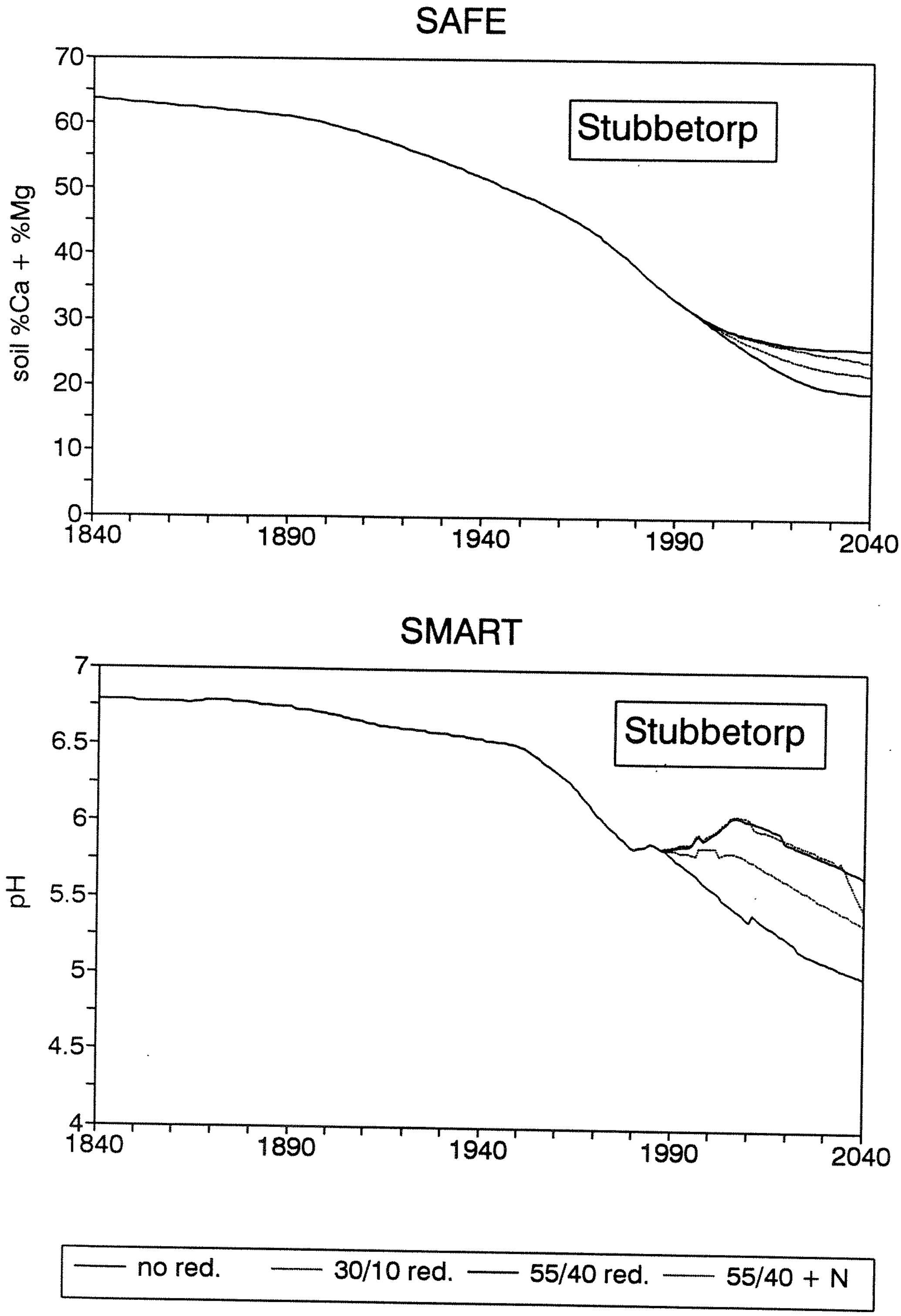

Figure 7. Stubbetorp. Soil base saturation (%) (top panel) as calculated by SAFE and stream pH as calculated by SMART (bottom panel) under 4 different scenarios for future deposition and nitrogen leaching.

Neither pH nor alkalinity is predicted to decrease to levels at which biological damage might be expected (Figure 8).

The 3 sites clearly differ in their present-day acidification status. Birkenes has highly acidic streamwater with pH 4.5 and alkalinity of -55 μeq/l, Stubbetorp is intermediate with pH 5.9 and 74 μeq/l alkalinity, and Yli-Knuutila is well-buffered with pH 6.0 and 180 μeq/l alkalinity (Figure 4).

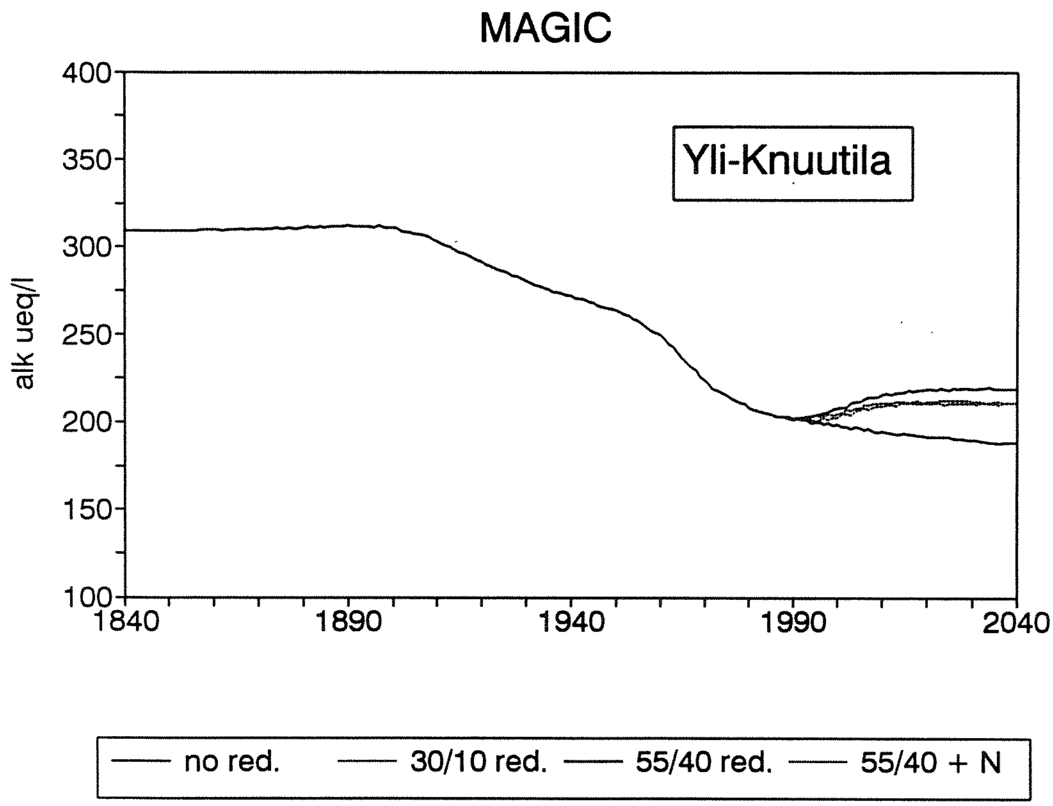

Figure 8. Yli-Knuutila. Streamwater alkalinity (μeq/l) as calculated by MAGIC under 4 different scenarios for future deposition and nitrogen leaching.

The differences in present-day streamwater chemistry reflect both inherent differences in catchment soils but also differences in amounts of acid deposition. Birkenes has the thinnest soils (0.4m), Stubbetorp is intermediate (0.8m), and Yli-Knuutila has the thickest (1.5m). Birkenes has the highest present-day loading of sulfate (161 meq/m^2/yr), about 3 times greater than Stubbetorp (53 meq/m^2/yr) and also higher than Yli-Knuutila (90 meq/m^2/yr). Thus although the soils at Birkenes and Stubbetorp started in pre-acidification times (1840) with roughly similar base saturation (Birkenes 45-65%, Stubbetorp 60-70%), the thinner soils and higher acid loading during the past 140 years caused Birkenes to loose a greater fraction of the pool of base cations in the soils. Stubbetorp is thus on the "Birkenes" path, but apparently has not progressed to the point at which alkalinity becomes negative (Figure 3).

5.4 Site comparisons

With this background it is perhaps not surprising that the predictions for Birkenes and Stubbetorp are quite similar. Both catchments require >55% reduction in sulfur loading to permit recovery. Yli-Knuutila is so well-buffered that although the soils will continue to loose base cations in the future, the change will be so slow that neither soil or streamwater appears threatened at current rates of acid loading.

5.5 Critical loads

Several of these models can be used to calculate critical and target loads for the 3 sites. Values can be estimated for critical load for soils and for surface waters. For soils we use the suggested criterion of Ca/Al molar ratio in soil solution, and for surface water the criterion of alkalinity (Sverdrup et al. 1990). We choose Ca/Al limits of 0.5 and 1 mol/mol, and alkalinity limits of 0, 20, and 50 μeq/l. These critical load estimates can be compared with the deposition under the 4 different scenarios to determine the exceeded amount.

Two factors have caused historical changes in the soils that have influenced the present-day acidification status. Acid deposition (Figure 1) has resulted in soil acidification -- a depletion in base saturation -- but also forest practices in the catchments may be responsible for removal of base cations and soil acidification (Figure 2). These factors are cumulative. Because of the soil acidification in the past, the ecological imapct of a given load of acid deposition is greater today.

The models demonstrate that the soils and streamwaters respond in a dynamic way to changes in deposition. The load a catchment can tolerate thus depends on the number of years for which the response can be awaited. For example, Stubbetorp can tolerate a much higher loading for 5 years than it can for 50 years. For our estimates of critical loads we set as a goal to maintain for 50 years into the future stream alkalinity above 0, 20 or 50 μeq/l and the molar Ca/Al ratio in soil solution above 0.5 or 1.

Under these conditions the critical load for sulfur at Birkenes for streamwater is about 40-50 meq/m^2/yr for alkalinity >0 and 140-160 meq/m^2/yr for Ca/Al > 1 (as calculated by both MAGIC and SMART) (Table 4, Figure 9a). If nitrate leaches as in the "nitrogen saturation" scenario, the critical load for sulfur is lower -- 0-10 meq/m^2/yr for alk >0 and 110-120 for Ca/Al >1 (Table 4).

Table 4. Critical load for sulfur (meq SO_4 m^{-2} yr^{-1}) calculated by the models under the condition that the criterion is met within 50 years. Criteria are streamwater alkalinity (SBC-SSA) (ueq l^{-1}) and molar ratio of Ca/Al in soil solution.

	Birkenes					
	MAGIC no red	MAGIC N-sat.	SAFE no red	SAFE N-sat	SMART no red	SMART N-sat
alk > 0	48	< 0	13	<0	44	10
alk > 20	< 0	< 0	<0	<0	< 0	< 0
alk > 50	< 0	< 0	<0	<0	< 0	< 0
Ca/Al > 1	161	121	145	118	142	110
Ca/Al > 0.5	250	204	230	188	200	160

Stubbetorp						
	MAGIC no red	MAGIC N-sat.	SAFE no red	SAFE N-sat	SMART no red	SMART N-sat
alk > 0	72	54	46	22	78	78
alk > 20	60	46	39	15	70	68
alk > 50	30	23	26	4	47	43
Ca/Al > 1	105	76	<0	<0	83	84
Ca/Al > 0.5	112	80	<0	<0	87	87

Yli-Knuutila						
	MAGIC no red	MAGIC N-sat.	SAFE no red	SAFE N-sat	SMART no red	SMART N-sat
alk > 0	1720		247	225	820	820
alk > 20	1570		241	219	450	450
alk > 50	1300		232	210	250	245
Ca/Al > 1	6600		300	290	5000	5000
Ca/Al > 0.5	7000		325	300	5000	5000

Critical loads at Stubbetorp are somewhat higher than at Birkenes, reflecting the thicker soils with higher base saturation (Figure 9b). Again the estimates obtained by MAGIC and SMART are similar. At Yli-Knuutila the critical load for sulfur calculated by MAGIC and SMART is very high, 5-100 times that of present-day deposition. SAFE gives a somewhat lower critical load because it assumes that 25% of the runoff comes directly from the 40 cm level in the soil rather than passing through the entire 1.5 m soil column.

At all three sites the critical load calculated by MAGIC and SMART for the soil criterion of Ca/Al in soil solution is less stringent than that for the water criterion of alkalinity. The values for soil solution is calculated as an aggregate for the entire soil. Surface horizons of the soil are probably more sensitive and thus have lower critical loads than deeper horizons. For forest soils only the uppermost soil -- the rooting zone -

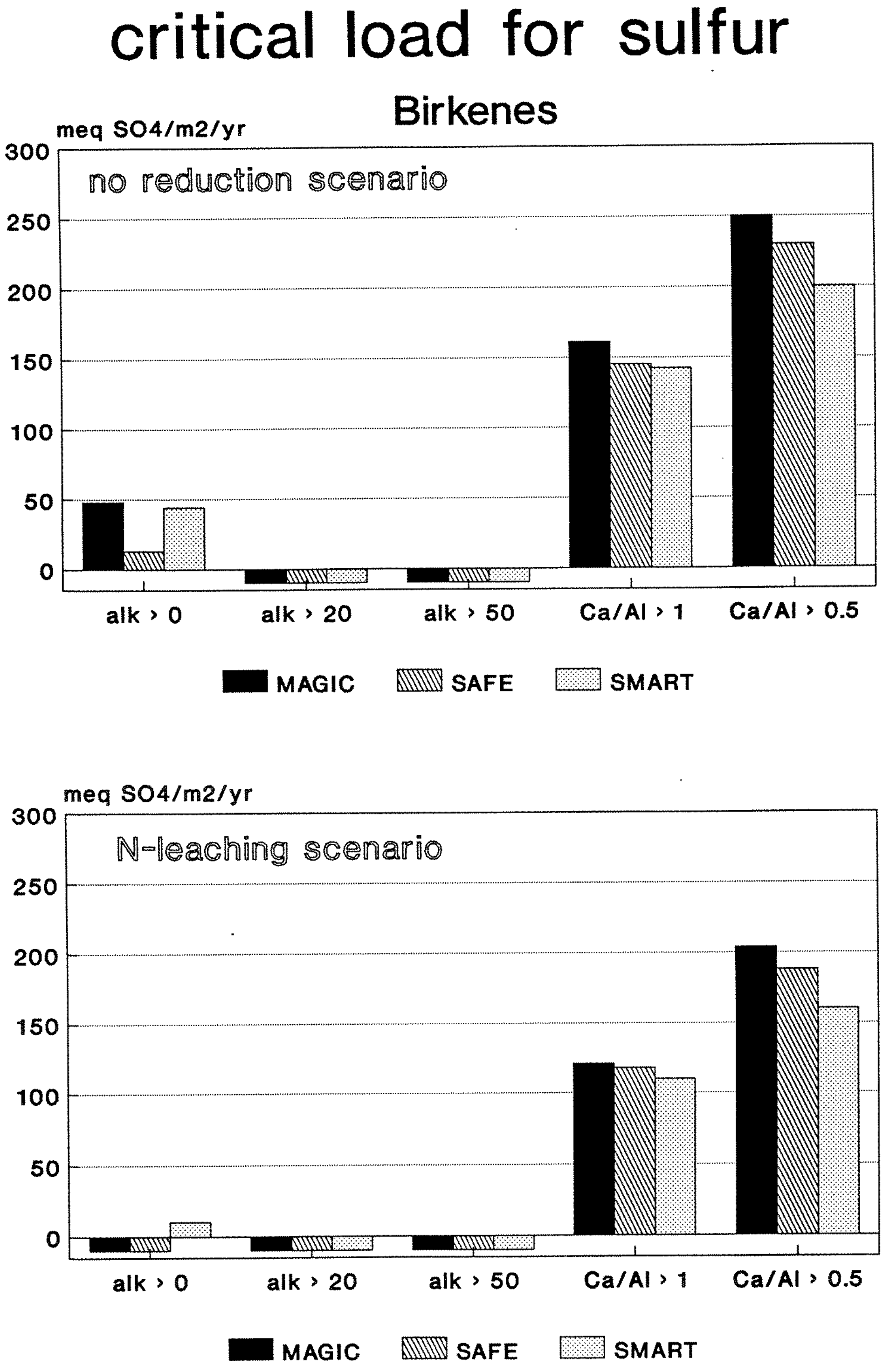

Figure 9a. Critical load for sulfur at Birkenes calculated by 3 models under 2 reduction scenarios and for 5 criteria.

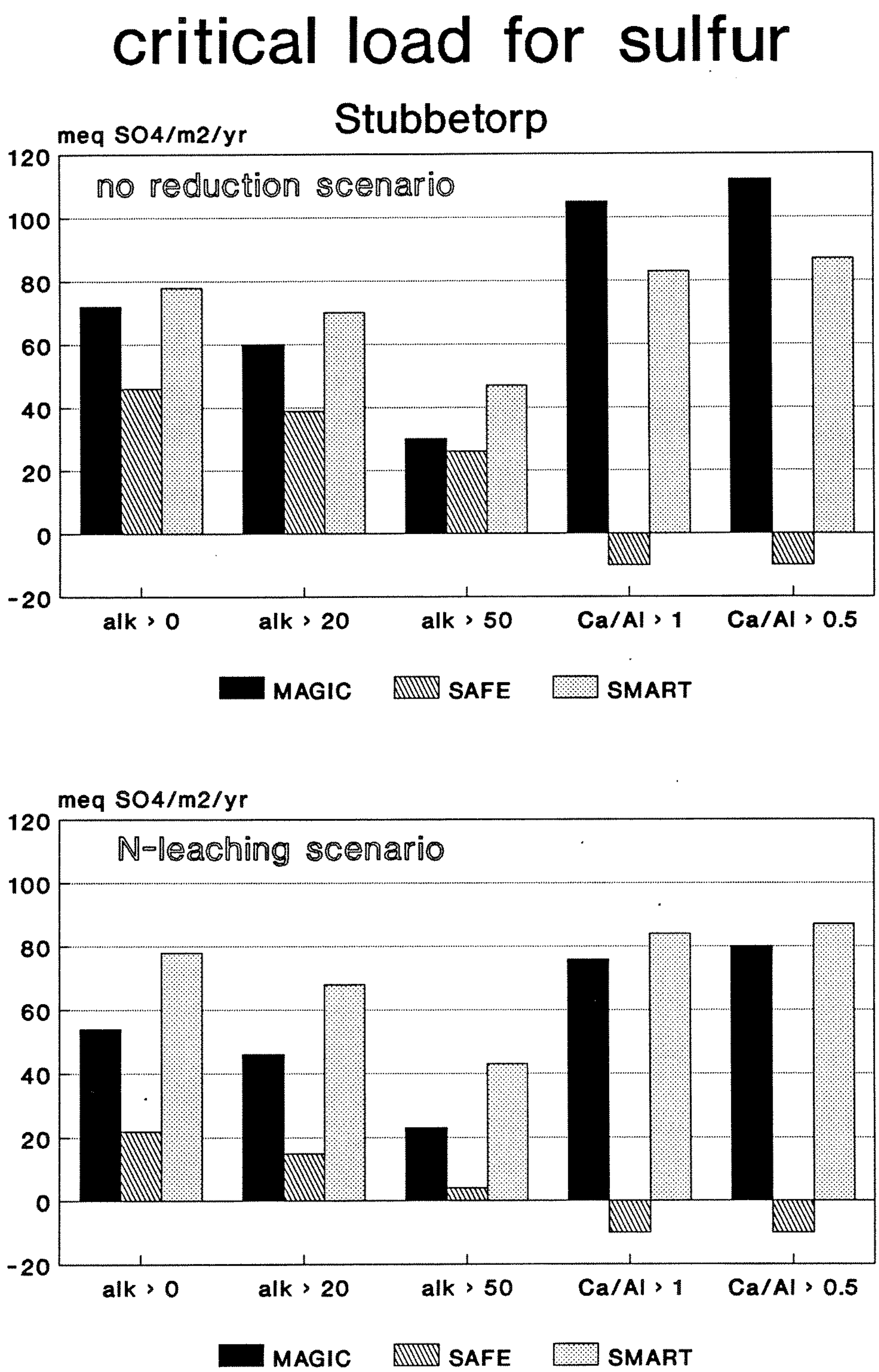

Figure 9b. Critical load for sulfur at Stubbetorp calculated by 3 models under 2 reduction scenarios and for 5 criteria.

- is of relevance with respect to critical loads. Thus at Yli-Knuutila although the soil as a whole appears well-protected, the uppermost soil horizons may be more sensitive than the streamwater. Here multi-layered acidification models such as SAFE are required to calculate the critical load for forest soils.

SAFE applied in a 3-layer version to Stubbetorp, for example, clearly illustrates the possible vertical gradients in soil profiles (Figure 10). The surface horizons can be quite acidic whereas the lower horizons continue to be well-buffered. Thus the critical load for soil at Stubbetorp will depend on the soil horizons for which the criterion is to be applied.

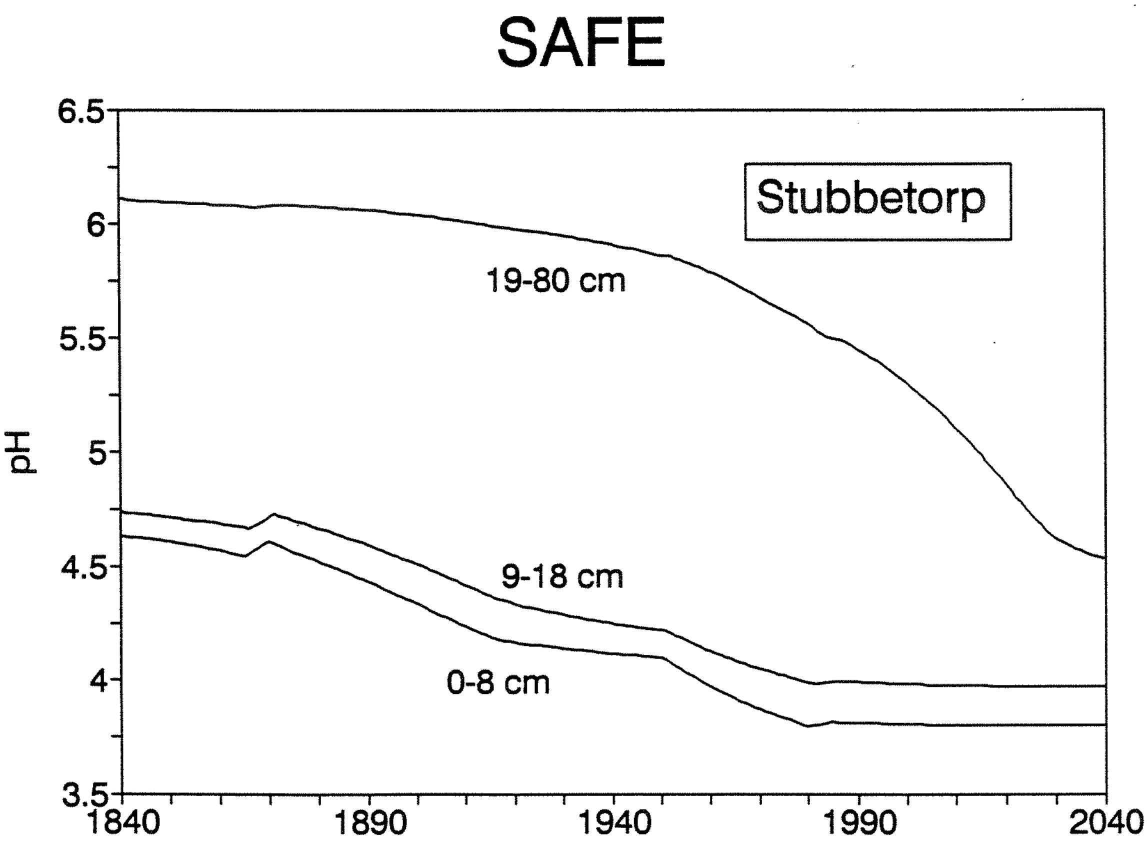

Figure 10. Stubbetorp. pH in soil solution at 3 depths in the soil as calculated by a 3-layer version of SAFE under the base case scenario.

6. DISCUSSION

All the models applied here give approximately the same acidification reconstructions and predictions for the 3 catchments. This is perhaps not so surprising because all 4 models incorporate many of the same processes. The models differ, however, in the structure and in the manner in which the different processes are coupled. In particular the SAFE model determines the key parameter weathering rate from measurements of soil texture and mineralogy, whereas the other 3 models use fitting routines to estimate weathering rates.

The similarity in predictions produced by these 4 models for three sites which differ in present-day acidification status is particularly reassuring as these models may be used in conjunction with international negotiations on emissions of acidifying compounds.

The predictions for the future indicate all sites will continue to deteriorate under all scenarios except perhaps the maximum reduction scenario. At Yli-Knuutila the deterioration will not be to levels at which biological damage is expected. But at Stubbetorp the models all suggest that although this catchment at present has positive alkalinity and pH about 6 (acceptable water quality for fish), the stream will acidify in the future such that alkalinity will decline past the 50 μeq/l level often used a critical load criterion. Damage to fish and other aquatic organisms can be expected. Stubbetorp thus falls into the category of "threatened, but not yet acidic".

Forestry practices involving clearcutting and removal of timber are responsible for a large fraction of the historical acidification to present-day and the predicted future acidification over the next 50 years. Growing forests remove base cations from the soil and may thus contribute to soil acidification.

Nitrogen inputs from the atmosphere are nearly entirely retained in the terrestrial catchment at Birkenes and Stubbetorp today. If in the future this retention becomes less complete and a greater fraction of nitrogen is leached to runoff in the form of nitrate, further acidification will result. Under these circumstances the target load for sulfur will be lower. For Birkenes nearly zero and for Stubbetorp about 50 meq/m^2/yr, approximately present-day levels (Table 4).

At present there are no satisfactory methods available to predict future nitrogen saturation. The importance of nitrogen and the critical load for nitrogen alone and nitrogen and sulfur in combination can be evaluated by means of the dynamic models applied here. These models can also be further developed to incorporate nitrogen processes and couple nitrogen with sulfur dynamics.

Future global change may also modify the degree and rates of acidification and recovery in acid-impacted terrestrial and aquatic ecosystems. These synergistic effects can be evaluated by means of models such as those used here using the same scenario approach for future climate change.

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APPENDIX

Parameter values used to calibrate the models. Tables A1, A2 and A3 list values for MAGIC, MIDAS and SMART all of which use 1 soil layer. Table A4 gives values for SAFE, which uses 3 soil layers for these calibrations.

Table A1. Parameter values used at Birkenes.

Parameter	Unit	MAGIC	MIDAS	SMART
Soil:				
temp	°C	5		
depth	cm	40	40	40
bulk dens.	kg m^{-3}	936		936
moisture cont.	$m^3 m^{-3}$		0.27	0.27
percolation	m	1.19	1.19	1.19
pCO_2 soil	atm	.005	.005	.005
log K_{gibb} soil		8.2	8.2	8.2
CEC	meq kg^{-1}	44		44.1
pK_1		4.4		4.5
pK_2		8.0		8
total org	mmol m^{-3}	65		65
DOC	mg l^{-1}			6.5
SO_4 halfsat	meq m^{-3}	110		
SO_4 maxcap	meq kg^{-1}	0.9		
nitrif. fact.				1
weathering	meq $m^{-3} yr^{-1}$			
Ca		12.4		
Mg		0.4		
Na		5.9		
K		1.0		
Ca+Mg		12.8	17	14
Na+K		6.9		11
selectivity coeff.				
log Al/Ca		-0.9		
log Al/Mg		-0.4		
log Al/Na		-1.7		
log Al/K		-6.2		
log H+Al/Ca+Mg			2.7	
log Al/Ca+Mg				0.5
log H/Ca+Mg				1.5
Stream:				
pCO_2 stream	atm	.0007		.0013
log K_{gibb} stream		8.6		8.2
pK_1		4.4		4.5
pK_2		8.0		8
total org.	mmol m^{-3}	61		65
DOC	mg l^{-1}			6.5

Table A2. Parameter values used at Stubbetorp.

Parameter	Unit	MAGIC	MIDAS	SMART
Soil:				
temp	°C	7.0		
depth	cm	80	80	80
bulk dens.	kg m^{-3}	1300		1300
moisture cont.	$m^3 m^{-3}$		0.27	0.27
percolation	m	0.28	0.28	0.28
pCO_2 soil	atm	.010	.009	.009
log K_{gibb} soil		8.6	9	9
CEC	meq kg^{-1}	10		10.0
pK_1		4.5		4.5
pK_2		8.0		
total org	mmol m^{-3}	70		70
DOC	mg l^{-1}			7
SO_4 halfsat	meq m^{-3}	100		
SO_4 maxcap	meq kg^{-1}	1		
nitrif. fact.				1
weathering	meq $m^{-3} yr^{-1}$			
Ca		16.5		
Mg		8.9		
Na		11.2		
K		2.6		
Ca+Mg		25.4	11	35
Na+K		13.8		20
selectivity coeff.				
log Al/Ca		2.5		
log Al/Mg		3.8		
log Al/Na		0.7		
log Al/K		-3.2		
log H+Al/Ca+Mg			5.0	
log Al/Ca+Mg				5.4
log H/Ca+Mg				4.0
Stream:				
pCO_2 stream	atm	.001		.001
log K_{gibb} stream		8.6		9
pK_1		4.5		4.5
pK_2		8.0		
total org.	mmol m^{-3}	60		70
DOC	mg l^{-1}			7

Table A3. Parameter values used at Yli-Knuutila.

Parameter	Unit	MAGIC	MIDAS	SMART
Soil:				
temp	°C	5.0		
depth	cm	150	150	150
bulk dens.	kg m^{-3}	1480		1480
moisture cont.	$m^3 m^{-3}$		0.27	0.27
percolation	m	.172	0.172	0.172
pCO_2 soil	atm	.01	.001	.009
log K_{gibb} soil		8.6	8.2	8
CEC	meq kg^{-1}	124		124
pK_1		4.5		4.5
pK_2		8.0		
total org	mmol m^{-3}	120		158
DOC	mg l^{-1}			10.2
SO_4 halfsat	meq m^{-3}	100		
SO_4 maxcap	meq kg^{-1}	1		
nitrif. fact.				1
weathering	meq $m^{-3} yr^{-1}$			
Ca		24.4		
Mg		16.8		
Na		8.9		
K		3.8		
Ca+Mg		41.2	30	48
Na+K		12.7		16
selectivity coeff.				
log Al/Ca		2.7		
log Al/Mg		2.9		
log Al/Na		1.2		
log Al/K		-1.6		
log H+Al/Ca+Mg			3.1	
log Al/Ca+Mg				3.5
log H/Ca+Mg				5.3
Stream:				
pCO_2 stream	atm	.001		.002
log K_{gibb} stream		8.6		8
pK_1		4.5		4.5
pK_2		8.0		
total org.	mmol m^{-3}	110		158
DOC	mg l^{-1}			10.2

Table A4. Parameter values used to calibrate SAFE at Birkenes, Stubbetorp and Yli-Knuttila.

Minerology data

Mineral content	% of total	Birkenes	Stubbetorp	Yli–Knuutila
K-feldspar		27	32	40
Oligoclase		25	16	25
Hornblende		0.25	1.6	1.7
Pyroxene				0.1
Garnet				0.2
Biotite		0.75	0.1	0.5
Chlorite			0.1	1.5
Vermiculite			10	
Apatite		0.3		
Quartz		45.7	50.2	31.2

Stream input data

Parameter	Unit	Birkenes	Stubbetorp	Yli–Knuutila
CO_2 pressure	atm	0.0008	0.0012	0.002
Dissolved organic carbon	mg/L	7	7	10
log Gibbsite eq. constant	$kmol^2/m^6$	8.5	8.5	8.5

Soil input data, Stubbetorp

Parameter	Unit	Soil layer		
		1	2	3
Soil layer height	m	0.09	0.09	0.62
Cation exchange capacity	kEq/kg	$250 \cdot 10^{-6}$	$21 \cdot 10^{-6}$	$6.3 \cdot 10^{-6}$
	% of total	20.2	23.4	56.4
Specific surface area	m^2/m^3	$7.3 \cdot 10^5$	$1.5 \cdot 10^6$	$2.9 \cdot 10^6$
Mg+Ca+K uptake †	% of total max	10	10	80
N uptake†	% of total max	10	10	80
Moisture content	m^3/m^3	0.25	0.20	0.27
Soil bulk density	kg/m^3	100	1230	1415
CO_2 pressure	atm	0.0012	0.002	0.004
Percolation	% of precipitation	84	71	43
Dissolved organic carbon	mg/L	10	10	10
log Gibbsite eq. constant	$kmol^2/m^6$	6.5	7.5	8.5

Soil input data, Yli–Knuutila

Parameter	Unit	Soil layer 1	Soil layer 2	Soil layer 3
Soil layer height	m	0.10	0.30	1.1
Cation exchange capacity	kEq/kg	$91.3 \cdot 10^{-6}$	$29.5 \cdot 10^{-6}$	$147 \cdot 10^{-6}$
	% of total	1.9	4.5	93.6
Specific surface area	m^2/m^3	$2.0 \cdot 10^5$	$2.5 \cdot 10^6$	$0.6 \cdot 10^6$
Mg+Ca+K uptake	% of total max	20	60	20
N uptake	% of total max	20	60	20
Moisture content	m^3/m^3	0.20	0.20	0.15
Soil bulk density	kg/m^3	560	1410	1580
CO_2 pressure	atm	0.0012	0.002	0.004
Percolation	% of precipitation	75	21	(21)
Horisontal flow	% of precipitation	0	7	21
Dissolved organic carbon	mg/L	10	10	10
log Gibbsite eq. constant	$kmol^2/m^6$	6.5	7.5	8.5

Soil input data, Birkenes

Parameter	Unit	Soil layer 1	Soil layer 2	Soil layer 3
Soil layer height	m	0.08	0.15	0.17
Cation exchange capacity	kEq/kg	$327 \cdot 10^{-6}$	$44 \cdot 10^{-6}$	$18 \cdot 10^{-6}$
	% of total	39.7	39.2	21.1
Specific surface area	m^2/m^3	$1.0 \cdot 10^5$	$0.96 \cdot 10^6$	$1.1 \cdot 10^6$
Mg+Ca+K uptake	% of total max	50	50	0
N uptake	% of total max	50	50	0
Moisture content	m^3/m^3	0.25	0.20	0.20
Soil bulk density	kg/m^3	266	1026	1235
CO_2 pressure	atm	0.0012	0.002	0.004
Percolation	% of precipitation	81	81	81
Dissolved organic carbon	mg/L	10	10	10
log Gibbsite eq. constant	$kmol^2/m^6$	6.5	7.5	8.5

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