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# Modeling Long-term Streamwater Chemistry in the Berg Catchment, Southwestern Sweden

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The geochemical model MAGIC (5.01) was applied to the Berg – Pipbäcken Nedre catchment in southwestern Sweden for the period 1846-2020. The major objective was to reconstruct historical acidification trends and predict the surface water and soil response to declining atmospheric deposition in the future based on the Gothenburg Protocol signed in 1999. Another task was to test the usefulness of Sweden's long-term small catchment monitoring program for a validation of the long-term acidification model. Berg has been one of fifteen catchments monitored very intensively by the Swedish Environmental Protection Agency during 1986-1993 and less intensively later by the Swedish University of Agricultural Sciences Department of Environmental Assessment. Anthropogenic atmospheric deposition was the dominant factor causing a decline of streamwater pH and especially soil base saturation. A scenario of future atmospheric deposition based on the emission reductions of S and N compounds agreed under the 1999 Gothenburg Protocol to the UNECE CLRTAP was modeled. This scenario stopped further acidification of soils, but recovery of streamwater pH and soil base saturation was slow and limited. Without further reductions from the deposition levels in 1995-1997, soil acidification would continue.

## Introduction

The rate of recovery is of great concern to the Nordic countries, since Europe's acidification strategy is designed to stop the spread of acidification, but does not direct-

ly consider the need for recovery in areas that are already acidified. Dynamic simulation of recovery subsequent to declining acid deposition using biogeochemical models such as MAGIC (Cosby *et al.* 1985; Wright *et al.* 1998), SMART (de Vries *et al.* 1989), NuCM (Liu *et al.* 1991), SAFE (Warfvinge *et al.* 1993), CHUM (Tipping 1996) or PnET-BGC (Krám *et al.* 1999; Gbondo-Tugbawa *et al.* 2001) is a valuable approach for assessing the "natural" potential for recovery. The Swedish national small catchment monitoring program (Aastrup *et al.* 1996), which comprised fifteen sites across Sweden between 1986 and 1993, is a resource that may be useful in developing a regionally nuanced understanding of the prospects for recovery.

The major objective is to assess the acidification history and to estimate the recovery prospects for the Berg catchment, SW Sweden given a partial removal of acid deposition based on the 1999 Gothenburg Protocol to the Geneva Convention on Long Range Transport of Air Pollutants. The Protocol sets emission ceilings for 2010 for sulfur and nitrogen (among other substances) based on scientific assessments of pollution effects and abatement options. The results of the reductions agreed to in this protocol are compared with the business as usual scenario of continued deposition at the 1995-97 level. Another objective was to increase the confidence in the MAGIC model predictions by comparison with the available long-term observations at Berg. Successful model validation helps to increase confidence in reconstructions of long-term historical acidification trends and predictions of the surface water response to international agreements limiting atmospheric emissions throughout Europe.

## Methods and Site Description

### Model Description

MAGIC (Model of Acidification of Groundwater in Catchments) was designed to reconstruct past and predict future drainage water as well as soil chemistry (Cosby *et al.* 1985; Cosby 1991). The MAGIC model uses a lumped representation of soil processes because understanding the catchment runoff chemistry may not require detailed knowledge of the spatial distribution of the parameters within a catchment. Water fluxes, atmospheric deposition, net vegetation uptake, weathering, and a description of organic acids are required as external inputs to MAGIC.

MAGIC has been used to simulate a large number of forest catchments, especially in north-western Europe. In Sweden, MAGIC was applied to simulate streamwater at the Stubbertorp catchment (Sandén *et al.* 1992) and at several sites in the vicinity of Lake Gårdsjön (the F1 catchment: Forsius *et al.* 1998; the G1 catchment: Beier *et al.* 1995; Moldan *et al.* 1998, and the lake itself: Wright *et al.* 1986).

### Swedish Integrated Monitoring

The Swedish National Environmental Monitoring Programme (PMK: Program för

övervakning av miljökvalitet) began in 1978. Intensive monitoring in selected forest catchments started in 1981 and included measurements of atmospheric deposition, groundwater, and streamwater. The monitoring also included observations of birds, rodents and fish, as well as periodic sampling of vegetation and soil. Fifteen integrated monitoring PMK catchments spread over Sweden were active in 1986. The Swedish small catchment monitoring was reduced to four catchments in 1993 which became part of the Intergovernmental Cooperative Program for Integrated Monitoring (IM) under the auspices of the United Nations Economic Commission for Europe. Only the Gårdsjön F1 catchment remained from the earlier PMK network, with the other three catchments newly established. Runoff chemistry monitoring by the Swedish University of Agricultural Sciences Department of Environmental Assessment, however, continues at the original PMK sites. Further information about the IM and PMK monitoring can be found on the Internet at http://info1.ma.slu.se/IM/Background.html. An extended description of integrated monitoring results in the PMK network are available in Aastrup *et al.* (1996) and Haglund and Bråkenhielm (1991). Examples of journal publications summarizing the PMK monitoring are Aastrup *et al.* (1995), Bringmark and Kvarnäs (1995), and Löfgren and Kvarnäs (1995).

## Study Area

The 93 ha Berg – Pipbäcken Nedre catchment (57°05'N, 12°46' E; 75-165 m a.s.l.) is situated near to the Ätran river in the province of Halland, southwestern Sweden (Fig. 1). It was one of the original PMK catchments. Berg was selected for this study to represent a region of high susceptibility to acid deposition. The site represents an area on the rainy western slope of Sweden's southern highlands. About 61% of the catchment is covered by century-old forest. More than 70% of the forest is formed by mixed coniferous trees composed of Norway spruce *(Picea abies)* and Scots pine *(Pinus sylvestris)*. Less than 30% of the forested area comprises deciduous trees like European beech *(Fagus sylvatica)*, black alder *(Alnus glutinosa),* sessile oak *(Quercus petraea),* silver and white birch *(Betula verrucosa* and *pubescens)* (Lindbergh and Nielsen 1986). One third of the catchment is a *Sphagnum* bog without closed-canopy forest (Fig. 1). Glacial till moraine covers 64% of the catchment and peat is developed on 31% of the catchment. The deposits are underlain by orthogneiss and amphibolite (Aastrup *et al.* 1996).

## Input Data Description

Table 1 shows the parameters used in the MAGIC application at Berg. Soil concentrations of exchangeable cations were weighted depending on the measured thickness of soil horizons and on estimated bulk density (L. Bringmark, personal communication 1999; Karltun 1995; E. Karltun, personal communication 1999). Partial pressure of $CO_2$ in the streamwater was assumed to be oversaturated with respect to the atmospheric partial pressure of $CO_2$ (by almost 3 times), a value which is within the observed range for streams. Total organic carbon (TOC) was estimated from the

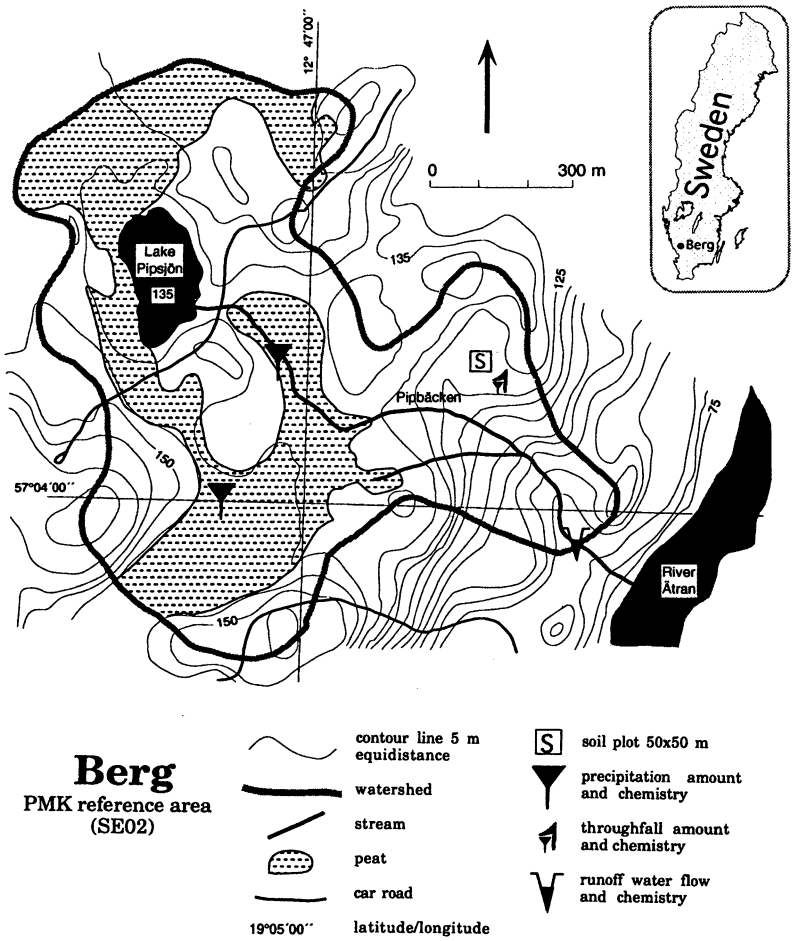



Fig. 1. Schematic map of Sweden, showing the location of the study site Berg. Topographic map of the Berg – Pipbäcken Nedre catchment, showing schematically the major landscape patterns and the most important monitoring plots (Sven Bråkenhielm, personal communication 2001). Peaty areas are without the closed canopy forest, the other parts of the catchment usually contain closed-canopy forest.

measured chemical oxygen demand (COD) of $KMnO_4$ (Wilander 1988). The volume-weighted mean streamwater concentration of TOC in 1986 was 0.6 mmol $l^{-1}$ (7.2 mg $l^{-1}$) in the calibration year (1986). Empirical dissociation constants (pKa's) of triprotic organic acids with a site density of 0.034 mol organic acids/mol TOC that Köhler *et al.* (1999) derived for Sweden were used. Sulfate adsorption parameters, the gibbsite solubility constant and weathering rates were fitted in the calibration procedure.

Table 1 – Selected parameter values for the MAGIC model calibrations at the Berg – Pipbäcken Nedre catchment.

| Parameter | Unit | Value |
|---|---|---|
| Soil depth | m | 0.69 |
| Soil bulk density | kg $m^{-3}$ | 1140 |
| Cation exchange capacity (CEC) | meq $kg^{-1}$ | 18.59 |
| Sulfate adsorption half saturation | meq $m^{-3}$ | 250 |
| Sulfate adsorption maximum capacity | meq $kg^{-1}$ | 1 |
| Partial pressure of $CO_2$ in soil water | atm | 0.005 |
| Partial pressure of $CO_2$ in stream water | atm | 0.001 |
| Organic acids in soil water | mmol $m^{-3}$ | 11.6 |
| Organic acids in stream water | mmol $m^{-3}$ | 20.4 |
| $pK_1$ of organic acids | -log 10 | 3.6 |
| $pK_2$ of organic acids | -log 10 | 4.2 |
| $pK_3$ of organic acids | -log 10 | 5.5 |
| Solubility of $Al(OH)_3$ in stream water | log 10 | 7.6 |
| Mean soil and stream temperature | °C | 6 |
| Precipitation volume | m $yr^{-1}$ | 1.008 |
| Catchment discharge | m $yr^{-1}$ | 0.59 |
| Weathering rate of Ca | meq $m^{-2}$ $yr^{-1}$ | 9.5 |
| Weathering rate of Mg | meq $m^{-2}$ $yr^{-1}$ | 6 |
| Weathering rate of Na | meq $m^{-2}$ $yr^{-1}$ | 8 |
| Weathering rate of K | meq $m^{-2}$ $yr^{-1}$ | 1.6 |

Prior to the afforestation around 1900, the Berg catchment was under grassland or bog vegetation. Net uptake of base cations was calculated from the history of afforestation and from the land use in the catchment using relative areas of forest and grassland. For the grassland period of the catchment in the 19th Century, net uptake of base cations was set to zero on the assumption that long-term accumulation in grassland vegetation is negligible (Fig. 2). An empirical curve for tree uptake of base cations (Moldan *et al.* 1999) where the maximum tree net uptake of base cations takes place at the time of canopy closure was adopted. Absolute values of the vegetation uptake and immobilization of inorganic nitrogen (N) species were calculated internally by the MAGIC using the fitted (for 1986) or estimated (for 1846) relative uptake of the available N from atmospheric deposition.

Background wet precipitation (in 1846) of base cations, sulfate, and chloride was set equal to the present concentration of sea salts. The hindcast deposition scenario for sulfate (Fig. 3) was based on the simplified scenario of Mylona (1996) for the EMEP square 20-21. The hindcast deposition of nitrate and ammonium was similar to the scenarios of Forsius *et al.* (1998) for the Gårdsjön catchment. Marine deposition of $Cl^-$, $Na^+$ and $Mg^{2+}$ predominated in wet precipitation over the entire period of the simulation. The contribution of the other sea salt ions was smaller but still

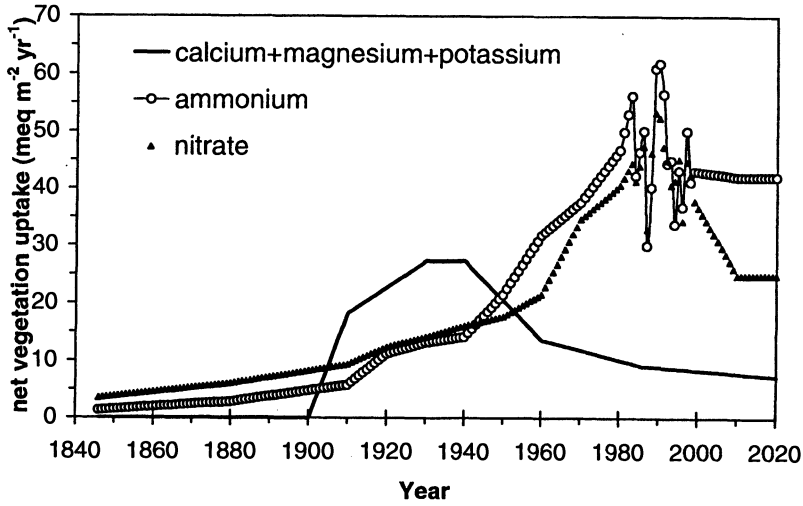



Fig. 2. Net vegetation uptake scenario for the nutrient base cations ($Ca^{2+}$, $Mg^{2+}$, $K^{+}$) and inorganic nitrogen species ($NH_4^{+}$, $NO_3^{-}$) used in modeling Berg – Pipbäcken Nedre 1846-2020.

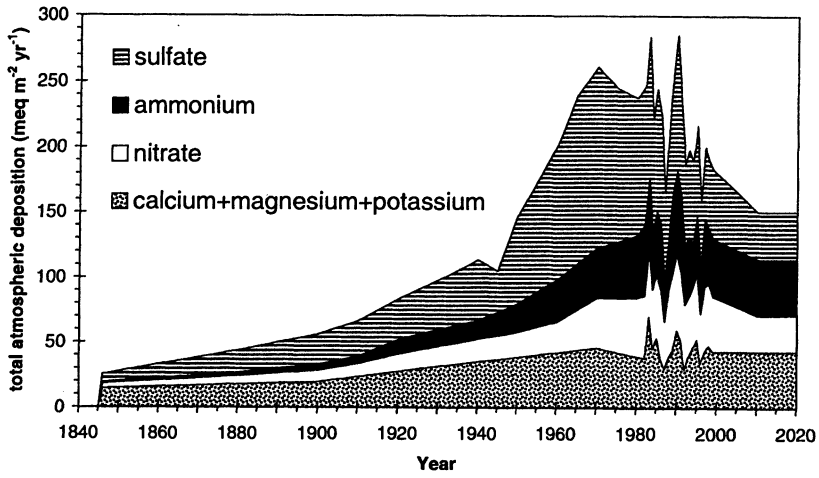



Fig. 3. Scenario of historical total atmospheric inputs of $NH_4^{+}$, $NO_3^{-}$, and $SO_4^{2-}$ and the nutrient base cations ($Ca^{2+}$, $Mg^{2+}$, $K^{+}$) from 1846 to 1982. The period 1983-1998 is based on the actual measurements at Berg. Reductions of the deposition of $NH_4^{+}$, $NO_3^{-}$, and $SO_4^{2-}$ in 1999-2010 are based on the implementation of the Gothenburg Protocol.

large for $K^+$ and $Ca^{2+}$. Dry deposition of base cations was set to increase linearly from zero (used in 1846-1900) to estimated canopy closure in 1930. Dry deposition of base cations and sulfate connected to industrial activities in Sweden and in Europe started to increase even more steeply in the end of the 1940's reaching the highest values in 1970 which is reflected by the total deposition fluxes (Fig. 3).

Actual wet deposition fluxes from the 16 year period (1983-1998) of observations at Berg (K. Kindbom 1999, unpublished) were used as inputs to MAGIC. Annual runoff for a 13-year period in 1986-1998 (H. Kvarnäs, personal communication 2000) was used to enhance the validation comparisons. Two scenarios of atmospheric deposition in 1999-2020 were simulated at Berg. In the first scenario the target deposition level of nitrate, ammonium and the non-marine sulfate in 2010 for the west coast of Sweden was based on the 1999 Gothenburg Protocol of the UN-ECE Convention on Long-Range Transport of Air Pollutants and the matrices of the EMEP model (G. Lövblad, personal communication 2000). Implementation of the Protocol for this simulation used the mean measured deposition in 1995-1997 at Berg as the baseline year. The target 2010 values represent a 49% decline in the non-marine sulfate (corresponds to a 37% decline of total sulfate), a 40% decline in nitrate and a 3% decline in ammonium relative to the 1995-1997 mean deposition. These 2010 values were then held constant to the end of the simulation in 2020. The second scenario used the 1995-1997 deposition of sulfate, nitrate and ammonium for the whole period 1999-2020. The deposition of chloride and base cations were kept constant from 1999-2020 in both scenarios.

## Results and Discussion

### Calibration and Validation

The MAGIC model was applied in a hindcast mode (1846-1986) and successfully calibrated against the mean concentrations in streamwater and soil for the reference year 1986 at Berg (Figs. 4-5). The calibration fit was evaluated by comparing observed concentrations with the model simulation in 1986. The difference between observed and simulated values of individual ions lay between ± 1 $\mu eq\ l^{-1}$ in streamwater and ± 0.05% in soil. The only exceptions with a larger range was $H^+$ (underpredicted by 5 $\mu eq\ l^{-1}$). Consequently the model calculated slighly higher pH (4.64) than the observed mean pH in 1986 (pH 4.56).

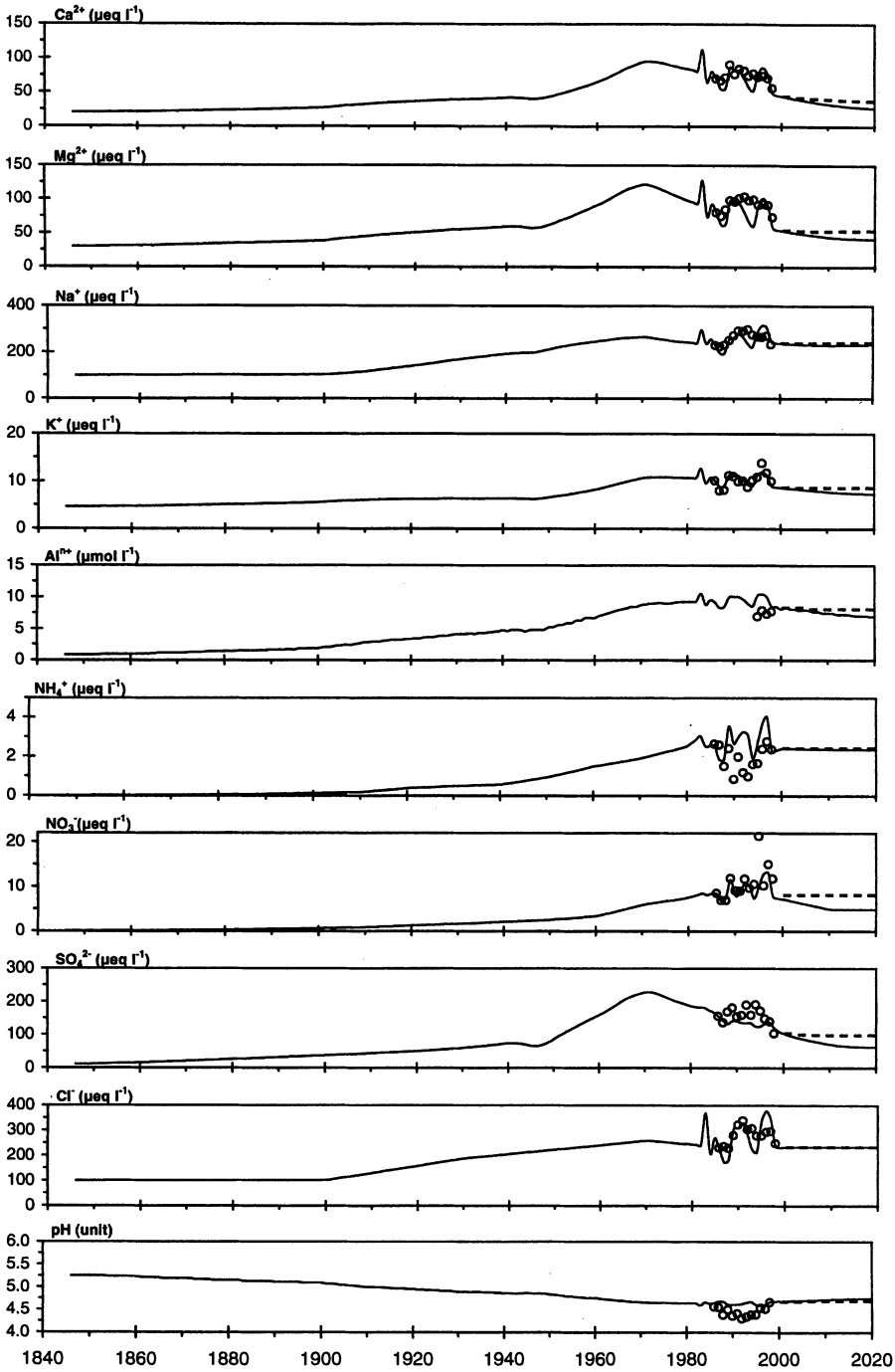
The model was then run in the predictive mode (1987-2020) to explore the time scale of potential acidification recovery given two scenarios of atmospheric deposition levels described in detail in the input data description chapter. Fig. 3 shows the deposition scenario based on the Gothenburg Protocol with significant declines of sulfate and nitrate deposition and a small decline in ammonium.

The simulations were compared to the measured streamwater data set (1987-1998) for the model evaluation (validation). The evaluation period showed some

slight underestimation in the concentrations of most simulated constituents (Figs. 4-5). The mean underprediction of the simulations for the sum of base cations (BC; $Ca^{2+}+Mg^{2+}+Na^{+}+K^{+}$) was 32.0 μeq $l^{-1}$ (or 7%) and 45.6 μeq $l^{-1}$ (or 10%) for the sum of strong acid anions (SAA; $SO_4^{2-}+Cl^{-}+NO_3^{-}$). Acid neutralizing capacity (ANC) defined as BC-SAA was very slightly negative at Berg, the ANC was -6 μeq $l^{-1}$ according to the model, the observed ANC was -16 μeq $l^{-1}$. The best validation results for the means of the annual concentrations in 1987-1998 of individual ions was recorded for $K^{+}$ (0% difference), very tiny relative underestimations were calculated for $Na^{+}$ and $Cl^{-}$ (both 3%). On the other hand larger relative underestimations of the model prediction in streamwater were found for $Ca^{2+}$ (8%), $Mg^{2+}$ (13%), $NO_3^{-}$ (14%) and $SO_4^{2-}$ (15%). MAGIC simulations also predicted higher pH (4.63) in 1987-1998 than the observed mean (4.44). The worst relative fit between observations and simulations was recorded for $Al^{n+}$ and $NH_4^{+}$. In the case of Al, the model overestimated the mean concentrations by almost 32% in 1995-1998 (Al was unfortunately not measured in 1986-1994 at Berg). A low gibbsite solubility constant (7.6) was used for the streamwater at Berg that decreased Al concentrations in the simulation. Higher values of gibbsite solubility (more in line with what is seen in some other applications of MAGIC to similar sites) would make the difference between the simulations and predictions even larger. Difficulties in simulating Al is a typical problem in the application of the MAGIC model. There is a hope that the brand new version of the MAGIC 7.07 (Cosby 2000) which allows a more flexible empirical approach (user-specification of the exponent in the gibbsite solubility equation) will improve Al modeling. Ammonium was overpredicted by the model by 52% in the validation period 1987-1998, but this has a negligible influence on the overall charge balance because the ammonium concentrations are very low in the stream. Better simulations of inorganic nitrogen species could be expected with the MAGIC 7.07 (Cosby 2000), because that version handles ammonium and nitrate in much more detail than the MAGIC 5.01 (Cosby 1991) used in this application.

Many factors can be involved in the discrepancies between simulations and observations for individual compounds in the validation years. These include unusual climatic patterns (*e.g.* dry years) and unusual atmospheric deposition patterns (*e.g.* sea salt episodes). In general, the simulated year-to-year variations are usually larger than those observed at Berg (Fig. 4). The major factor could be much smaller observed variation of the major anion, chloride. There appears to be short-term reten-

Fig. 4. MAGIC model simulation results of the annual mean streamwater concentrations for calcium, magnesium, sodium, potassium, aluminum, ammonium, nitrate, sulfate, and chloride, as well as the pH values at Berg – Pipbäcken Nedre from 1846 to 2020. The impact of implementing the Gothenburg Protocol on atmospheric deposition reductions of sulfate, nitrate and ammonium in 1999-2010 are shown in full lines. Results of the "business as usual" scenario are in dashed lines. Thirteen years of the observed annual mean concentrations (1986-1998) are shown in circles.



tion processes that are not accounted for in MAGIC that dampen the response of $Cl^-$ outputs to large annual changes in $Cl^-$ inputs. This problem was mentioned by Wright (2001) in his modeling of the Vikedal and Tovdal catchments in southern Norway.

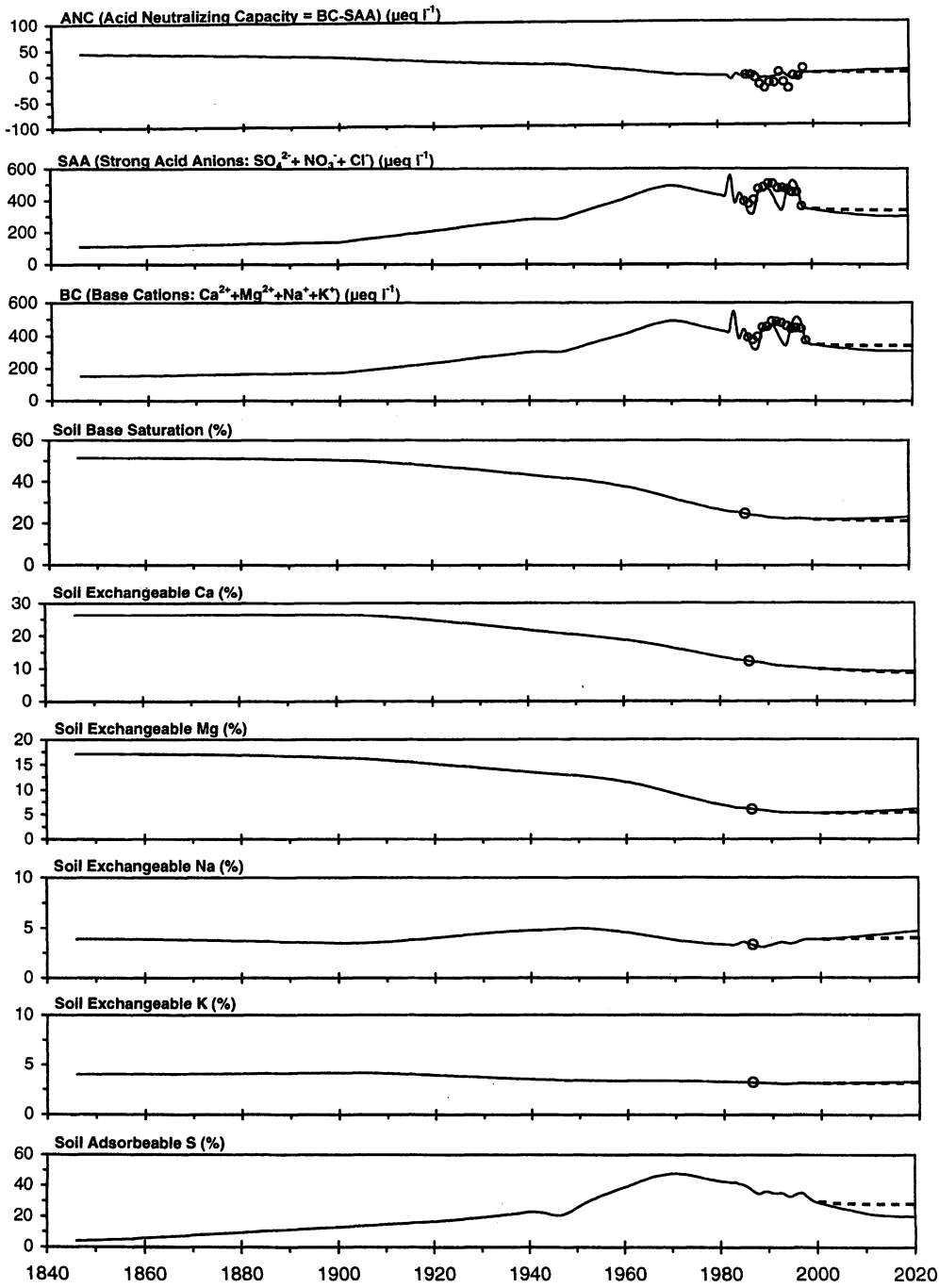
**Hindcast and Forecast**

Large increases of the streamwater base cations in the hindcast period (Fig. 4) were partially caused by the increase of the dry deposition of sea-salts related to the development of forest canopy in the first half of the 20th Century. Enhanced leaching of the exchangeable base cations from the soil (Fig. 5) was responsible for the increase of the streamwater BC concentrations in the 1950's and 1960's. The peak of the simulated base cation concentrations was in 1970-1974, and 1971 for sulfate. The maximum increase of the streamwater concentrations (and fluxes) for 1846-1970's was as follows: $Ca^{2+}$ 4.8 fold, $Mg^{2+}$ 4.1 fold, $Na^+$ 2.7 fold, $K^+$ 2.4 fold, and $SO_4^{2-}$ 21.9 fold.

Implementation of the Gothenburg Protocol will cause a significant decrease of sulfate and the base cations, especially the divalent ones. Aluminum should decrease according to the Gothenburg protocol future scenario, but it will still remain well above the potentially dangerous level in long-term exposure (3.2 $\mu mol\ l^{-1}$) for sensitive aquatic biota (Gensemer and Playle 1999). Increase of $Cl^-$ in the first half of the 20th Century was caused entirely by the enhanced dry deposition of the sea-salt. Higher values around 1970 reflect a possible contribution of HCl emitted from industrial sources. Nitrate and ammonium concentrations reflect entirely the atmospheric deposition scenario (Fig. 3) and the N uptake calculated by the model (Fig. 2) using the fitted percentage vegetation uptake (nitrogen processes are treated in a very simplistic way in the MAGIC 5.01 model). The percentages of the nitrate and ammonium uptake were kept constant in 1987-2020, exactly on the fitted level from the calibration year 1986. Note that the predictions of the streamwater $NO_3^-$ and $NH_4^+$ changes have very large uncertainty.

Streamwater acidification was present at Berg since the beginning of the model simulation, even though the pre-industrial pH was quite low (Fig. 4) despite a soil base saturation of ca 50% (Fig. 5). Simulated pH of streamwater decreased from 5.25 in 1846 to 5.1 in 1900 and further to 4.9 in 1929. In the beginnig of the 20th Century, the acidification was exacerbated by base cation uptake by the growing

Fig. 5. Simulation results of the annual mean streamwater concentrations of the combined cations and anions and the annual mean soil concentrations of the individual base cations and sulfur at Berg – Pipbäcken Nedre from 1846 to 2020. The impact of implementing the Gothenburg Protocol on atmospheric deposition reductions of sulfate, nitrate and ammonium in 1999-2010 are shown in full lines. Results of the "business as usual" scenario are in dashed lines. Observed annual mean concentrations are shown in circles.



young forest (Fig. 2). Streamwater acidified slightly faster in the 1950's and 1960's (1953 pH 4.8, 1972 pH 4.65). The main reason for this was the enhancement of the total atmospheric deposition of strong acid anions, mainly anthropogenic sulfate. In the period after 1970, sulfur deposition decreased substantially (Fig. 3). Streamwater acidification did not proceed much further after the peak of anthropogenic deposition (1970) (Fig. 3) in the 1970's and the first half of the 1980's. The minimum simulated pH was 4.64 using the long-term input scenarios (and only 4.59 in 1996 using the real inputs). In 1999-2010, the pH reversal is slow for both future deposition scenarios. The more optimistic (Gothenburg Protocol based) scenario showed a streamwater pH value of 4.72 in 2010, while the scenario "business as usual" simulated only a slightly lower pH (4.69) in 2010.

The soil chemistry results showed a large decline of soil base saturation (BS), from 51.4% in 1846 to 24.6% in 1986 (Fig. 5). The rate of decrease was variable throughout the simulation. Negligible decline (to 50.4% in 1900) was apparent throughout the 19th Century. The steepest decline was simulated in the 1970's (1970 BS 32.6%, 1980 BS 27%). Both scenarios produced further BS decline in the very near future but diverged in the prediction of the trends in 2004-2020. The Gothenburg scenario produced the lowest BS (22%) in 2003 and then slight improvements of BS (22.2% in 2010 and 23.1% in 2020). However the "business as usual" scenario showed a continuous decline as the lowest BS was simulated in the last year (BS in 2010 21.6%, BS in 2020 21.2%).

Given the deposition and land use history at the Berg catchment, most of the soil BS decline is due to acidic deposition. A smaller portion of the BS decline resulted from the incorporation of base cations into tree biomass as the site changed from pasture to forest early in the 20th century.

There are obvious differences in the behavior of the individual soil exchangeable base cations (Fig. 5). Recovery of soil $Mg^{2+}$ and $Na^{+}$ is predicted by MAGIC but $K^{+}$ should remain relatively constant. Moreover, the exchangeable $Ca^{2+}$ will continuously decline in the future even in the more optimistic scenario. Pools of the adsorbed sulfate (Fig. 5) will still be relatively large in 2020 in the business as usual scenario creating further oportunity to slow down chemical recovery of the soil and stream water. However, the implementation of the Gothenburg Protocol will decrease the adsorbed sulfate considerably.

Predictions of just annual mean concentrations by MAGIC are valuable but do not allow complete assessment of all potential threats to aquatic ecosystems. Spring floods profoundly effect the biodiversity of aquatic biota. Other models, like the Boreal Dilution Model (BDM) have to be used for evaluation of such important factors (Laudon 2000).

## Conclusions

This paper shows simulations of streamwater and soil chemistry from the Berg – Pipbäcken Nedre forest catchment. It appears that anthropogenic deposition was the dominant factor causing streamwater pH decline and very large soil base saturation decline at Berg in the 20th Century. Two simulation forecasts were used for 1999-2020. 1) Implementation of the Gothenburg Protocol for further significant declines in the atmospheric deposition of major anthropogenic acidifying compounds ($SO_4^{2-}$ and $NO_3^-$) and a small decline of $NH_4^+$ for the target year 2010. 2) The business as usual scenario. Both simulations showed very limited prospects for recovery of soil base saturation and streamwater pH. The key difference between the two scenarios was that soil acidificaiton was reversed in the Gothenburg Protocol scenario, while it continued if deposition was held constant at 1995-1997 levels (business as usual scenario).

This application of MAGIC also gave some indication of the value of Sweden's long-term monitoring in small forest catchments for validation of long-term biogeochemical models. Moreover the network of catchments spread over Sweden has a potential for detailed site-specific applications in regions with different deposition histories and land use.

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