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Critical Loads, Target Load Functions and Dynamic Modelling for Surface Waters and ICP Waters Sites

Norwegian Institute for Water Research

REPORT

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Abstract

Critical load functions (CLF) for ICP Water sites estimated with the FAB model and based on recent data (2000-02) do not differ greatly from those based on earlier data (1992-94/5). The ICP Waters sites in Europe are largely characteristic of the most sensitive waters within the EMEP 50 km grid cell. Target load functions (TLF) require use of a dynamic model. For acidified waters the TLF for sulphur (Smax) is often lower than the CLF, because of time lags in the recovery of catchment soils in response to reduced S deposition. On the other hand the TLF for nitrogen (Nmax) is often higher than the CLF, because most catchments presently are not N-saturated and retain a large fraction of N-deposition. Scientific understanding of processes governing N-retention and loss from catchments remains insufficient and is the major source of uncertainty in modelling acidification of soils and waters. Climate change will also affect the uncertainty with respect to N.

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CONVENTION ON LONG-RANGE
TRANSBOUNDARY AIR POLLUTION

INTERNATIONAL COOPERATIVE PROGRAMME ON
ASSESSMENT AND MONITORING OF ACIDIFICATION
OF RIVERS AND LAKES

Dynamic Modelling, Target Load Functions and Critical Loads for Surface Waters and ICP Waters Sites

ICP Waters Programme Centre,
Norwegian Institute for Water Research
Oslo, December 2005

Preface

The International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes (ICP Waters) was established under the Executive Body of the Convention on Long-range Transboundary Air Pollution at its third session in Helsinki in July 1985. The Executive Body has also accepted Norway's offer to provide facilities for the Programme Centre, which has been established at the Norwegian Institute for Water Research, NIVA. A programme subcentre is established at the Laboratory of Freshwater Ecology and Inland Fisheries at University of Bergen. The ICP Water programme is lead by Berit Kvæven, Norwegian Pollution Control Authority (SFT).

The Programme objective is to establish an international network of surface water monitoring sites and promote international harmonization of monitoring practices. One of the aims is to detect long-term trends in effects of acidic deposition on surface water chemistry and aquatic biota, and to reveal the dose/response relationship between water chemistry and aquatic biota.

The data collected within ICP Waters are well-suited for calculations of critical loads for freshwaters. An ICP Waters report in 1998 presented critical loads and exceedances for ICP Waters sites calculated using the static models SSWC (steady-state water chemistry model) and FAB (first-order acidity balance model) (Henriksen and Posch 1998). The present report is an update of the 1998-report with new data. Additionally, target loads which take into account time lags in ecoystem response to deposition reduction are calculated with dynamic models and compared with critical loads.

The report has been discussed and approved by the ICP Waters 21st Task Force meeting, held in Tallin 17-19 October 2005.

ICP Waters Programme Centre,
Oslo, December 2005

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Summary

Critical load functions (CLF) for ICP Water sites estimated with the FAB model, a static (steady-state) model, and based on recent data (2000-2002) do not differ greatly from those based on earlier data (1992-1994/95). The ICP Waters sites in Europe are largely characteristic of the most sensitive waters within the EMEP 50 km grid cell.

Dynamic models provide an extension to critical loads by predicting the timescale of chemical recovery to emission reductions. They can also be used to determine the deposition levels required to achieve a prescribed target chemistry within a given timescale and so have direct utility in the formulation of further emission reductions.

Target load functions (TLF) require use of a dynamic model. For acidified waters the TLF for sulphur (S_{max}) is often lower than the CLF, because of time lags in the recovery of catchment soils in response to reduced S deposition. On the other hand the TLF for nitrogen (N_{max}) is often higher than the CLF, because most catchments presently are not N saturated and retain a large fraction of N deposition.

Scientific understanding of processes governing N-retention and loss from catchments remains insufficient and is the major source of uncertainty in modelling acidification of soils and waters. Climate change will also affect the uncertainty with respect to N.

1. Introduction

The link between the emissions of sulphur (S) and nitrogen (N) species to the atmosphere and the acidification of soils and surface waters is now well established and understood. These links form the basis for calculation of critical loads for surface waters and soils. Critical loads have contributed to the scientific basis for the 1994 Oslo Protocol and the 1999 Gothenburg Protocol to the UN-ECE Convention of Long-Range Transboundary Air Pollution.

In 1998 Henriksen and Posch (1998) calculated critical loads for acidity and their exceedances for ICP-Waters sites using static (steady-state) models. They also compared the site critical loads with values for the EMEP grid cells in which the sites were located. The estimates were based on data for 1992-94/5.

The Gothenburg protocol is now subject to revision as part of the ongoing work with the Convention. In recent years it has become increasingly clear that the static model approach used to estimate critical loads may be insufficient to determine the reduction in level of acid deposition necessary to allow recovery of acidified ecosystems within acceptable time. Deposition may have to be reduced to a target load below the critical load.

In November 2004 the Coordination Center for Effects (CCE) of the International Cooperative Programme on Modelling and Mapping (ICP M&M) issued a call for data for target load functions for soils and surface waters. In March 2005 three countries, Norway, Sweden and the UK, submitted data for surface waters to the CCE as requested. The new call for data adhered to the precautionary principle by specifying that for a given ecosystem the value to be used will always be the lesser of the target load and the critical load. In the process the work with dynamic models and target load functions revealed a number of phenomena not apparent from static models and critical loads.

This report first updates the critical loads estimates for ICP Waters sites and the comparison with the gridded data for waters held by the CCE. Data for the period 2000-02 are used. This report also deals with features in determining target load functions for surface waters, and sources of uncertainty in critical load and target load functions.

2. Critical loads for ICP Waters sites

2.1 Critical load functions with the static models SSWC and FAB

The Steady-State Water Chemistry (SSWC) model allows the calculation of critical loads of acidity (Henriksen and Posch 2001). Exceedance of the critical load using SSWC assumes no net retention of S deposition and present-day percent retention of N deposition. The First-order Acidity Balance (FAB) model is based on SSWC for S but introduces new formulations for N. Nitrogen retention is assumed to be the long-term steady-state value rather than the present-day value (Posch et al. 1997). FAB allows the simultaneous calculation of critical loads of acidifying N and S deposition and their exceedances. When considering the effects of both sulphur and nitrogen simultaneously, there is no unique value for the critical load of S and N, since there is a trade-off between reduction in the deposition of sulphur and reduction in deposition of acidifying nitrogen compounds (Posch et al. 1997).

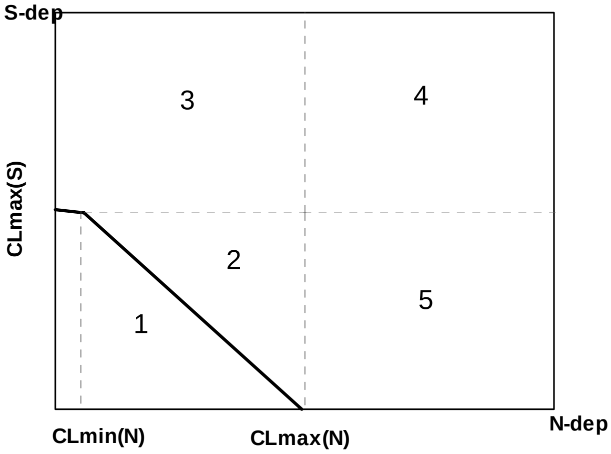

***Figure 1**. The critical load function. Each surface water has its own diagram. The critical load function (thick line) is given by the calculated values for CLmax(S), CLmin(N) and CLmax(N). The area below the thick line (1) indicates N and S deposition values not causing exceedance. The location of current N and S deposition in the diagram gives the reduction requirements necessary to meet the critical limit criterion: 1. no reductions required, 2. N or S reduction, 3. S-reduction required, 4. S and N reduction required, 5. N-reduction required (from Posch et al. 1997).*

Static models do not take into account time delays between change in deposition of S and N and the response in sulphate (SO_4) and nitrate (NO_3) in runoff. Thus, for example, if current deposition is in sector 4 (both S and N exceeded) and the proposed reduction in deposition will fall into sector 1 (ecosystem is protected), the SSWC and FAB models cannot say whether the recovery of the ecosystem will take 1, 10, 100, or 1000 years.

2.2 Update of critical loads for ICP Waters sites

Critical loads has been a subject of work within ICP Waters, as the data collected are well-suited for calculating critical loads using static models. Henriksen and Posch (1998) calculated critical loads and exceedances for ICP Waters sites using the static models SSWC (steady-state water chemistry model) and FAB (first-order acidity balance model). They used water chemistry data for the period 1992-94/95 from ICP Waters sites. For the sites in Europe they showed that the critical loads at the ICP Waters sites compared well with the 5th percentile of critical loads in the contiguous 50x50 km^2 EMEP grid cells derived from the European critical load database maintained at the CCE.

More recent data from the ICP Waters sites provides a basis for an update of these critical load estimates. In principle the critical load for a given site should not vary with time, but year-to-year variations in water chemistry occur due to natural causes such as climatic variations. We use data for ICP Waters sites for the period 2000-2002 to make new calculations of critical load using the First-order Acidity Balance (FAB) model (Posch et al. 1997, Henriksen and Posch 2001, UBA 2004).

The results show that the critical load estimates based on the 2000-02 data are very similar to those based on the 1992-94/5 data (Figure 2). For S the points fall very close to the 1:1 line. As expected the deviation is largest for the sites with highest critical load (S_{max}). This is because these sites have the highest concentrations of ions and thus most subject to natural variations in concentration due to dilution effects caused by variations in discharge. For N the points also fall close to the 1:1 line, but in many cases slightly below the line. This is because more conservative values for N uptake (set to zero) were used for the 2000-02 estimates.

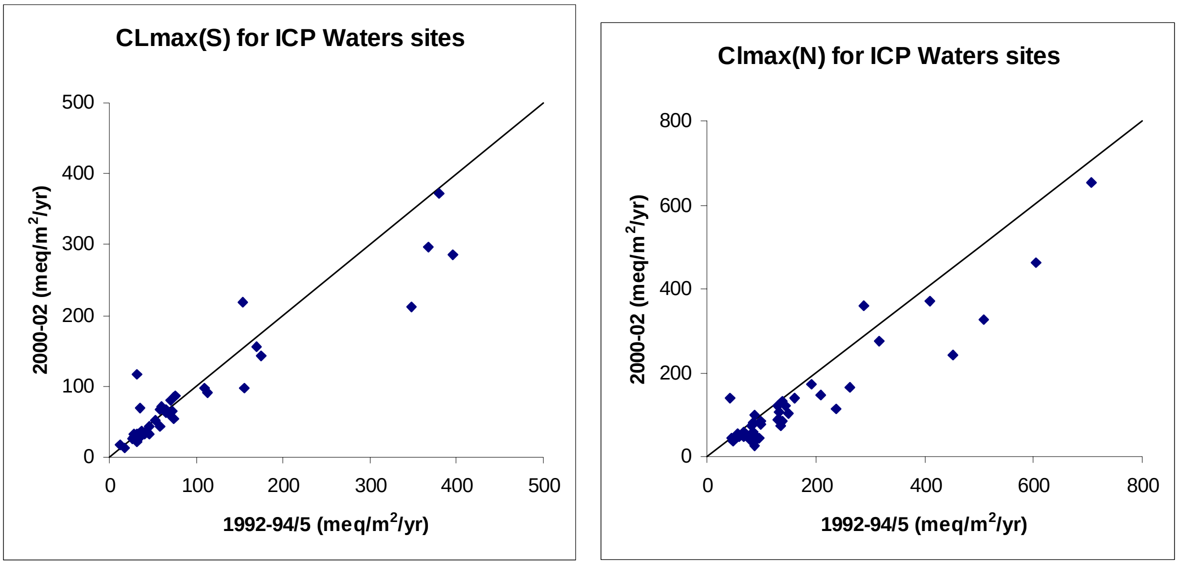

Figure 2. *Critical loads for ICP Waters sites calculated using the FAB model and based on data for the periods 1992-94/95 (from Henriksen and Posch 1998) and 2000-02. Left-hand panel: critical load for S (maximum); right-hand panel: critical load for N (maximum).*

2.3 Comparison of critical loads for ICP Waters sites with gridded data from the CCE database

The critical loads for the ICP Waters sites in Europe lie close to the 5th-percentile value for the EMEP 50 km grid cell in which the site lies (Figure 3). This is the same result as found by Henriksen and Posch (1998) for the 1992-94/5 data. ICP Waters sites thus generally lie at the most sensitive end of the range of waters within each EMEP grid cell. Both the critical load estimates for the ICP Waters sites and the 5th-percentile are based on new data; the ICP Waters data are from 2000-02 and the EMEP grid cell data are the results for surface waters from the CCE’s 2005 call for data.

Two sites are exceptions to this general result. The Italian site IT02 (Lago di Mergozzo) has much higher critical load for both S and N as compared to the 5th-percentile of its EMEP grid cell. Lago di Mergozzo is a high pH, well-buffered lake and lies within a region with many waters which are much more sensitive to acidification. The Swedish site SE 09 (River Fiolen) is a well-buffered large river (catchment area nearly 2000 km^2), and presumably the EMEP grid cell here contains many more sensitive smaller lakes and streams.

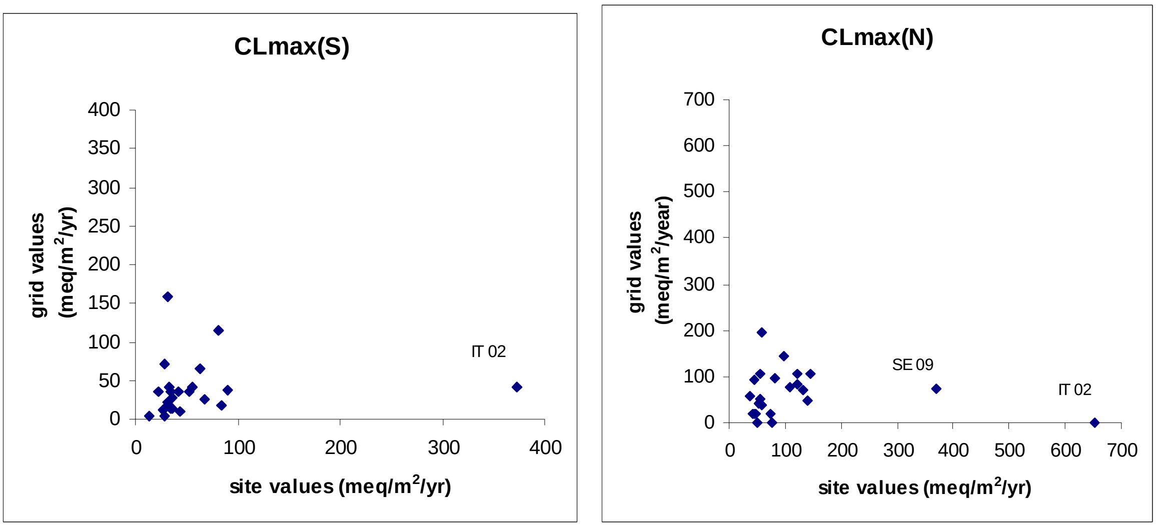

Figure 3*. Critical loads for ICP Waters sites in Europe as compared to the CCE’s 5th percentile contiguous grid values of* $Cl_{max}(S)$ *(left-hand panel) and* $Cl_{max}(N)$ *(right-hand panel).*

The comparison shows that most of the ICP Waters sites lie in the sensitive end of the range within each grid cell. This is of course intended, as the ICP Waters sites were largely originally chosen to monitor the acidification process. The sites are similarly well-suited to follow the recovery in response to decreases in S and N deposition. The ICP Waters data are an important supplement to the CCE database.

3. Dynamic modelling and target loads

3.1 Critical loads vs. target loads

The report on critical loads for ICP Waters sites (Henriksen and Posch 1998) was followed by an introduction to the use of dynamic models to derive target load functions for surface waters (Jenkins et al. 2002, Skjelkvale 2003). ICP-IM has also conducted work with dynamic models (Jenkins et al. 2003).

Static (steady-state) models are used to calculate critical loads whereas dynamic models are used to calculate target loads. The key difference between these approaches is related to the response times between changes in acid deposition and damage to biota. Just as the damage to biota was delayed beyond the onset of acid deposition, so the recovery from acidification will also be delayed. In the chain of events from the deposition of strong acids to the damage to key indicator organisms there are two major factors that can give rise to time delays. Biogeochemical processes can delay the chemical response in the catchment soils and consequently surface waters, and biological processes can further delay the response of indicator organisms, such as damage to fish. The static models to determine critical loads consider only the steady-state condition, in which the chemical and biological response to a change in deposition is complete. Dynamic models, on the other hand, attempt to estimate the time required for a new state to be achieved.

With critical loads, i.e. in the steady-state situation, only two cases can be distinguished when comparing them to deposition: (1) the deposition is below or equal to critical load, and (2) deposition is greater than critical load, i.e. exceedance. In the first case there is no problem, and no reduction in deposition is deemed necessary. In the second case there is, by definition, a risk of damage to the ecosystem, and therefore the deposition should be reduced.

The critical load concept assumes that acidification of soils and surface waters is fully reversible and that reducing deposition to or below the critical load removes the risk of harmful effects. In the case of surface waters this means that the chemical parameter (such as acid neutralising capacity ANC) that links the critical load to the biological effect attains a safe value and that there is biological recovery as well. The time required to achieve both these steps is not addressed. The removal of the risk of further damage, however, does not imply that recovery will necessarily occur. In addition, the reaction to changes in deposition is delayed by finite buffers, such as the cation exchange capacity in catchment soils. These buffers can delay the attainment of a critical chemical parameter by decades or even centuries. These finite buffers are not included in the critical load formulation, since they do not influence the steady state, but only the time to reach it.

Dynamic models, therefore, are needed to estimate the times involved in attaining a certain chemical state in response to given deposition scenarios (i.e. the target load).

The possible development of a chemical and biological variable in response to a ‘typical’ temporal deposition pattern can be summarised into five stages (Figure 4) (Jenkins et al. 2003):

Stage 1: Deposition was and is below the critical load (CL) and the chemical and biological variables do not violate their respective criteria. As long as deposition stays below the CL, this is the 'ideal' non-acidified situation.
Stage 2: Deposition has increased to above the CL, but chemical and biological criteria are not yet violated due to the time delay in response. No damage is likely to occur at this stage, therefore, despite exceedance of the CL. The time between the first exceedance of the CL and first violation of the biological criterion (the first occurrence of actual damage) is termed the *Damage Delay Time* ($DDT=t_3–t_1$).
Stage 3: The deposition is still above the CL and both the chemical and biological criteria are violated. Measures (emission reduction) have to be taken to avoid a (further) deterioration of the ecosystem status. This is the damaged acidified situation.
Stage 4: Deposition has been reduced to below the CL, but the chemical and biological criteria are still violated, and thus recovery can not yet occur. The time between the first non-exceedance of the CL and the subsequent non-violation of both criteria is termed the *Recovery Delay Time* ($RDT=t_6–t_4$).
Stage 5: Deposition is below the CL, and both criteria are no longer violated. This stage is similar to Stage 1, and only at this stage can the ecosystem be considered to have recovered.

Stages 2 and 4 can each be further subdivided into two sub-stages: Chemical delay times ($DDT_c=t_2–t_1$ and $RDT_c=t_5–t_4$; dark grey in Figure 4) and additional biological delay times ($DDT_b=t_3–t_2$ and $RDT_b=t_6–t_5$; light grey). Very often, due to the lack of operational biological response models, damage and recovery delay times mostly refer to chemical recovery alone and this is used as a surrogate for overall recovery.

Dynamic models may have a key role to play in the review of the Gothenburg Protocol and can provide a new effects driven basis to underpin any further negotiations in the future. They can be applied to determine the timing of ecosystem recovery in response to the Gothenburg Protocol and their capabilities in this respect are documented in this report.

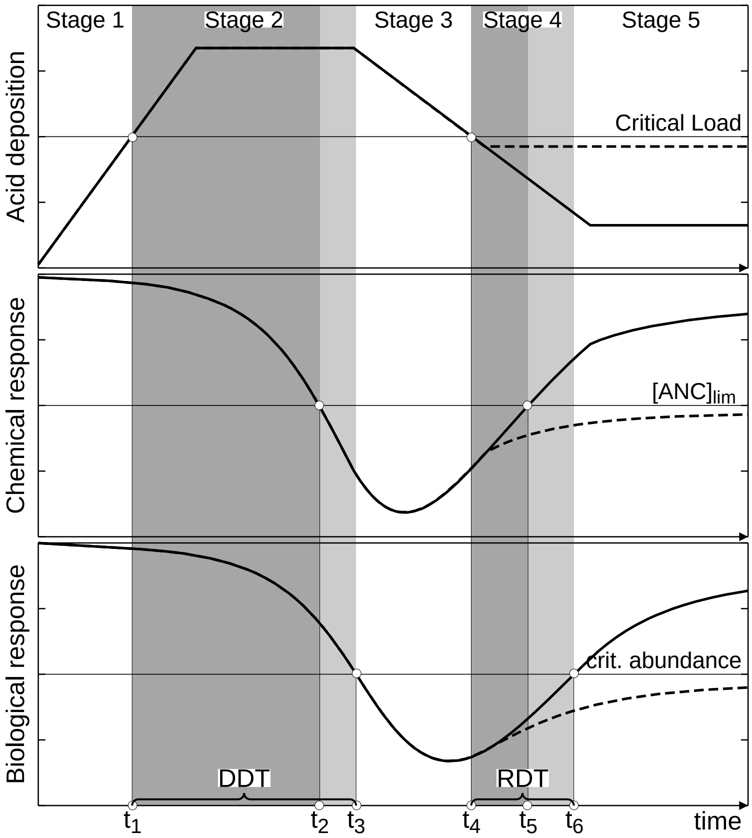

Figure 4. *'Typical' past and future development of the acid deposition affects on a lake chemical variable (ANC concentration) and the corresponding biological response in comparison to the critical values of those variables and the critical load derived from them. The delay between the (non-) exceedance of the critical load, the (non-) violation of the critical chemical criterion and the crossing of the critical biological response is indicated in grey shades, highlighting the Damage Delay Time (DDT) and the Recovery Delay Time (RDT) of the system. If deposition is reduced only to the critical load, recovery takes much longer (dashed lines) (from Jenkins et al. (2003).*

3.2 Modelling target load functions for surface waters

Four dynamic models have been identified as being widely used, documented and tested with respect to the requirement of the Convention (Posch et al. 2003). Several of these are simple enough to be applied on a regional scale. In response to the CCE's latest call for data the three countries that supplied target load function data for surface waters all used the MAGIC model. MAGIC focuses on surface water chemistry and is generally applied at catchment scale (Cosby et al. 1985).

MAGIC (Model of Acidification of Groundwater In Catchments) is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on soils and surface water chemistry (Cosby et al. 1985, Cosby et al. 2001). The model simulates soil solution chemistry and surface water chemistry to predict the monthly and annual average concentrations of the major ions in lakes and streams. MAGIC represents the catchment with aggregated, uniform soil compartments (one or two) and a surface water compartment that can be either a lake or a stream. MAGIC consists of (1) a section in which the concentrations of major ions are assumed to be governed by simultaneous reactions involving sulphate adsorption, cation exchange, dissolution-precipitation-speciation of aluminium and dissolution-speciation of inorganic and organic carbon, and (2) a mass balance section in which the flux of major ions to and from the soil is assumed to be controlled by atmospheric inputs, chemical weathering inputs, net uptake in biomass and losses to runoff. At the heart of MAGIC is the size of the pool of exchangeable base cations in the soil. As the fluxes to and from this pool change over time owing to changes in atmospheric deposition, the chemical equilibria between soil and soil solution shift to give changes in surface water chemistry. The degree and rate of change in surface water acidity thus depend both of flux factors and the inherent characteristics of the affected soils.

The soil layers can be arranged vertically or horizontally to represent important vertical or horizontal flowpaths through the soils. If a lake is simulated, seasonal stratification of the lake can be implemented. Time steps are monthly or yearly. Time series inputs to the model include annual or monthly estimates of (1) deposition of ions from the atmosphere (wet plus dry deposition; (2) discharge volumes and flow routing within the catchment; (3) biological production, removal and transformation of ions; (4) internal sources and sinks of ions from weathering or precipitation reactions; and (5) climate data. Constant parameters in the model include physical and chemical characteristics of the soils and surface waters, and thermodynamic constants. The model is calibrated using observed values of surface water and soil chemistry for a specific period.

MAGIC has been modified and extended several times from the original version of 1984. In particular, organic acids have been added to the model (version 5; (Cosby et al. 1995)) and most recently nitrogen processes have been added (version 7; (Cosby et al. 2001)). The MAGIC model has been extensively applied and tested over a 17 year period at many sites and in many regions around the world (Cosby et al. 2001).

4. Catchment processes leading to time lags

4.1 Time delay: sulphur

It has long been recognised that in some ecosystems S is strongly retained in soil, and thus there is a long time delay between changes in S deposition and response in SO_4 concentrations in runoff (Reuss et al. 1987). Sulphate adsorption is the main cause for this delay. The SO_4 adsorption capacity of soil is related to the amount of sequioxides (mostly Al and Fe secondary minerals) in the soil. Old, highly-weathered soils often have high SO_4 adsorption capacity. These soils are found in regions that were not glaciated during the Pleistocene.

The acidification history at White Oak Run, Virginia, USA, well-illustrates the time delay caused by SO_4 adsorption. White Oak Run (WOR) is a second-order stream which drains a 5.15 km^2 catchment in the Shenandoah National Park. This is the “type locality” for MAGIC, the catchment at which MAGIC was originally developed (Cosby et al. 1985). WOR is characterised by soils of deeply-weathered rocky silts or silty clay loams on steep hillslopes (Cosby et al. 1985). WOR is covered with second-growth forest 70-100 years old. A mixed stand of chestnut and white oak covers most of the catchment with scrub pines on drier south-facing slopes. Bedrock is quartzites and shales. WOR has temperate climate with precipitation about 1000 mm/yr and runoff 420 mm/yr. The site receives significant deposition of sulphur (about 100 meq/m^2/yr) and nitrogen (65 meq/m^2/yr). WOR is typical for sites with highly-weathered old soils (that is, on terrain not glaciated during the Pleistocene) in that during much of the 1900s a major fraction of the incoming S was adsorbed in the soil. The high SO_4 adsorption means that over the short term the catchment can accommodate large rates of SO_4 deposition without low ANC concentrations in streamwater, but not over the long term.

The MAGIC calibration to WOR indicates that S deposition has been above 50 meq/m^2/yr since about 1920, and yet the stream in 1990 still had ANC of nearly 20 µeq/l (**Figure 5**). The high sulphate adsorption capacity has caused the soil to retain a large fraction of the incoming SO_4. As the capacity gradually became filled, the concentration of SO_4 in streamwater rose and during the past 20-30 years led to the paradoxical situation in which SO_4 concentrations show a increasing trend despite the decreasing trend in SO_4 deposition.

The large amount of S stored in the soil can also delay the recovery of surface waters in response to reduction in S deposition. SO_4 adsorption is generally thought to be reversible and the response to increase or decrease in S deposition is symmetric. It is modelled in this way in MAGIC and other dynamic models.

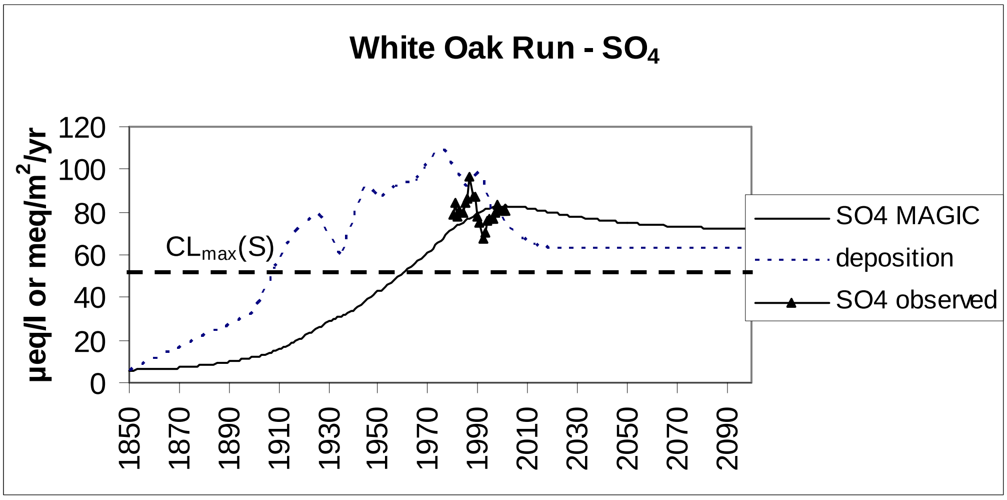

***Figure 5**. MAGIC simulation of streamwater chemistry at White Oak Run, Virginia, USA, with future conditions assuming implementation of the US Clean Air Act (CAA). Total SO_4 deposition (meq/m^2/yr) and SO_4 concentrations (µeq/l) in streamwater. Also shown is the CL for S (meq/m^2/yr) calculated by FAB.*

In young soils developed on glaciated terrain sulphate adsorption is usually minor, and SO_4 concentrations in surface waters follow S deposition closely, with lag times of a few years. This is well-illustrated by the data from Birkenes, Norway (Figure 6).

Birkenes is a 0.41 km^2 coniferous-forested catchment located about 20 km from the south coast of Norway. Birkenes has been a research and monitoring site since 1972 and is part of the Norwegian programme for monitoring long-range transported air pollution (SFT 2004). The chemical composition of precipitation and air are determined on samples collected daily by NILU (Aas et al. 2004), and for runoff on samples collected weekly by NIVA (SFT 2004). Soils have been sampled three times at 8-year intervals and analysed by Skogforsk (SFT 2004). The MAGIC model has been calibrated to this 30-year record by (Larssen 2005).

The Birkenes data clearly show that runoff responds rapidly to the decrease in S deposition. This is the case in most surface waters in glaciated terrain, as shown by the trend analyses conducted as part of the ICP Waters programme (Stoddard et al. 1999, Skjelkvåle 2003) as well as other studies (Evans et al. 2001, Prechtel et al. 2001).

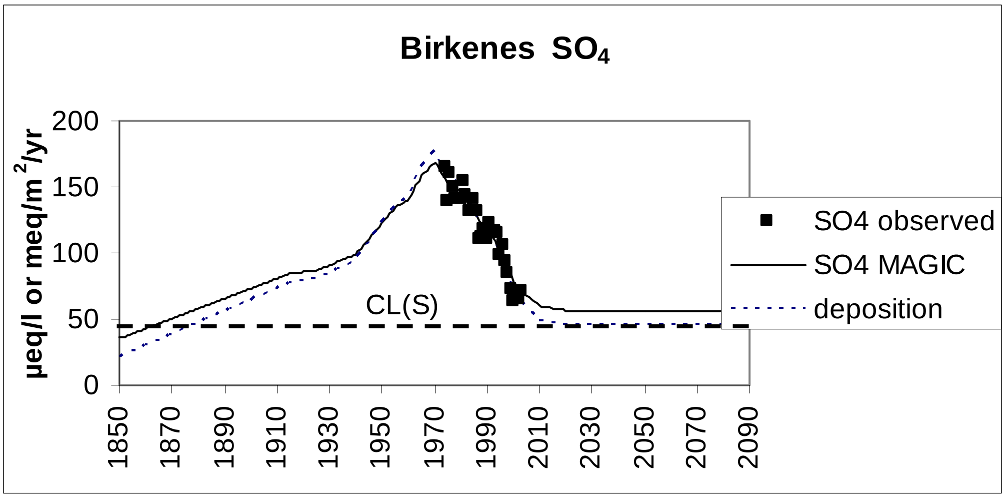

Figure 6*. Observed and MAGIC simulated streamwater chemistry at Birkenes, Norway, with future conditions assuming implementation of the UN-ECE LTRAP Gothenburg protocol and other legislation (the CLE scenario). Total SO_4 deposition (meq/m^2/yr) and SO_4 concentrations (µeq/l) in streamwater. Also shown are the $CL_{max}(S)$ calculated by FAB. Deposition data from Aas et al. (2004), streamwater data fromn SFT (2004), and MAGIC calibration from Larssen (2005).*

4.2 Time delay: nitrogen

Nitrogen is also strongly retained in terrestrial ecosystems. Here the usual situation is that N deposition (sum of reduced NH_y and oxidised NO_x species) greatly exceeds the output of inorganic N (mostly as NO_3) in runoff. Ammonium (NH_4) concentrations in runoff are usually negligible. Most of this N is retained in the soil and vegetation, as the gaseous losses due to denitrification are generally of minor importance. This situation is clearly not sustainable over the long term, as terrestrial ecosystems must have a finite capacity to store N. Terrestrial ecosystems subjected to chronically high levels of N deposition thus begin to leach more and more NO_3 with time. This condition is conventionally termed “nitrogen saturation” (Aber et al. 1989).

Birkenes provides an example of a forested catchment which retains nearly all of incoming N deposition. The flux of NO_3 in runoff amounts to only about 10% of the deposition flux of NH_4+NO_3 (Figure 7). Although the forest vegetation is slowly accumulating N in biomass, this accounts for only a very small fraction of the N retained. Most of the N must be stored in the soil, bound in soil organic matter.

The 30-year record of deposition and runoff chemistry at Birkenes indicates that the catchment is proceeding very slowly towards N saturation (Figure 7). Annual mean NO_3 concentrations in runoff are less than 20 µeq/l, whereas at steady-state concentrations above 100 µeq/l would be expected. In MAGIC the accumulation of N in the soil is assumed to lead to decreased C/N ratio in soil organic matter, and this in turn is assumed to regulate the fraction of incoming N released to runoff. At Birkenes, this MAGIC calibration suggests that several hundred years of N deposition will be required to reach the FAB steady-state condition in which all incoming N is lost from the catchment.

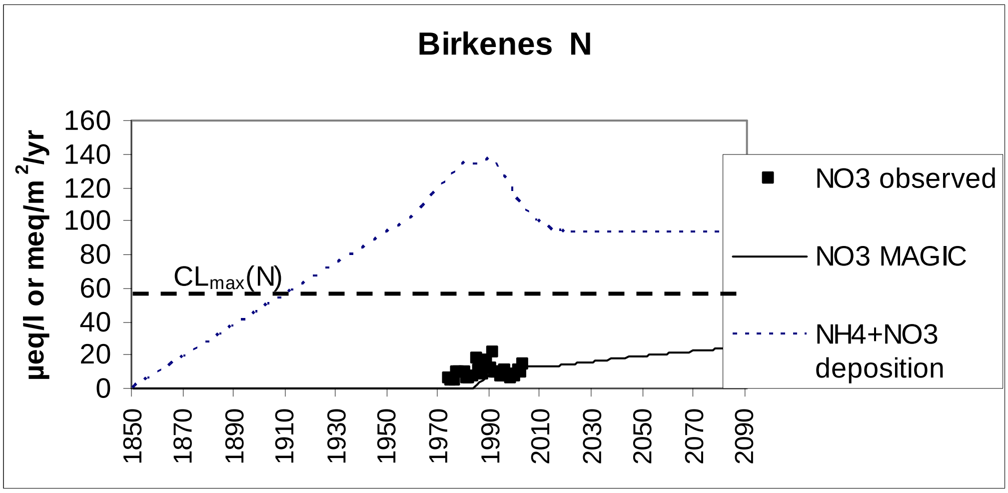

Figure 7. *Observed and MAGIC simulated streamwater chemistry at Birkenes, Norway, with future conditions assuming implementation of the CLE scenario.* NH_4+NO_3 *deposition* ($meq/m^2/yr$) *and* NO_3 *concentrations in streamwater. Also shown are the* CL_{max} *for N calculated by FAB, and the* ANC_{limit} *for damage to fish. Deposition data from Aas et al. (2004), streamwater data from SFT (2004), and MAGIC calibration from Larssen (2005).*

Whereas the response of runoff to increased N deposition may be delayed by many decades, the response to decreased N deposition can be quite rapid. In contrast to the situation for S deposition, N deposition has not decreased dramatically during the past 20 years, and thus there are few good examples of ecosystem response to decreased N deposition. Here the best evidence is from large-scale whole ecosystem experiments with roofs, such as those conducted as part of the RAIN project at Risdalsheia, Norway (Wright et al. 1993) and the NITREX project at Ysselsteyn, the Netherlands (Boxman et al. 1998). At these sites a dramatic decrease in N deposition resulted in rapid decrease in NO_3 in runoff and soil solution (Figure 8).

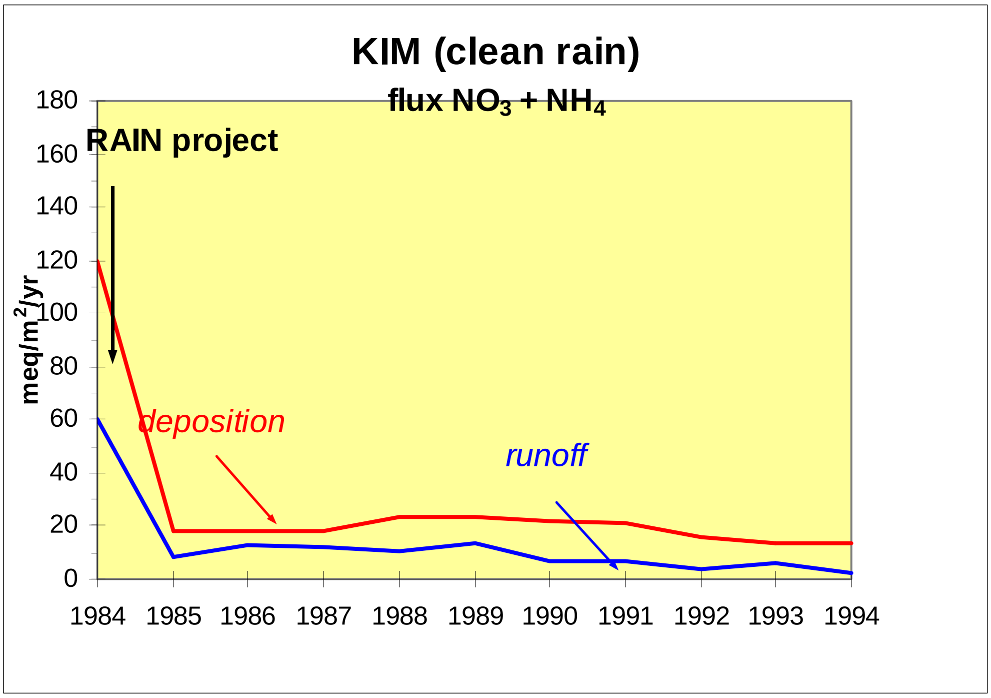

Figure 8*. Nitrogen deposition and runoff at KIM catchment, Risdalsheia, Norway. N deposition was excluded beginning 1985 by roof over the catchment (arrow indicates start of treatment). Data from Wright et al.(1993) and Wright (1998).*

4.3 Time delay: base cations

Acid deposition causes a long-term slow depletion of base cations for the soil. The increased flux of strong acid anions (SO_4 and NO_3) from the soil is accompanied by cations, the acid cations H^+ and Al^{n+} and the base cations Ca, Mg, Na, and K. The relative concentration of these two groups of cations depends largely on the base saturation (%BS) of the soil. Soil with high %BS releases more base cations relative to soil with low %BS (Reuss et al. 1987).

This is well-illustrated by the 30-year record of observed concentrations of sum of base cations (SBC) in streamwater at Birkenes, Norway. The concentrations of SBC in streamwater rose during the period of high acid deposition and high concentrations of SO_4 to peak in the 1970s. As SO_4 deposition and SO_4 concentrations in streamwater declined from the mid-1980’s, the concentration of SBC also declined (Figure 9).

Recovery of soil %BS can first begin when the leaching rate of base cations falls below the rate of base cation supply from deposition and weathering. In acidified catchments, therefore, a moderate decrease in acid deposition may simply slow the rate of depletion of base cations from the soil, not reverse the trend. The long-term pattern of soil %BS is often asymmetric; the decrease can be rapid (halving the %BS in only several decades), while the replenishment following major decline in acid deposition may require centuries.

The MAGIC reconstructed %BS in the soil at Birkenes shows a long-term depletion of base cations in the soil caused by the chronic high levels of acid deposition. The model indicates that 140 years of acid deposition removed about 1/3 of the base cations form the ion-exchange complex in the soil. The 70-80% decrease in acid deposition from the mid-1980s to 2010 is sufficient to stop further decline, but the model indicates that the replenishment of the BS will proceed very slowly and require much longer time (Figure 9). The build-up of the soil %BS means that there will be a gradual increase in the SBC and decrease in acid cations in streamwater, and thus a gradual increase in ANC.

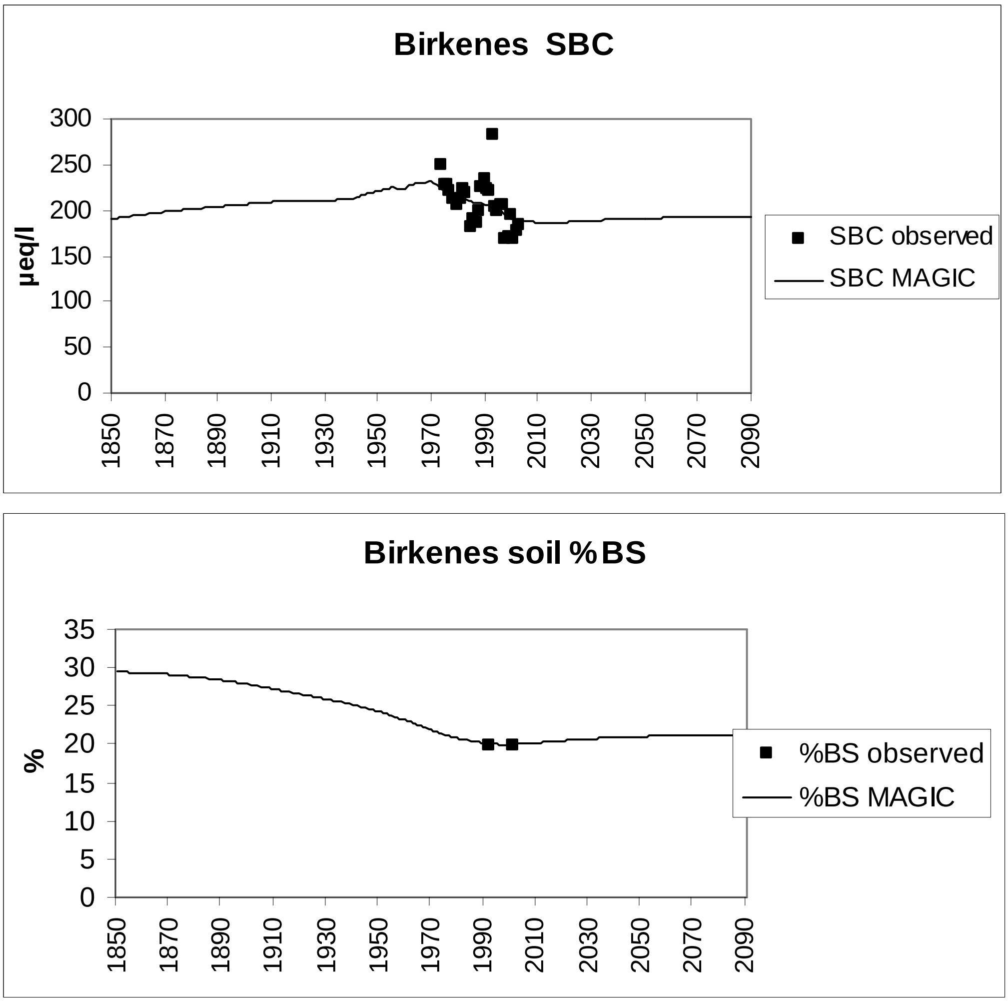

Figure 9. *Observed and MAGIC simulated sum of base cations (SBC) in streamwater and base saturation (%BS) in soil at Birkenes, Norway, with future conditions assuming implementation of the CLE scenario. Streamwater and soil data from SFT (2004), and MAGIC calibration from Larssen (2005).*

5. Target load functions vs. critical load functions

5.1 Example Birkenes

Birkenes is a typical acidified surface water which requires substantial reduction in acid deposition to allow recovery. The MAGIC simulation of streamwater chemistry given implementation of the CLE scenario indicates that ANC will remain below 0 µeq/l unless additional emission reductions are made (Figure 10). The simulation suggests that ANC will improve until about 2020 and then begin to decline once again. The improvement is due to the reduced SO_4 deposition which leads to decreased SO_4 concentrations in streamwater, and also to the slow increase in %BS in the soil. The downturn from about 2020 is due to the gradual increase in NO_3 leaching due to the slow progression of N saturation in the catchment.

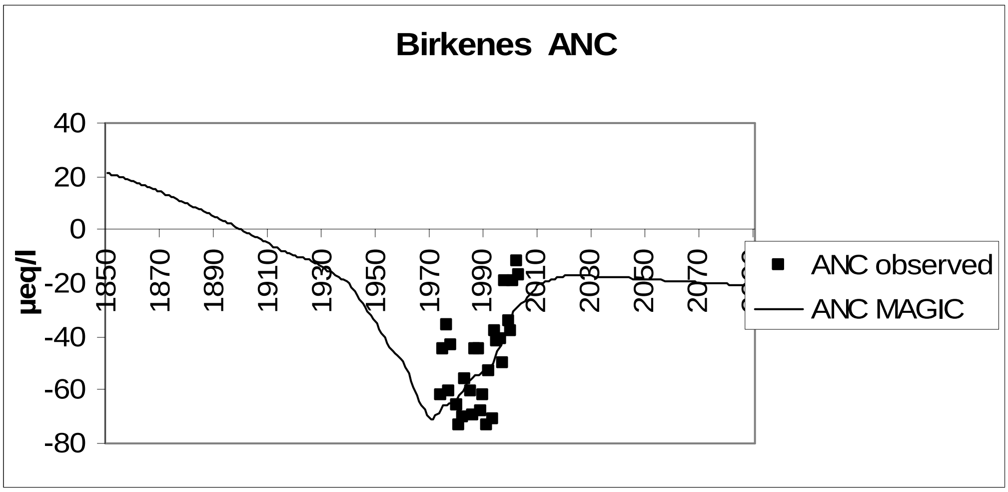

***Figure 10**. Historical and future ANC at Birkenes as simulated by MAGIC. Future conditions assume implementation of the CLE scenario. Also shown are observed volume-weighted annual ANC. Streamwater data from SFT (2004), and MAGIC calibration from Larssen (2005).*

At Birkenes the target load for sulphur lies below the critical load (Figure 11). Recovery of the stream ANC to the target of ANC=0 µeq/l requires larger reduction in S deposition if the target is to be achieved soon (i.e. year 2030) rather than later (e.g. 2090). As very long time scales the target load for sulphur approaches the steady-state critical load.

For N the situation is the opposite (Figure 11). Over the short term the site can accommodate large amounts of N deposition as most is retained in the soil and does not contribute to acidification of runoff and the low ANC. But over the long term, if the concept of N saturation holds, the catchment ecosystem will retain less and less of incoming N and the target load will be lower. At very long times, the target load will approached the steady-state critical load. The target load functions thus cross the critical load function (TL < CL for S and TF > CL for N).

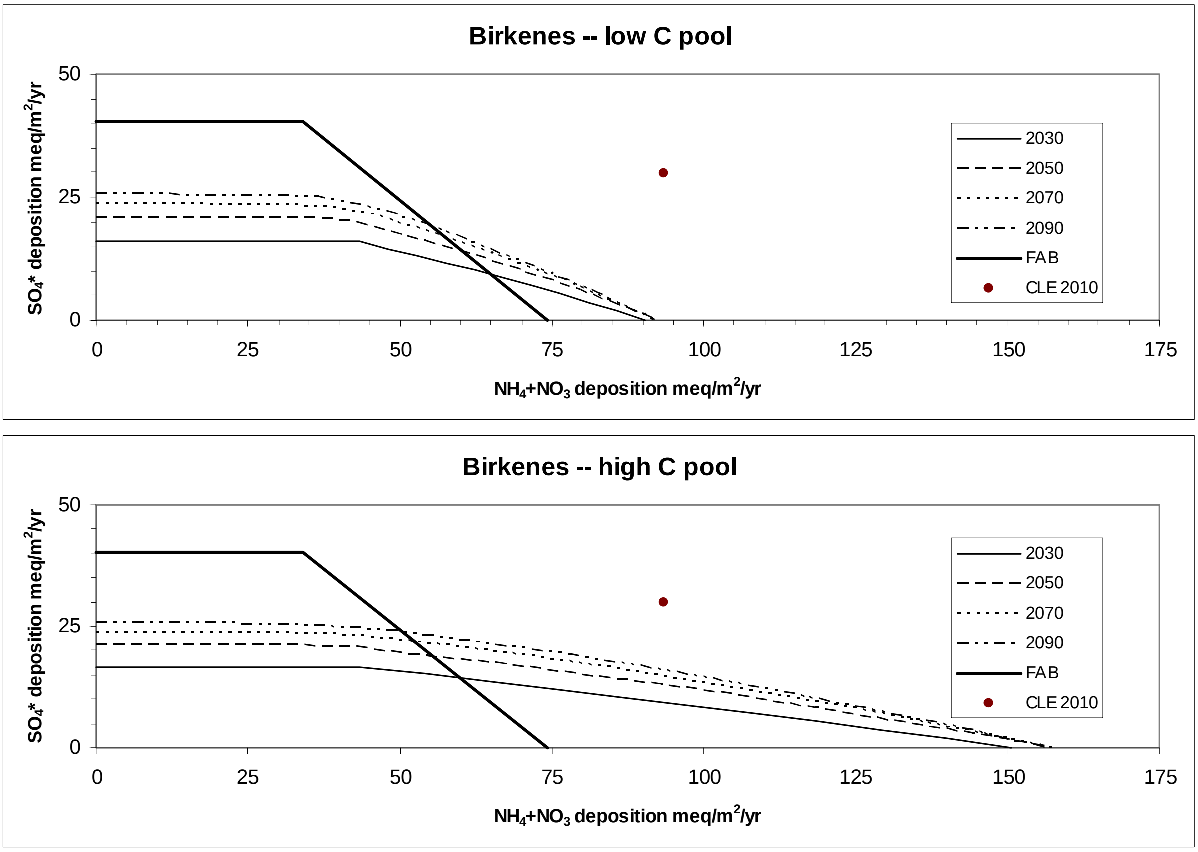

Figure 11. *Target load functions and critical load function for surface water at Birkenes, Norway. The TLF are calculated by MAGIC. The critical load function is calculated by the FAB model. Four TLFs are calculated for achievement of the criterion of ANC= 0 µeq/l by the year 2030, 2050, 2070 and 2090, respectively. Also shown is the deposition estimated for the year 2010 assuming that the CLE scenario is fully implemented. Upper panel: TLFs assuming a low C pool (only the organic horizons 593 molC/m^2) in the soil. Lower panel: TLFs assuming a high C pool (the entire soil profile 2933 molC/m^2).*

The larger the active C pool in the soil the greater amount of N that the soil can retain. The path to N saturation is lower, and thus the TL for N is higher (Figure 11). At Birkenes if only the C in the organic soil horizons is considered active, then the TL for N for the year 2030 is about 90 meq/m^2/yr, whereas if the entire soil C pool is active, the TL for N for year 2030 is 150 meq/m^2/yr.

The TLF crosses the CLF. The protocol used by the CCE is based on the precautionary principle and thus in all cases the minimum of the TLF and CLF is used. In this example for Birkenes this means that for low values of N deposition the TLF will be used, whereas for high values of N deposition the CLF will be used.

5.2 Example White Oak Run

At White Oak Run the target load function lies above the critical load function both with respect to S and to N (Figure 12). Over the short term S deposition can be much larger without causing critically low ANC levels; the target load for S is higher than the critical load. Only after nearly 800 years does the target load approach the critical load. Again based on the precautionary principle the minimum of the TLF and CLF will be used. In this case for WOR then the CLF will be used for all pairs of S and N deposition.

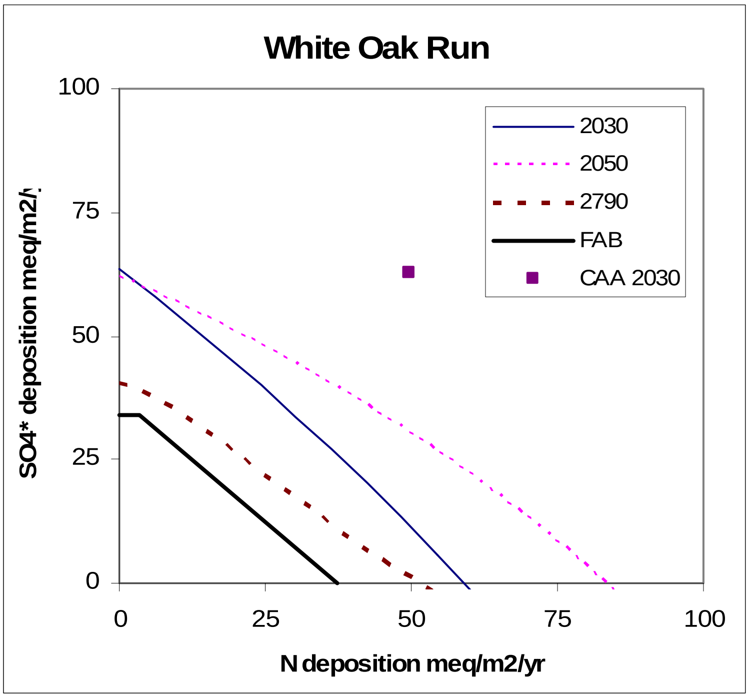

Figure 12. *Target load functions and critical load function for surface water White Oak Run, Virginia, USA. Three TLFs are calculated by MAGIC for achievement of the criterion of ANC= 20 µeq/l by the year 2030, 2050, and 2790 (800 years in the future). The critical load function is calculated by the FAB model. Also shown is the deposition expected in year 2030 if the Clean Air Act is fully implemented (solid square). The target load functions were calculated assuming implementation of the Clean Air Act by year 2010, and further reductions implemented in 2010.*

5.3 Seasons vs. year

Critical loads and target load functions supplied by the National Focal Centres to the CCE are based on yearly data, usually estimates of volume-weighted concentrations or fluxes for deposition and surface waters. The calculations use an ANC_{limit} set to protect an indicator organism, such as brown trout. The use of yearly values has been adequate in the negotiations of the 1994 Oslo Protocol and the 1999 Gothenburg Protocol, in that these protocols were aimed at reducing the deposition of S and N such that exceedance in Europe was minimised. These protocols took a big step closing the gap between critical load and deposition.

But as deposition approaches the level of critical load, and surface waters recover to the point at which indicator organisms can begin to reappear, the seasonal and episodic variations in water quality become increasingly important. Many of the chemical species that enter into the critical load and target load calculations show large annual variations due to such phenomena as snowmelt and high flow events. The natural biological cycle in vegetation and soils of the catchment also affects the seasonal pattern in concentrations of many chemical species.

Nitrate is one of the key parameters that typically has large seasonal variations. The situation at Øygardsbekken, Norway, well-illustrates the seasonal pattern observed in streams in areas receiving chronic high N deposition and exhibiting signs of increased leaching of NO_3 (Figure 13).

Øygardsbekken is one of seven catchments monitored as part of the Norwegian Monitoring Programme for Long-Range Transported Air Pollutants (SFT 2004). The stream drains 2.55 km^2 of heathland including a small lake. Bedrock is gneissic, with thin and patchy organic-rich soils. N deposition is high (for Norway) and was about 120 $meq/m^2/yr$ in the mid-1990s.

Several catchment processes act to give the seasonal pattern in NO_3 concentrations in streamwater at Øygardsbekken. Uptake of N species by vegetation is linked to the growing season (temperature) and highest during the summer months. Litterfall in the autumn returns N to the soil. Decomposition of litter and soil organic matter is a function of temperature and moisture. And finally immobilisation of inorganic N compounds is temperature dependent. All these processes can be simulated by MAGIC. Simulation of the monthly variations requires monthly timesteps in MAGIC. Such a calibration has been made for Øygardsbekken by Sjøeng and Kaste (2005).

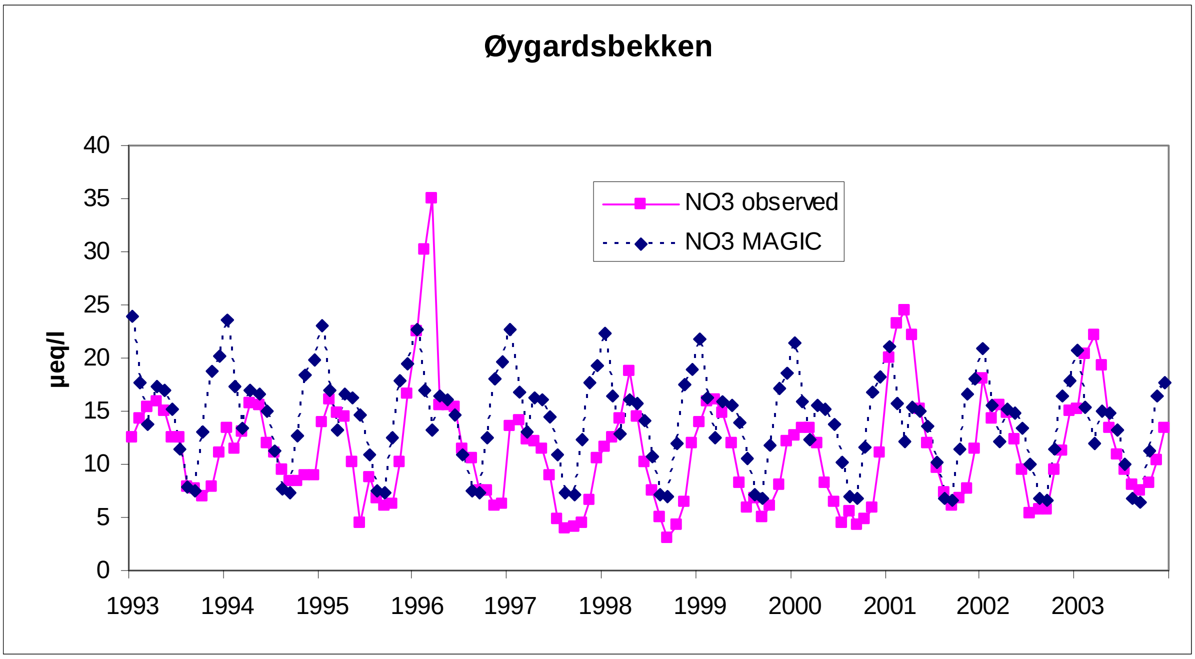

***Figure 13**. Monthly volume-weighted concentrations of NO_3 in runoff at Øygardsbekken observed and simulated by MAGIC. Observed data from SFT (2004); MAGIC calibration from Sjøeng and Kaste (2005).*

When based on yearly data the target load functions and critical load function for Øygardsbekken (Figure 14) are quite similar to those for Birkenes (Figure 11). The TLFs cross the CLF and the TLF moves towards the CLF with longer and longer time for achievement of the ANC criterion.

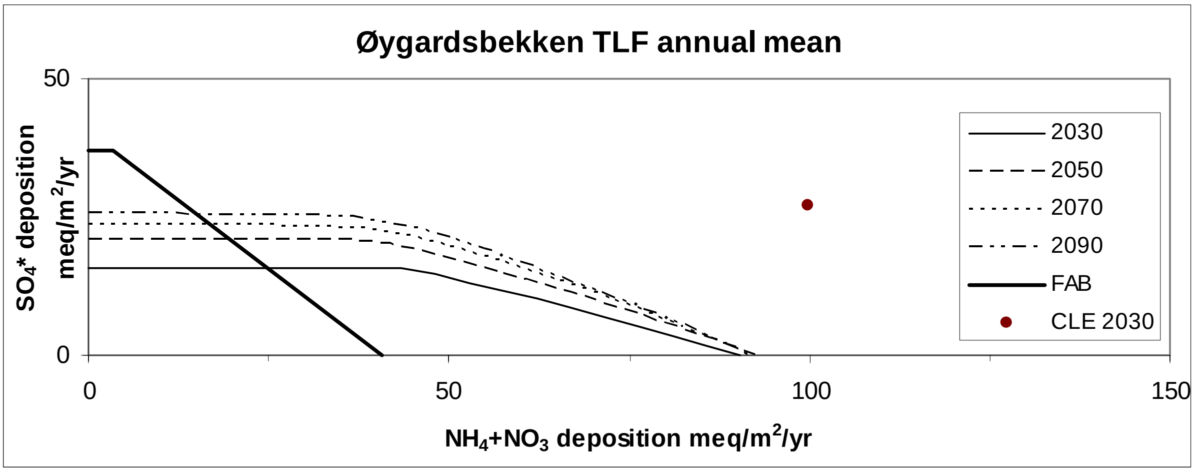

***Figure 14.** Target load functions and critical load function for surface water at Øygardsbekken, Norway. Four TLFs are calculated for achievement of the criterion of ANC= 0 µeq/l by the year 2030, 2050, 2070 and 2090, respectively. The critical load function is calculated by the FAB model. Also shown is the deposition estimated for the year 2010 assuming that the CLE scenario is fully implemented.*

But TLFs differ greatly depending upon which month the goal is to be achieved (Figure 15). If the goal is to be achieved for the month of September when NO_3 concentrations are at the lowest for the year, the TLF is considerably higher than that of the yearly mean. On the other hand, if the goal is to achieve the target in March when NO_3 concentrations peak, then the TLF is considerably lower (in this case zero; cannot be achieved even with zero S and N deposition).

The CCE database is generally based on annual estimates. Response to changes in acid deposition, however, may be most pronounced in one or more seasons. The ICP Waters monitoring at many sites is conducted with sufficient frequency to detect changes in seasonal patterns. A future challenge is to model effects at timesteps shorter than one year.

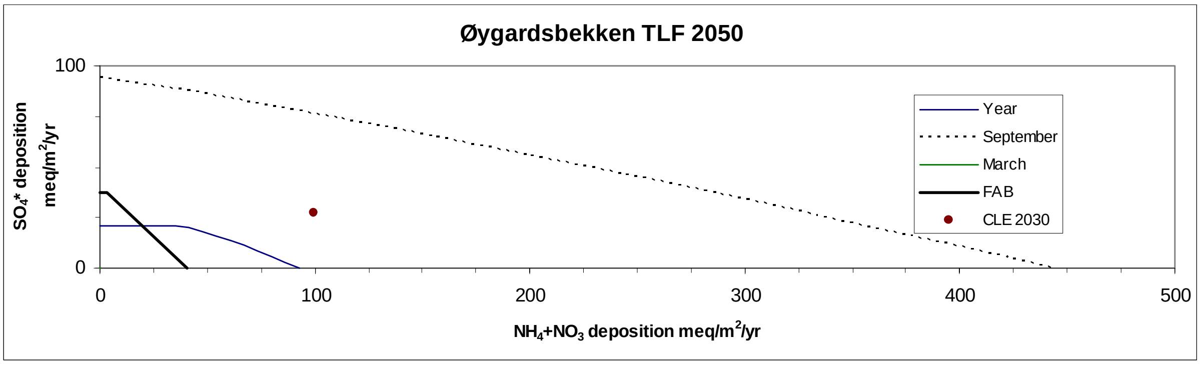

***Figure 15**. Target load functions for the year 2050 and critical load function for surface water at Øygardsbekken, Norway, calculated for yearly mean data (solid line), September data (dotted line), and March data (zero, zero; does not appear on the graph). Also shown is the critical load function is calculated by the FAB model and the deposition estimated for the year 2010 given the CLE scenario.*

5.4 Climate change

Climate change is one of several factors that can affect acidification of freshwaters in the future. Climate change might affect the amount and timing of deposition of acid compounds. More storms with high winds will increase the deposition of seasalts in coastal areas. Many catchment processes are affected by changes in temperature and moisture. These include growth of forest, mineralization of soil organic matter, immobilisation of N compounds, rate of weathering, and production of dissolved organic matter. The relative importance of each of these potential effects on streamwater chemistry will, of course, vary from site-to-site. The predicted future climate will also vary from site to site. And the future climate change predicted by global climate models (GCMs) varies among the GCMs and depends on the scenario for emissions of greenhouse gases.

As a first step in evaluating the potential impact of climate change on recovery of surface waters from acidification, a series of trials with MAGIC has been conducted at 14 sites in Europe and North America (Wright et al. In press). Birkenes, Norway, is one of the sites included. The trials at Birkenes assumed a future in crease in temperature, precipitation and storminess. The magnitude of the response was dependent on the size of the active C pool in the soil. The simulations suggested that with a high C pool (i.e. the entire C in the soil is active), the effect of future climate change on streamwater ANC will be small, but with a low C pool (only the C in the organic horizons active) the re-acidification of the streamwater will be major over the next 50 years (***Figure 16)***. The most important effect here is the acceleration of N saturation due to the increased mineralization of soil organic mater caused by the higher temperature in the future.

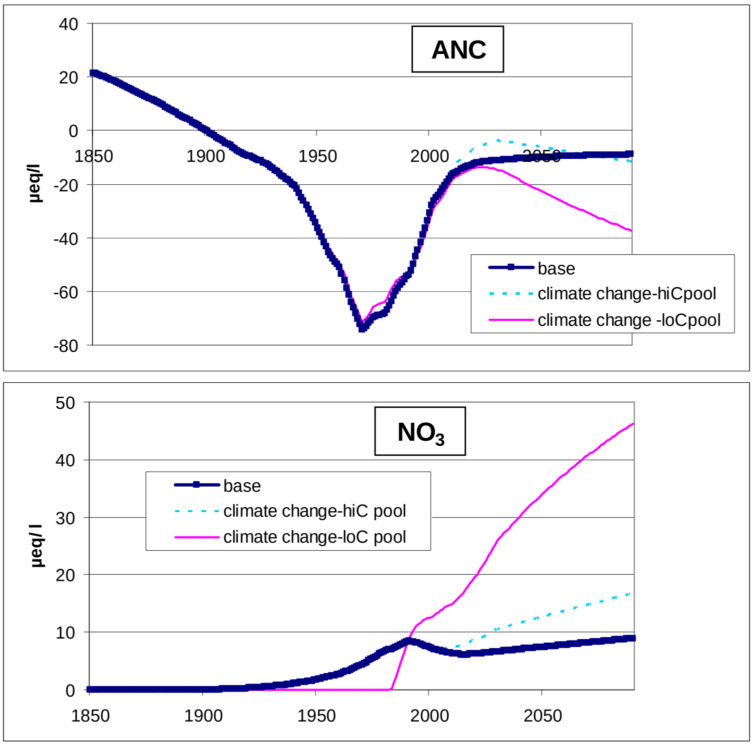

Figure 16. *ANC (top panel) and NO$_3$ concentrations at Birkenes simulated by MAGIC for a future scenario of climate change and with high and low C pool in the soil. The climate change scenario is the combination of seven possible effects of a warmer, wetter and windier climate during the next 100 years. Modified from Wright et al. (In press).*

The scenario with future climate change alters the TLF (Figure 17). The alteration is pronounced for both the case with high soil C pool and low soil C pool. In both cases the target load is lower with climate change, and is more pronounced for N than for S.

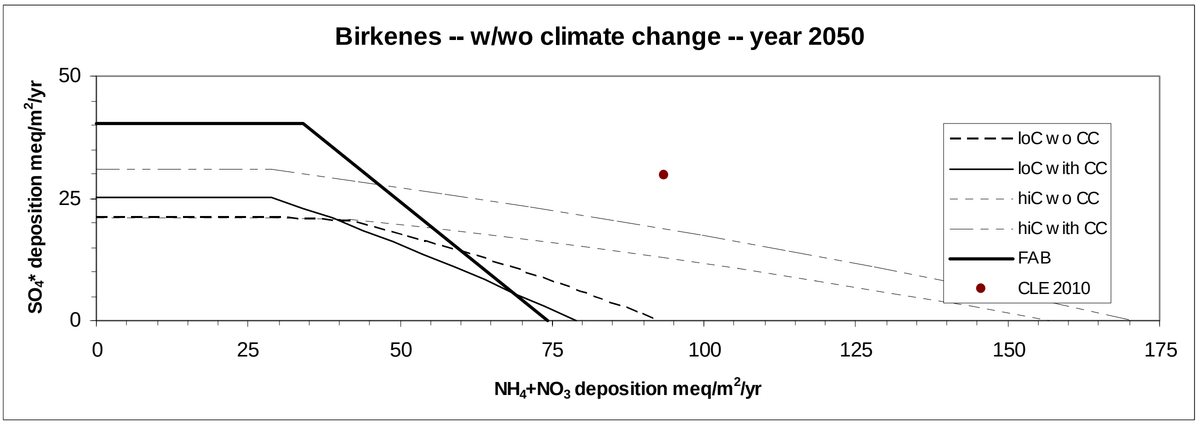

Figure 17. *Target load functions and critical load function for surface water at Birkenes, Norway, for achievement of ANC = 0 µeq/l by the year 2050 assuming present-day climate conditions (without CC) and given a scenario of future climate change (with CC). The critical load function is calculated by the FAB model. The TLF are calculated for two sizes of soil C pool: a high C pool (the entire soil profile 2933 molC/m2) and a low C pool (only the organic horizons 593 molC/m2). Also shown is the deposition estimated for the year 2010 assuming that the CLE scenario is fully implemented.*

6. Concluding remarks

The updated CL for the ICP Waters sites based on 2000-02 data were very similar to those calculated based on 1992-94/5 data. The comparison with the 5th percentile from the EMEP 50 km grid cells for water was also very similar. This is reassuring and lends confidence in the robustness of the methods used to calculate critical loads and to upscale to the European EMEP grid. With only a few exceptions the ICP Waters sites tested lie at the most sensitive end of the scale within each respective grid cell. They are thus well-suited for monitoring response to changes in S and N deposition.

Use of dynamic models to set target loads represents a new degree of complexity in the scientific underpinning of the work conducted with the UN-ECE Convention on Long-range Transboundary Air Pollutants. The MAGIC model was used to calculate TLFs for freshwaters in three countries in response to the call for data issued by the CCE in November 2004. Freshwaters in sensitive regions elsewhere in Europe and North America have been modelled using MAGIC, and these applications have been used to assess the effects of scenarios for future S and N deposition (Wright et al. 2005).

The addition of the time dimension necessitates facing several major uncertainties in the understanding of biogeochemical processes which affect surface water chemistry. Of these N retention and loss in catchments continues to pose a major unknown, despite extensive scientific research on the subject in recent decades. While it is clear that chronic high levels of N deposition cannot be sustained in catchments over the long run, the rate of transition from a high degree of retention (the present-day situation in many catchments) to the steady-state condition of low retention with no change in pools of N in soil and vegetation (as assumed by the FAB model and in accordance with the precautionary principle) is simply not known in most cases. The now 30+ year records of N deposition and leaching from small catchments do not yield a consistent pattern pointing to progressive degree of N saturation (Wright et al. 2001). The large-scale whole ecosystem experiment with N addition (NITREX) at Gårdsjön, Sweden, shows that even 13 years of very high N deposition (50 kg/ha/yr) only caused the fraction of N leached to increase from 2 to 14% (Moldan et al. in press). This experiment shows that for this region of Sweden in any case, over the next several decades there is little risk of increased N leaching due to continued N deposition.

The large gap between the long-term steady-state and the present-day N retention means that the TLF differs from the CLF, especially with respect to N deposition. Over periods of several decades catchments not currently showing N saturation can receive large N deposition with little adverse effects on the ANC of surface water.

For sites located in young soils with low S adsorption capacity, the TLF typically crosses the CLF; the TLF indicates that S deposition must be decreased below the CL, whereas for N the TLF indicates that for many decades sites can tolerate higher deposition than the CL. This is because for catchments with high present-day N retention, NO_3 concentrations are low and SO_4 comprises most of the sum of non-marine acid anions. Because the recovery of soil base saturation is slow, S deposition must be decreased to levels below the CL to achieve the ANC target within the next several decades.

The effects of future climate change on catchments and freshwaters add new levels of complexity to the evaluation of scenarios of acid deposition. The research fields of acid deposition and climate change have many areas of overlap. For example, emission of S gases affects the atmospheric energy balance and global warming affects the release of NO_3 from soils. Although many of the possible effects of climate change can be simulated by MAGIC, these exists a requirement for further development of the model. These developments are required in the near future if the full potential of dynamic models is to be realised within the timescale of the Convention, i.e. the forthcoming review of the Gothenburg protocol as scheduled by the medium term strategy of the Working Group on Effects.

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Appendix A. Publications from ICP Waters

All reports from the ICP Waters programme from 1987 up to present are listed below. All reports are available from the Programme Centre. Publications from 2002 up to present can be found at http://www.iis.niva.no/ICP%2Dwaters

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