Convention on Long-range Transboundary Air Pollution

International Cooperative Programme on Assessment and Monitoring of Acidification of Rivers and Lakes

ICP-WATERS REPORT 70/2002

Dynamic Modelling of Surface Waters:

Impact of emission reduction – possibilities and limitations







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Title Dynamic Modelling of Surface Waters: Impact of emission reduction - possibilities and limitations	Serial No. SNO 4598-2002	Date 9.12.2002	
	Report No. Sub-No. ICP Waters report 70/2002	Pages Price 42	
Author(s) Jenkins, Alan (CEH Wallingford, UK); Larssen, Thorjørn; Moldan, Filin (IVI, Sweden): Posch Maximilian (RIVM/CCE, The	Topic group Distribution Acid Rain		
<i>Netherlands),</i> Wright, Richard F.	Geographical area Europe	Printed NIVA	

Client(s)

UK Department of the Environment, Food and Rural Affairs (Contract No. EPG 1/3/194), ASTA International and National Abatement Strategies for Transboundary Air Pollution, Norwegian Pollution Control Authorities (SFT). Client ref.

Abstract

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.

The MAGIC model has been tested extensively against long-term diatom reconstructions, experimental manipulation experiments and long-term chemistry observations and has in all cases been found to successfully capture the dynamics of chemical change. This also provides confidence in the predictions from the model.

Methods for summarising regional model predictions are summarised and a methodology for the derivation of 'target' loads is proposed for use within the Convention. Requirements for further research, including model uncertainty and the link between surface water chemistry and biological receptors, are outlined.

4 keyw	ords, Norwegian	4 keywords, English	
1. 2. 3. 4.	Forsuring Overflatevann Dynamisk modellering Regional modellanvendelse	 Acidification Surface waters Dynamic modelling Regional model application 	

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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 of Surface Waters: Impact of emission reduction possibilities and limitations

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ICP Waters Programme Centre, Norwegian Institute for Water Research Oslo, November 2002

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 has been lead by Berit Kvæven, Norwegian Pollution Control Authority.

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 aim of this report is to evaluate the possibilities and limitations in using dynamic modelling of surface water to predict future water quality due to emission reductions of sulphur and nitrogen.

The report is written by experts in the field of dynamic modelling of surface waters as a contribution in kind to the ICP Waters Programme.

The report has been discussed and approved by the ICP Waters 18th Task Force meeting, held in Moscow 7-9 October 2002.

ICP Waters Programme Centre Oslo, November, 2002

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Summary

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.

The MAGIC model has been tested extensively against long-term diatom reconstructions, experimental manipulation experiments and long-term chemistry observations and has in all cases been found to successfully capture the dynamics of chemical change. This also provides confidence in the predictions from the model.

Methods for summarising regional model predictions are summarised and a methodology for the derivation of 'target' loads is proposed for use within the Convention. Requirements for further research, including model uncertainty and the link between surface water chemistry and biological receptors, are outlined.

1. Introduction

The link between the emission of oxides of sulphur and nitrogen (and reduced nitrogen) to the atmosphere and the acidification of soils and surface waters is now well established and understood. The impact of the chemical changes on biota is also sufficiently understood such that chemical targets aimed at protecting both aquatic and terrestrial biota have been established to form the basis of international agreements on emission reductions within the UNECE and EU. The link between deposition of acidic pollutants and the loss of or damage to biota, however, is not immediate.

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 (steady) state to be achieved. This report describes the possibilities and limitations of using dynamic models to better define the limits and timescales of the recovery processes.

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 (and or equal to) critical loads, i.e. does not exceed critical loads, and (2) the deposition is greater than critical loads, i.e. there is critical load exceedance. In the first case there is no (apparent) problem, i.e. no reduction in deposition is deemed necessary. In the second case there is, by definition, an increased risk of damage to the ecosystem, and therefore the deposition should be reduced. A critical load serves as a warning as long as there is exceedance, since it tells that deposition should be reduced. However, it is often assumed that acidification of soils and surface waters is fully reversible and that reducing deposition to (or below) critical loads immediately removes the risk of 'harmful effects', i.e. the chemical parameter (e.g. the [ANC]-limit) that links the critical load to the biological effect(s), immediately attains a non-critical ('safe') value and that there is immediate biological recovery as well. The removal of the risk of further damage, however, does not necessarily imply that recovery will occur. In addition, the reaction to changes in deposition is delayed by (finite) buffers, such as the cation exchange capacity (CEC) in catchment soils. These buffers can delay the attainment of a critical chemical parameter and it might take decades or even centuries, before a (new) equilibrium (steady state) is reached. 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. It is also likely that the desirable or critical chemical target will be achieved prior to a new steady state and so the concept of equilibrium in the long term becomes irrelevant. Dynamic models, therefore, are needed if we wish to estimate the times involved in attaining a certain chemical state in response to given deposition scenarios, e.g., the consequences of 'gap closures' in emission reduction negotiations. In addition to the delay in chemical recovery, there is likely to be a further delay before the 'original' biological state is reached, i.e. even if the chemical criterion is met (e.g. [ANC]>0), it will take time before full biological recovery is achieved as a result of the dispersion characteristics of the species, for example. On the other hand, the possibility remains that the original biological status will not be recovered but this possibility is common to both critical load and dynamic approaches.

The possible development of a chemical and biological variable in response to a 'typical' temporal deposition pattern can be summarised into five stages (Figure 1):

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' situation.

Stage 2: Deposition is above the CL, but (chemical and) biological criteria are not violated because there is a time delay before this happens. 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 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.

Stage 4: Deposition is below the CL, but the (chemical and) biological criteria are still violated and thus recovery has not yet occurred. 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 $(DDT_c=t_2-t_1 \text{ and } RDT_c=t_5-t_4; \text{ dark grey in Figure 1})$ and (additional) biological delay times $(DDT_b=t_3-t_2 \text{ and } RDT_b=t_6-t_5; \text{ light grey})$. 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.



Figure 1. '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).

The speed of recovery also depends on the extent of the deposition reduction (Figure 1). If deposition is reduced to (or just below) the critical load only, chemical and biological recovery might take a very long time (see dashed lines in Figure 1). The determination of delay times (DDT and RDT) as a function of the deposition reductions is an important task of dynamic modelling since these strongly influence target loads and are helpful to assess the likely chemical response of freshwaters to an agreed emission reduction within a given timescale. These delay times are new quantitative information which is complementary to the critical loads concept. Modelling of DDT and RDT allows assessment of deposition scenarios which are, from a critical loads point of view, equal but which may have very different biological consequences.



Figure 2. Surface water critical load exceedances at EMEP grid scale, 1980 (maximum deposition) to 2010 (assuming implementation of the Gothenburg Protocol). Dynamic modelling is appropriate in almost all squares for which data was reported to the CCE.

Dynamic models have a key role to play in the review of the latest (Gothenburg) Protocol for emission reductions 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. Models can be used to predict recovery from acidification of soils and surface waters since it is not possible to decouple the soil and water system with respect to chemistry, but this report is focused only on surface

waters as model outputs. Dynamic models for assessing soil responses are fully documented elsewhere (Posch *et al.* 2002).

Dynamic model applications are to a certain extent limited by the availability of suitable data to describe the physico-chemical characteristics of surface waters and their terrestrial catchment areas, especially soil chemistry. Given this requirement, it is clear that the focus of dynamic model applications should be on areas that are considered to be acidified or acid 'sensitive'. This makes sense within the framework of the Convention since emissions across Europe are declining and will continue to decline into the foreseeable future under the Gothenburg Protocol and so the speed of recovery from acidification is the key question. An attempt to identify acid sensitive regions has been initiated by the Joint Expert Group on Dynamic Modelling (UNECE 2002) and this implies a wider extent of the problem (Table 1) than might be inferred from an analysis of critical load exceedances (Figure 2).

2. Dynamic Models of Surface Waters

Four models have been identified as being widely used, documented and tested with respect to the requirement of the Convention (Posch *et al.* 2002) and which are simple enough to be applied on a regional scale; SAFE (Warfvinge *et al.* 1993), SMART (De Vries *et al.* 1989), VSD (Posch and Reinds 2003) and MAGIC (Cosby *et al.* 1985a). An overview of the first three models, all of which are effectively plot scale soil models is given in Posch *et al.* (2002). The MAGIC model, however, focuses on surface water chemistry and is generally applied at catchment scale. The catchment implicitly modelled by MAGIC is defined by the location of the surface water sampling point and can, therefore, vary considerably in size. A key difference, therefore, is that weathering rates must be calibrated from surface water and soil chemistry data rather than estimated empirically or from mineralogy (e.g. using PROFILE; Warvfinge and Sverdrup 1992).

2.1 The MAGIC Model

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. 1985a,b,c, 1986). 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, dissolutionprecipitation-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). Overall, the model has proven to be robust, reliable and useful in a variety of scientific and environmental management activities (Ferrier *et al.* 1995, Jenkins *et al.* 1998, Cosby *et al.* 1995, Wright *et al.* 1998).

 Table 1. Acid sensitive regions receiving significant inputs of acid deposition identified by the UNECE
 Joint Expert Group on Dynamic Modelling and the current status of dynamic modelling (UNECE 2002)

Region	Country	ICP Waters	Soil data	Modelling	Comments
		Sites			(project/contact)
Fenno-Scandian	Norway	YES	YES	YES	RECOVER:2010 + national
shield					project + EMERGE
	Sweden	YES	YES	YES	RECOVER:2010 + national
					project
	Finland	YES	YES	YES	NMR - project
	Russian Federation - Kola	YES	NO	?	
	Russian Federation - Karelia	NO	?	NO	
Upland areas in the	Scotland	YES	YES	YES	RECOVER:2010 + national
British Isles					project + EMERGE
	Wales	YES	YES	YES	RECOVER:2010 + national
					project
	Northern England	YES	YES	YES	RECOVER:2010 + national
					project
	Ireland	YES	NO	NO	ž
Lowland	SE England	NO	YES	NO	
Heaths/Forests	C C				
	Denmark	NO	YES	NO	
	Germany – northern	YES	YES	YES	RECOVER:2010 + national
					project
	Netherlands	YES	?	?	· · ·
	Belgium	NO		NO	
Mid-European	France – Vosges	NO	?	NO	
Forests	C C				
	Belgium – Ardennes	NO	?	NO	
	Germany – Black Forest, Harz	YES	YES	YES	RECOVER:2010 + national
	Mountains				project
	Germany – Bavarian Forest,	YES	YES	YES	RECOVER:2010 + national
	Mittelgebirge				project
	Czech Republic	YES	YES	YES	RECOVER:2010 + national
	*				project
Pyrenees	Spain	YES	YES	YES	EMERGE
•	France	YES	?	?	
Alps	Italy	YES	YES	YES	RECOVER:2010 +
1					EMERGE
	Switzerland	YES	NO	NO	
	Austria	YES	NO	NO	
Tatras	Slovakia	NO	YES	YES	EMERGE
	Poland	YES	?	YES	-
S. Europe	Bulgaria	NO	?	NO	
5. Lutope	Duigana	no	-	no	1

2.2 List of required data

The data requirement to run MAGIC must be spatially and temporally averaged (or 'lumped') to represent the whole catchment area (divided into one or several soils and waters compartments) and the time step of the model (annual or monthly). If physical and chemical data are available at only one point in the catchment or from one point in time, it must be assumed that this is representative of the whole catchment at that time step. Any uncertainty in these data and in the representativeness is incorporated into the model and the prediction must be interpreted accordingly. The minimum data requirement is given below:

Hydrological parameters:

- Annual runoff (m/yr)
- Annual precipitation volume (m/yr)

Soil parameters:

- Soil depth (m)
- Porosity (%)
- Bulk density (kg/m^3)
- CEC (meq/kg)
- SO₄ maximum adsorption capacity (meq/kg)
- SO₄ half-saturation coefficient (meq/m³)
- Dissociation constant for aluminium hydroxide solid phase, K_{Al} (log₁₀)
- Temperature (annual average) (°C)
- pCO₂ (%)
- Organic acids (mmol C/m³)
- Dissociation constants for organic acids (pK)
- Nitrification (% of input)

Surface water parameters:

- Retention time (Yr)
- Relative area (%)
- Temperature (annual average) (°C)
- Dissociation constant for aluminium hydroxide solid phase, K_{Al} (log₁₀)
- pCO₂ (%)
- Organic acids (mmol C/m³)
- Dissociation constants for organic acids (pK)
- Nitrification (% of input)

Surface water/soil chemistry in calibration year (i.e. 2001):

- Concentration of major ions (Ca, Mg, Na, K, NH₄, SO₄, Cl, NO₃) (meq/m³)
- Soil exchangeable base cations (Ca, Mg, Na, K) (meq/kg)

Deposition parameters in calibration year (i.e. 2001):

- Concentration of major ions (Ca, Mg, Na, K, NH₄, SO₄, Cl, NO₃)
- Time sequence of change in:
 - deposition concentration
 - dry deposition factors for each ion (= total deposition/wet deposition)

The historical sequences of changing deposition of sulphur and nitrogen are usually derived from estimates made by EMEP (Mylona 1993). These historical 'trajectories' at the scale of the EMEP grid are usually modified at a site or regional scale to incorporate more detailed measurements or estimates, particularly in more recent years. Further updated deposition histories for the EMEP grid are currently being derived by the CCE/IIASA (Posch, *et al.* 2003).

More detailed data is desirable to describe:

- dry deposition flux
- uptake to plant biomass in soil and water
- in-lake processes (stratification, sedimentation etc)
- forest growth history

These data, if available, can improve the model performance against observations and serve to increase confidence in model predictions. In addition to the data required to parameterise the model, information is also required to enable model calibration.

2.3 Model Calibration

The calibration of MAGIC is a sequential process whereby firstly the input and output of those ions assumed to act conservatively in the catchment are balanced (usually only Cl); next, the anion concentrations in surface waters are matched by adjusting catchment net retention (of N) and soil adsorption (of S) if appropriate. Thirdly, the four individual major base cation concentration in the stream and on the soil solid phase (expressed as a percentage of cation exchange capacity) are matched by adjusting the cation exchange selectivity coefficients and the base cation weathering rates. Finally, surface water pH, Al and organic anion concentrations are matched by adjusting the aluminium solubility coefficient and total organic acid concentration in surface water.

The first step is achieved through comparison of the present day wet (or bulk) deposition concentration of Cl, the rainfall amount (which together provide the input flux), the surface water concentration of Cl and the runoff (which together provide the output flux). In most cases the input based on wet deposition only is less than the output and it is assumed that the extra Cl is deposited as a dry deposition of sea-salt that is not represented by the wet deposition concentration. This extra Cl is added as a neutral salt by also adding base cations in their sea-salt ratio to the deposition flux. Similarly, there is a need to estimate the dry deposition of S if the measurements are not available. In catchments where the net output flux of SO_4 is greater than the input flux at present day, a similar calculation is performed assuming this to represent dry deposition of SO_2 and also assuming no S adsorption within the catchment.

Observed inputs of N are usually much higher than observed N concentrations in surface waters. Indeed, NH_4 concentrations are usually zero. For calibration, nitrification of NH_4 is set such that simulated concentrations of NH_4 match observed. Then net catchment retention required to match the observed NO_3 concentration in the surface water is calculated. This percentage retention is usually assumed to be constant throughout the model simulation. For SO_4 , if surface water data is available for only one point in time the adsorption parameters (Maximum adsorption capacity = E_{mx} and Half-saturation constant = C) if required, must be estimated from soils data. For regions with geologically 'young' soils, SO_4 adsorption is generally small. For the applications with long time-series the C and E_{mx} can be calibrated to match the observed trend in surface water SO_4 given an observed trend in SO_4 deposition.

The base cation calibration follows an iterative process whereby the base cation selectivity coefficients are set, values are chosen for base cation weathering, the model is run from some background, preacidification condition, the simulated values of base cations in soil and surface waters are compared with observed. This process is repeated, adjusting the selectivities and weathering rates until the observed target concentrations are achieved. If time-series data are available further adjustment may be undertaken to match trends. This part of the calibration procedure can be undertaken automatically using an appropriate numerical optimisation procedure.

This calibration procedure is performed to determine the catchment weathering rate of base cation and the initial fraction of base cations held on the soil solid phase (cf ion exchange selectivities). These parameters can be estimated or measured but given that they vary greatly in space across the catchment and during the year in response to other factors, notably soil moisture in the case of weathering, their calibration provides pragmatic values. In any case, comparison of calibrated weathering rates from MAGIC with those estimated using other weathering models or catchment mass-balance studies show an acceptable consistency (Warfvinge *et al.* 1992).

2.4 Model Output

The basic model output (Figure 3) provides a reconstruction of the key chemical variables from a historical or background condition which represents a pre-acidification condition to the present day under assumed (up to c.1970) and measured (c.1970 to present) emission/deposition levels. This pre-acidification condition also potentially provides the ultimate target for the future chemistry of the water, i.e. it is not possible to achieve better conditions than were present prior to the onset of acidic deposition by merely reducing the acid deposition in the future. The timing and magnitude of the historical acidification response at any given site, indicated by the ANC (Figure 3), is determined mainly by the flux of S and N deposition over time, as reflected by the change in surface water SO_4 concentration (Figure 3) and the capability of the catchment soils to buffer the strong acids with base cations from weathering and the soil exchange complex, reflected by the Ca concentrations (Figure 3).



Figure 3. The MAGIC model applied to Lochnagar, NE Scotland, driven using the historical (1848-1986) S deposition from EMEP (Mylona 1993), observed S deposition at the site (1986-2000) and modelling deposition in response to the Gothenburg Protocol (2000-2020). Also shown are the observed annual mean surface water concentrations (1988-2000) from the UK Acid Waters Monitoring Network.

Beyond the present day, the model is driven forward under some assumptions regarding the deposition of strong acids from the atmosphere. Assuming the emission reductions agreed under the Gothenburg Protocol are achieved by 2010, the model predicts a rapid recovery of water chemistry (Figure 3) in response but a much slower response thereafter as emissions are assumed to remain constant at that level beyond 2010. In most acid sensitive regions of Europe, water chemistry data is available to describe the current status of many surface waters and so the model can be calibrated to multiple sites (typically in order of tens to hundreds of sites) and thereby enable this question to be asked at a regional scale.

2.5 Model Testing

Clearly, the future predictions from a model are subject to uncertainties in the data used in the processes represented within the model and the mathematical representation of those processes. The degree to which the model fits against observations, therefore, indicates the confidence with which we can interpret those predictions. Dynamic models of surface water chemistry have key advantages over soil models in that they can be tested against three independent data sources: historical pH trends inferred from diatom reconstructions; long time-series data of direct measurements; and, data from ecosystem manipulation experiments.

2.5.1 Comparison with Observed Data

Deposition and water chemistry time-series data are available for many sites covering 10-30 years (e.g. Stoddard *et al.* 1999, Moldan *et al.* 2001, Evans and Monteith 2001, Evans *et al.* 2001a). These data provide the opportunity to compare the model simulation for the last decade or so against observations to ensure that the model is capable of capturing the dynamics of the system (Figure 4). At nearly all sites across Europe the past two decades have seen a marked reduction in S deposition and so the observations of water chemistry reflect recovery from acidification and the model can closely match observed trends (Figure 4). The MAGIC model has been tested at many sites spanning a range of environments, deposition levels and deposition reductions and matched the observations well in all cases (e.g. Jenkins and Cullen 2001, Hruška *et al.* 2002, Jenkins *et al.* 2003)



Figure 4. Comparison of MAGIC simulated SO_4 (left) and ANC (right) and observed annual mean chemistry at Stavsvaten, S Norway. The model simulation beyond 2000 is driven assuming implementation of the Gothenburg Protocol.



Figure 5. Measured and predicted pH and concentrations of sulphate, aluminium, sum of base cation, chloride and ANC at the Gardsjon experimental roof catchment (after Beier et al. 2003). The observations (squares) and MAGIC predict (solid line) represent annual means.

2.5.2 Comparison with Experimental Data

Since the mid-1980s, a number of whole ecosystem manipulation experiments have been undertaken to assess the acidification and recovery process. Whilst these experiments necessarily accelerate the rate of change in the ecosystem by significantly changing the input of S and N relative to the slower changes in response to emissions increase and reduction, they offer a unique test for the dynamic models. The processes operating to produce the surface water chemistry in the manipulated catchments are the same as those operating under ambient conditions and so the dynamic models should be capable of reproducing the observations. At the roof covered catchment experiment in Sweden (Gårdsjön), the MAGIC model successfully reproduced (Figure 5) the effects of 'clean rain' treatment (Beier *et al.* 2003). Similar successful applications of the model to experimental catchment manipulations have been undertaken at Risdalsheia (Norway) and Klosterhede (Denmark) (Beier *et al.* 1995). The MAGIC model also captured the acidification response induced by addition of S and N at catchments in Norway (Sogndal) (Cosby *et al.* 1995). Further detailed comparisons between the MAGIC model and experimental data have been undertaken by Moldan *et al.* (1998) and Wright *et al.* (1998).

2.5.3 Comparison with Diatom Reconstructions

A key method by which the long-term acidification of surface waters has been demonstrated is the reconstruction of surface water pH based on diatom assemblages from lake sediment cores (Battarbee *et al.* 1990). There have been several attempts to compare these pH reconstructions with the historical simulated pH change from dynamic models (Jenkins *et al* 1990, Wright *et al.* 1986). The similarity in the predicted and reconstructed timing of pH change and the background pH inferred from both techniques has generally been found to be good. Recently both the diatom reconstructions and the dynamic models have been changed to incorporate new processes and data; there remains a reasonable match between the two techniques (Figure 6).



Figure 6. A comparison of the diatom pH reconstruction and MAGIC model pH simulation at the Round Loch of Glenhead, SW Scotland, UK.

3. The Contribution of Dynamic Modelling of Surface Waters to the Work under the Convention

Dynamic models can contribute to the Convention in two important areas; firstly, they can provide an estimate of the expected surface water chemistry at any time in the future in response to the implementation of the Gothenburg Protocol (assessment of the impact of emission reductions) and secondly, they can be used to assist in the calculation (optimisation) of further emission reductions (input to the process of Integrated Assessment Modelling).

Regional assessment of surface water response to the implementation of the Gothenburg Protocol has been conducted under the EU RECOVER:2010 project (Ferrier *et al.* 2001) as well as numerous national studies (e.g. Evans *et al.* 2001b) for Southern Norway, Southern Sweden, Galloway, Wales and the Pennines in the UK, Central Germany, Northern Italy and the Czech Republic. Regional modelling produces a time series output (e.g. Figure 3) for each of the sites under consideration and these can be summarised to provide regional responses through time (Figure 7). Presented in this form, however, only regional questions can be addressed and the model applications lose their spatial resolution. Clearly, it is possible to summarise the regional output and still maintain the spatial distribution of the data in map form. In the simplest form the regional assessment can take the form of maps (Figure 8). These show the chemistry of the region at any given time and provide an indication of the areas within the wider region which require more detailed assessment possibly with a view to further emission reductions or management intervention such as liming. This is potentially very useful with respect to sites of special scientific interest or special conservation areas such as those identified under the EU Habitats Directive.

A more quantitative summary of the regional response to deposition reductions can be provided using the same "time slice" data from the model predictions but presented as frequency distributions (Figure 9) or cumulative frequency distributions (Figure 10). These provide a clear quantification of how the mean ANC has changed across the region in response to the change in SO_4 and also, perhaps more importantly, provide an estimate of the percentage of surface waters in the region which do not reach the specified target ANC within a given timescale. This "tail" of the distribution is potentially of most significance since it represents the most acidified sites (Figure 11).



Figure 7. Time series output from MAGIC for many sites in a region (left) which can be summarised to provide regional statistics through time (right).



Figure 8. The MAGIC model applied regionally in S Norway. The left panels show the modeled excess sulfate concentrations in lakes; the right panels the modeled ANC in lakes.

Figure 9. Frequency distributions from MAGIC model simulations for S Norway. Deposition to 2010 assumes implementation of the Gothenburg Protocol and held constant thereafter to 2050.

Most crucially then, dynamic models are currently capable of providing an assessment of the implications of achieving the emission reductions agreed under the Gothenburg Protocol. They are capable of addressing the question of whether the current agreements promote an appropriate response in surface water chemistry within a given timeframe and in helping to identify regions where further reductions might be required to achieve the specified target chemistry at some specified time. If it is determined that further emission reductions are required and desirable, the models can be put to a further use, that is, to determine the level of deposition that must be met within a certain time to reach the specified target chemistry within a specified time. This capability provides a clear advantage over the concept of critical loads in that these provide the deposition required to reach the specified target chemistry at some un-specified time in the future when the aquatic system has reached an equilibrium with the deposition chemistry. This is likely to take several decades or even hundreds of years. The link between the dynamic model approach and the critical loads is clear, however, in that if the model is run with the calculated critical load it will be possible to specify the time required to reach the chemistry target. Conversely, if the target year in which the specified chemistry is required is set to several hundred years from present, the calculated deposition required in the dynamic model to meet that target should approximate closely to the critical load. In the context of management and policy making, however, such long time scales (typically required for recovery of soils) are difficult to accommodate in current legislative plans and so the use of dynamic models for surface waters is crucial in providing the reductions required over a timescale of a few decades.

Outputs from dynamic models can be summarised and presented as a logical extension to the critical loads approach already used in the Integrated Assessment process. For example, in the same way in which the critical loads of S and N are represented as a function for input to the Integrated Assessment Models (Figure 12), the deposition required to achieve a given chemistry within a specified time can be calculated from the dynamic models and expressed as a target load function (TLF) (Figure 12). At every combination of S and N deposition on the target load function, a target ANC will be reached in the specified target year. The only difference between the TLF and the critical load function (CLF) being the concept of time to reach the target chemistry since for the CLF, the time to reach the chemical target is infinite. Note that the shape of both the CLF and TLF are similar, the 'shelf' at low N deposition representing the long-term capability of the system to utilise N, but the TLF will always be lower than the CLF. For a target chemistry to be reached in the very long term (infinite timescale) the TLF and CLF are the same.

Figure 11. Ranked change plots over time from the regional MAGIC application to S Norway.

Figure 12. The critical load function (upper) as constructed and used by the Integrated Assessment Models for calculation of optimal deposition reduction scenarios. Emission reductions must be achieved at some future time to achieve the target chemistry at some (unknown) point in the future. The target load function is essentially the same <u>but</u> the timing of emission reductions must be specified to achieved the specified chemistry in a given year.

Figure 13. The effect of different target years on the target load function for a given target chemistry at an individual site.

Clearly and implicitly, at a currently acidified site the deposition reduction required to reach a specified target within 15 years will be greater than that required to reach the same target over a longer timescale. This is because the deposition flux over the whole period (i.e. integral under the deposition curve over time) is largely responsible for the chemistry predicted for a given year (Figure 13). Assuming that costs increase with greater reduction of N and S, this implies, therefore, that there is an increased cost associated with selecting a more immediate ecosystem recovery. Additionally, it is also clear that the deposition reduction required to reach a less stringent water chemistry target (for example ANC=0 ueq/l) over the same timescale will be less than for the more stringent target (for example, ANC=30 ueq/l) (Figure 14). Again, the economic implication is that the stricter the target chemistry, the larger emission reductions required and the higher the cost. It should be noted that the slopes of the TLFs under different target years and target chemistry will vary from site to site as a function of the buffering capacity of the soils (weathering rate, soil exchangeable base cations) the deposition (historical and current) and the predicted level of future deposition.

The specification of the TLF from a dynamic model requires one further set of assumptions regarding the timing of further emission reductions. For example, the start and end years of the required reductions must be specified since any delay in emission reduction and the time period over which they are achieved will affect the position of the TLF (Figure 15).

In terms of achieving an optimal solution to emission reduction, therefore, the policy maker needs to make key decisions regarding: (i) the target chemistry required to protect the chosen biological receptor (target chemistry) (note also that this will also influence the critical load); (ii) the year in which the target chemistry is required (target year); (iii) the year in which emission reductions will start to be implemented (implementation year); and (iv) the year in which the emission reduction must be completed (completion year). Clearly, the regional TLF is constructed for use by the IAM will incorporate the result of these four assumptions. In effect, therefore, there is a possibility to 'optimise' these four assumptions/decisions prior to application of the IAM.

Figure 14. The effect of different target chemistry on the target load function for a given target year at an individual site.

Figure 15. The effect of different years of implementation of the emission reductions to meet the specified target chemistry in 2025 at a site. The two cases shown here represent the reductions beginning in 2010 and being completed by 2015 and 2010.

Figure 16. Calculation of a regional target load function. This would also represent a method for calculating the TLF within grid squares for a given target year, target chemistry (ANC) and emission reduction implementation and completion years.

These target load functions can be accumulated for all of the sites within a given region and analysed to provide a regional TLF which represents the combination of S and N deposition required to achieve the target chemistry at all sites within that region or some percentage of them (Figure 16). This 'minimum' regional TLF may be further constrained by the current deposition of N and S on the basis that it is unlikely that increased emission of either pollutant will be acceptable. This methodology can also be developed to incorporate the "gap closure" concept used in the IAMs for the development of the Gothenburg Protocol.

4. Future Developments

The MAGIC model is currently capable of wide application across acid sensitive areas of Europe in support of the objectives of the Convention. There still exists, however, a requirement for further developments with respect to the model, its interpretation and its application. These developments are required in the near future if the full potential of dynamic models are to be realised within the timescale of the Convention, i.e. the forthcoming review of the Gothenburg Protocol as scheduled by the Working Group on Effects medium term strategy.

4.1 Model Uncertainties

There are uncertainties related in the application of all mathematical models. Different types of uncertainties exist depending on the model, its application and the information available for running the model. One way of grouping uncertainties is to consider them according to whether they can be quantified. This provides three groups: technical uncertainties; methodological uncertainties; and, epistemological uncertainties (Funtowicz and Ravetz, 1990). The latter group are, by definition, impossible to quantify and difficult to include in model applications. Such uncertainties relate to the fact that unforeseen events may occur to cause model predictions that do not match reality. Such events are not included in the models (if they were, these are no longer epistemological uncertainties) and are only imaginable to a limited extent. Accordingly, epistemological uncertainties are not discussed further.

Technical uncertainties in dynamic modelling of surface waters are related to estimation of model inputs and parameters from observed data and relate mainly to measurement errors and variability in both time and space. These can in principle be quantified if enough measurements are available and hence it may be possible to quantify the uncertainty in model predictions (Larssen *et al.* 2003). The representativity of available data used for modelling is important for the overall uncertainty in model predictions. The natural variation in, for example, deposition or runoff composition throughout a year may be substantial and the uncertainty in model prediction will typically increase with decreased sampling frequency. Similarly, the spatial variability, especially in soil data, results in increasing uncertainty in predictions as the spatial resolution of the sampling decreases. Hence, for regional model applications, the uncertainty in predictions will increase due to decreased resolution. The technical uncertainties in model outputs can be summarised and presented as probability distributions (Figure 17).

The methodological uncertainties are more difficult to assess and can only partly be quantified. This group of uncertainties relates to the fact that models are necessarily simplifications of nature and that important processes may be excluded or inappropriately described in the model. Examples of factors giving methodological uncertainties in dynamic modelling of freshwaters are nitrogen dynamics, the role of organic matter and climate impacts including sea salt events (Ferrier *et al.* 2001). The lack of scientific understanding is an important factor for this group of uncertainties, in particular the current lack of understanding of the controls on N leakage from the terrestrial to the aquatic system has been intensively studied, but remain is a major source of uncertainty in model predictions.

Episodic impacts from sea salt episodes are important in delaying recovery from acidification in areas located relatively close to the sea. Severe storms have been shown to kill aquatic biota due to sea salt induced acidification peaks (Hindar *et al.* 1994). Models can reproduce such events, but the likely frequency and magnitude of such events in the future is not known. A practical approach to address this is to repeat the observed frequency of sea salt deposition into the future and in this way illustrate the importance and magnitude of such events relative to the predicted general trend. In this way the importance of naturally extreme years in combination with reduced anthropogenic deposition can be shown (Figure 18).

Figure 17. The technical uncertainties in model predictions can be calculated and presented as probability distributions over time. This example show the probability distribution for ANC into the future at Birkenes (S. Norway), given implementation of the Gothenburg protocol and taking technical uncertainties into account in the model inputs and parameter estimates (Larssen et al. 2003).

Figure 18. Illustration of the response in ANC at Birkenes (S. Norway) when taking the variation in sea salt deposition into account in combination with decreased sulphur and nitrogen deposition under the Gothenburg protocol. The observed sea salt deposition frequency for the period 1973-2000 is repeated into the future (Larssen et al. 2002).

As the deposition of S decreases in Europe, the relative importance of N increases since the reduction of N emission is comparatively very small. Most semi-natural terrestrial systems, however, are Nlimited and the vegetation and soils retain most of the N deposited with only a very small flux to surface waters in most areas. Nevertheless, where NO_3 is observed in surface water, this acts as a strong acid anion to promote increased acidity and decreased ANC. The question that remains to be answered is what controls the degree of N leakage in the long term? A 'worst case' assumption is that when N deposition exceeds the uptake requirement of the vegetation and microbial biomass in the soil, or when root damage caused by acidification reduces the N uptake capacity, N leakage to surface waters increases (Aber et al. 1989; Stoddard 1994). This saturation and breakthrough response is further strengthened by observations of C/N ratios and soilwater NO₃ at forested sites across Europe within the NITREX project (Gundersen et al. 1998a). In this respect, an empirical relationship between soil litter layer C/N ratio and NO₃ (Figure 19) leakage from the terrestrial system has been established (Gundersen et al. 1998b). There currently exists, however, little empirical evidence of changes in surface water NO₃ concentrations over time to support the concept of N saturation (Wright et al. 2001). At Lange Bramke, central Germany, for example (Figure 20) streamwater NO₃ concentrations rose steadily through the 1980s but have since declined again indicating that climatic influences may have been responsible. The potential for climate induced changes in N dynamics has been demonstrated from catchment manipulation studies (e.g. Wright and Jenkins, 2001). This does not mean, however, that N saturation does not occur in the long-term but perhaps indicates that the process takes a very long time (several decades).

In terms of critical loads, the steady state FAB model (Henriksen and Posch 2001) embraces the concept of N-saturation and assumes that at some point in the future, the terrestrial system will be incapable of immobilising incoming N other than some small fraction that is required for net plant uptake, lost to denitrification or immobilised as soil builds up in the long-term. In this case, the derived critical load incorporates a maximum level of N leakage and hence NO₃ concentration. This N saturation concept has also been incorporated into MAGIC by assuming the relationship between C/N in soils and NO_3 concentration in water (Figure 19) to be a time dependent process (Cosby *et al.* 2001). Given the lack of observed time-series data describing increased NO_3 concentrations in surface waters, however, it is also possible to model N dynamics by calculating the current proportion of N immobilised in the terrestrial system and assuming this to remain constant into the future, irrespective of N deposition changes. Clearly, the result of this assumption is that NO₃ concentration decreases proportionally with decreased N deposition and this may be considered a 'best case'. The net effect of these 'best' and 'worst' cases on the model predictions in the longer term (Figure 21) and under the Gothenburg Protocol reductions in N and S are not appreciable with respect to ANC over a time-scale of c.30 years (Jenkins and Cullen 2001, Jenkins et al. 2001). Beyond this, however, NO₃ leakage could have a significant impact on the acidification status as the proportion of N retained declines even under constant N deposition.

A more detailed analysis of the uncertainty related to N dynamics and the impact of episodic sea-salt inputs in relation to uncertainty in data for calibration and parameterisation has been undertaken at Birkenes, Norway (Larssen *et al.* 2002) and is summarised in Jenkins *et al.* (2003).

total N retention ECOFEE database

Figure 19. The observed relationship between C/N of the forest floor and NO₃ input/output flux for sites across Europe (Gunderson et al. 1998a).

Figure 20. Long-term changes in stream NO₃ concentration at Lange Bramke, Germany (Wright et al. 2001).

Figure 21. MAGIC predicted NO_3 (top) and ANC (bottom) at Lolchnagar, Scotland, assuming a best case N leakage (dotted line) and a worst case (solid line) (see Jenkins et al. 2001).

4.2 Inclusion of Biological Responses

With respect to biological status, the link between surface water ANC and brown trout population has been established in Norway (Lien *et al.* 1992; Figure 22). This relationship is derived from chemistry and fish population status data of the Norwegian 1000-lake survey (Henriksen *et al.* 1989). Lakes with ANC < 20 µeq Γ^1 generally had damaged fish populations and most lakes with ANC < 0 were barren of fish. On this basis, a simple probability algorithm has been incorporated into MAGIC to link a given ANC with three categories of brown trout status; healthy, sparse and extinct (Wright *et al.* 1994). A comparison of the MAGIC output and historical information on fish catch from Loch Riecawr in Galloway, Scotland (Harriman *et al.* 2001) demonstrates the utility of this approach (Figure 23), whereby the predicted decline in the probability of a healthy fish population correlates closely with the historical fish catch record (Helliwell and Juggins 2002). More work is clearly required to relate the probability of fish status to population numbers but the utility of the approach with respect to target loads for emission reduction is clear.

From the relationship detailed by Lien *et al.* (1996) it is also possible to define critical 'target' ANC concentrations to summarise the regional response of brown trout to emission reduction scenarios on a static basis. In simple terms, for example, it is possible to define ANC < 0 as being an unacceptable chemistry with brown trout essentially absent; ANC 0 to 20 as being intermediate with the brown trout population 'at risk'; and ANC > 20 where the chemistry is acceptable as a healthy population is expected. The predicted chemistry for a region can then be used to represent biological status at any point in time. For the S. Norwegian lakes (e.g. Figures 8, 9, 10 and 11), for example, this information can be readily summarised to represent the consequences of the Gothenburg Protocol implementation over the next 50 years (Figure 24).

Figure 22. The observed relationship between brown trout status and surface water ANC at 827 sites in Norway (from Lien et al. 1996).

Figure 23. Comparison of MAGIC predicted probability of healthy brown trout population (Helliwell and Juggins 2002) and observed fish catch (Harriman et al. 2001) at Loch Riecawr, Galloway, Scotland.

The widespread improvement in surface water quality during the past 15-20 years should give rise to biological recovery. Thus far, however, there are relatively few documented examples of biological recovery. The reason might be lags in biological response but also a lack of appropriate long-term monitoring data.

The time lag to biological recovery can be divided into two parts. The first is the lag for return of extirpated species. The second is for sensitive species to achieve densities and distributions typical of undamaged lakes and rivers. For some species of macroinvertebrates and fish the second is longer than the first. The lag times for return of fish depend on connections to source populations. If the lake or river is isolated from sources of colonisers, the return of fish will depend on stocking (at least if we want them to return within a timescale of a hundred years or so). Stocking means that the fish can return as soon as chemistry recovers, essentially a lag time of zero. Most of the other taxa (algae, macroinvertebrates and zooplankton) are more mobile and many species will return without human intervention.

Figure 24. The predicted surface water status in S Norway lakes prior to acidification, at peak deposition, present day and into the future assuming full implementation of the Gothenburg Protocol by 2010. ANC < 0 implies an extinct brown trout population; ANC 0-20 implies a population 'at risk'; and ANC > 20 implies a viable population.

- Low or scattered occurrence, present in 50% of suitable limed habitation recorded in 100 % of suitable limed habitati

Figure 25. Summary of the development of sensitive taxa of invertebrates in the River Audna during the 15 years following liming (from Raddum and Fjellheim, 2003).

Once the chemical threshold is reached, lag times for common, widely distributed species might be:

- 1. Algae: 1-2 years
- 2. Macroinvertebrates: 1-3 years in streams (for first appearance of sensitive species; normal populations 5-10 years). 1-10 years in lakes.
- 3. Zooplankton: 1 year (species with resting stages in the sediments) >10 years (for whole communities).
- 4. Fish: 2-20 years.

Biological recovery following liming of acidified waters gives a good indication of the potential lag times involved in biological recovery (Figure 25). For example, during the 15 years following liming of the River Audna, southern Norway, the reappearance of sensitive species of invertebrates required 1-10 years, depending on such factors as the proximity of refuge populations and the life cycles and dispersal mechanisms of the various species (Raddum and Fjellheim, 2003).

4.3 Wider Application in Europe

The application of MAGIC to those areas of Europe where surface waters are considered to be acid sensitive (Table 1) depends primarily on the availability of soil and surface water chemistry data with which to parameterise and calibrate the model. The role of the ICPs (Forests, Mapping and Modelling, Waters and Integrated Monitoring) is crucial to providing this information. Furthermore, each ICP is often represented by a different Institution as the National Focal Point and it is not always the NFCs who are best placed for undertaking the modelling work. It is clear that each country needs to enforce integration of data and expertise if they are to undertake a site-specific or regional modelling exercise. There also exists a number of European scale databases which may provide data for new parameterisation. These include:

Soils: The Soil Geographical Data Base of Europe (SGDBE) is now available (in digital form) on a scale 1:1,000,000. Apart from the map, which uses the FAO soil classification, it also contains pedo-transfer rules, a soil profile analytical database and a database of hydraulic properties (European Soil Bureau 1999).

Land use/land cover: The most detailed European database is the CORINE land use database. However, CORINE does not (yet) cover the whole of Europe (mostly EU countries). A pan-European land cover map/database is the one prepared by the PELCOM project (Mücher *et al.* 2000). Another European land use/cover database, used also by EMEP/MSC-W, is held at the Stockholm Environment Institute (SEI) in York (www2.york.ac.uk/inst/sei/fsheets). A short comparison of these three European databases can be found in De Smet and Hettelingh (2001).

Deposition: Present (and more recent past) deposition fields of S and N covering Europe can be obtained from EMEP/MSC-W (www.emep.int). Historical depositions, mostly based on work by EMEP and IIASA, are available from Posch *et al.* (2003).

Obviously, none of these databases contains the information (input data) in the form needed to run dynamic models. But they often do contain information which can be used in conjunction with so-called (pedo-)transfer functions to get an estimate of the desired input data. Examples of such transfer function can be found in the Mapping Manual (UBA 1996) and in Posch *et al.* (2002).

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Appendix A. Reports and publications from the ICP-Waters Programme

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