Article (refereed) - postprint

Posch, Maximilian; Aherne, Julian; Moldan, Filip; Evans, Chris D.; Forsius, Martin; Larssen, Thorjørn; Helliwell, Rachel; Cosby, B. Jack. 2019. **Dynamic modeling and target loads of sulfur and nitrogen for surface waters in Finland, Norway, Sweden, and the United Kingdom**. *Environmental Science & Technology*, 53 (9). 5062-5070. https://doi.org/10.1021/acs.est.8b06356

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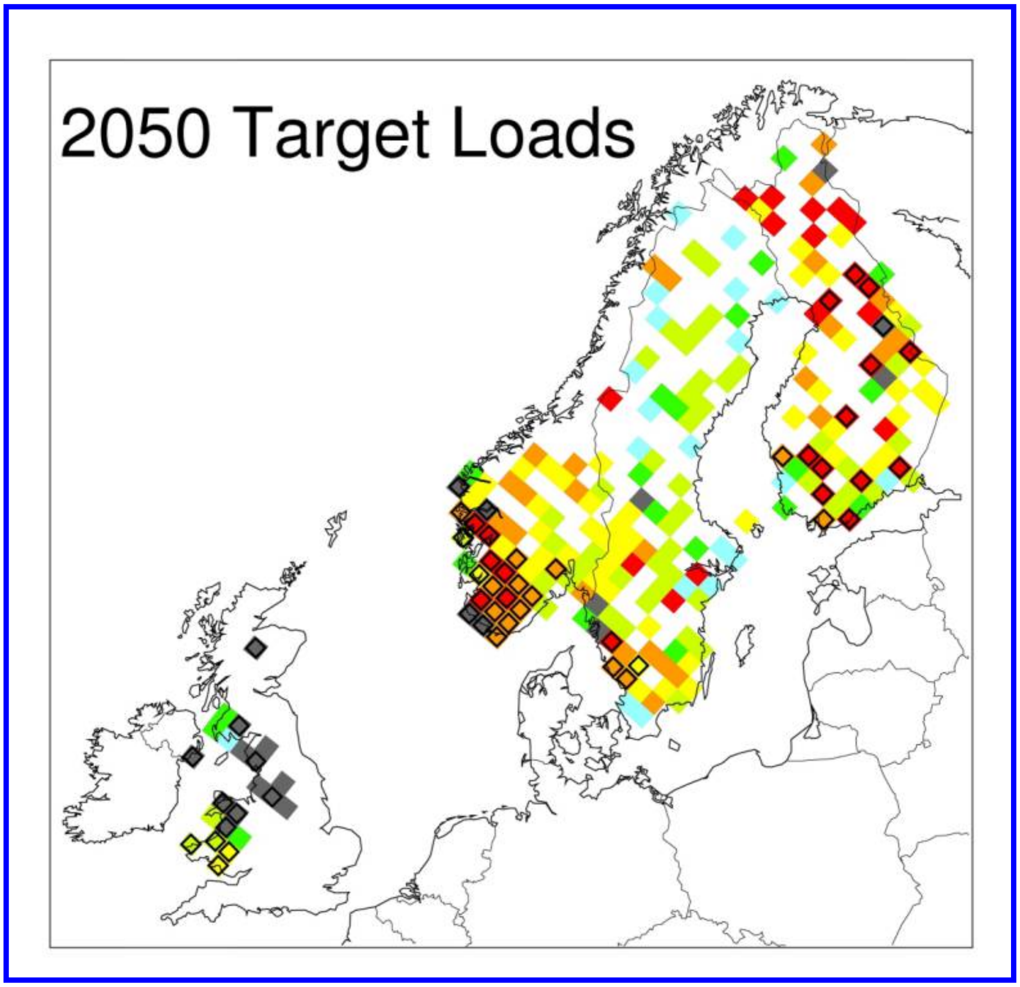

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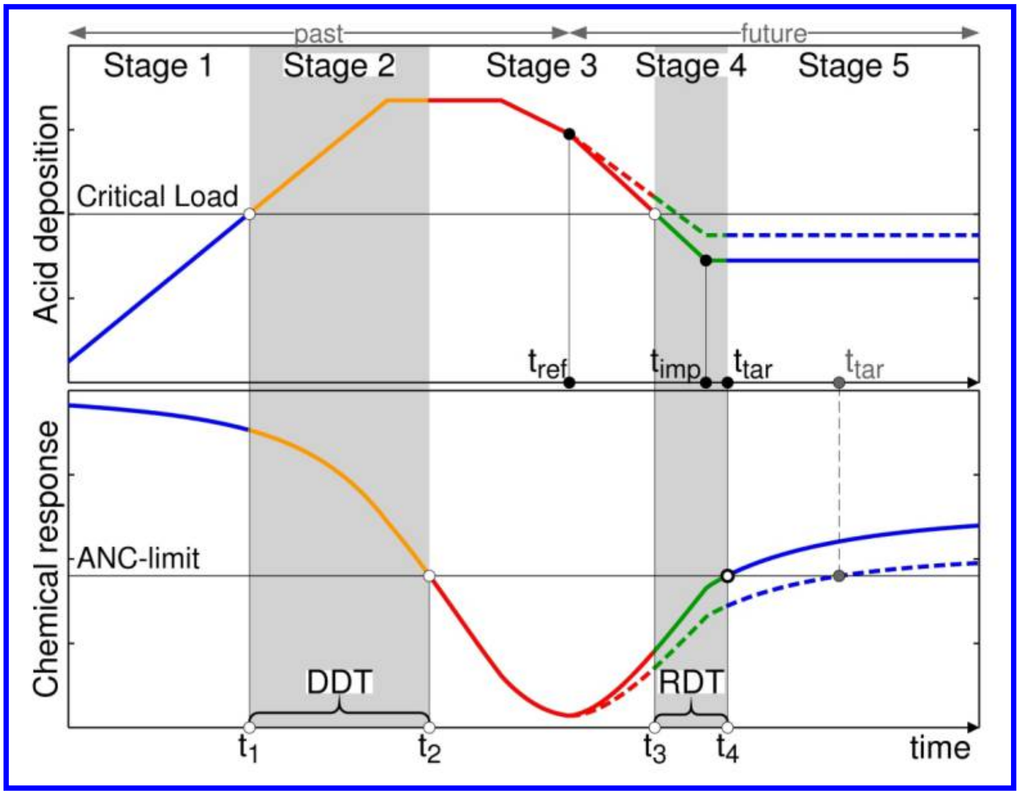

Figure 1: Generalised past and future development stages (indicated by different colours) of acidic deposition and lake chemical criterion response (here: ANC) in comparison to the critical chemical value (ANC-limit) and the critical load derived from it (i.e., the determination of critical load is based on a critical limit for a specified chemical criterion, which protects the biological indicator from deleterious effects). The delay between the (non-)exceedance of the critical load and the (non-)violation of the critical chemical value is indicated in grey shades, highlighting the Damage Delay Time (DDT) and the Recovery Delay Time (RDT) of the system. Also shown are the points in time (tref, timp) relevant for defining a target load (< critical load) to reach non-violation of the chemical value at a pre-specified time ttar. The dashed lines show the temporal development for a later target year (labelled in grey).

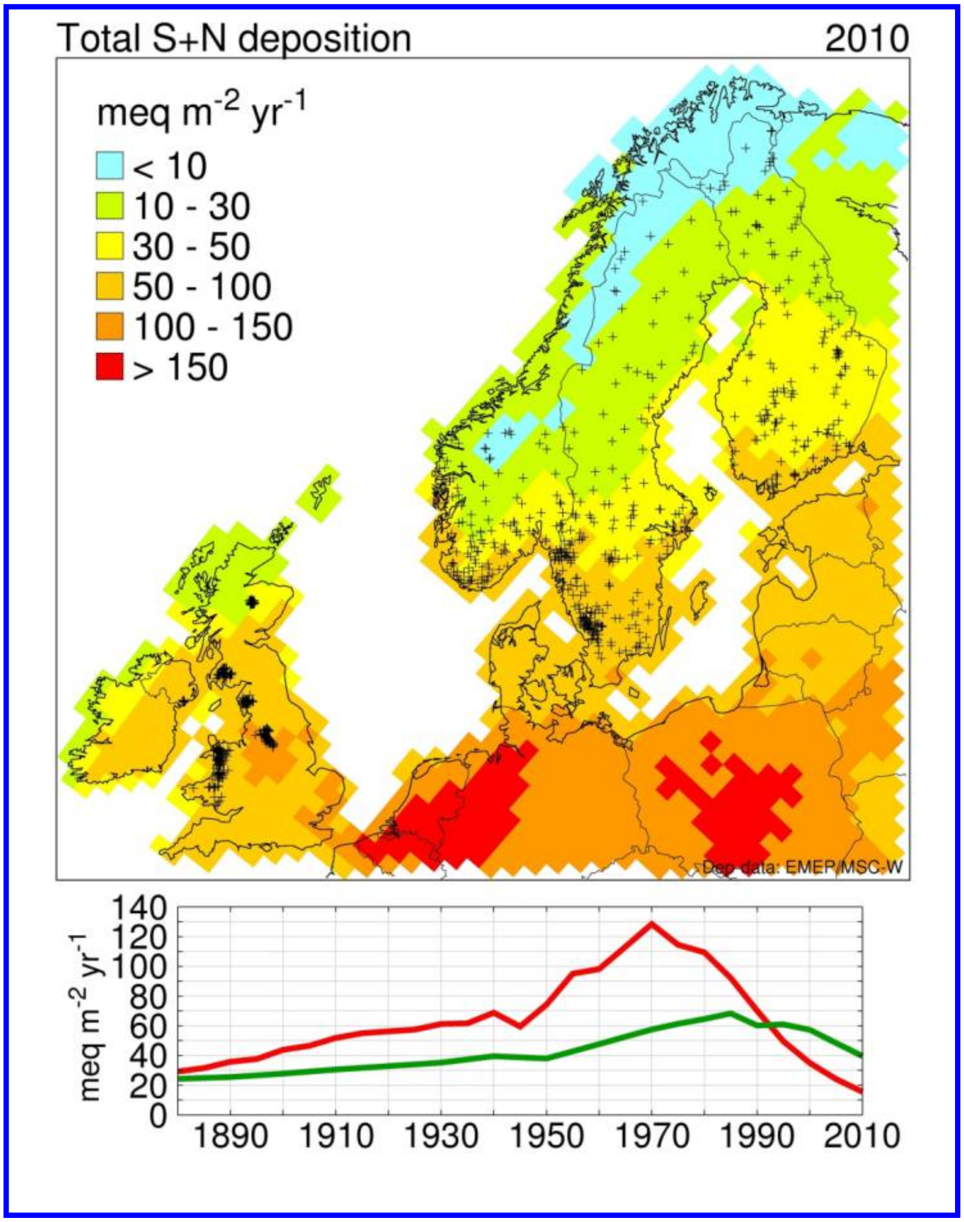

Figure 2: Location of the study sites used for dynamic modelling (n = 848; black crosses) overlaid upon modelled total sulphur (S) plus nitrogen (N) deposition in 2010 (meq m−2 yr−1) on the EMEP 50 km × 50 km grid (for further details on the EMEP model see Simpson et al.57). The temporal development (1880–2010) of the total S (red) and N (green) deposition averaged over the 848 study sites is also shown.

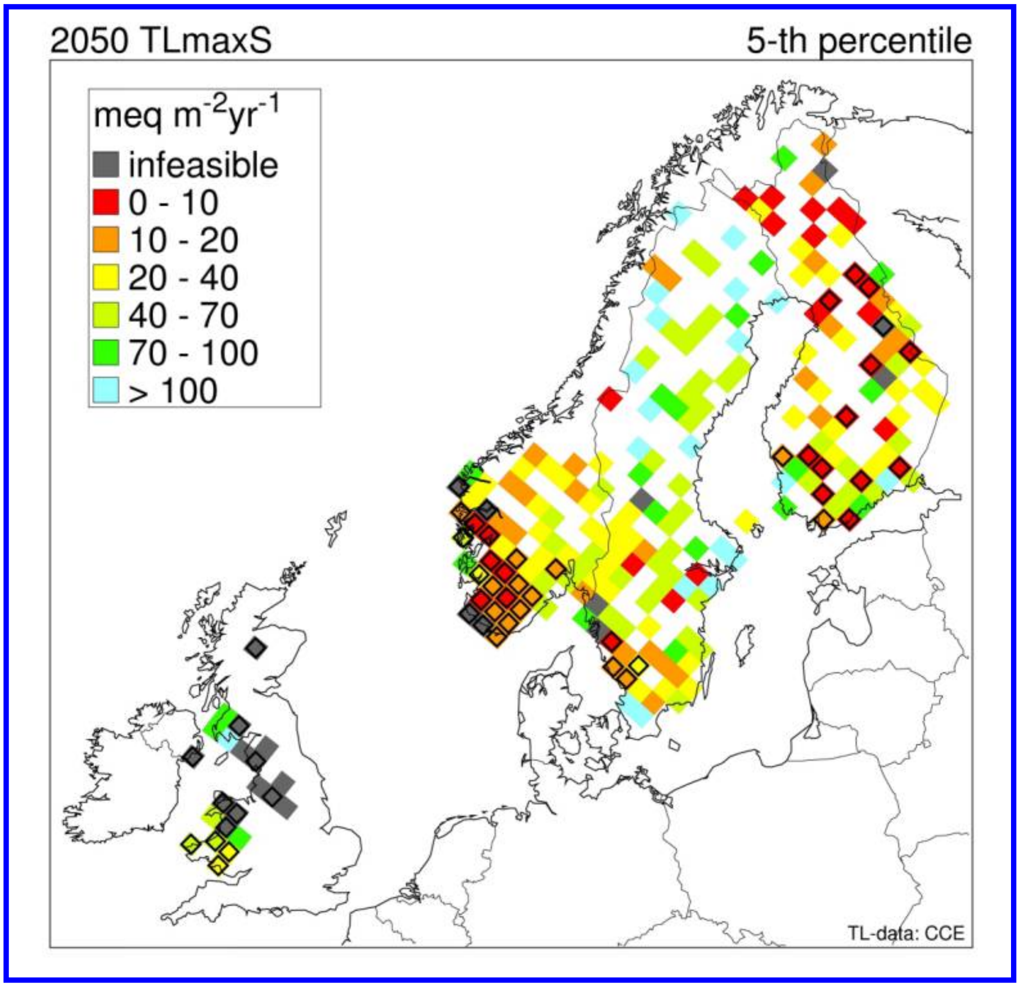

Figure 3: Map of the 5-th percentile of the 2050 maximum target load of sulphur (TLmaxS) on the EMEP 50 km × 50 km grid for 848 catchments in Finland, Norway, Sweden and the United Kingdom. Note: TLmaxS was set to the maximum critical load of S (CLmaxS) where TLmaxS > CLmaxS (i.e., the critical load is sufficient for non-violation of the ANC-limit by 2050). Grey-filled cells (label 'infeasible') denote grids containing at least one lake where the simulated ANC does not meet the specified limit by 2050, even under zero deposition after 2020 (FI = 3, NO = 4, SE = 4, UK = 35; Table 1). Black diamonds frame grids with at least one 'true' TL, i.e., where a TL exists and is lower than the CL (FI = 21, NO = 52, SE = 10, UK = 44; Table 1).

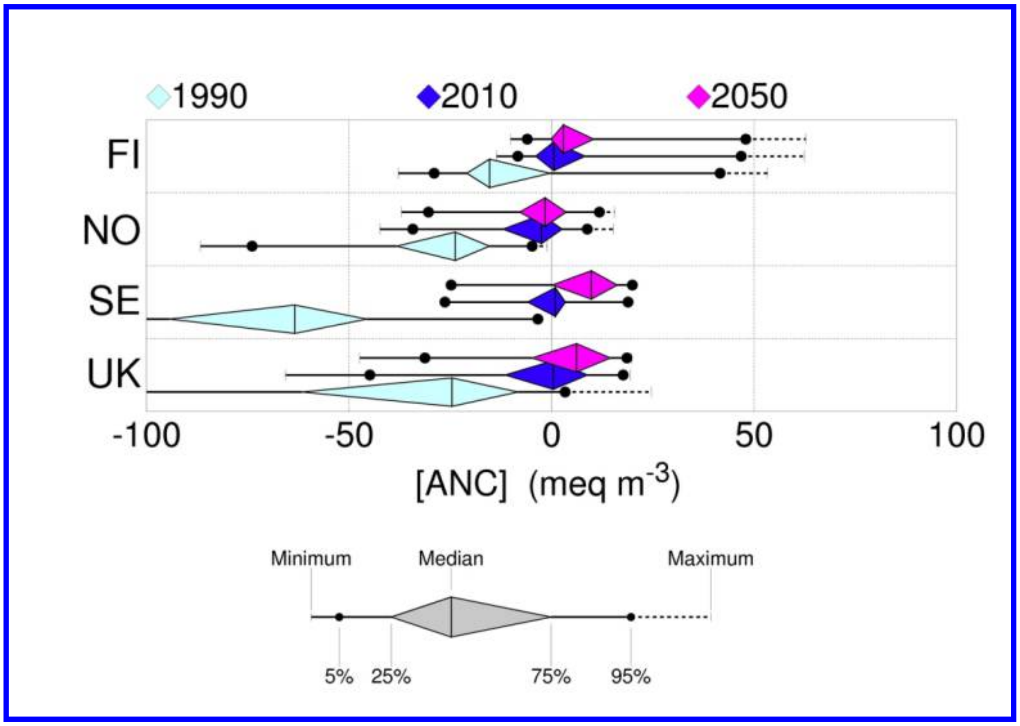

Figure 4: Percentile statistics ('diamond plots') of simulated annual average lake acid neutralising capacity (ANC) in 1990, 2010 and 2050 in Finland (FI), Norway (NO), Sweden (SE) and the United Kingdom (UK). Data are only shown for lakes for which target loads were determined (FI = 24, NO = 56, SE = 14, UK = 79; see columns 'INF' plus 'TL' in Table 1).

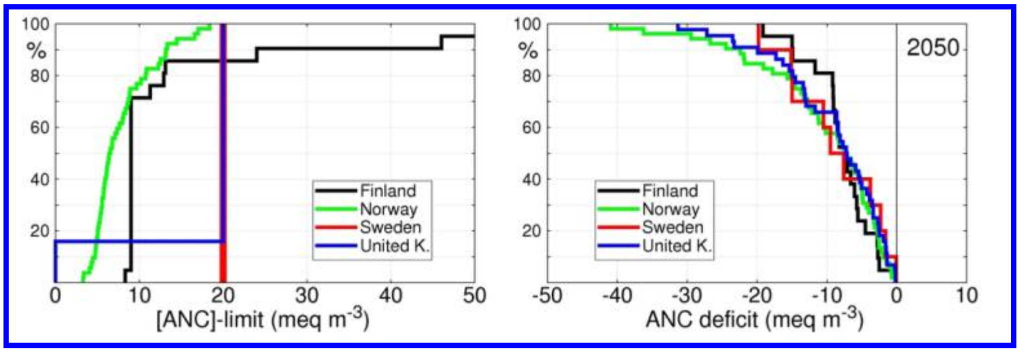

Figure 5: Left: Cumulative distribution functions of the ANC-limits for the lakes with target loads in Finland (21), Norway (52), Sweden (10) and the United Kingdom (44). Right: Inverse cumulative distribution functions of the 'ANC deficit' in 2050 for the same lakes in those countries. The ANC deficit shows the difference between simulated ANC in 2050 under the 2010 Gothenburg Protocol (see Figure 2) and the specified critical ANC-limit (left Figure).

Dynamic Modelling and Target Loads of Sulphur and Nitrogen for Surface Waters in Finland, Norway, Sweden and the United Kingdom

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Abstract

The target load concept is an extension of the critical load concept of air pollution inputs to ecosystems. The advantage of target loads over critical loads is that one can define the deposition *and* the point in time (target year) when the critical (chemical) limit is no longer violated. This information on the timing of recovery requires dynamic modelling. Using a well-documented dynamic model, target loads for acidic deposition were determined for 848 surface waters across Finland, Norway, Sweden and the United Kingdom for the target year 2050. In the majority of sites (n = 675), the critical ANC-limit was predicted to be achieved by 2050; however, for 127 sites target loads were determined. In addition, 46 sites were infeasible, i.e., even a deposition reduction to zero would not achieve the limit by 2050. The average maximum target load for sulphur was 38% lower than the respective critical load across the study lakes (n = 127). Target loads on a large regional scale can inform effects-based emission reduction policies; the current assessment suggests that reductions beyond the Gothenburg Protocol are required to ensure surface water recovery from acidification by 2050.

Keywords. Acidification; critical load; recovery; Acid Neutralising Capacity (ANC); Model of Acidification of Groundwaters in Catchments (MAGIC)

1. Introduction

During the 1970s it was recognized that surface waters in large parts of Europe and North America were being influenced by air pollution, i.e., acidic deposition, owing to anthropogenic emissions of sulphur (S) and nitrogen (N) oxides.[1,2] Shortly thereafter, empirical[3] and steady-state[4-6] models were developed and applied to predict the impacts of acidic deposition on surface waters. In concert, it was recognised that time-dependent processes could buffer (delay) ecosystem (soil and surface water) response to acidic deposition. The incorporation of these processes required time-dependent or 'dynamic' modelling frameworks. The earliest dynamic simulation models incorporated established relationships from soil and water chemistry to predict the most likely effects of acidic deposition on surface waters.[7] Ultimately, these dynamic models provided a quantitative framework to assess whether (and how quickly) a decrease in acidic stress would result in a recovery of ecosystems.[8] Since the 1980s, several dynamic (hydro-chemical) models have been developed and extensively applied at site-specific and regional scales to predict changes in soil and surface water chemistry due to acidic deposition.[9-15] Moreover, dynamic models can provide a quantitative estimate of the time lag between a reduction in deposition and the attainment of 'acceptable' ecosystem status (based on a threshold, or 'critical value', for a specified chemical criterion, e.g., surface water pH=6.0). This time lag has been denoted as the damage time lag[16] or recovery delay time.[17-19]

The assessment of impacts of acidic deposition on terrestrial and aquatic ecosystems has supported policies to reduce anthropogenic S and N emissions. In Europe, the critical loads approach is widely accepted as the basis for negotiating effects-based control strategies for air pollution. A critical load is defined as 'a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge'.[20] The approach is based on setting a critical limit for a chemical criterion (e.g., Acid Neutralising Capacity [ANC]) to protect a specified biological indicator for a chosen receptor ecosystem (e.g., fish species for surface waters, or tree roots in forest soils), and via inverse modelling a deposition (the critical load) is derived to ensure the limit is not violated and thus 'harmful effects' avoided.[21-23] Critical loads have been used in the negotiations of several protocols to the United Nations Economic Commission for Europe's (UNECE[24]) Convention on Long-range

Transboundary Air Pollution (LRTAP) and the European Union's National Emission Ceilings (NEC) Directive[25] and its revision.

However, by definition, critical loads are steady-state quantities and as such they do not provide information on the time involved for a system (e.g., surface water) to reach a certain chemical (or biological) state. Dynamic models are needed to calculate recovery times under prescribed emission reductions. As such, dynamic modelling has also become an important part of the effects-oriented work under the LRTAP Convention.[26] If a desired chemical state of a surface water is defined for a given year, dynamic models can be used in an inverse mode to compute the deposition path leading to that desired state (if feasible). Such a deposition is called a target load, and the year in which the desired state is to be reached is called the target year.[18,19] There have been few published studies describing the use of target loads in Europe[16,17,27] and fewer for surface waters.[28-30] It is important to note that in North America the term 'target load' has also been used to refer to a 'target' deposition, determined by political (or management) agreement, that can be higher or lower than the critical load[31-35], often based on arbitrary interpretations of the impacts data rather than the avoidance of specific deleterious ecological effects.[32]

The objective of this study was to establish target loads for European regions dominated by acid-sensitive surface waters, which ensure acidification recovery by the year 2050 (target year). The target loads go beyond deposition reductions under the Gothenburg Protocol (implemented in 2010), to ensure chemical recovery in surface water ANC (chemical criterion). The target loads were determined using the Model of Acidification of Groundwaters in Catchments (MAGIC) for lakes in Finland, Norway, Sweden, and the United Kingdom. Further, the conceptual basis for the determination of a target load from a dynamic model is also provided.

2. Dynamic Modelling and Target Loads

With critical loads, i.e., in the steady-state situation, only two cases can be distinguished when evaluated against deposition: (1) deposition is below the critical load, or (2) deposition is greater than the critical load, i.e., there is an exceedance of the critical load. 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. Thus, a critical load serves as a warning as long as there is exceedance, since it indicates that deposition should be reduced. However, it is often (implicitly) assumed that reducing deposition to (or below) the critical load immediately removes the risk of 'harmful effects', i.e., the critical chemical criterion (e.g., the ANC-limit) that links the critical load to the (biological) effect, immediately attains a non-critical ('safe') value (and that there is immediate biological recovery as well). However, the reaction of an ecosystem (or catchment) to changes in deposition is delayed by (finite) buffers, e.g., the cation exchange capacity of catchment soils. These buffer mechanisms can delay the attainment of the critical chemical value, and it might take decades or even centuries, before 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.

Therefore, dynamic models are needed to estimate the time involved in attaining a certain chemical state in response to different deposition scenarios. Five stages can be distinguished in the possible development of a (lake) chemical variable in response to a 'typical' temporal deposition pattern (see Figure 1):

Stage 1: Deposition is below the critical load (CL) and the chemical criterion (ANC) does not violate its critical limit. As long as deposition stays below the CL, this is the 'ideal' situation (blue lines in Figure 1).

Stage 2: Deposition is above the CL, but the critical chemical criterion is not (yet) violated because there is a time delay of ecosystem response before adverse effects occur. Therefore, no damage is likely to occur at this stage, despite exceedance of the CL. The time between the first exceedance of the CL and the first violation of the chemical criterion is termed the *Damage Delay Time* (DDT=t_2–t_1).

Stage 3: The deposition is above the CL and the critical chemical criterion is violated. Measures (emission reductions) have to be taken to avoid a (further) deterioration of the ecosystem (biological indicator linked to the chemical criterion).

Stage 4: Deposition is below the CL, but the chemical criterion is still violated and thus (full) recovery has not yet occurred. The time between the first non-exceedance of the CL and the subsequent non-violation of the criterion is termed the *Recovery Delay Time* (RDT=t_4–t_3). Note: RDT is not necessarily the same (or even similar) to DDT due to hysteresis effects in certain (soil) processes (e.g., cation exchange); the schematic is purely for illustration and does not reflect the relative temporal processes.

Stage 5: Deposition is below the CL and the critical chemical criterion is no longer violated. This stage is analogous to Stage 1, and the ecosystem is considered to have recovered. In practice it might happen that the critical limit cannot be achieved within a reasonable (policy-relevant) timeframe, even for zero N and S deposition, e.g., due to the depletion of exchangeable base cations.

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, it will take time before biological recovery is achieved (if at all).

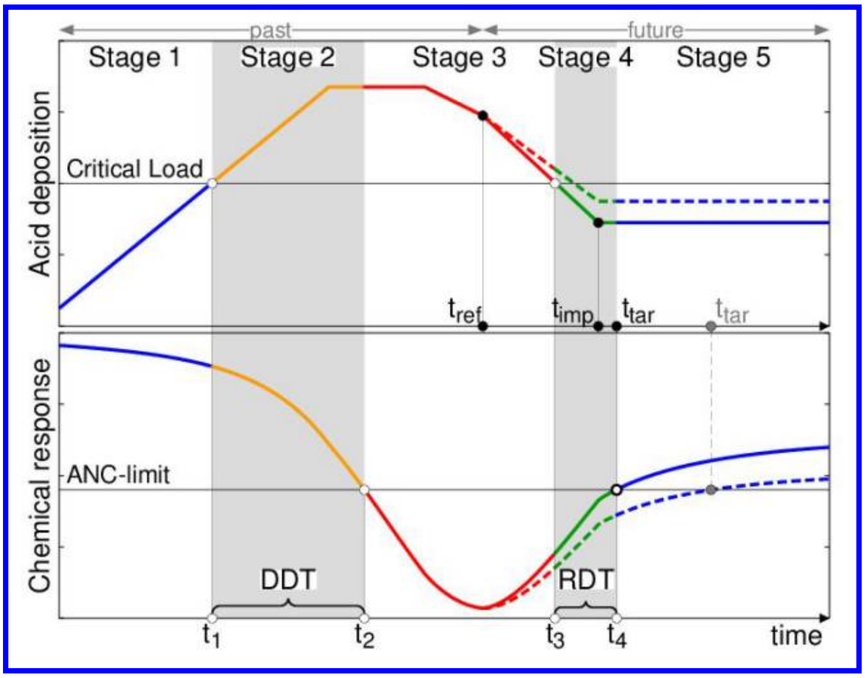

Figure 1: Generalised past and future development stages (indicated by different colours) of acidic deposition and lake chemical criterion response (here: ANC) in comparison to the critical chemical value (ANC-limit) and the critical load derived from it (i.e., the determination of critical load is based on a critical limit for a specified chemical criterion, which protects the biological indicator from deleterious effects). The delay between the (non-)exceedance of the critical load and the (non-)violation of the critical chemical value is indicated in grey shades, highlighting the Damage Delay Time (DDT) and the Recovery Delay Time (RDT) of the system. Also shown are the points in time (t_{ref}, t_{imp}) relevant for defining a target load (< critical load) to reach non-violation of the chemical value at a pre-specified time t_{tar}. The dashed lines show the temporal development for a later target year (labelled in grey).

The most straightforward use of a dynamic model is scenario analysis, i.e., first a future deposition scenario is assumed, and then the (chemical) consequences for the ecosystem (e.g., lake) are evaluated. A target load, on the other hand, is the deposition path that ensures that a prescribed value of the chemical criterion is achieved in a given year. Here we define a target load as a deposition path characterised by three numbers (years): (i) the reference year, (ii) the implementation year, and (iii) the target year (see Figure 1). The *reference year*, t_{ref}, is the year (time) up to which the (historic) deposition path is given and cannot be changed. The *implementation year*, t_{imp}, is the year by which all reduction measures, needed to reach the final deposition (the target load), are implemented. Between the reference year and the implementation year depositions are assumed (assured) to change linearly (see Figure 1). Finally, the *target year* is the year in which the critical chemical criterion (e.g., the ANC-limit) is met (for the first time). The above three years define a unique deposition path, the final value of which is referred to as a target load. The earlier the target year, the lower the target load (at sites where the chemical criterion is violated – for other sites a target load is not relevant), since higher deposition reductions are needed to achieve the desired status within a shorter timeframe (see Figure 1). In extreme cases, a target load might not exist at all, i.e., even reduction to zero deposition in the implementation year will not result in the desired ecosystem status within the prescribed time; in this instance the target load is termed 'infeasible'. For more information on target loads and related topics see Posch et al.[22], Jenkins et al.[19] or Bonten et al.[27]

3. Materials and Methods

The current study focused on surface waters (lakes and streams) with long-term observations of chemistry in acid-sensitive regions of Europe[36], i.e., Finland, Norway, Sweden, and the United Kingdom. In general, these sites are considered to be sentinel indicators of acidification impacts, and their recovery is assumed to reflect wider ecosystem acidification recovery across the entire study region; as such, they are well suited for the determination of regionally representative target loads. All surface waters were part of routine acidification monitoring networks since the 1980s and 1990s, typically located in regions with acid-sensitive geology. For example, the Finnish acidification monitoring network, maintained by the Finnish Environment Institute, consisted of 163 lakes located throughout Finland, subjectively chosen by expert judgement from a national acidification survey during 1987 for use in acidification assessments.[37] Similarly, the Norwegian study lakes (n = 131) were a subset of the national monitoring programme, confined to lakes south of 62.5° latitude, with observations suitable for dynamic modelling. The study sites are predominantly small acid-sensitive headwater lakes and streams, with low base cation concentrations, low alkalinity and low (charge balance) ANC.[38] All surface waters have been widely used in acidification assessments evaluating long-term trends in surface water chemistry[39-41], and the prediction of future chemistry using dynamic (hydro-chemical) models, specifically MAGIC.[42-46] The study sites have played a central role in European-scale projects, such as 'Recover:2010'[47] and 'Eurolimpacs'[48], focused on model simulations of surface water response to European emissions reduction policies. The process limitations and predictive uncertainty of MAGIC in isolation, and compared with other models, e.g., PnET-BGC (photosynthesis and evapotranspiration-biogeochemistry), SAFE (soil acidification in forest ecosystems), and VSD (very simple dynamic), have been widely published.[12,15,49,50] Similarly, the influence of climate change on model predictions for MAGIC have been widely assessed[43,46,51,52]. As such, herein we focus on the determination of target loads using MAGIC, which (hitherto for) have not been reported for the study sites and refer the reader to previous publications for detailed information regarding model calibration and process uncertainty for MAGIC. Nonetheless, we provide a brief overview of MAGIC, its application, calibration and simulation for the study sites, specifically with respect to target loads.

MAGIC is a lumped-parameter model of intermediate complexity, developed to predict the long-term effects of acidic deposition on soil and surface water chemistry.[53,54] The model predicts monthly and annual average concentrations of the major ions for soil solution and surface water chemistry. MAGIC represents the catchment with aggregated, uniform soil compartments (up to three), and a surface water compartment that can be either a lake or a stream. Time series inputs to the model include: deposition of ions from the atmosphere (wet plus dry deposition); discharge volume and flow routing within the catchment; biological production and removal of ions; internal sources and sinks of ions from weathering or precipitation reactions; and climate data. Constant parameters in the model include physical and chemical characteristics of the soils and surface waters, and thermodynamic constants. Soil base cation weathering rate and initial base saturation are calibrated using observed values of surface water and soil chemistry for a specified period. In this instance, calibration refers to an automated optimisation procedure that is a component of the MAGIC suite (i.e., MAGICOPT), generally used for regional applications. The minimum required site-measured variables for calibration are: surface water concentrations for the major ions and soil exchangeable fractions for base cations: calcium (Ca^{2+}), magnesium (Mg^{2+}), sodium (Na^+) and potassium (K^+). The MAGIC suite also includes an iteration routine for the determination of target loads. In this study, the deposition path was optimised between 2010 (Gothenburg Protocol) and 2020 (the implementation year) to ensure the ANC-limit was achieved by 2050.

In the current study, the application of MAGIC (by national experts) across the study lakes followed a common (prescribed) procedure as described in Posch et al.[38]; for further details on the application and calibration of MAGIC see, for example, Aherne et al.[44] In brief, all catchments were represented by one soil compartment receiving deposition and releasing discharge to the lake compartment. The soil compartment represented the aggregated horizons of the catchment soils (mineral and organic), with exchangeable base cation data taken from national and focused surveys.[38] Simulations were carried out using an annual time-step, with a number of simplifying assumptions applied consistently across all study lakes. Discharge volume and flow routing within the catchments were not varied; discharge was described using long-term means with 100% routed to the lake. Detailed process-oriented N dynamics were not modelled, i.e., the coefficient for in-lake N retention was set to a similar value for all lakes[36], and terrestrial N retention was set to match observed lake concentrations. To account for uncertainty in a number of the fixed parameters (lake retention, soil depth, soil bulk density, cation exchange capacity, etc.), a 'fuzzy' optimisation method was employed. Ten calibrations were carried out for each study lake using MAGICOPT; for each simulation the fixed parameters were randomly varied within specified uncertainty bands (±10%). Uptake of base cations from forested catchments were modelled using a simplified regional sequence, based on a regional average planting date, constant nutrient concentrations (from literature), and species composition and coverage from national forest inventories (for further details see, e.g., Aherne et al.[52,55] for Finland and Moldan et al.[46] for Sweden).

In the current study, MAGIC was calibrated to 848 lakes (see Figure 2) across Finland (FI=163), Norway (NO=131), Sweden (SE=234), and the United Kingdom (UK=320), spanning a range in deposition (S plus N) from <10 meq m^{-2} yr^{-1} in northern Sweden and Finland to 150 meq m^{-2} yr^{-1} in the South Pennines, UK (Figure 2). Historic and future surface water chemistry for each lake were simulated for the period 1880–2100 under modelled anthropogenic S and N deposition[56] from the EMEP model.[57] Future lake chemical recovery under the Gothenburg Protocol (which came into force in 2010) was evaluated using charge balance ANC (defined as the difference between the sum of the concentrations of base cations and strong acid anions); lake ANC is an established acidification indicator, as it is strongly indicative of biological recovery.[58,59] In addition, ANC is the most widely used chemical criterion in critical load calculations for surface waters.[60] Target loads were estimated for the target year 2050, with the implementation year 2020, i.e., the year in which deposition reductions beyond the Gothenburg Protocol (year 2010) are fully implemented to ensure attainment of the critical chemical criterion by the target year.[38] While dynamic modelling was carried out on every study site (n = 848), target loads were only calculated for each surface water that did not meet the specified critical chemical criterion (ANC-limit) by 2050 under the Gothenburg Protocol, i.e., those lakes that still violated ANC-limits or with a recovery delay (Note: lakes were the only study sites that still violated the ANC-limit). The specified ANC-limit varied among countries (based on national management objectives); all Swedish lakes had a fixed value of 20 meq m^{-3}, similarly the UK surface waters (lakes and streams) had a value of 20 meq m^{-3}, except for a small number of naturally acidic sites that had a limit of zero (n=21; 6.5%). Organic acids can act as strong acid anions reducing the acid neutralizing (buffering) capacity of a lake to incoming acidity[61]; to accommodate this, Finland and Norway used an organic acid adjusted ANC-limit[62], which, for example, resulted in an average ANC-limit of 14 meq m^{-3} for the Finnish lakes.

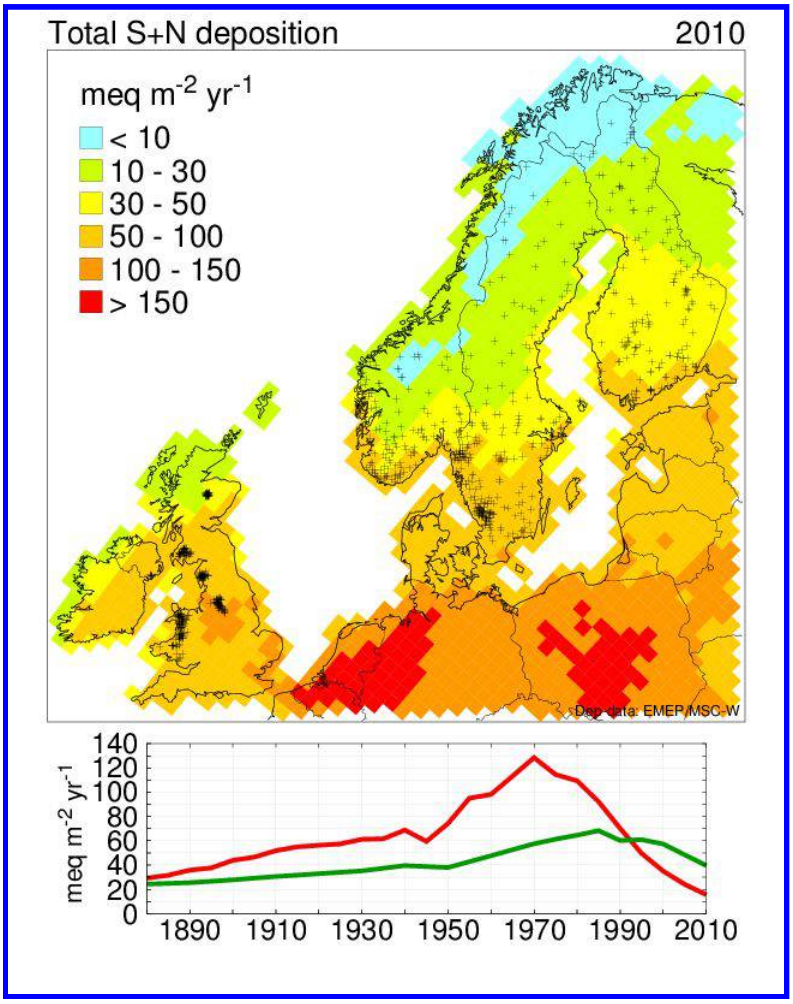

Figure 2: Location of the study sites used for dynamic modelling (n = 848; black crosses) overlaid upon modelled total sulphur (S) plus nitrogen (N) deposition in 2010 (meq m^{-2} yr^{-1}) on the EMEP 50 km × 50 km grid (for further details on the EMEP model see Simpson et al.[57]). The temporal development (1880–2010) of the total S (red) and N (green) deposition averaged over the 848 study sites is also shown.

Target load functions were estimated for each calibrated surface water that did not meet the specified critical chemical criterion (ANC-limit) by 2050 under the Gothenburg Protocol, i.e., every pair of N and S deposition that met the ANC-limit in 2050 under further (beyond Gothenburg) emission reductions defined a target load function of acidity (TLF), similar to a critical load function[21] (see also Supporting Information for further details) for each study lake (catchment). The piece-wise linear function in the (N, S) deposition-plane is delineated by the maximum target load of S, TLmaxS (for N deposition = 0) and the maximum target load of N, TLmaxN (for S deposition = 0).

4. Results and Discussion

Regional dynamic modelling results have been reported for individual countries.[42,45,46,52,63] However, previous assessments primarily focused on scenario analyses, i.e., simulations to answer the question: ‘what is the future chemical status of a surface water under various deposition scenarios?’ In contrast, the current study addresses the inverse question: ‘what deposition, called target load, is required to obtain a specified lake chemical status within a given time period (if feasible)?’.

Dynamic model simulations were carried out for 848 surface waters, but target load calculations were only necessary for 173 lakes (Table 1). The simulated water chemistry for the target year 2050 was predicted to be greater than (or equal to) the chemical criterion (ANC-limit) for 675 surface waters. Target loads, i.e., loads below the respective critical loads, were determined for 24 lakes in Finland, 56 in Norway, 14 in Sweden and 79 in the United Kingdom. Of these 173 lakes, 46 were ‘infeasible’ (Table 1), i.e., even reducing anthropogenic deposition to zero by 2020 would not result in an ANC greater than or equal to the ANC-limit in 2050. In general, infeasible lakes occurred in < 3% of the study sites per country; however, in the United Kingdom, infeasible lakes occurred in ~11% (n = 35) of the study sites likely reflecting their higher cumulative historic deposition (Figure 2). Neglecting infeasible sites, ‘true’ target loads were determined for127 lakes (Table 1), 21 in Finland, 52 in Norway, 10 in Sweden and 44 in the United Kingdom; the highest proportion occurred in Norway (40%) followed by Finland (13%).

The average TLmaxS (see Supporting Information) per country ranged from 7.5 meq m^{-2} yr^{-1} (Finland) to 38.9 5 meq m^{-2} yr^{-1} (United Kingdom). Note, for all study sites the maximum critical load of S (CLmaxS) was also computed as the steady-state solution of the dynamic model; compared with the average CLmaxS, the average TLmaxS was 53% lower in Finland, 40% in Norway, 20% in Sweden and 36% in the United Kingdom. Across all lakes, average TLmaxS (24.1 meq m^{-2} yr^{-1}; n = 127) was 38% lower than the respective CLmaxS (39.1 meq m^{-2} yr^{-1}; Table 1).

Table 1: Number of lakes in each country with dynamic model (DM) simulations, divided into the number of lakes for which the critical load is sufficient to achieve the ANC-limit in 2050 (ANC-2050), number of infeasible sites (INF) and ‘true’ target loads (TL). Also given are the averages of TLmaxS and CLmaxS for lakes under ‘TL’.

Country	DM	ANC-2050	INF	TL	TLmaxS*	CLmaxS
					meq m^{-2} yr^{-1}	
Finland (FI)	163	139	3	21	7.45	15.94
Norway (NO)	131	75	4	52	19.17	31.94

Sweden (SE)	234	220	4	10	19.91	25.02
United Kingdom (UK)	320	241	35	44	38.87	61.72
Sum/Average	848	675	46	127	24.11	39.07

*See Supporting Information for further description of TLmaxS (and CLmaxS).

To provide greater regional coverage, target loads were mapped on the EMEP 50 km × 50 km grid (Figure 3) by setting TLmaxS to CLmaxS where TLmaxS > CLmaxS (since the critical load is already sufficient for non-violation of the ANC-limit by 2050). To account for all TLs within a grid cell, the 5-th percentile of the cumulative distribution function for all target loads in that grid cell was mapped.[23] Overall, no clear pattern can be discerned in the mapped target loads. In general, the critical load is sufficient for achieving non-violation of the ANC-limit in most areas; nevertheless 'true' target loads are concentrated in southern Norway and Finland, and in northern Wales in the United Kingdom (Figure 3).

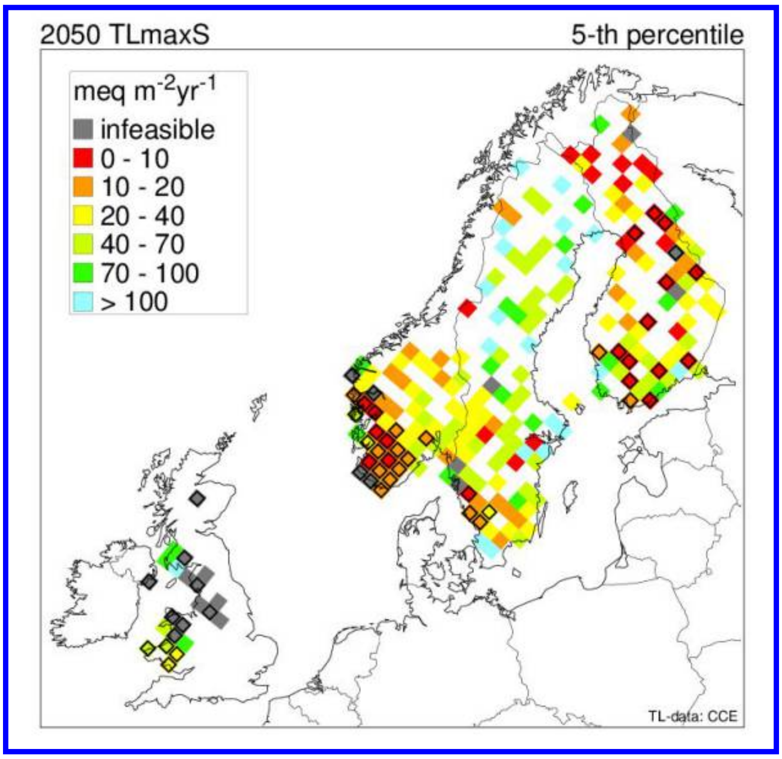

Figure 3: Map of the 5-th percentile of the 2050 maximum target load of sulphur (TLmaxS) on the EMEP 50 km × 50 km grid for 848 catchments in Finland, Norway, Sweden and the United Kingdom. Note: TLmaxS was set to the maximum critical load of S (CLmaxS) where TLmaxS > CLmaxS (i.e., the critical load is sufficient for non-violation of the ANC-limit by 2050). Grey-filled cells (label 'infeasible') denote grids containing at least one lake where the simulated ANC does not meet the specified limit by 2050, even under zero deposition after 2020 (FI = 3, NO = 4, SE = 4, UK = 35; Table 1). Black diamonds frame grids with at least one 'true' TL, i.e., where a TL exists and is lower than the CL (FI = 21, NO = 52, SE = 10, UK = 44; Table 1).

The key chemical variable of interest was ANC, as it is used as a chemical criterion linking water chemistry to the biological (fish) status of the lakes; as such, time series of ANC were simulated to illustrate the timing and rate of chemical changes during acidification and recovery. The general pattern of predicted ANC in the study lakes is similar in the four countries (Figure 4), driven by the deposition of S and N (Figure 2). The differences between the regions were due to proximity to emission sources, acid sensitivity of regions, differences in land use and the selected lakes.

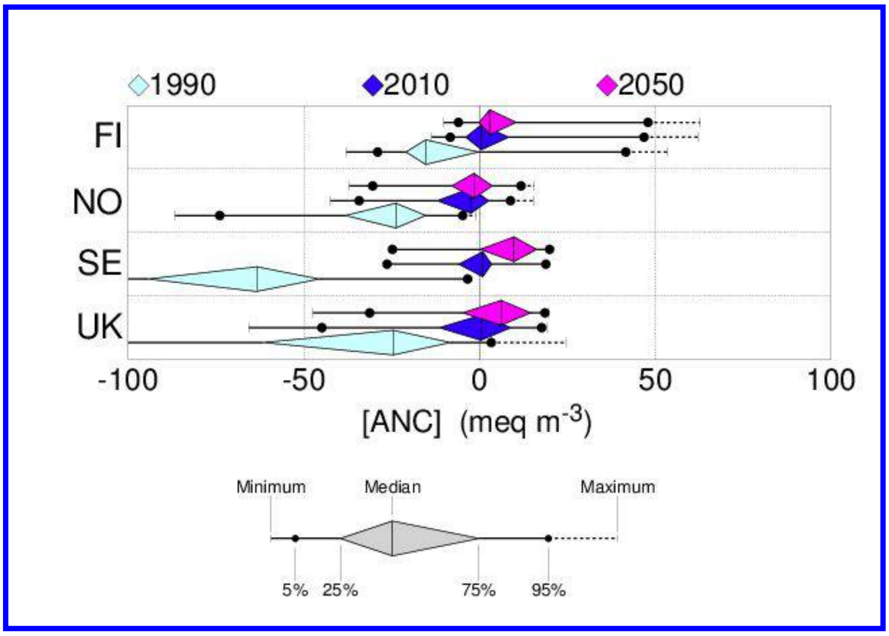

Figure 4: Percentile statistics ('diamond plots') of simulated annual average lake acid neutralising capacity (ANC) in 1990, 2010 and 2050 in Finland (FI), Norway (NO), Sweden (SE) and the United Kingdom (UK). Data are only shown for lakes for which target loads were determined (FI = 24, NO = 56, SE = 14, UK = 79; see columns 'INF' plus 'TL' in Table 1).

Implementation of the Gothenburg Protocol resulted in a significant increase in ANC from 1990 to 2010 (paired t-test, $p < 0.001$) in all four countries (average increase of 32.6 meq m^{-3}; Figure 4) and is predicted to significantly improve by 2050 albeit by a smaller amount (average increase of 4.2 meq m^{-3}; Figure 4). This is due to the fact that the deposition is kept at the 2010 level after that year, and that only (slow) improvements in the soil and water conditions, e.g., replenishment of base cation pools, will raise the ANC.[64] Surface waters in Sweden showed the greatest improvement in ANC between 1990 and 2010 (Figure 4) owing to the concentration of sensitive lakes in south-western Sweden[1,14] and the large reduction in acidic deposition in that region (Figure 2).

Despite the predicted improvements in ANC, ultimately, we are concerned with the 'distance' between the predicted chemical status of a lake and its desired status (note: the desired ANC-limit is variable in all countries except in Sweden, see Figure 5a). A better characterisation of the 'distance' of a lake's chemical status from the desired one is through the so-called ANC-deficit, i.e., the difference between simulated ANC (under the 2010 Gothenburg Protocol deposition) and the (lake-specific) ANC-limit. The inverse cumulative distributions of the ANC-deficit for the year 2050 were quite similar in the four countries up to the 80-th percentile (Figure 5b); notably a maximum deficit around 40 meq m^{-3} was simulated for some Norwegian lakes and 30 meq m^{-3} for some lakes in the United Kingdom. Implementation of target load depositions would ensure that all lakes reach their specified ANC-limit by 2050, i.e., all ANC deficits reach zero by 2050.

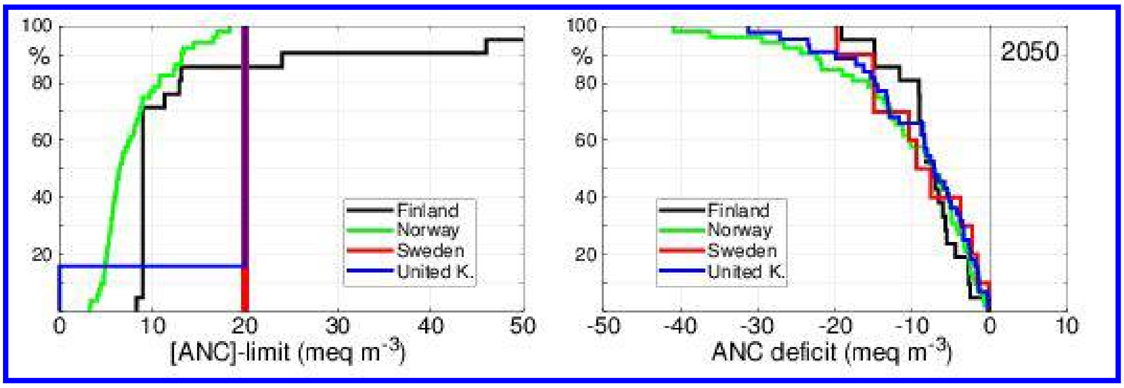

Figure 5: Left: Cumulative distribution functions of the ANC-limits for the lakes with target loads in Finland (21), Norway (52), Sweden (10) and the United Kingdom (44). Right: Inverse cumulative distribution functions of the 'ANC deficit' in 2050 for the same lakes in those countries. The ANC deficit shows the difference between simulated ANC in 2050 under the 2010 Gothenburg Protocol (see Figure 2) and the specified critical ANC-limit (left Figure).

Dynamic modelling was carried out for 848 lake catchments in Finland, Norway, Sweden and the United Kingdom. Given the large number of acid-sensitive lakes in each country, a larger number would be desirable, but the chosen study lakes were deemed to be a representative sample (as outlined in Materials and Methods) and (more importantly) have the inputs required for dynamic modelling. It was predicted that 675 of these 848 lakes will achieve their critical ANC-limit by the year 2050 under the Gothenburg Protocol, which leaves 173 lakes (20%), for which emission reductions beyond Gothenburg are required, if one wants to achieve non-violation of the ANC-limit by 2050. However, for 46 of these lakes (~5% of the total), even a reduction to zero deposition by 2020 would not be sufficient to achieve the ANC-limit by 2050. This does not mean that those lakes would never recover (chemically), only that recovery would occur (maybe long) after 2050.

In the current study, model simulations have been conducted without consideration of future climate change, as the primary objective was to support emissions reduction polices (irrespective of climate perturbations). Nonetheless, several (regional) studies have been conducted using MAGIC that explore the direct and indirect effects of climate change on lake chemistry[43,51,52]. Although the (indirect) effects can be great for individual lakes, the overall effects on lake chemistry are not huge, considering all other (model) uncertainties.[51] Reductions of S and N deposition are the most important of determinants of future lake (acid) status in European surface waters.

While target loads have been discussed and determined in Europe under the LRTAP Convention, they have not been used explicitly to guide emission reduction policies. One reason might be that it requires dynamic modelling – and thus more input data and expertise to determine target loads – compared to critical loads that are ‘easily determined’ steady-state quantities. However, lack of information on time needed for achieving the desired chemical status under critical loads should ultimately encourage the determination of target loads to provide policy advisors with guidance on the timing of ecosystem recovery. While acidification is generally assumed to be ‘solved’ in Europe, there is growing recognition that surface waters in some regions are still acidified[2]; the current assessment suggests that emissions reductions beyond the Gothenburg Protocol are required to ensure surface water recovery from acidification by 2050.

Supporting Information

Description of the Target Load Function (TLF)

Notes

The authors declare no competing financial interest.

Acknowledgements

M.P. thanks the Trust Fund for the effect-oriented activities under the Convention on Long-range Transboundary Air Pollution. This research was undertaken, in part, thanks to funding to J.A. from an NSERC Discovery grant and the Irish EPA (2012-CCRP-MS.7 and 2016-CCRP-MS.43). M.F. thanks the Strategic Research Council at the Academy of Finland for financial support (decision 312559).

References

(1) Garmo, Ø. A.; Skjelkvåle, B. L.; De Wit, H. A.; Colombo, L.; Curtis, C.; Fölster, J.; Hoffmann, A.; Hruška, J.; Høgåsen, T.; Jeffries, D. S.; Keller, W. B.; Krám, P.; Majer, V.; Monteith, D. T.; Paterson, A. M.; Rogora, M.; Rzychon, D.; Steingruber, S.; Stoddard, J. L.; Vuorenmaa, J.; Worsztynowicz, A. Trends in surface water chemistry in acidified areas in Europe and North

America from 1990 to 2008. *Water Air and Soil Pollution* **2014**, 225(3), 1880.

(2) Austnes, K.; Aherne, J.; Arle, J.; Čičendajeva, M.; Couture, S.; Fölster, J.; Garmo, Ø.; Hruška, J.; Monteith, D.; Posch, M.; Rogora, M.; Sample, J.; Skjelkvåle, B. L.; Steingruber, S.; Stoddard, J. L.; Ulańczyk, R.; Van Dam, H.; Toro Velasco, M.; Vuorenmaa, J.; Wright, R. F.; De Wit, H. Regional assessment of the current extent of acidification of surface waters in Europe and North America. NIVA Report 7268 (ICP-Waters Report 135), Norwegian Institute for Water Research, Oslo, Norway, **2018**; 134 pp; www.niva.no

(3) Small, M. J.; Sutton, M. C. A direct distribution model for regional aquatic acidification. *Water Resources Research* **1986**, 22(13): 1749–1758.

(4) Henriksen, A. A simple approach for identifying and measuring acidification of freshwater. *Nature* **1979**, 278: 542–545.

(5) Thompson, M. E. The cation denudation rate as a quantitative index of sensitivity of Eastern Canadian rivers to acidic atmospheric precipitation. *Water, Air, & Soil Pollution* **1982**, 18, 215–226.

(6) Schnoor, J. L.; Lee, S.; Nikolaidis, N. P.; Nair, D. R. Lake resources at risk to acidic deposition in the eastern United States. *Water, Air, & Soil Pollution* **1986**, 31, 1091–1101.

(7) Reuss, J. O. Simulation of soil nutrient losses resulting from rainfall acidity. *Ecological Modelling* **1980**, 11, 15–38.

(8) Kauppi, P.; Kämäri, J.; Posch, M.; Kauppi, L.; Matzner, E. Acidification of forest soils: Model development and application for analyzing impacts of acidic deposition in Europe. *Ecological Modelling* **1986**, 33, 231–253; DOI: 10.1016/0304-3800(86)90042-6

(9) De Vries, W.; Reinds, G. J.; Posch, M. Assessment of critical loads and their exceedance on European forests using a one-layer steady-state model. *Water, Air, & Soil Pollution* **1994**, 72, 357–394; DOI: 10.1007/BF01257134

(10) Alveteg, M.; Sverdrup, H.; Warfvinge, P. Regional assessment of the temporal trends in soil acidification in southern Sweden, using the SAFE model. *Water, Air, & Soil Pollution* **1995**, 85, 2509–2514.

(11) Aherne, J.; Dillon, P. J.; Cosby, B. J. Acidification and recovery of aquatic ecosystems in south-central Ontario, Canada: regional application of the MAGIC model. *Hydrology and Earth System Sciences* **2003**, 7, 561–573.

(12) Wright, R. F.; Larssen, T.; Camarero, L.; Cosby, B. J.; Ferrier, R. C.; Helliwell, R.; Forsius, M.; Jenkins, A.; Kopáček, J.; Majer, V.; Moldan, F.; Posch, M.; Rogora, M.; Schöpp, W. Recovery of acidified European surface waters. *Environmental Science & Technology* **2005**, 39, 64A-72A; DOI: 10.1021/es0531778

(13) Gbondo-Tugbawa, S. S.; Driscoll, C. T.; Aber, J. D.; Likens, G. E. Evaluation of an integrated biogeochemical model (PnET-BGC) at a northern hardwood forest ecosystem. *Water Resources Research* **2001**, 37(4): 1057–1070.

(14) Helliwell, R. C.; Wright, R. F.; Jackson-Blake, L. A.; Ferrier, R. C.; Aherne, J.; Cosby, B. J.; Evans, C. D.; Forsius, M.; Hruska, J.; Jenkins, A.; Krám, P.; Kopáček, J.; Majer, V.; Moldan, F.; Posch, M.; Potts, J. M.; Rogora, M.; Schöpp, W. Assessing recovery from acidification of European surface waters in the year 2010: Evaluation of projections made with the MAGIC model in 1995. *Environmental Science & Technology* **2014**, 48(22), 13280–13288; DOI: 10.1021/es502533c

(15) Tominaga, K.; Aherne, J.; Watmough, S. A.; Alveteg, M.; Cosby, B. J.; Driscoll, C. T.; Posch, M.; Pourmokhtarian A. Predicting acidification recovery at the Hubbard Brook Experimental Forest, New Hampshire: Evaluation of four models. *Environmental Science & Technology* **2010**, 44(23): 9003–9009; DOI: 10.1021/es102243j

(16) Hettelingh, J.-P.; Posch, M. Critical loads and a dynamic assessment of ecosystem recovery. In *Predictability and Nonlinear Modelling in Natural Sciences and Economics*; Grasman, J., Van Straten, G., Eds.; Kluwer: Dordrecht, Netherlands, **1994**; pp. 439–446.

(17) Warfvinge, P.; Holmberg, M.; Posch, M.; Wright, R. F. The use of dynamic models to set target loads. *Ambio* **1992**, 21(5), 369–376.

(18) Posch, M.; Hettelingh, J.-P.; Slootweg, J., Eds.. Manual for dynamic modelling of soil response to atmospheric deposition. RIVM Report 259101012, Coordination Centre for Effects, Bilthoven, Netherlands, **2003**; 71 pp; www.rivm.nl

(19) Jenkins, A.; Cosby, B. J.; Ferrier, R. C.; Larssen, T.; Posch, M. Assessing emission reduction targets with dynamic models: deriving target load functions for use in integrated assessment. *Hydrology and Earth System Sciences* **2003**, 7(4), 609–617; DOI: 10.5194/hess-7-609-2003

(20) Nilsson, J.; Grennfelt, P., Eds. Critical loads for sulphur and nitrogen. Nord 1988:97, Nordic Council of Ministers, Copenhagen, Denmark, **1988**; 418 pp.

(21) Posch, M.; Aherne, J.; Forsius, M.; Rask, M. Past, present, and future exceedance of critical loads of acidity for surface waters in Finland. *Environmental Science & Technology* **2012**, 46, 4507–4514; DOI: 10.1021/es300332r

(22) Posch, M.; De Vries, W.; Sverdrup, H. U. Mass balance models to derive critical loads of nitrogen and acidity for terrestrial and aquatic ecosystems. In *Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems*; De Vries, W., Hettelingh, J.-P., Posch, M., Eds.; Springer: Dordrecht, **2015**; pp. 171–205; DOI: 10.1007/978-94-017-9508-1_6

(23) UBA. Manual on methodologies and criteria for modelling and mapping critical loads & levels and air pollution effects, risks and trends. Umweltbundesamt Texte 52/04, Berlin, Germany, **2004**; www.icpmapping.org

(24) UNECE. The 1999 Gothenburg Protocol to Abate Acidification, Eutrophication and Ground-level Ozone. United Nations Economic Commission for Europe (UNECE), **2015**; www.unece.org/env/lrtap/multi_h1.html

(25) EC. Directive 2001/81/EC of the European Parliament and of the Council of 23 October 2001 on National Emission Ceilings for Certain Atmospheric Pollutants, **2001**; http://eur-lex.europa.eu

(26) Hettelingh, J.-P.; Posch, M.; Slootweg, J.; Reinds, G. J.; Spranger, T.; Tarrason, L. Critical loads and dynamic modelling to assess European areas at risk of acidification and eutrophication. *Water, Air, & Soil Pollution: Focus* **2007**, 7(1–3), 379–384; DOI: 10.1007/s11267-006-9099-1

(27) Bonten, L. T. C.; Reinds, G. J.; Groenenberg, J. E.; De Vries, W.; Posch, M.; Evans, C. D.; Belyazid, S.; Braun, S.; Moldan, F.; Sverdrup, H. U.; Kurz, D. Dynamic geochemical models to assess deposition impacts and target loads of acidity for soils and surface waters. In *Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems*; De Vries, W., Hettelingh, J.-P., Posch, M., Eds.; Springer: Dordrecht, **2015**; pp. 225-251; DOI: 10.1007/978-94-017-9508-1_8

(28) Sullivan, T. J.; Cosby, B. J.; Jackson, W. A. Target loads of atmospheric sulfur deposition for the protection and recovery of acid-sensitive streams in the Southern Blue Ridge Province. *Journal of Environmental Management* **2011**, 92, 2953–2960.

(29) Sullivan, T. J.; Cosby, B. J.; Driscoll, C, T.; McDonnell, T. C.; Herlihy, A. T.; Burns, D. A. Target loads of atmospheric sulfur and nitrogen deposition for protection of acid sensitive aquatic resources in the Adirondack Mountains, New York, *Water Resources Research* **2012**, 48, W01547, DOI: 10.1029/2011WR011171

(30) Fakhraei, H.; Driscoll, C. T.; Renfro, J. R.; Kulp, M. A.; Blett, T. F.; Brewer, P. F.; Schwartz, J. S. Critical loads and exceedances for nitrogen and sulfur atmospheric deposition in Great Smoky Mountains National Park, United States. *Ecosphere* **2016**, 7(10): e01466; DOI: 10.1002/ecs2.1466

(31) US-Canada. Memorandum of intent on transboundary air pollution. Report of the impact assessment working group I, Section 3: Aquatic effects, **1983**.

(32) Grigal, D. F. The concept of target and critical loads. Topical Report EPRI EN-7318, Electric Power Research Institute, Palo Alto, USA, **1991**; 30 pp.

(33) Jeffries, D. S.; Lam, D. C. L. 1993. Assessment of the effect of acidic deposition on Canadian lakes: Determination of critical loads for sulphate deposition. *Water Science & Technology* **1993**, 28(3–5), 183–187.

(34) Foster, K. R.; McDonald, K.; Eastlick, K. Development and application of critical, target and monitoring loads for the management of acid deposition in Alberta, Canada. *Water, Air, & Soil Pollution Focus* **2001**, 1, 135–151.

(35) Porter, E.; Blett, T.; Potter, D. U.; Huber, C. Protecting resources on federal lands: Implications of critical loads for atmospheric deposition of nitrogen and sulfur. *BioScience* **2005**, 55(7): 603–612.

(36) Curtis, C, J.; Posch, M.; Aherne, J.; Fölster, J.; Forsius, M.; Larssen, T.; Moldan, F. Assessment of critical loads of acidity and their exceedances for European lakes. In *Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems*; De Vries, W., Hettelingh, J.-P., Posch, M., Eds.; Springer: Dordrecht, **2015**; pp. 439–462; DOI: 10.1007/978-94-017-9508-1_17

(37) Forsius, M.; Malin, V.; Mäkinen, I.; Mannio, J.; Kämäri, J.; Kortelainen, P.; Verta, M. Finnish lake acidification survey: design and random selection of lakes. *Environmetrics* **1990**, 1, 79–99.

(38) Posch, M.; Slootweg, J.; Hettelingh, J.-P., Eds. European critical loads and dynamic modelling: CCE Status Report 2005. Coordination Centre for Effects, MNP Report 259101016, Bilthoven, Netherlands, ISBN 90-6960-128-1, **2005**; 171 pp. www.rivm.nl

(39) Skjelkvåle, B. L.; Mannio, J.; Wilander, A.; Andersen, T. Recovery from acidification of lakes in

Finland, Norway and Sweden 1990-1999. *Hydrology and Earth System Sciences* **2001**, 5(3), 327-337.

(40) Vuorenmaa, J.; Forsius, M. Recovery of acidified Finnish lakes: trends, patterns and dependence of catchment characteristics. *Hydrology and Earth System Sciences* **2008**, 12, 465–47.

(41) Monteith, D. T.; Evans, C. D.; Henrys, P. A.; Simpson, G. L.; Malcolm, I. A. Trends in the hydrochemistry of acid-sensitive surface waters in the UK 1988-2008. *Ecological Indicators* **2014**, 37, 287-303.

(42) Helliwell, R. C.; Jenkins, A.; Ferrier, R. C.; Cosby, B. J. Modelling the recovery of surface water chemistry and the ecological implications in the British uplands. *Hydrology and Earth System Sciences* **2003**, 7, 456–466.

(43) Posch, M.; Aherne, J.; Forsius, M.; Fronzek, S.; Veijalainen, N. Modelling the impacts of European emission and climate change scenarios in acid-sensitive catchments in Finland. *Hydrology and Earth System Sciences* **2008**, 12, 449–463; DOI: 10.5194/hess-12-449-2008

(44) Aherne, J.; Posch, M.; Forsius, M.; Vuorenmaa, J.; Tamminen, P.; Holmberg, M.; Johansson, M. Modelling the hydro-geochemistry of acid-sensitive catchments in Finland under atmospheric deposition and biomass harvesting scenarios. *Biogeochemistry* **2008**, 88(3), 233–256; DOI: 10.1007/s10533-008-9206-7

(45) Larssen, T.; Cosby, B. J.; Lund, E.; Wright, R. F. Modeling future acidification and fish populations in Norwegian surface waters. *Environmental Science & Technology* **2010**, 44, 5345–5351.

(46) Moldan, F.; Cosby, B. J.; Wright, R. F. Modeling past and future acidification of Swedish lakes. *Ambio* **2013**, 42, 577–586.

(47 Ferrier, R. C.; Wright, R. F.; Jenkins A.; Barth, H. Predicting recovery of acidified freshwaters in Europe and Canada: an introduction. *Hydrology and Earth System Sciences* **2003**, 7(4): 431–435; DOI: 10.5194/hess-7-431-2003

(48) Wright, R. F.; Dillon, P. J. Role of climate change in recovery of acidified surface waters. *Hydrology and Earth System Sciences* **2008**, 12(2): 333–335; DOI: 10.5194/hess-12-333-2008

(49) Tominaga, K.; Aherne, J.; Watmough, S. A.; Alveteg, M.; Cosby, B. J.; Driscoll, C. T.; Posch, M. Voyage without constellation: evaluating the performance of three uncalibrated process-oriented models. *Hydrology Research* **2009**, 40(2-3): 261–272; DOI: 10.2166/nh.2009.085

(50) Forsius, M.; Alveteg, M.; Jenkins, A.; Johansson, M.; Kleemola, S.; Lükewille, A.; Posch, M.; Sverdrup, H.; Walse, C. MAGIC, SAFE and SMART model applications at integrated monitoring sites: Effects of emission reduction scenarios. *Water, Air, & Soil Pollution* **1998**, 105: 21–30; DOI: 10.1023/A:1005099930089

(51) Wright, R. F.; Aherne, J.; Bishop, K.; Camarero, L.; Cosby, B. J.; Erlandsson, M.; Evans, C. D.; Forsius, M.; Hardekopf, D. W.; Helliwell, R.; Hruska, J.; Jenkins, A.; Kopáček, J.; Moldan, F.; Posch, M.; Rogora, M. Modelling the effect of climate change on recovery of acidified freshwaters: Relative sensitivity of individual processes in the MAGIC model. *Science of the Total Environment* **2006**, 365, 154–166; DOI: 10.1016/j.scitotenv.2006.02.042

(52) Aherne, J.; Posch, M.; Forsius, M.; Lehtonen, A., Härkönen, K. Impacts of forest biomass removal on soil nutrient status under climate change: a catchment-based modelling study for Finland. *Biogeochemistry* **2012**, 107(1–3), 471–488; DOI: 10.1007/s10533-010-9569-4

(53) Cosby, B. J.; Hornberger, G. M.; Galloway, J. N.; Wright, R. F. Modeling the effects of acid deposition: Assessment of a lumped parameter model of soil water and streamwater chemistry. *Water Resources Research* **1985**, 21(1), 51-63.

(54) Cosby, B. J.; Ferrier, R. C.; Jenkins, A.; Wright, R. F. Modelling the effects of acid deposition: refinements, adjustments and inclusion of nitrogen dynamics in the MAGIC model. *Hydrology and Earth System Sciences* **2001**, 5(3), 499-517.

(55) Aherne, J.; Posch, M.; Forsius, M.; Vuorenmaa, J.; Tamminen, P.; Holmberg, M.; Johansson, M. Modelling the hydro-geochemistry of acid-sensitive catchments in Finland under atmospheric deposition and biomass harvesting scenarios. *Biogeochemistry* **2008**, 88(3): 233–256; DOI: 10.1007/s10533-008-9206-7

(56) Schöpp, W.; Posch, M.; Mylona, S.; Johansson, M. Long-term development of acid deposition (1880–2030) in sensitive freshwater regions in Europe. *Hydrology and Earth System Sciences* **2003**, 7(4), 436–446; DOI: 10.5194/hess-7-436-2003

(57) Simpson, D.; Benedictow, A.; Berge, H.; Bergström, R.; Emberson, L. D.; Fagerli, H.; Flechard, C. R.; Hayman, G. D.; Gauss, M.; Jonson, J. E.; Jenkin, M. E.; Nyiri, A.; Richter, C.; Semeena, V. S.; Tsyro, S.; Tuovinen, J.-P.; Valdebenito, A.; Wind P. The EMEP MSC-W chemical transport model - technical description. *Atmospheric Chemistry and Physics* **2012**, 12, 7825–

7865.

(58) Hutchinson, N. J.; Holtze, K. E.; Munro, J. R.; Pawson, T. W. Modifying effects of life stage, ionic strength and postexposure mortality on lethality of H+ and Al to lake trout and brook trout. *Aquatic Toxicology* **1989**, 15(1), 1–25; DOI: 10.1016/0166-445X(89)90002-7

(59) Lien, L.; Raddum, G. G.; Fjellheim, A.; Henriksen, A. A critical limit for acid neutralizing capacity in Norwegian surface waters, based on new analyses of fish and invertebrate responses. *Science of the Total Environment* **1996**, 177, 173-193.

(60) Henriksen, A.; Posch, M.; Hultberg, H.; Lien, L. Critical loads of acidity for surface waters - Can the ANClimit be considered variable? *Water, Air, & Soil Pollution* **1995**, 85, 2419-2424; DOI: 10.1007/BF01186196

(61) Fakhraei, H.; Driscoll, C. T. Proton and aluminum binding properties of organic acids in surface waters of the northeastern U.S. *Environmental Science & Technology* **2015**, 49: 2939–2947; DOI: 10.1021/es504024u

(62) Lydersen, E.; Larssen, T.; Fjeld, E. The influence of total organic carbon (TOC) on the relationship between acid neutralizing capacity (ANC) and fish status in Norwegian lakes. *Science of the Total Environment* **2004**, 326, 63–69.

(63) Forsius, M.; Moldan, F.; Larssen, T.; Posch, M.; Aherne, J.; Lund, E.; Wright, R. F.; Cosby, B. J. National-scale dynamic model applications for Nordic lake catchments. In *Critical Loads and Dynamic Risk Assessments: Nitrogen, Acidity and Metals in Terrestrial and Aquatic Ecosystems*; De Vries, W., Hettelingh, J.-P., Posch, M., Eds.; Springer: Dordrecht, **2015**; pp. 463–484; DOI: 10.1007/978-94-017-9508-1_18

(64) Reuss, J. O.; Johnson, D. W. *Acid Deposition and the Acidification of Soils and Waters.* Springer: Berlin, **1986**; vi+119 pp.