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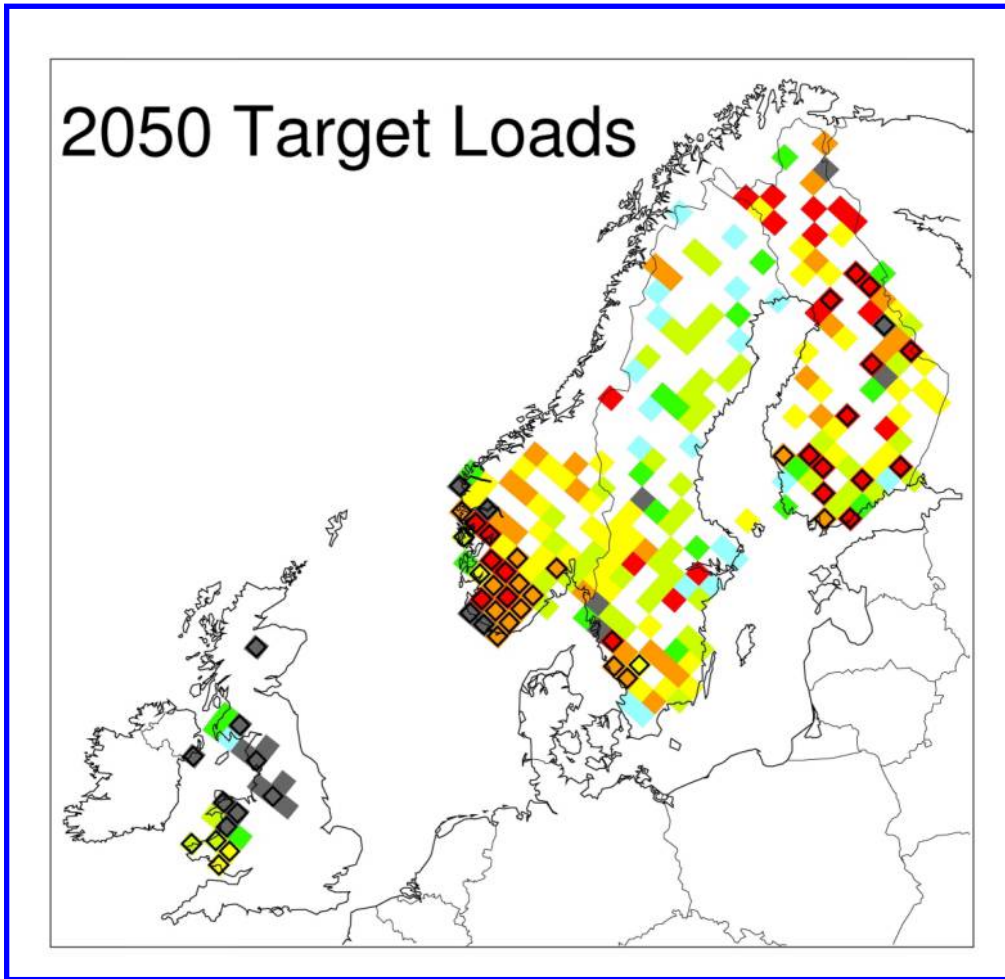
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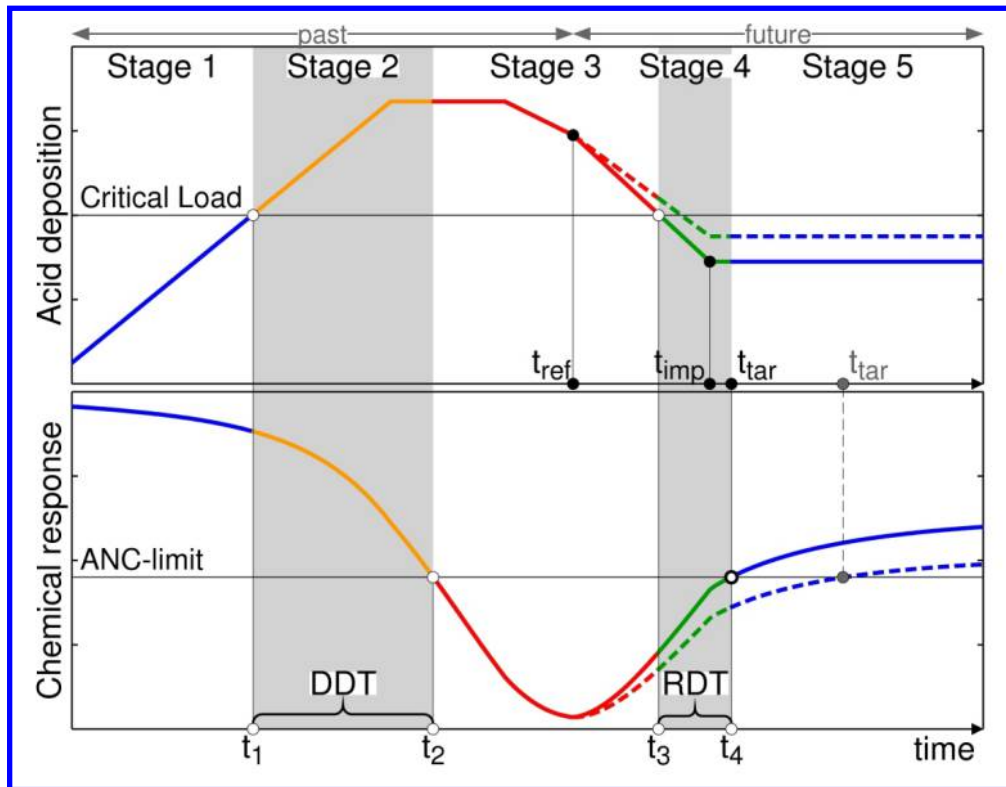


Figure 1: Generalised past and future development stages (indicated by different colours) of acidic deposition and lake chemical criterion response (here: ANC) in comparison to the critical chemical value (ANC-limit) and the critical load derived from it (i.e., the determination of critical load is based 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 (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).

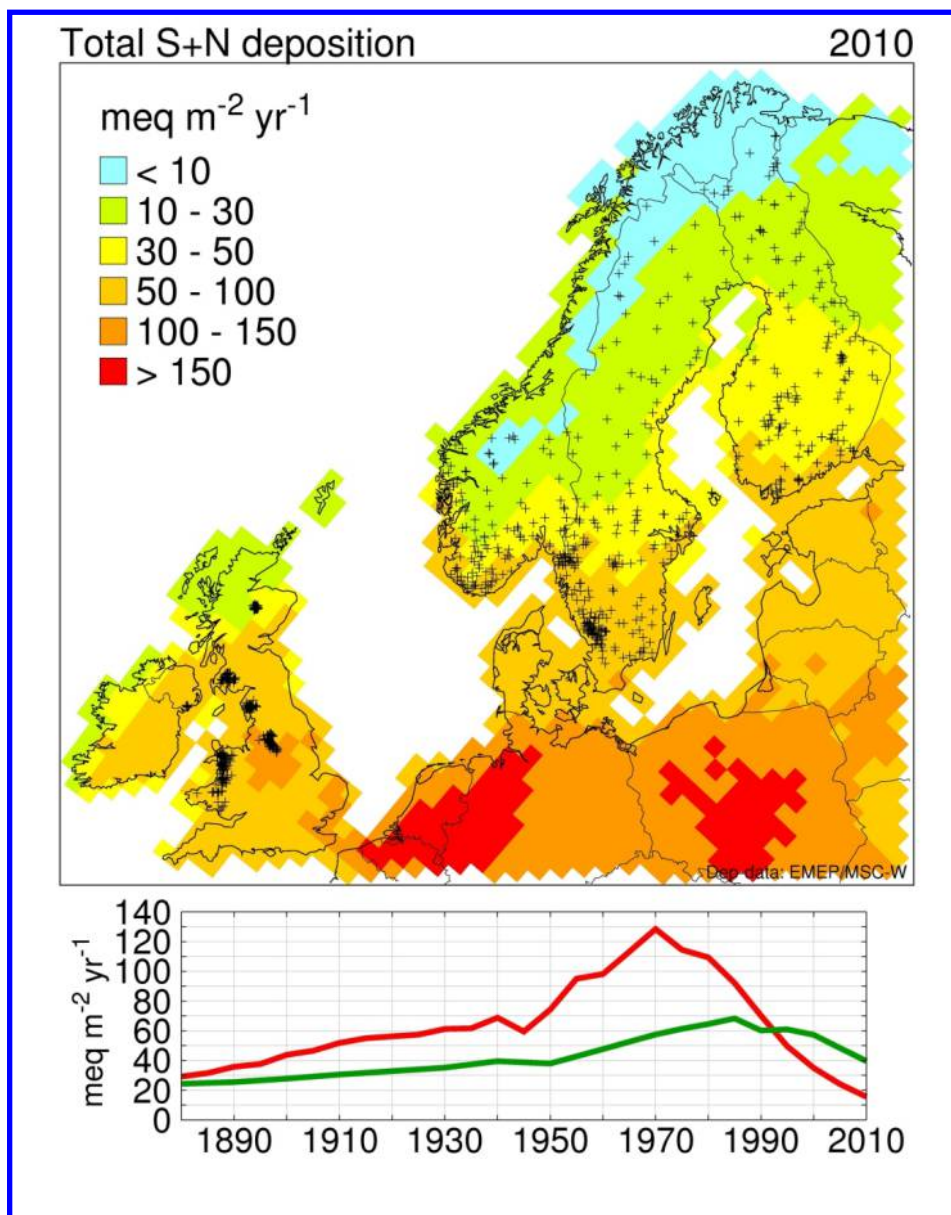


Figure 2: Location of the study sites used for dynamic modelling ($n = 848$; black crosses) overlaid upon modelled total sulphur (S) plus nitrogen (N) deposition in 2010 ($\text{meq m}^{-2} \text{ yr}^{-1}$) on the EMEP $50 \text{ km} \times 50 \text{ km}$ grid (for further details on the EMEP model 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.

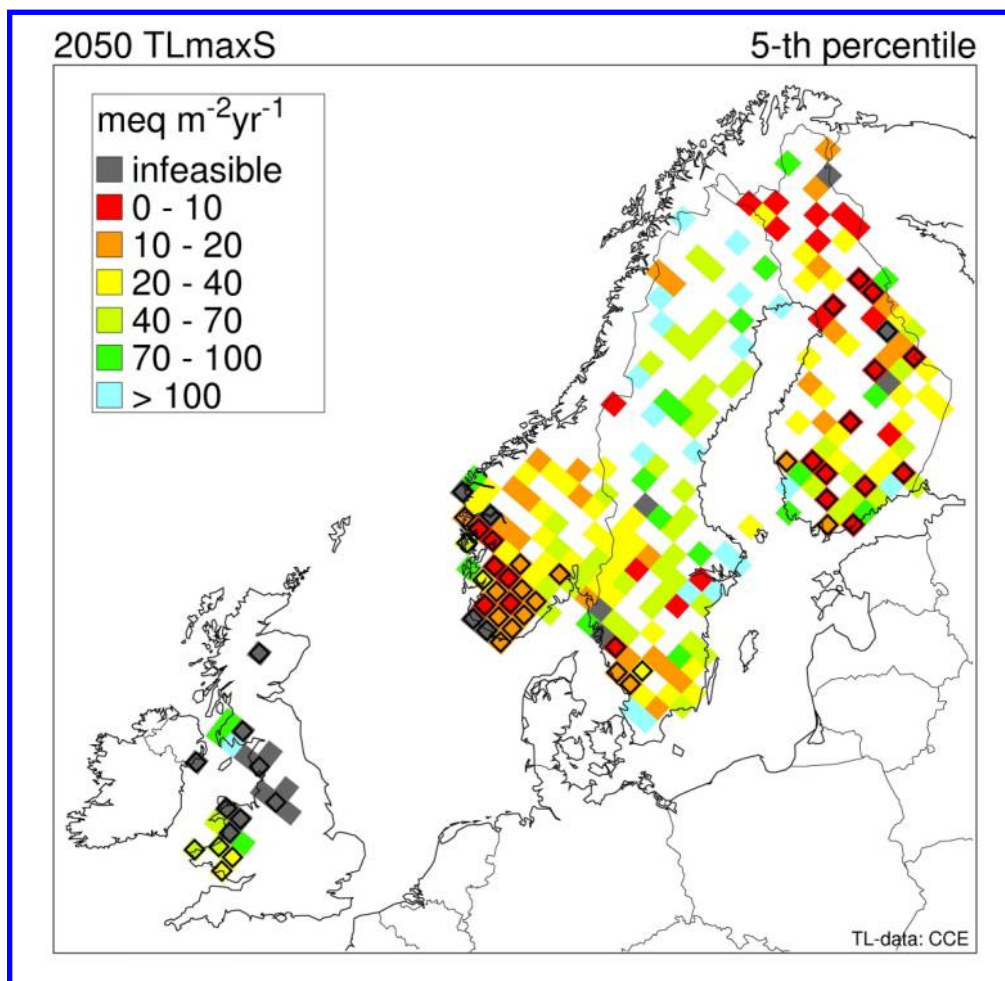


Figure 3: Map of the 5-th percentile of the 2050 maximum target load of sulphur (TLmaxS) on the EMEP 50 km × 50 km grid for 848 catchments in Finland, Norway, Sweden and the United Kingdom. Note: TLmaxS was set to the maximum critical load of S (CLmaxS) where TLmaxS > CLmaxS (i.e., the critical load 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).

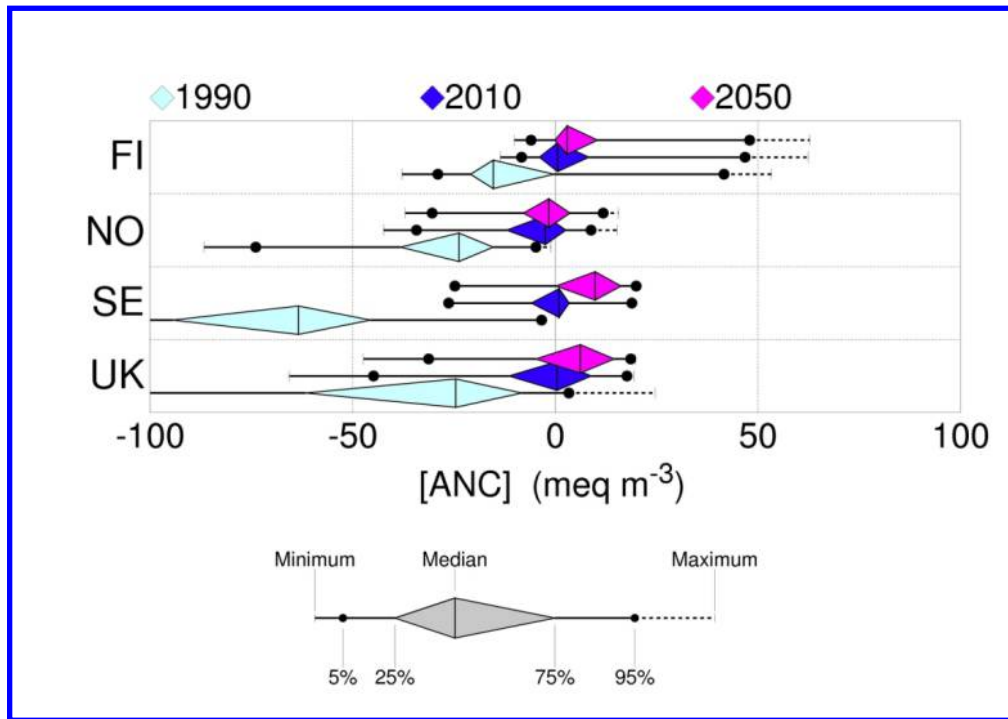


Figure 4: Percentile statistics ('diamond plots') of simulated annual average lake acid neutralising capacity (ANC) in 1990, 2010 and 2050 in Finland (FI), Norway (NO), Sweden (SE) and the United Kingdom (UK). Data are only shown for lakes for which target loads were determined (FI = 24, NO = 56, SE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

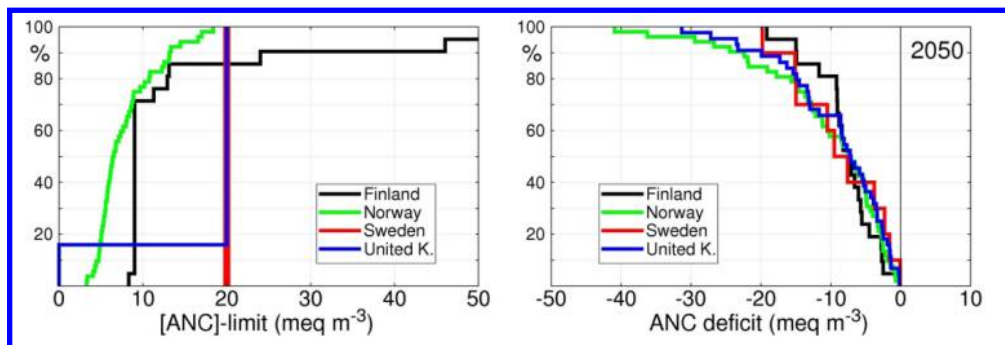


Figure 5: Left: Cumulative distribution functions of the ANC-limits for the lakes with target loads in Finland (21), Norway (52), Sweden (10) and the United Kingdom (44). Right: Inverse cumulative distribution functions of the 'ANC deficit' in 2050 for the same lakes in those countries. The ANC deficit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

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

3
4
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18
19
20 **Abstract**

21 The target load concept is an extension of the critical load concept of air pollution inputs to
22 ecosystems. The advantage of target loads over critical loads is that one can define the
23 deposition *and* the point in time (target year) when the critical (chemical) limit is no longer
24 violated. This information on the timing of recovery requires dynamic modelling. Using a
25 well-documented dynamic model, target loads for acidic deposition were determined for 848
26 surface waters across Finland, Norway, Sweden and the United Kingdom for the target year
27 2050. In the majority of sites (n = 675), the critical ANC-limit was predicted to be achieved
28 by 2050; however, for 127 sites target loads were determined. In addition, 46 sites were
29 infeasible, i.e., even a deposition reduction to zero would not achieve the limit by 2050. The
30 average maximum target load for sulphur was 38% lower than the respective critical load
31 across the study lakes (n = 127). Target loads on a large regional scale can inform effects-
32 based emission reduction policies; the current assessment suggests that reductions beyond the
33 Gothenburg Protocol are required to ensure surface water recovery from acidification by
34 2050.

35

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

38

39

40 **1. Introduction**

41 During the 1970s it was recognized that surface waters in large parts of Europe and North
42 America were being influenced by air pollution, i.e., acidic deposition, owing to
43 anthropogenic emissions of sulphur (S) and nitrogen (N) oxides.^{1,2} Shortly thereafter,
44 empirical³ and steady-state⁴⁻⁶ models were developed and applied to predict the impacts of
45 acidic deposition on surface waters. In concert, it was recognised that time-dependent
46 processes could buffer (delay) ecosystem (soil and surface water) response to acidic
47 deposition. The incorporation of these processes required time-dependent or ‘dynamic’
48 modelling frameworks. The earliest dynamic simulation models incorporated established
49 relationships from soil and water chemistry to predict the most likely effects of acidic
50 deposition on surface waters.⁷ Ultimately, these dynamic models provided a quantitative
51 framework to assess whether (and how quickly) a decrease in acidic stress would result in a
52 recovery of ecosystems.⁸ Since the 1980s, several dynamic (hydro-chemical) models have
53 been developed and extensively applied at site-specific and regional scales to predict changes
54 in soil and surface water chemistry due to acidic deposition.⁹⁻¹⁵ Moreover, dynamic models
55 can provide a quantitative estimate of the time lag between a reduction in deposition and the
56 attainment of ‘acceptable’ ecosystem status (based on a threshold, or ‘critical value’, for a
57 specified chemical criterion, e.g., surface water pH=6.0). This time lag has been denoted as
58 the damage time lag¹⁶ or recovery delay time.¹⁷⁻¹⁹

59

60 The assessment of impacts of acidic deposition on terrestrial and aquatic ecosystems has
61 supported policies to reduce anthropogenic S and N emissions. In Europe, the critical loads
62 approach is widely accepted as the basis for negotiating effects-based control strategies for air
63 pollution. A critical load is defined as ‘a quantitative estimate of an exposure to one or more
64 pollutants below which significant harmful effects on specified sensitive elements of the
65 environment do not occur according to present knowledge’.²⁰ The approach is based on
66 setting a critical limit for a chemical criterion (e.g., Acid Neutralising Capacity [ANC]) to
67 protect a specified biological indicator for a chosen receptor ecosystem (e.g., fish species for
68 surface waters, or tree roots in forest soils), and via inverse modelling a deposition (the
69 critical load) is derived to ensure the limit is not violated and thus ‘harmful effects’
70 avoided.²¹⁻²³ Critical loads have been used in the negotiations of several protocols to the
71 United Nations Economic Commission for Europe’s (UNECE²⁴) Convention on Long-range

72 Transboundary Air Pollution (LRTAP) and the European Union's National Emission Ceilings
73 (NEC) Directive²⁵ and its revision.

74

75 However, by definition, critical loads are steady-state quantities and as such they do not
76 provide information on the time involved for a system (e.g., surface water) to reach a certain
77 chemical (or biological) state. Dynamic models are needed to calculate recovery times under
78 prescribed emission reductions. As such, dynamic modelling has also become an important
79 part of the effects-oriented work under the LRTAP Convention.²⁶ If a desired chemical state
80 of a surface water is defined for a given year, dynamic models can be used in an inverse mode
81 to compute the deposition path leading to that desired state (if feasible). Such a deposition is
82 called a target load, and the year in which the desired state is to be reached is called the target
83 year.^{18,19} There have been few published studies describing the use of target loads in
84 Europe^{16,17,27} and fewer for surface waters.²⁸⁻³⁰ It is important to note that in North America
85 the term 'target load' has also been used to refer to a 'target' deposition, determined by
86 political (or management) agreement, that can be higher or lower than the critical load³¹⁻³⁵,
87 often based on arbitrary interpretations of the impacts data rather than the avoidance of
88 specific deleterious ecological effects.³²

89

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

98

99 **2. Dynamic Modelling and Target Loads**

100 With critical loads, i.e., in the steady-state situation, only two cases can be distinguished when
101 evaluated against deposition: (1) deposition is below the critical load, or (2) deposition is
102 greater than the critical load, i.e., there is an exceedance of the critical load. In the first case
103 there is no (apparent) problem, i.e., no reduction in deposition is deemed necessary. In the
104 second case there is, by definition, an increased risk of damage to the ecosystem. Thus, a
105 critical load serves as a warning as long as there is exceedance, since it indicates that
106 deposition should be reduced. However, it is often (implicitly) assumed that reducing
107 deposition to (or below) the critical load immediately removes the risk of 'harmful effects',
108 i.e., the critical chemical criterion (e.g., the ANC-limit) that links the critical load to the

109 (biological) effect, immediately attains a non-critical ('safe') value (and that there is
110 immediate biological recovery as well). However, the reaction of an ecosystem (or
111 catchment) to changes in deposition is delayed by (finite) buffers, e.g., the cation exchange
112 capacity of catchment soils. These buffer mechanisms can delay the attainment of the critical
113 chemical value, and it might take decades or even centuries, before steady state is reached.
114 These finite buffers are not included in the critical load formulation, since they do not
115 influence the steady state, but only the time to reach it.

116

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

121

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

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

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

133 **Stage 4:** Deposition is below the CL, but the chemical criterion is still violated and thus (full)
134 recovery has not yet occurred. The time between the first non-exceedance of the CL and the
135 subsequent non-violation of the criterion is termed the *Recovery Delay Time* ($RDT=t_4-t_3$).

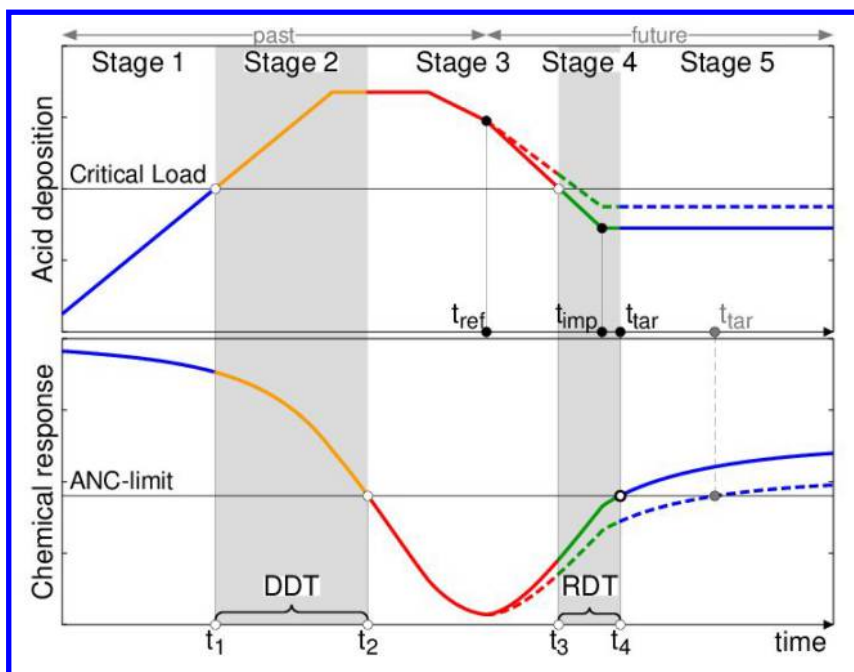
136 Note: RDT is not necessarily the same (or even similar) to DDT due to hysteresis effects in
137 certain (soil) processes (e.g., cation exchange); the schematic is purely for illustration and
138 does not reflect the relative temporal processes.

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

144

145 In addition to the delay in chemical recovery, there is likely to be a further delay before the
 146 'original' biological state is reached, i.e., even if the chemical criterion is met, it will take
 147 time before biological recovery is achieved (if at all).

148



149

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

160

161 The most straightforward use of a dynamic model is scenario analysis, i.e., first a future
 162 deposition scenario is assumed, and then the (chemical) consequences for the ecosystem (e.g.,
 163 lake) are evaluated. A target load, on the other hand, is the deposition path that ensures that a
 164 prescribed value of the chemical criterion is achieved in a given year. Here we define a target
 165 load as a deposition path characterised by three numbers (years): (i) the reference year, (ii) the
 166 implementation year, and (iii) the target year (see Figure 1). The *reference year*, t_{ref} , is the
 167 year (time) up to which the (historic) deposition path is given and cannot be changed. The
 168 *implementation year*, t_{imp} , is the year by which all reduction measures, needed to reach the
 169 final deposition (the target load), are implemented. Between the reference year and the

170 implementation year depositions are assumed (assured) to change linearly (see Figure 1).
171 Finally, the *target year* is the year in which the critical chemical criterion (e.g., the ANC-
172 limit) is met (for the first time). The above three years define a unique deposition path, the
173 final value of which is referred to as a target load. The earlier the target year, the lower the
174 target load (at sites where the chemical criterion is violated – for other sites a target load is
175 not relevant), since higher deposition reductions are needed to achieve the desired status
176 within a shorter timeframe (see Figure 1). In extreme cases, a target load might not exist at
177 all, i.e., even reduction to zero deposition in the implementation year will not result in the
178 desired ecosystem status within the prescribed time; in this instance the target load is termed
179 ‘infeasible’. For more information on target loads and related topics see Posch et al.²², Jenkins
180 et al.¹⁹ or Bonten et al.²⁷

181

182 **3. Materials and Methods**

183 The current study focused on surface waters (lakes and streams) with long-term observations
184 of chemistry in acid-sensitive regions of Europe³⁶, i.e., Finland, Norway, Sweden, and the
185 United Kingdom. In general, these sites are considered to be sentinel indicators of
186 acidification impacts, and their recovery is assumed to reflect wider ecosystem acidification
187 recovery across the entire study region; as such, they are well suited for the determination of
188 regionally representative target loads. All surface waters were part of routine acidification
189 monitoring networks since the 1980s and 1990s, typically located in regions with acid-
190 sensitive geology. For example, the Finnish acidification monitoring network, maintained by
191 the Finnish Environment Institute, consisted of 163 lakes located throughout Finland,
192 subjectively chosen by expert judgement from a national acidification survey during 1987 for
193 use in acidification assessments.³⁷ Similarly, the Norwegian study lakes (n = 131) were a
194 subset of the national monitoring programme, confined to lakes south of 62.5° latitude, with
195 observations suitable for dynamic modelling. The study sites are predominantly small acid-
196 sensitive headwater lakes and streams, with low base cation concentrations, low alkalinity and
197 low (charge balance) ANC.³⁸ All surface waters have been widely used in acidification
198 assessments evaluating long-term trends in surface water chemistry³⁹⁻⁴¹, and the prediction of
199 future chemistry using dynamic (hydro-chemical) models, specifically MAGIC.⁴²⁻⁴⁶ The study
200 sites have played a central role in European-scale projects, such as ‘Recover:2010’⁴⁷ and
201 ‘Eurolimpacs’⁴⁸, focused on model simulations of surface water response to European
202 emissions reduction policies. The process limitations and predictive uncertainty of MAGIC in
203 isolation, and compared with other models, e.g., PnET-BGC (photosynthesis and
204 evapotranspiration-biogeochemistry), SAFE (soil acidification in forest ecosystems), and
205 VSD (very simple dynamic), have been widely published.^{12,15,49,50} Similarly, the influence of
206 climate change on model predictions for MAGIC have been widely assessed^{43,46,51,52}. As such,

207 herein we focus on the determination of target loads using MAGIC, which (hitherto for) have
208 not been reported for the study sites and refer the reader to previous publications for detailed
209 information regarding model calibration and process uncertainty for MAGIC. Nonetheless,
210 we provide a brief overview of MAGIC, its application, calibration and simulation for the
211 study sites, specifically with respect to target loads.

212

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

232

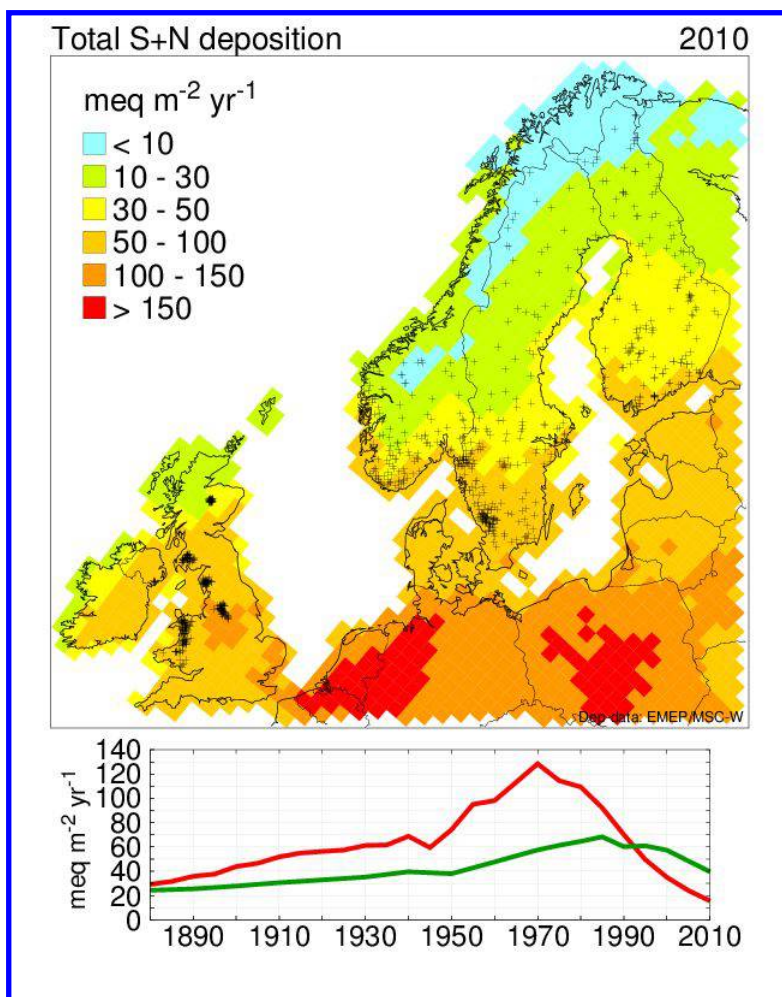
233 In the current study, the application of MAGIC (by national experts) across the study lakes
234 followed a common (prescribed) procedure as described in Posch et al.³⁸; for further details
235 on the application and calibration of MAGIC see, for example, Aherne et al.⁴⁴ In brief, all
236 catchments were represented by one soil compartment receiving deposition and releasing
237 discharge to the lake compartment. The soil compartment represented the aggregated horizons
238 of the catchment soils (mineral and organic), with exchangeable base cation data taken from
239 national and focused surveys.³⁸ Simulations were carried out using an annual time-step, with a
240 number of simplifying assumptions applied consistently across all study lakes. Discharge
241 volume and flow routing within the catchments were not varied; discharge was described
242 using long-term means with 100% routed to the lake. Detailed process-oriented N dynamics
243 were not modelled, i.e., the coefficient for in-lake N retention was set to a similar value for all

244 lakes³⁶, and terrestrial N retention was set to match observed lake concentrations. To account
245 for uncertainty in a number of the fixed parameters (lake retention, soil depth, soil bulk
246 density, cation exchange capacity, etc.), a ‘fuzzy’ optimisation method was employed. Ten
247 calibrations were carried out for each study lake using MAGICOPT; for each simulation the
248 fixed parameters were randomly varied within specified uncertainty bands ($\pm 10\%$). Uptake of
249 base cations from forested catchments were modelled using a simplified regional sequence,
250 based on a regional average planting date, constant nutrient concentrations (from literature),
251 and species composition and coverage from national forest inventories (for further details see,
252 e.g., Aherne et al.^{52,55} for Finland and Moldan et al.⁴⁶ for Sweden).

253

254 In the current study, MAGIC was calibrated to 848 lakes (see Figure 2) across Finland
255 (FI=163), Norway (NO=131), Sweden (SE=234), and the United Kingdom (UK=320),
256 spanning a range in deposition (S plus N) from $<10 \text{ meq m}^{-2} \text{ yr}^{-1}$ in northern Sweden and
257 Finland to $150 \text{ meq m}^{-2} \text{ yr}^{-1}$ in the South Pennines, UK (Figure 2). Historic and future surface
258 water chemistry for each lake were simulated for the period 1880–2100 under modelled
259 anthropogenic S and N deposition⁵⁶ from the EMEP model.⁵⁷ Future lake chemical recovery
260 under the Gothenburg Protocol (which came into force in 2010) was evaluated using charge
261 balance ANC (defined as the difference between the sum of the concentrations of base cations
262 and strong acid anions); lake ANC is an established acidification indicator, as it is strongly
263 indicative of biological recovery.^{58,59} In addition, ANC is the most widely used chemical
264 criterion in critical load calculations for surface waters.⁶⁰ Target loads were estimated for the
265 target year 2050, with the implementation year 2020, i.e., the year in which deposition
266 reductions beyond the Gothenburg Protocol (year 2010) are fully implemented to ensure
267 attainment of the critical chemical criterion by the target year.³⁸ While dynamic modelling
268 was carried out on every study site ($n = 848$), target loads were only calculated for each
269 surface water that did not meet the specified critical chemical criterion (ANC-limit) by 2050
270 under the Gothenburg Protocol, i.e., those lakes that still violated ANC-limits or with a
271 recovery delay (Note: lakes were the only study sites that still violated the ANC-limit). The
272 specified ANC-limit varied among countries (based on national management objectives); all
273 Swedish lakes had a fixed value of 20 meq m^{-3} , similarly the UK surface waters (lakes and
274 streams) had a value of 20 meq m^{-3} , except for a small number of naturally acidic sites that
275 had a limit of zero ($n=21$; 6.5%). Organic acids can act as strong acid anions reducing the acid
276 neutralizing (buffering) capacity of a lake to incoming acidity⁶¹; to accommodate this, Finland
277 and Norway used an organic acid adjusted ANC-limit⁶², which, for example, resulted in an
278 average ANC-limit of 14 meq m^{-3} for the Finnish lakes.

279



280

281 **Figure 2:** Location of the study sites used for dynamic modelling ($n = 848$; black crosses) overlaid
 282 upon modelled total sulphur (S) plus nitrogen (N) deposition in 2010 ($\text{meq m}^{-2} \text{yr}^{-1}$) on the EMEP 50
 283 $\text{km} \times 50 \text{ km}$ grid (for further details on the EMEP model see Simpson et al.⁵⁷). The temporal
 284 development (1880–2010) of the total S (red) and N (green) deposition averaged over the 848 study
 285 sites is also shown.

286

287 Target load functions were estimated for each calibrated surface water that did not meet the
 288 specified critical chemical criterion (ANC-limit) by 2050 under the Gothenburg Protocol, i.e.,
 289 every pair of N and S deposition that met the ANC-limit in 2050 under further (beyond
 290 Gothenburg) emission reductions defined a target load function of acidity (TLF), similar to a
 291 critical load function²¹ (see also Supporting Information for further details) for each study
 292 lake (catchment). The piece-wise linear function in the (N, S) deposition-plane is delineated
 293 by the maximum target load of S, TL_{maxS} (for N deposition = 0) and the maximum target
 294 load of N, TL_{maxN} (for S deposition = 0).

295

296 **4. Results and Discussion**

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

303

304 Dynamic model simulations were carried out for 848 surface waters, but target load
 305 calculations were only necessary for 173 lakes (Table 1). The simulated water chemistry for
 306 the target year 2050 was predicted to be greater than (or equal to) the chemical criterion
 307 (ANC-limit) for 675 surface waters. Target loads, i.e., loads below the respective critical
 308 loads, were determined for 24 lakes in Finland, 56 in Norway, 14 in Sweden and 79 in the
 309 United Kingdom. Of these 173 lakes, 46 were ‘infeasible’ (Table 1), i.e., even reducing
 310 anthropogenic deposition to zero by 2020 would not result in an ANC greater than or equal to
 311 the ANC-limit in 2050. In general, infeasible lakes occurred in < 3% of the study sites per
 312 country; however, in the United Kingdom, infeasible lakes occurred in ~11% (n = 35) of the
 313 study sites likely reflecting their higher cumulative historic deposition (Figure 2). Neglecting
 314 infeasible sites, ‘true’ target loads were determined for 127 lakes (Table 1), 21 in Finland, 52
 315 in Norway, 10 in Sweden and 44 in the United Kingdom; the highest proportion occurred in
 316 Norway (40%) followed by Finland (13%).

317

318 The average TLmaxS (see Supporting Information) per country ranged from 7.5 meq m⁻² yr⁻¹
 319 (Finland) to 38.9 5 meq m⁻² yr⁻¹ (United Kingdom). Note, for all study sites the maximum
 320 critical load of S (CLmaxS) was also computed as the steady-state solution of the dynamic
 321 model; compared with the average CLmaxS, the average TLmaxS was 53% lower in Finland,
 322 40% in Norway, 20% in Sweden and 36% in the United Kingdom. Across all lakes, average
 323 TLmaxS (24.1 meq m⁻² yr⁻¹; n = 127) was 38% lower than the respective CLmaxS (39.1 meq
 324 m⁻² yr⁻¹; Table 1).

325

326 **Table 1:** Number of lakes in each country with dynamic model (DM) simulations, divided into the
 327 number of lakes for which the critical load is sufficient to achieve the ANC-limit in 2050 (ANC-2050),
 328 number of infeasible sites (INF) and ‘true’ target loads (TL). Also given are the averages of TLmaxS
 329 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 |

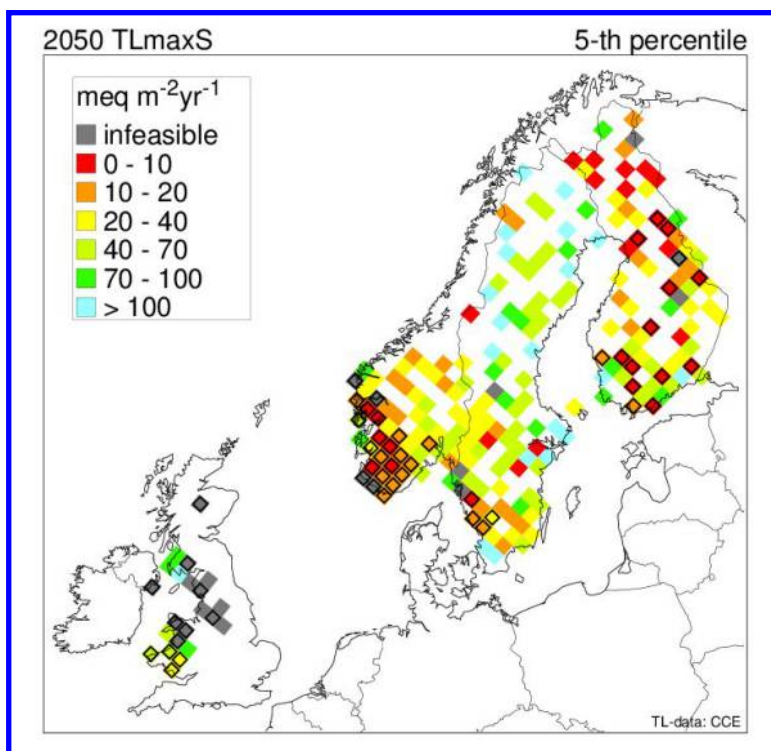
| | | | | | | | |
|-----|---------------------|-----|-----|----|-----|-------|-------|
| 330 | Sweden (SE) | 234 | 220 | 4 | 10 | 19.91 | 25.02 |
| | United Kingdom (UK) | 320 | 241 | 35 | 44 | 38.87 | 61.72 |
| | Sum/Average | 848 | 675 | 46 | 127 | 24.11 | 39.07 |

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

332

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

341

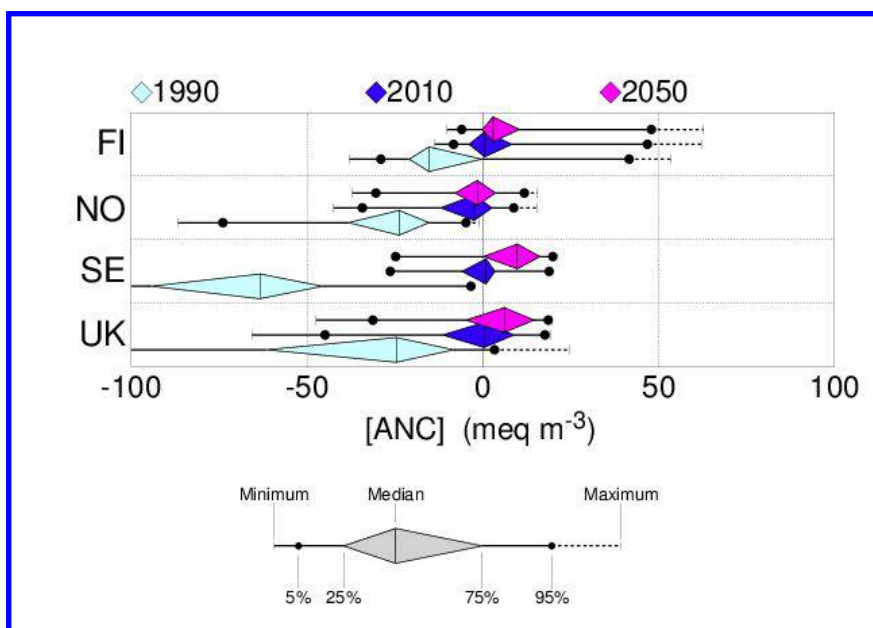


342

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

351

352 The key chemical variable of interest was ANC, as it is used as a chemical criterion linking
 353 water chemistry to the biological (fish) status of the lakes; as such, time series of ANC were
 354 simulated to illustrate the timing and rate of chemical changes during acidification and
 355 recovery. The general pattern of predicted ANC in the study lakes is similar in the four
 356 countries (Figure 4), driven by the deposition of S and N (Figure 2). The differences between
 357 the regions were due to proximity to emission sources, acid sensitivity of regions, differences
 358 in land use and the selected lakes.
 359



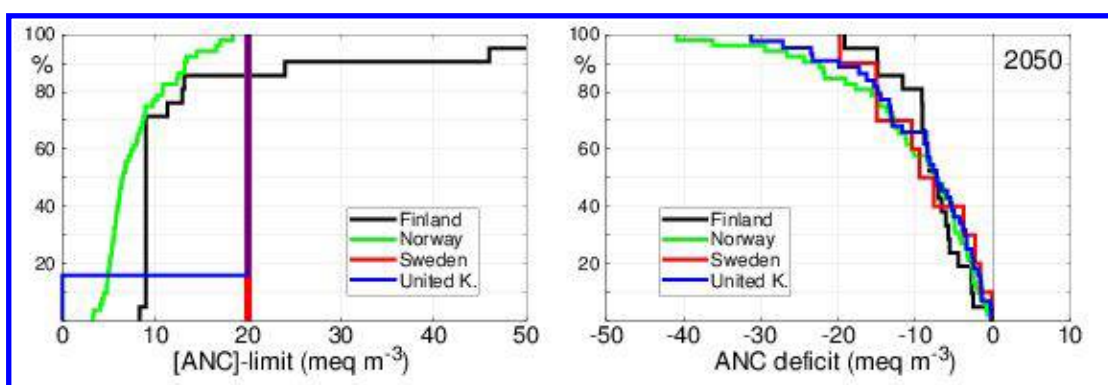
360
 361 **Figure 4:** Percentile statistics ('diamond plots') of simulated annual average lake acid neutralising
 362 capacity (ANC) in 1990, 2010 and 2050 in Finland (FI), Norway (NO), Sweden (SE) and the United
 363 Kingdom (UK). Data are only shown for lakes for which target loads were determined (FI = 24, NO =
 364 56, SE = 14, UK = 79; see columns 'INF' plus 'TL' in Table 1).

365
 366 Implementation of the Gothenburg Protocol resulted in a significant increase in ANC from
 367 1990 to 2010 (paired t-test, $p < 0.001$) in all four countries (average increase of 32.6 meq m^{-3} ;
 368 Figure 4) and is predicted to significantly improve by 2050 albeit by a smaller amount
 369 (average increase of 4.2 meq m^{-3} ; Figure 4). This is due to the fact that the deposition is kept
 370 at the 2010 level after that year, and that only (slow) improvements in the soil and water
 371 conditions, e.g., replenishment of base cation pools, will raise the ANC.⁶⁴ Surface waters in
 372 Sweden showed the greatest improvement in ANC between 1990 and 2010 (Figure 4) owing
 373 to the concentration of sensitive lakes in south-western Sweden^{1,14} and the large reduction in
 374 acidic deposition in that region (Figure 2).

375
 376 Despite the predicted improvements in ANC, ultimately, we are concerned with the 'distance'
 377 between the predicted chemical status of a lake and its desired status (note: the desired ANC-

378 limit is variable in all countries except in Sweden, see Figure 5a). A better characterisation of
 379 the ‘distance’ of a lake’s chemical status from the desired one is through the so-called ANC-
 380 deficit, i.e., the difference between simulated ANC (under the 2010 Gothenburg Protocol
 381 deposition) and the (lake-specific) ANC-limit. The inverse cumulative distributions of the
 382 ANC-deficit for the year 2050 were quite similar in the four countries up to the 80-th
 383 percentile (Figure 5b); notably a maximum deficit around 40 meq m⁻³ was simulated for some
 384 Norwegian lakes and 30 meq m⁻³ for some lakes in the United Kingdom. Implementation of
 385 target load depositions would ensure that all lakes reach their specified ANC-limit by 2050,
 386 i.e., all ANC deficits reach zero by 2050.

387



388

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

394

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

406

407 In the current study, model simulations have been conducted without consideration of future
 408 climate change, as the primary objective was to support emissions reduction policies

409 (irrespective of climate perturbations). Nonetheless, several (regional) studies have been
410 conducted using MAGIC that explore the direct and indirect effects of climate change on lake
411 chemistry^{43,51,52}. Although the (indirect) effects can be great for individual lakes, the overall
412 effects on lake chemistry are not huge, considering all other (model) uncertainties.⁵¹
413 Reductions of S and N deposition are the most important of determinants of future lake (acid)
414 status in European surface waters.

415

416 While target loads have been discussed and determined in Europe under the LRTAP
417 Convention, they have not been used explicitly to guide emission reduction policies. One
418 reason might be that it requires dynamic modelling – and thus more input data and expertise
419 to determine target loads – compared to critical loads that are ‘easily determined’ steady-state
420 quantities. However, lack of information on time needed for achieving the desired chemical
421 status under critical loads should ultimately encourage the determination of target loads to
422 provide policy advisors with guidance on the timing of ecosystem recovery. While
423 acidification is generally assumed to be ‘solved’ in Europe, there is growing recognition that
424 surface waters in some regions are still acidified²; the current assessment suggests that
425 emissions reductions beyond the Gothenburg Protocol are required to ensure surface water
426 recovery from acidification by 2050.

427

428

429 **Supporting Information**

430 Description of the Target Load Function (TLF)

431

432 **Notes**

433 The authors declare no competing financial interest.

434

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441

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