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Past, Present, and Future Exceedance of Critical Loads of Acidity for Surface Waters in Finland

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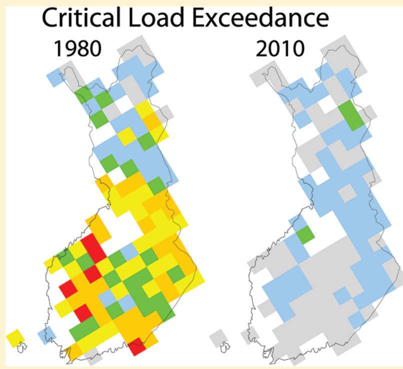
Supporting Information

ABSTRACT: A critical load is a deposition limit below which harmful effects for a given ecosystem do not occur; the approach has underpinned European sulfur (S) and nitrogen (N) effects-based emission reduction policies during the last two decades. Surface waters are an important resource in Finland, as such the development of models and determination of critical loads has played a central role in supporting their recovery from acidification or preservation of ecosystem health. Critical loads of acidity for Finnish lakes were determined using the steady-state First-order Acidity Balance (FAB) model in conjunction with comprehensive national surveys of surface waters (headwater lakes; n = 1066) and soils. In the 1980s almost 60% of the study lakes were exceeded, impacting brown trout and perch populations. The steep decline in emissions and acidic (S and N) deposition during the last two decades has reduced exceedance to <10%, and by 2020 exceedance is predicted to reach preindustrial (1880) levels. In concert with these reductions, chemical and biological recovery has been observed. The critical load approach has been instrumental in assessing impacts to surface waters in Finland and directing effects-based emission reduction policies.



INTRODUCTION

The impact of anthropogenic sulfur (S) and nitrogen (N) depositions on terrestrial and aquatic ecosystems is widely assessed using the critical load approach. In Europe the approach underpins effects-based emission reduction policies and is the key international instrument under the UNECE Convention on Long-range Transboundary Air Pollution (LRTAP) and the EU National Emission Ceilings (NEC) Directive.[1,2] 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'.[3] The approach is based on setting a critical chemical limit to protect a specified biological indicator for a chosen receptor ecosystem (e.g., fish species for surface waters), and via inverse modeling a deposition load (the critical load) is derived to ensure the limit is not violated and thus 'harmful effects' avoided.

Two steady-state models are widely used for calculating critical loads for surface waters:[4] the Steady-State Water Chemistry (SSWC) model and the First-order Acidity Balance (FAB) model. The SSWC model requires volume-weighted mean annual water chemistry and runoff volume to calculate critical loads of S acidity. The FAB model allows the simultaneous calculation of critical loads of acidifying S and N deposition similar to the Simple Mass Balance (SMB) model widely used for forest soil critical loads.[5,6] In addition to processes in the terrestrial catchment soils, such as uptake, immobilization, and denitrification, the FAB model includes in-lake retention of N and S. Both models have been applied to many regions of Europe,[7−10] North America,[11−13] and Asia.[14] In Fennoscandia, critical loads have been continually assessed owing to the preponderance of surface waters and regional concerns on acidification.[15] In Finland, there has been ongoing refinement of the modeling approach,[16,17] in conjunction with the assessment of critical loads of acidity.[8,18]

The critical loads approach has provided a direct link, or feedback, between effects-based science and the emissions policy process under the LRTAP Convention and EU negotiations through an integrated assessment modeling framework (RAINS/GAINS model[1]). This framework has allowed the development of cost-effective effects-based options for emission reductions of different air pollutants, guiding and motivating the international policy negotiations. Accordingly, S emission reductions have been recognized as one of the great environmental 'success stories'; S emissions have been reduced by ∼67% in Europe between 1980 and 2000, with reductions of

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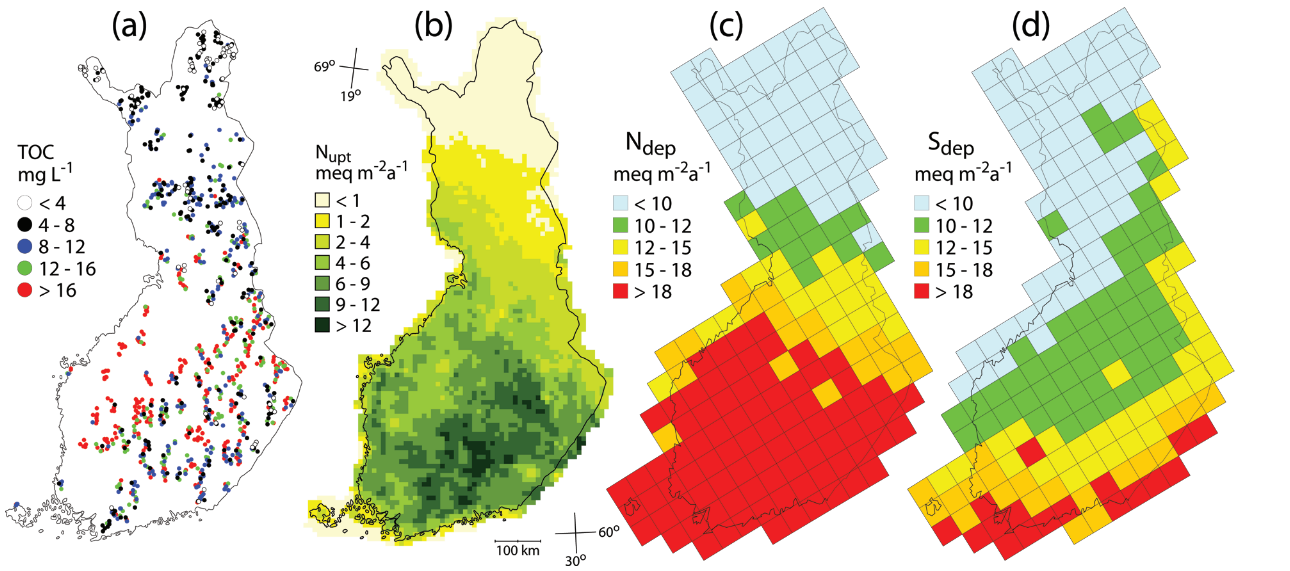

Figure 1. (a) Location of the study lakes (n = 1066) stratified according to observed (1987) lake TOC; (b) gridded (10 km × 10 km) annual mean nitrogen net uptake by forests (removal by harvesting) in 2010; (c) total nitrogen and (d) sulfur deposition in 2010 on the EMEP 50 km × 50 km grid.

almost 90% in many countries.[19] In concert, there has been a shift from assessing impacts to evaluating chemical and biological recovery.[20−24] The objective of this study was to assess the evolution of critical load exceedances in Finland during the period 1880−2020 through national-scale determination of critical loads of acidity for surface waters. In addition, the association between exceedances and biological impacts on fish (perch) was evaluated. Critical loads were determined using the FAB model in conjunction with comprehensive national surveys of surface waters (headwater lakes; n = 1066) and soils.

MATERIAL AND METHODS

Study Sites. During 1987 a country-wide lake survey was conducted under the Finnish Research Project on Acidification (HAPRO[25]). A random sample of 987 lakes, from the ∼56,000 lakes in Finland >1 ha (with ∼15,700 > 10 ha), were sampled during fall overturn and analyzed for all major ions. Statistical procedures for lake selection, sampling protocols, analytical methods, and quality control procedures are described in detail by Forsius et al.[26] An analysis and discussion of the lake water chemistry is given by Kämäri et al.[27] North of latitude 66.13° (Finnish Lapland) only lakes >10 ha were sampled;[26] therefore, the data set was supplemented by a comprehensive set of Lappish lakes sampled around the same time, resulting in a total of 1066 lake catchments (Figure 1). Catchment characteristics and soils for the study lakes were derived from topographic maps and national-scale data sets[26,28−30] (see Table 1).

The First-Order Acidity Balance (FAB) Model. The original version of the FAB model was developed during the early 1990s to calculate critical loads of acidity (S and N) for (headwater) lakes in the Nordic countries.[8] Here we provide a description of the current model equations and input data used in this study (see Part 1 of the Supporting Information for a detailed derivation of the model from 'first principles'). A comparison with other surface water critical load models can be found in Henriksen and Posch.[4]

Table 1. Statistical Summaries (5th Percentile, Median, 95th Percentile) of Catchment Characteristics and Lake Chemistry (1987) for the Study Catchments (n = 1066; see Figure 1)

variable	unit	5%ile	median	95%ile
catchment area	ha	28.0	185.5	5932.5
forest area	% terrestrial area	34.3	70.8	84.1
peatland area	% terrestrial area	0.9	19.3	63.7
lake area	ha	1.8	8.3	120.8
maximum lake depth	m	2.2	4.8	18.4
retention time	a	0.03	0.33	2.72
catchment discharge	$m\ a^{-1}$	0.233	0.321	0.414
calcium	$\mu eq\ L^{-1}$	29.94	119.76	349.30
magnesium	$\mu eq\ L^{-1}$	18.51	65.82	172.77
sodium	$\mu eq\ L^{-1}$	26.10	60.90	130.49
potassium	$\mu eq\ L^{-1}$	2.56	12.79	46.04
ammonium	$\mu eq\ L^{-1}$	0.14	0.71	5.84
sulfate	$\mu eq\ L^{-1}$	22.91	62.49	207.77
chloride	$\mu eq\ L^{-1}$	8.46	25.39	117.77
nitrate	$\mu eq\ L^{-1}$	0.00	0.86	10.00
acid neutralizing capacity[a]	$\mu eq\ L^{-1}$	35.38	164.99	385.31
pH	pH units	4.84	6.30	7.09
labile aluminum	$\mu eq\ L^{-1}$	0.00	1.45	8.78
total organic carbon	$mg\ L^{-1}$	3.06	10.90	27.38
gran alkalinity	$\mu eq\ L^{-1}$	−16.0	75.0	278.5
conductivity	$\mu S\ cm^{-1}$	13.0	29.0	72.0

[a]Acid neutralizing capacity (ANC) was estimated as the sum of base cations minus the sum of strong acid anions.

The FAB model incorporates several simplifying assumptions, in concert with other mass balance models.[4,5] The catchment is assumed small enough to be properly characterized by average soil and lake properties. Since it is a steady-state model, pools described by equilibrium equations, such as cation exchange and sulfate sorption, are not considered. For S no (other) long-term sources or sinks are considered. The input of ammonium is assumed to be completely nitrified or taken up by vegetation, i.e., only nitrate is leached to the lake.

The FAB model accounts for the long-term net removal of N by harvesting, N_u, and net immobilization, N_i, denitrification as a fraction, f_{de}, of the net N input, and in-lake retention of N and S.

Starting from the charge balance at the outlet of a lake and using steady-state mass balances of N, S, and base cations, one arrives at the following equation[4] (see also Part 1 of the Supporting Information)

$$(1-\rho_S)\cdot S_{dep}+(1-\rho_N)\cdot b_N\cdot N_{dep}=M_N+L_{crit} \quad (1)$$

Every pair of N and S deposition (N_{dep}, S_{dep}) fulfilling eq 1 defines a critical load function (CLF, see Figure 2) of acidity for the specified lake (catchment). The trapezoidally shaped function in the (N_{dep}, S_{dep})-plane is delineated by the maximum critical load of S, $CL_{max}S$ (for N_{dep} = 0) and the maximum critical load of N, $CL_{max}N$ (for S_{dep} = 0). Both the dimensionless coefficient b_N and the flux M_N depend on N_{dep}: If $N_{dep} \leq N_u + N_i$, i.e., N deposition is smaller than the sum of average net N uptake and immobilization in the terrestrial catchment, then $M_N = 0$ and $b_N = r$, where r is the lake-to-catchment area ratio, reflecting the fact that only N deposited directly onto the lake contributes to leaching. If, however, $N_{dep} > N_u + N_i$, then $M_N = (1-\rho_N)\cdot(1-f_{de})\cdot(1-r)\cdot(N_u+N_i)$ and $b_N = 1-(1-r)\cdot f_{de}$.

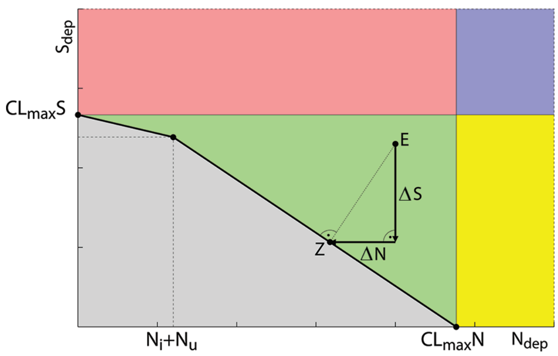

Figure 2. Piece-wise linear critical load function (CLF) of acidifying N and S for a lake defined by its catchment properties. For a given deposition pair (N_{dep}, S_{dep}) the critical load exceedance is calculated by adding the N and S deposition reductions needed to reach the CLF via the shortest path (E→Z): $Ex = \Delta S + \Delta N$. The gray area below the CLF denotes deposition pairs resulting in nonexceedance of critical loads. If a deposition pair is located in the green area (such as E), nonexceedance can be achieved by reducing N *or* S deposition (or both); in the red (yellow) area S_{dep} (N_{dep}) *has to be* reduced to achieve nonexceedance; and in the blue area *both* N_{dep} *and* S_{dep} *have to be* reduced.

A constant N immobilization in the catchment soils of N_i = 0.5 kgN ha^{-1} a^{-1} was used in the current study, based on Swedish data.[31] The denitrification fraction in the catchment soils was estimated as $f_{de} = 0.1 + 0.7\cdot f_{peat}$, where f_{peat} is the fraction of peatlands in the terrestrial catchment.[8] Nitrogen removal in harvested biomass, N_u (Figure 1), was based on uptake by three (groups of) species: pine, spruce, and broad-leaved (mostly birch). Business-as-usual projections of forest growth and harvesting were obtained from the management-oriented large-scale forestry model MELA.[32] The projections assumed no change in policies and climate (i.e., continued cuttings at present-day level). The removal (harvest) of N was estimated using element concentrations from a compilation of Nordic sources.[30]

The dimensionless quantities ρ_N and ρ_S account for in-lake retention of N and S, respectively. The retention factor for N was modeled as a kinetic equation[33]

$$\rho_N=\frac{s_N}{s_N+Q/r} \quad (2)$$

where Q is the catchment runoff (m a^{-1}), described in Aherne et al.[29] for the study lakes, and s_N is the net mass transfer coefficient. A value of s_N = 6.5 m a^{-1} was chosen for all lakes using data from Kaste and Dillon.[34] An analogous equation holds for ρ_S; and s_S = 0.5 m a^{-1} was used for all lakes.[35] The importance of in-lake retention has been evaluated by Hindar et al.[36]

The term L_{crit} (see eq 1) denotes the difference between the net base cation leaching and the critical acid neutralizing capacity (ANC) leaching. Since base cation fluxes, especially those due to weathering of the catchment soils, are poorly known, this term is modeled by the SSWC model[4]

$$L_{crit}=Q\cdot([BC]_0{}^*-ANC_{\lim}) \quad (3)$$

where $[BC]_0{}^*$ is the sea salt corrected preacidification concentration of base cations in the lake water, and ANC_{lim} is the ANC (concentration) limit above which no damage to the specified biological indicator (fish) occurs. The sea salt correction — denoted by a superscript asterisk — assumes all chloride originates from sea salt; $[BC]_0{}^*$ is estimated according to Henriksen,[37] who found that the change over time t in base cation concentration was proportional to the change in the concentration of strong acid anions (originally only sulfate)

$$\begin{aligned}&[BC]_t{}^*-[BC]_0{}^*\\&\quad=F\cdot([SO_4]_t{}^*+[NO_3]_t-[SO_4]_0{}^*-[NO_3]_0)\end{aligned} \quad (4)$$

with F denoting the proportionality factor (the 'F-factor'); Henriksen[37] suggested a value in the range 0.2–0.4 for the F-factor. Later, Brakke et al.[38] suggested a sine-function depending on $[BC]_t{}^*$. To avoid time-dependence, Posch et al.[17] derived an F-factor as a function of $[BC]_0{}^*$ using the following relationship

$$F=1-\exp(-[BC]_0{}^*/B) \quad (5)$$

The scaling factor B was estimated from paleo-limnological and water chemistry data for Finnish lakes[17] with an optimal value of B = 131 μeq L^{-1}; a comparison with the 'traditional' F-factor can be found in Henriksen and Posch.[4] Preacidification lake nitrate was set to zero, and preacidification lake sulfate was derived from 1880 modeled S deposition[39] and runoff: $[SO_4]_0{}^* = S_{dep,1880}/Q$. Subsequently $[BC]_0{}^*$ is estimated by inserting $[SO_4]_0{}^*$, measured concentrations of sulfate and nitrate, and F from eq 5 into eq 4. Since F depends on $[BC]_0{}^*$ (and not $[BC]_t{}^*$ as in Brakke et al.[38]), this requires the solution of a nonlinear equation to obtain $[BC]_0{}^*$ (see Part 2 of the Supporting Information).

The ANC limit is generally based on regional-scale assessments of the selected biological indicator.[40−44] Nonetheless, a 'default' limit of ANC_{lim} = 20 meq m^{-3}, derived from an empirical relationship between lake water chemistry and fish status in Norway,[41] is widely applied (China;[14] Southern Central Alps, Italy;[9] Kola, Northern Russia[45]). However, ANC is highly influenced by organic acids; as such a variable ANC limit taking into account the lake's concentration of total organic carbon (TOC, mg L^{-1}) was used in the current study following Lydersen et al.[43] The ANC limit was computed as

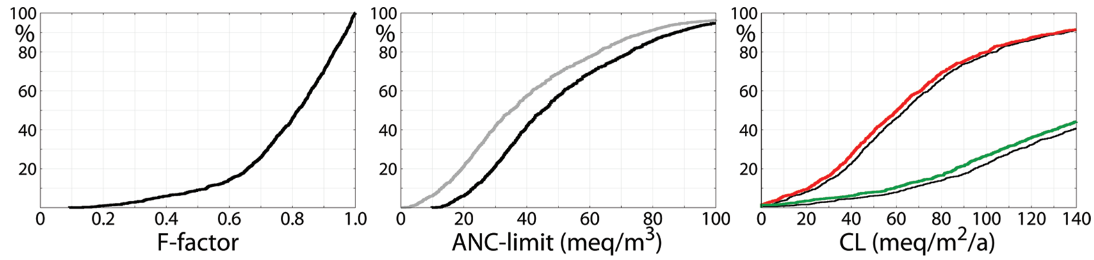

Figure 3. (a) Cumulative distribution function (CDF) of lake-specific F-factors; (b) CDFs of the ANC limits (black: brown trout, gray: perch); (c) CDFs of $CL_{max}S$ (red) and $CL_{max}N$ (green) for brown trout, as well as for perch (thin black lines).

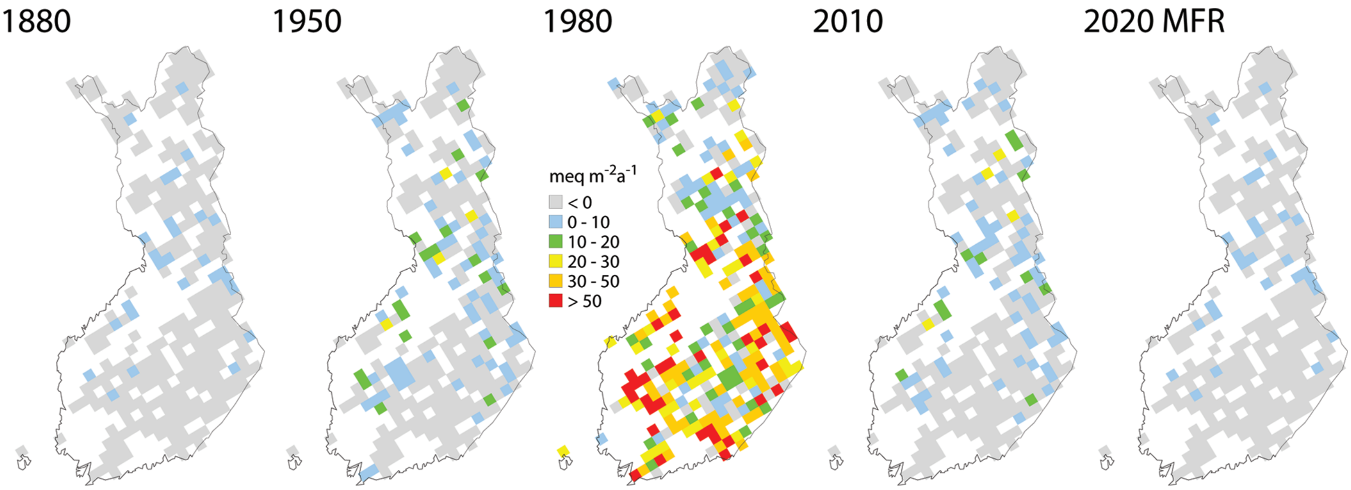

Figure 4. Spatial distribution and evolution of the average exceedance on the EMEP 25 km × 25 km grid for depositions in 1880, 1950, 1980, 2010, and MFR 2020. Exceedance was estimated by combining critical loads summarized on the EMEP 25 km × 25 km grid with total deposition on the EMEP 50 km × 50 km grid (see Figure 1).

$ANC_{lim} = ANC_{oaa,lim}+3.4\cdot TOC$, incorporating an 'organic-acid-adjusted' ANC limit, $ANC_{oaa,lim}$, which depends on the fish species to be protected. This limit was set at 8 meq m^{-3} for brown trout[43] (*Salmo trutta*) and −2 meq m^{-3} for perch[43] (*Perca fluviatilis*).

The risk of 'harmful effects' is quantified by the so-called exceedance of a critical load. Both N and S deposition (see Figure 1) can contribute to exceedance, and there are, in general, infinitely many ways of achieving nonexceedance (i.e., a point on the CLF). To be able to evaluate 'the' exceedance, it has become customary to define it as the sum of N and S deposition reduction required to reach the CLF on the shortest path:[6,46] $Ex(N_{dep},S_{dep}) = \Delta N + \Delta S$ (see Figure 2). If (N_{dep},S_{dep}) lies below the CLF, then there is nonexceedance, and Ex is set to zero.

Deposition Data and Scenarios. Past, present, and future exceedances of critical loads were estimated using modeled N and S deposition on a 50 km × 50 km grid resolution (Figure 1) for the years 1990−2010 from the EMEP/MSC-W eulerian dispersion model,[47] in combination with modeled deposition history for the period 1880−1990 from Schöpp et al.[39] Two future (2020) scenarios for S and N deposition were used in the current study: (i) a 'Current Legislation' (CLE) scenario, which assumed the implementation of the 1999 Gothenburg Protocol of the LRTAP Convention as well as the EU NEC Directive; and (ii) a 'Maximum Feasible Reductions' (MFR) scenario, which assumed implementation of all technically feasible emission reduction measures by 2020.[48] All deposition scenarios followed a common sequence between 1880 and 2010; from 2010 the two scenarios were phased in linearly until 2020. The MFR scenario showed considerably greater reductions; relative to 2000, S deposition in 2020 was reduced by 67%, compared with 28% under CLE. Exceedances were mapped on a 25 km × 25 km grid with critical loads summarized at this resolution.

Response of Fish Populations. The link between regional-scale critical loads modeling and observed changes in water chemistry and biological impacts were demonstrated at two lakes in acid-sensitive areas in southern Finland (Munajärvi and Orajärvi). Perch populations in both lakes were seriously affected and even close to extinction in the late 1980s due to acidification induced disturbances in reproduction.[49,50] The lakes were sampled for relative abundance and population structure of perch during 1985−2007 by gill net test fishing on a three-year interval. For details on sampling as well as the age and growth determination of perch see Tammi et al.[51]

RESULTS AND DISCUSSION

Regional-Scale Modeling of Critical Loads and Their Exceedance. The proportionality factor (F-factor) is a key parameter in critical loads modeling (eq 4) as it defines long-term ecosystem sensitivity to acidic deposition. It represents the proportion of base cations in surface waters that are leached from catchment soils (i.e., from the cation exchange complex) owing to elevated acidic deposition, with higher buffering capacity leading to higher values of the F-factor. In the present study, values for the F-factor varied between 0.1 and 1.0 with a median of $F = 0.82$ (Figure 3a). In general, values were high compared with studies in other regions (e.g., Sweden[52] ~0.2, Norway[8] ~0.45) suggesting that Finnish lakes were well

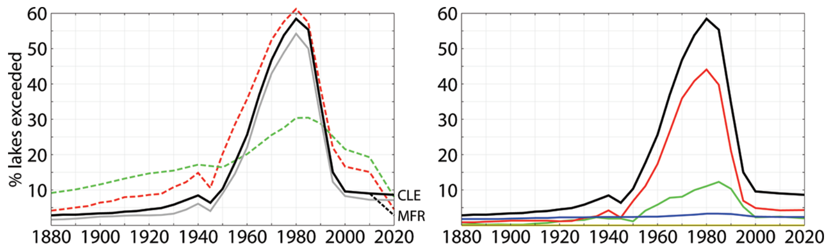

Figure 5. Temporal development of percentage of lakes with critical loads exceeded for brown trout (black line in both panels). Left: The gray line shows the exceedance percentages for perch; after 2010 exceedances are shown for both the CLE and MFR deposition scenarios. The dashed lines show the total N (green) and S (red) deposition (in meq m^{-2} a^{-1}; mean over the 1066 catchments). Right: Exceedance percentages split according to type (see colored regions in Figure 2): red: $S_{dep} > CL_{max}S$ but $N_{dep} < CL_{max}N$; green: $N_{dep} < CL_{max}N$ and $S_{dep} < CL_{max}S$; blue: $N_{dep} > CL_{max}N$ and $S_{dep} > CL_{max}S$. There are no cases with $N_{dep} > CL_{max}N$ and $S_{dep} < CL_{max}S$ (yellow line at the bottom).

buffered by catchment soils. In most regional studies, water chemistry based methods have been used to derive the F-factor,[11,16] because the approach requires limited input data. However, several studies have also suggested that these empirical estimates are a significant source of uncertainty in surface water critical loads[52,53] owing to declining soil base cation pools. If sufficient information is available, the F-factor can be estimated using dynamic hydrogeochemical models. In the current study, the F-factor was also derived from an extensive hydrogeochemical model application[30] using the MAGIC model[54] (version 777ext). There was a highly significant ($p < 0.0001$) correlation ($r = 0.96$) between critical loads ($CL_{max}S$) computed with the FAB model and MAGIC-derived base cation fluxes, giving confidence in the simpler approach (see Figure S2, Supporting Information, *Part 3*). Moreover, while long-term changes in catchment soil base cation pools at individual study lakes may influence estimates of the F-factor (and consequently critical loads), at the national level uncertainty assessments have shown that the F-factor has limited influence on the proportion of exceeded lakes.[4,10] Notwithstanding the uncertainties associated with the simpler empirical approach, it is noteworthy that dynamic hydrogeochemical models are also not free from uncertainties.[55]

Finnish lakes have high concentrations of TOC (median value for the 1066 lakes = 10.9 mg L^{-1}, Table 1), as such, an ANC limit incorporating lake-specific TOC was used in the current study[43] for both perch and brown trout to accommodate the sensitivity of different fish species. The lake-specific ANC limits ranged from close to zero to about 160 meq m^{-3} with a median value of 45 meq m^{-3} for brown trout and 35 meq m^{-3} for perch (Figure 3b), both higher than the 'default' 20 meq m^{-3}. The distribution of the resultant critical loads ($CL_{max}S$ and $CL_{max}N$) was similar for the two fish species (Figure 3c); the median $CL_{max}S$ and $CL_{max}N$ for brown trout (and perch) was 60 (64) and 156 (164) meq m^{-2} a^{-1}, respectively.

The evolution of exceedance of critical loads for Finnish lakes during the period 1880−2020 clearly shows the successful implementation of control measures for the emissions of acidifying pollutants (Figure 4). During the peak emission/deposition years in the 1980s, critical loads of acidity for surface waters were exceeded across large areas in different parts of the country. A large reduction in the exceeded area was predicted for 2020 (under CLE), and deposition according to the 2020 MFR scenario resulted in an exceedance of 1.8% for perch and 2.8% for brown trout, close to preacidification conditions during the 1880s (1.6% and 2.8%; Figures 4 and 5). In contrast, critical loads were exceeded in 54% (perch) and 58% (brown trout) of the lakes during the peak (S) deposition year 1980. The highest deposition of N compounds occurred a few years later than S and subsequently dominated total deposition until 2020 owing to greater reductions in S emissions (Figure 5, left).

Nitrogen and S deposition both contribute to exceedance; accordingly the CLF allows for an assessment of required emission reduction measures for S and N for each of the individual exceedance cases (see Figure 2). The most common of these exceedance cases for the study lakes was $S_{dep} > CL_{max}S$ and $N_{dep} < CL_{max}N$, indicating mandatory S reduction (Figure 5, right). Mandatory reductions in either N or S occurred in <15% of the study lakes (during peak emission/deposition years). The case where mandatory reductions of both S and N were required ($N_{dep} > CL_{max}N$ and $S_{dep} > CL_{max}S$) was quite rare.

Several studies have evaluated uncertainty in the critical loads approach (see the literature review in Skeffington[56]) and the potential influence of climate change.[57] Parametric uncertainty was estimated at about ±20% of the median critical load for Finnish lakes.[17,18] Moreover, critical loads have been shown to be far more sensitive to lake base cation concentration than the other parameters,[4,10] owing to the importance of base cations in defining long-term sensitivity to acidification.

The significant reduction in critical load exceedance and associated risk of damage to Finnish surface waters is strongly supported by empirical observations. Regional-scale chemical recovery of acidified lakes has been documented in several studies; decreasing lake sulfate concentrations and increasing alkalinity were first observed in the early 1990s.[58] Forsius et al.[59] estimated that 1400 lakes of size 4−100 ha (27% of Finnish headwaters) increased in alkalinity during the 1990s. The chemical recovery of lakes from acidification has been strongest and most consistent in southern Finland, where lakes were exposed to the highest S deposition load (Figure 1). This region has also shown the strongest emission reduction responses in deposition.[60,61] Dynamic hydrogeochemical model simulations (using MAGIC) for Finnish lakes under current emission reductions (CLE scenario) suggest that future chemical recovery of surface water chemistry will continue but more slowly than during previous decades.[29,30,62] In addition, future emissions reductions will likely halt soil acidification (i.e., base cation depletion); however, more stringent reductions are required to improve soil and lake water chemistry in the long term.

Critical Load Exceedance and Recovery of Fish Populations. Earlier assessments of the impacts of acidic deposition on fish status focused on perch and roach populations in Finnish lakes. The number of fish populations

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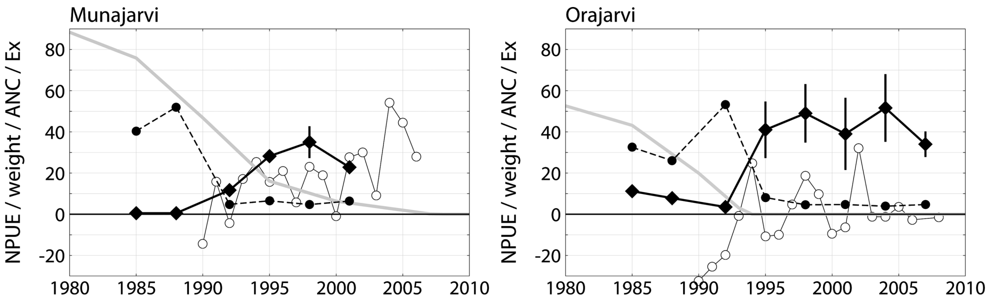

Figure 6. Response of perch to recovery from acidification in lakes Munajärvi (left) and Orajärvi (right). Diamond symbols: NPUE (= mean number of perch in one NORDIC survey net in one night) with standard deviation; Full circles: Mean weight (in dag; 1 dag = 10 g) of perch in the gill net catches; Open circles: 1990−2006 observed annual average ANC (in μeq L^{-1}); Gray line: Exceedance of acidity critical load for the respective lake (in eq $ha^{-1}a^{-1}$).

for which acidification had affected growth or population structure was estimated at between 2200 and 4400. Out of these, the number of populations that had disappeared due to acidic deposition was estimated at 1000−2000.[40] A later survey[63] indicated that the number of lakes with lost or affected roach populations was ∼1000, and 410 lakes had affected perch stocks. The first signs of recovery in affected perch populations were observed at the same time or in some cases even earlier than chemical changes.[49,50]

Long-term monitoring of fish status at selected Finnish lakes (e.g., Munajärvi and Orajärvi) showed new strong year-classes of perch in response to chemical recovery, resulting in higher fish abundance and dominance of young and small perch (Figure 6). Further, the growth of perch became slower due to increased intraspecific food competition in denser populations.[49,50] The changes were reflected as increased catches per unit effort (NPUE, mean number of fish per gill net in one night) and as decreased mean weight of perch. The recovery of the perch populations in Munajärvi and Orajärvi occurred following reduction in the exceedance of the critical load (Figure 6).

The age distribution of the two perch populations indicated that the first strong year classes of perch were born during 1990 and 1991 in Munajärvi and during 1991 and 1992 in Orajärvi. In Munajärvi, no successful reproduction of perch took place during the 1980s; in Orajärvi, only occasional reproduction took place during that time, for example in 1983 and 1986.[49]

The positive signals in perch populations for both lakes were strongly related to observed changes in lake chemistry and reduced exceedance of critical load. Nonetheless, factors other than decreased acidic deposition affected the occurrence of suitable conditions for the reproduction of perch. Favorable thermal conditions, such as warm springs and growing seasons and the mild winters of 1989, 1990, and 1992, and consequent changes in hydrology and weaker acidification pulses during spring melt runoff may have had a beneficial impact on the reproduction of perch.[50] This suggests an important role of climatic factors in determining biological responses. Successful introduction of perch into lakes that had lost their original populations during the 1980s[64] further emphasizes the ecological significance of the chemical recovery of lakes from acidification.

The critical loads methodology has provided an efficient scientific effects-based approach for assessing the environmental consequences of air pollution at large regional or national scales. The linking of critical load exceedance to ecosystem impacts has consequently directed emission reductions policies, leading to recovery of important biological indicator species in Finnish surface waters. Simplifications and uncertainties associated with surface water critical load models are well acknowledged;[4,10] nonetheless, critical loads have proven instrumental in directing effects-based emission reduction policies.

■ ASSOCIATED CONTENT

Supporting Information

Part 1: detailed derivation of the FAB model; Part 2: methods to compute the preacidification base cation concentration; Part 3: comparison with critical loads derived from MAGIC simulations. This material is available free of charge via the Internet at http://pubs.acs.org.

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Notes

The authors declare no competing financial interest.

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