Chapter 17
Assessment of Critical Loads of Acidity and Their Exceedances for European Lakes

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17.1 Introduction

Lakes and rivers provided some of the earliest evidence of the effects of acidic deposition on natural ecosystems. Early warnings of the potential impacts of 'acid rain' came from Scandinavian scientists such as Odén (1968) and Jensen and Snekvik (1972), drawing attention to the acidification of lakes and rivers in Sweden and Norway, respectively. By the 1980s, the decline and loss of fish populations in Scandinavian surface waters provided one of the most visible signs of acidification and a compelling political driver for international action on transboundary air pollution. In concert, palaeolimnological studies of lake sediments provided some of the strongest evidence linking chronic acidic deposition to surface water acidification (Fig. 17.1, Battarbee et al. 1985; Flower and Battarbee 1983). Critical loads

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Fig. 17.1 The Round Loch of Glenhead, Galloway, Scotland, is perhaps the most intensively studied site in palaeolimnology. It provided key evidence linking changes in reconstructed lake acidity, based on preserved diatom assemblages, with patterns of fossil fuel combustion and acid deposition. Despite some recovery from acidification since the 1980s, it remains acidified and continues to exceed critical loads beyond 2020. (Photo: C.J. Curtis)

for freshwaters were developed in response to the growing political controversy, when the most efficient methods of addressing acidic deposition issues (then primarily associated with sulphur (S)) at the international level were in dispute (see Chap. 1).

For freshwaters, the Steady-State Water Chemistry (SSWC) model was initially used to generate critical load and exceedance maps of S for Scandinavia and the UK (CLAG 1995; Henriksen et al. 1992, 1995). The SSWC model was employed because of its modest data requirements, needing only water chemistry and an estimate of annual runoff to calculate a critical load, and an estimate of S deposition to determine whether the critical load was exceeded. The key to the simplicity of the model is the assumption that the sulphate anion is mobile in catchments (Henriksen 1984; Seip 1980), so that input fluxes of sulphate as acidic deposition will equal output fluxes of sulphate in the runoff from a catchment. Empirical relationships are employed to derive the original, pre-industrial leaching rate of base cations from a lake or stream catchment, using only measured water chemistry. A critical acid neutralising capacity (ANC) limit is specified to ensure an acceptable level of protection to a selected biological indicator, and converted into a critical flux in runoff to derive the critical load (Henriksen et al. 1992). Details of the model are provided in Chap. 6.

While S deposition declined dramatically during the 1980s and 1990s across much of Europe, scientists in various countries had begun to notice that nitrate concentrations in acid-sensitive lakes appeared to be increasing from their very low 'pre-acidification' levels (e.g. Brown 1988; Grennfelt and Hultberg 1986; Henriksen and Brakke 1988; Sullivan et al. 1997). During the same period, emissions of oxidized forms of nitrogen (N) from vehicles and industry along with ammonia emissions from intensive agriculture had at best remained constant (INDITE 1994).

Furthermore, in the late 1980s and early 1990s the concept of N saturation was proposed (Aber et al. 1989; Aber 1992; Ågren and Bosatta 1988; Dise and Wright 1995; Skeffington and Wilson 1988; Stoddard 1994). The hypothesis states
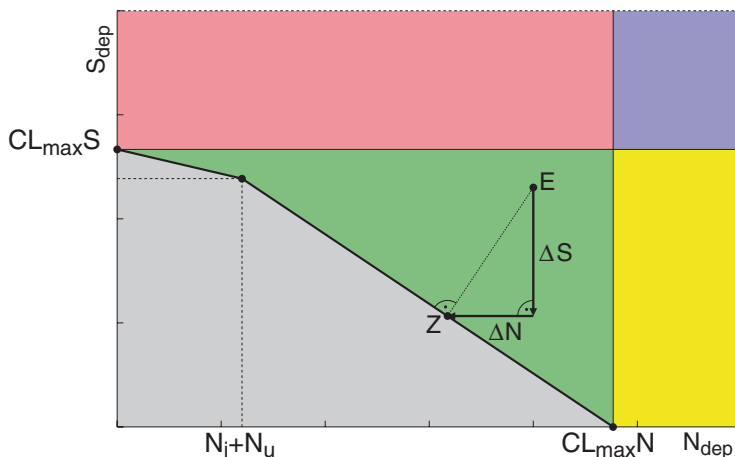
Fig. 17.2 Piece-wise linear critical load function (CLF) of acidifying N and S for a lake defined by its catchment properties. For a given deposition pair (N_{dep}, S_{dep}) the critical load exceedance is calculated by adding the N and S deposition reductions needed to reach the CLF via the shortest path (E → Z): $Ex = \Delta S + \Delta N$. The *grey* area below the CLF denotes deposition pairs resulting in non-exceedance of critical loads. If a deposition pair is located in the *green* area (such as E), non-exceedance 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 non-exceedance; and in the *blue* area both N_{dep} and S_{dep} have to be reduced

that in N-limited terrestrial ecosystems, atmospheric N deposition can be assimilated until biological N demand is met entirely by internal supply through re-mineralization and nitrification. In this situation, nitrate leaching into surface waters increases. Although this process may be interpreted as a decline in retention of atmospheric inputs, rapid cycling of deposited N has been demonstrated using stable isotope methods which show that most leached nitrate from semi-natural catchments has been microbially produced (Curtis et al. 2012). A critical load model for N and S therefore has to account for the steady-state rates of N retention, removal and leaching to determine the potential contribution of enhanced nitrate leaching to acidification.

The principal freshwater model incorporating linked critical loads of N and S is the First-order Acidity Balance (FAB) model (Henriksen and Posch 2001; Posch et al. 1997, 2012), which is analogous to the Simple Mass Balance (SMB) model for terrestrial ecosystems (see Chap. 6). The FAB model employs a charge balance for N and S, which includes the base cation leaching from the SSWC model (see above) together with a critical chemical criterion, to construct a 'critical load function' (CLF; Fig. 17.2). This criterion is set to protect a chosen biological indicator, commonly brown trout (*Salmo trutta*) using a dose-response relationship between ANC and probability of occurrence, derived for Norwegian lakes (Lien et al. 1996; Lydersen et al. 2004). A description of the FAB model can be found in Chap. 6.

A requirement for the FAB model is the parameterisation of steady-state rates for the key N retention processes, which has largely been based on simplified models

and literature default values. Application of the FAB model to regional and national datasets using these literature-based values (e.g. Curtis et al. 1998, 2000; Kaste et al. 2002; Posch et al. 2012) suggests that N deposition could offset any recovery anticipated from reductions in S deposition.

The FAB model provided critical loads of S and N deposition which were used in the negotiation of the multi-pollutant, multi-effect protocol for emissions reductions under the auspices of the Convention on Long-range Transboundary Air Pollution (LRTAP), signed in Gothenburg in 1999 and known as the Gothenburg Protocol. In this chapter we describe the various approaches taken to the collation of national datasets and application of the FAB model by five countries (Finland, Norway, Sweden, the United Kingdom and Ireland), with a comparison of critical load exceedances and the role of modelled N fluxes in national predictions of continued surface water acidification.

17.2   National Applications of the FAB Model in Europe

17.2.1   Finland

Critical loads of acidity and their exceedances for Finnish lakes have previously been determined by Henriksen et al. (1990), Forsius et al. (1992), Kämäri et al. (1993) and Posch et al. (1997, 2012).

Site Selection and Sampling: During 1987, a country-wide lake survey was conducted under the Finnish Research Project on Acidification (HAPRO; see Kauppi et al. 1990). A random sample of 987 lakes, from the ~56,000 lakes in Finland >1 ha (with ~15,700 >10 ha), were sampled during autumn overturn and analysed for all major ions in the laboratories of the (then) National Board of Waters and the Environment. Statistical procedures for lake selection, sampling protocols, analytical methods and quality control procedures are described in detail by Forsius et al. (1990). An analysis and discussion of the lake water chemistry is given by Kortelainen et al. (1989) and Kämäri et al. (1991).

North of 66.13º (7340 km in the Finnish coordinate system) only lakes >10 ha were sampled, owing to logistical limitations and the very large number of small lakes in Finnish Lapland (Forsius et al. 1990). Thus, the data set was supplemented by a comprehensive survey of lakes sampled around the same time by the Water and Environment District Office of Lapland. This combined set of lakes constitutes the basis for the assessment of critical loads for surface waters in Finland under the LRTAP Convention (Posch et al. 1997). In the current study, only lakes north of the 7340-line and <10 ha were selected from the Lappish lake set. Moreover, the number of (small) lakes was limited so that their proportion was the same as that south of the 7340-line, resulting in a total of 1066 lakes (see Aherne et al. 2012). Not all variables were available for the additional Lappish lakes, e.g., total organic carbon

Table 17.1 FAB model parameters employed in Finland

Variable	Description	Value	Source
ANC_{limit}	ANC limit	Variable	$ANC_{limit} = ANC_{oaa} + (10.2/3) \cdot TOC$, with ANC_{oaa} (organic acid adjusted ANC) $= 8$ µeq l^{-1} (for brown trout) (Lydersen et al. 2004)
F	F-factor	Variable	$F = 1 - \exp(-[BC^*]_0/B)$ where B is estimated as 131 µeq l^{-1} based on paleo-limnological and water chemistry data of Finnish lakes (Posch et al. 1993)
$[SO_4^*]_0$	Pre-industrial sulphate conc.	Variable	Derived from 1880 modelled EMEP background sulphate deposition (Schöpp et al. 2003) and discharge: $[SO_4^*]_0 = S_{dep,1880}/Q$
N_{upt}	N uptake in the catchment	Variable	Based on pine, spruce and broad-leaved (mostly birch) harvest data and element contents (see Aherne et al. 2012)
N_{imm}	N immobilisation	0.5 kg N ha^{-1} yr^{-1}	Mapping Manual (www.icpmapping.org)
f_{de}	Denitrification factor	$0.1 + 0.7f_{peat}$	Mapping Manual (www.icpmapping.org)
s_N	In-lake mass transfer coefficient for N	6.5 m yr^{-1}	Kaste and Dillon (2003)
s_S	In-lake mass transfer coefficient for S	0.5 m yr^{-1}	Baker and Brezonik (1988)

(TOC) was estimated from a linear regression with measured chemical oxygen demand (COD; Kortelainen 1993).

Derivation of FAB Datasets: The FAB model methoCy for Finland was initially described by Henriksen et al. (1993) and Posch et al. (1997) with recent advances described by Posch et al. (2012). The base cation fluxes were estimated with the SSWC model using the observed sea-salt corrected (chloride as tracer) base cation concentrations. Long-term normals for discharge (runoff) were obtained from the FINESSI project (www.finessi.info), which provided data for all of Finland on a 10 km × 10 km grid resolution (see Aherne et al. 2008). Pre-acidification lake nitrate was set to zero. A description of the FAB model parameters for Finland is provided in Table 17.1. Nitrogen removal in harvested biomass from Finnish forests was based on three species/groups: pine, spruce and broad-leaved (mostly birch). Business-as-usual (BAU) projections of forest growth and harvesting were obtained from the MELA model (Kärkkäinen et al. 2008; Redsven et al. 2004). 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 (Bringmark 1977; Finér 1989; Finér and Brække 1991; ICP Integrated Monitoring 2004; Mälkönen 1977). For further details see Aherne et al. (2012).

Table 17.2 FAB model parameters employed in Norway

Variable	Description	Value	Source
ANC_{limit}	ANC limit	Variable	Henriksen and Posch (2001), Lydersen et al. (2004)
F	F-factor	Variable	$F=\sin((\pi/2)Q[BC^*]/S)$, where $S=400$ meq $m^{-2}yr^{-1}$ is the base cation flux at which $F=1$ (Henriksen and Posch 2001)
$[SO_4^*]_0$	Pre-industrial sulphate conc.	Variable	Derived from 1880 modelled EMEP background sulphate deposition (Schöpp et al. 2003) and discharge: $[SO_4^*]_0=S_{dep,1880}/Q$
N_{upt}	N uptake in the catchment	0–50 meq m^{-2} yr^{-1}	Forest inventory data from Norwegian Forest and Landscape Institute (Frogner et al. 1994).
N_{imm}	N immobilisation	0.5 kg N ha^{-1} yr^{-1}	Mapping Manual (www.icpmapping.org)
f_{de}	Denitrification factor	0.1	Larssen et al. (2008)
s_N	In-lake mass transfer coefficient for N	5 m yr^{-1}	Dillon and Molot (1990)
s_S	In-lake mass transfer coefficient for S	0.5 m yr^{-1}	Baker and Brezonik (1988)

17.2.2 Norway

Site Selection and Sampling: The official Norwegian database for critical loads for surface waters is based on a 0.5° latitude by 1° longitude grid, with each grid square divided into 16 sub-grids (Henriksen 1998). The surface water chemistry within a sub-grid was derived from available observations for lakes and rivers, which includes results from the national 1500-lake survey conducted in 1995 (Skjelkvåle et al. 1996). The chemistry of the lake that was judged to be the most typical was chosen to represent the grid. If there were wide variations within a sub-grid, the most sensitive area was selected if it was >25 % of the grid's area. Sensitivity was evaluated on the basis of water chemistry, topography and bedrock geology. Geology was determined from the geological map of Norway (1:1 million) prepared by the Norwegian Geological Survey.

Derivation of FAB Datasets: The methodology for Norway was described by Henriksen (1998) and later updated by Larssen et al. (2005, 2008). The base cation fluxes were estimated with the SSWC model using the observed sea-salt corrected (chloride as tracer) base cation concentrations. Mean annual runoff data were taken from runoff maps prepared by the Norwegian Water Resources and Energy Directorate. Land type characteristics (lake area, catchment area, forest area, bare rock area) were measured from maps. A description of the FAB model parameters for Norway is provided in Table 17.2.

A variable ANC limit as described by Henriksen and Posch (2001) was used, but adjusted for the strong acid anion contribution from organic acids after Lydersen et al. (2004). For the F-factor, the sine function of Brakke et al. (1990) has typically been used, but in recent applications $[BC]_0^*$ has instead been taken from hindcasts from MAGIC model runs used for calculating target loads (Larssen et al. 2005). Nitrogen removal in harvested biomass was estimated by Frogner et al. (1994) and mapped for the whole of Norway according to forest cover and productivity. All uptake rates were kept constant and assume constant removal from harvest and no change from climate, eutrophication or other factors. The denitrification factor (f_{de}) was kept constant at 0.1. In-lake mass transfer coefficients were kept constant at 5 m yr^{-1} for N and 0.5 m yr^{-1} for S, chosen as the mid-value of the ranges proposed by Dillon and Molot (1990) and Baker and Brezonik (1988), respectively.

17.2.3   Sweden

Site Selection and Sampling: The Swedish critical load lakes are part of the national surveillance monitoring of lakes sampled during 2007, 2008 and 2009 (Grandin 2007). They include 2410 lakes >1 ha selected by a stratified random selection and thus representing the lake ecosystems of the whole area of Sweden. The nine largest lakes were excluded to avoid overlap of lake catchments. Lake water was sampled in the centre of the lake during autumn circulation and analysed for water chemistry. Lakes affected by liming ($n = 458$) were corrected by using the average Ca:Mg ratio from non-limed reference lakes within 20 km distance and the Mg concentration of the liming agent (Fölster et al. 2011).

Derivation of FAB Datasets: The critical loads were calculated using the FAB model as described in Henriksen et al. (1993), Posch et al. (1997) and Rapp et al. (2002), with some modifications described below (Table 17.3). The base cation leaching used in the FAB-model was taken from (MAGIC) simulated BC concentration in 2100 under the Cost Optimised Baseline (COB) deposition scenario. Thus the F-factor for estimating the weathering rate was not used. The year 2100 was used instead of 1860 for steady-state, since simulations indicated that the BC concentration of 1860 was not possible to reach within a reasonable time. The estimates of N immobilisation were based on Gundersen et al. (1998). Nitrogen immobilisation was set to 100 % for deposition up to 2 kg N ha^{-1} yr^{-1}, 50 % for deposition from 2 kg N ha^{-1} yr^{-1} up to 10 kg N ha^{-1} yr^{-1} and zero for deposition exceeding 10 kg N ha^{-1} yr^{-1}. In addition to this, leaching of organic N calculated from the lake concentration of Total Organic Nitrogen (TON) was regarded as non-acidifying. The chemical threshold, ANC limit, was calculated for each lake individually corresponding to a change in pH of 0.4 units from reference conditions estimated using the MAGIC model (Moldan et al. 2004). This threshold is used as a definition of acidification in the Swedish Environmental Quality Criteria and for the fulfilment of 'Good Ecological Status' within the EU Water Framework Directive (Fölster et al. 2007). Where insufficient data for MAGIC were available, parameters

Table 17.3 FAB model parameters employed in Sweden

Variable	Description	Value	Source
ANC_{limit}	ANC limit	0–300 μeq l^{-1} (exceeded lakes)	SEPA (2010) (derived from $\Delta pH = 0.4$)
F	F-factor	–	Steady-state BC is calculated by MAGIC
$[SO_4^*]_0$	Pre-industrial sulphate conc.	–	Not required when using MAGIC
N_{upt}	N uptake in the catchment	2.5–43.2 meq m^{-2} yr^{-1}	Calculated from the national monitoring of forests. Uptake depends on species composition and productivity of the forest
N_{imm}	N immobilisation	0–114.7 meq m^{-2} yr^{-1}	Gundersen et al. (1998)
f_{de}	Denitrification factor	$0.1 + 0.7 f_{peat}$	Mapping Manual (www.icpmapping.org)
s_N	In-lake mass transfer coefficient for N	5 m yr^{-1}	Dillon and Molot (1990)
s_S	In-lake mass transfer coefficient for S	0.5 m yr^{-1}	Baker and Brezonik (1988)

used in FAB were taken from a similar lake within a database of 2900 MAGIC simulated lakes using an analogue matching procedure, i.e. the MAGIC library, a web-based acidification assessment tool (see www.ivl.se/magicbibliotek).

The 2410 lakes were distributed over 2106 of the 5 km × 5 km squares used in national FAB applications. In most cases there was one modelled lake per 5 km × 5 km square and the ecosystem area was then set to 25 km². For lakes within squares with more than one lake, the ecosystem area was set to 25 km² divided by the number of lakes within that square.

17.2.4   United Kingdom

Site Selection and Sampling: Prior to 2004, the UK freshwater critical loads dataset comprised a grid-based survey of lakes and streams judged to be the most acid-sensitive in either 10 km grid squares for acid-sensitive regions (mainly in the uplands) or 20 km grid squares in less acid-sensitive lowland regions (Curtis et al. 2000; Kreiser et al. 1995). These lakes were sampled between 1991 and 1993. In 2004, additional freshwater sites from later, intensive regional studies were incorporated into the national dataset (described in Curtis et al. 2005a) with subsequent additions to the national dataset in 2008 (Curtis and Simpson 2011; Hall 2008). The resultant composite dataset includes 1752 sites which are neither random nor grid-based, but a combination of the original gridded survey sites with spatially intensive, complementary datasets in the most impacted regions. Furthermore, the UK dataset also contains stream sites as well as standing waters, due to known acidification problems in regions without lakes, such as the North York Moors (Evans et al. 2014).

Table 17.4 FAB model parameters employed in the United Kingdom

Variable	Description	Value	Source
ANC_{limit}	ANC limit	20 or 0 µeq l⁻¹	20 µeq l⁻¹ except where site-specific MAGIC modelling or palaeolimno-logical reconstruction indicates a lower pre-industrial value, when zero is used (Curtis and Simpson 2011)
F	F-factor	Variable	Estimated following Brakke et al. (1990) as described by Curtis et al. (2000)
$[SO_4^*]_0$	Pre-industrial sulphate conc.	Variable	Calculated as $15 + 0.16[BC^*]_t$ (Henriksen and Posch 2001)
N_{upt}	N uptake in the catchment	21–42 meq m⁻² yr⁻¹	Managed coniferous wood-land = 21 meq m⁻² yr⁻¹ Managed broad-leaf woodland = 42 meq m⁻² yr⁻¹
N_{imm}	N immobilisation	1–3 kg N ha⁻¹ yr⁻¹	Weighted by soil type; see Curtis et al. (2000), Hall et al. (1997)
N_{de}	Denitrification flux	1–4 kg N ha⁻¹ yr⁻¹	Weighted by soil type; see Curtis et al. (2000), Hall et al. (1997)
s_N	In-lake mass transfer coefficient for N	5 m yr⁻¹	Dillon and Molot (1990)
s_S	In-lake mass transfer coefficient for S	0.5 m yr⁻¹	Baker and Brezonik (1988)

Derivation of FAB Datasets: While most aspects of model parameterisation were comparable with other national applications (Table 17.4), a key difference in the UK application is the use of a constant (i.e. deposition-independent) term for denitrification, based on empirically derived values for each soil type. This decision was taken on the basis of field and laboratory studies of actual and potential denitrification rates which suggested that the published method (Posch et al. 1997) would greatly over-estimate denitrification fluxes from UK peatland soils (Curtis et al. 2006). The range of N immobilisation values is also higher than used in other regions or recommended in the Mapping Manual (www.icpmapping.org).

17.2.5   Ireland

Critical loads of acidity and exceedances for Irish lakes have been previously described by Aherne et al. (2002) and Aherne and Curtis (2003). The current study incorporated an extended number of study lakes (n = 221) sampled during the period 2000–2010.

Site Selection and Sampling: During 1997, a survey of upland headwater lakes (n = 200) was carried out in predominantly acid-sensitive coastal regions of Ireland; site selection was pseudo-randomly weighted on acid-sensitive regions based on mapped soil characteristics and bedrock geology. A sub-set of these lakes were re-sampled during 2007 (Burton and Aherne 2012) and 2008 (n = 139). In addition, the current study included 'acid lakes' routinely sampled by the Irish Environmental Protection Agency (EPA) under the Water Framework Directive monitoring

Table 17.5 FAB model parameters employed in Ireland

Variable	Description	Value	Source
ANC_{crit}	ANC limit	Variable	$ANC_{limit}=ANC_{oaa}+(10.2/3)\cdot TOC$, with ANC_{oaa} (organic acid adjusted ANC)=8 meq m^{-3} for brown trout (Lydersen et al. 2004)
F	F-factor	Variable	Estimated following Henriksen and Posch (2001) as previously described by Aherne et al. (2002) and Aherne and Curtis (2003)
$[SO_4^*]_0$	Pre-industrial sulphate conc.	Variable	Derived from 1880 modelled EMEP background sulphate deposition (Schöpp et al. 2003) and discharge: $[SO_4^*]_0=S_{dep,1880}/Q$
N_{upt}	N uptake in the catchment	31 meq m^{-2} yr^{-1}	Based on Sitka spruce (*Picea sitchensis*)
N_{imm}	N immobilisation	0.5 kg N ha^{-1} yr^{-1}	Mapping Manual (www.icpmapping.org)
f_{de}	Denitrification factor	$0.1+0.7f_{peat}$	Mapping Manual (www.icpmapping.org)
s_N	In-lake mass transfer coefficient for N	6.5 m yr^{-1}	Kaste and Dillon (2003)
s_S	In-lake mass transfer coefficient for S	0.5 m yr^{-1}	Baker and Brezonik (1988)

programme ($n=41$) and other national monitoring programmes ($n=41$). All study lakes ($n=221$) were sampled during the period 2000–2010, with an average of 4 years of chemistry data for each lake (and up to 12 observations per year for some lakes).

Derivation of FAB Datasets: The lakes ranged in size from 0.08–862.58 ha, with an average area of 28.75 ha, and average elevation of 186 m a. s. l. (max=710.7 m a. s. l.). Catchment land cover (derived from the National Teagasc Land Cover Map 1995 [TLC95]; Green and Fealy 2010) was dominated by peatlands (mean=53 % across all catchments), exposed rock (25 %), forest (7 %) and grasslands (15 %). Not all variables were available for the EPA lakes, i.e., dissolved organic carbon was only measured on a sub-set of the lakes, as such it was estimated from a linear regression with measured colour (available for all EPA lakes). Further details on FAB model parameters are provided in Table 17.5.

The methodology followed Aherne et al. (2002) with recent updates following Posch et al. (2012). The base cation fluxes were estimated with the SSWC model using the observed sea-salt corrected (chloride as tracer) base cation concentrations. Long-term normals for discharge (runoff) on a 1 km × 1 km grid resolution were estimated using MetHyd (a meteo-hydrological model; Slootweg et al. 2010), and monthly climate normals for rainfall volume, average temperature and sunshine hours (Met Eireann [URL: www.met.ie]). Pre-acidification lake nitrate was set to zero. Nitrogen removal in harvested biomass assumed that managed Irish forests were dominated by Sitka spruce (*Picea sitchensis*), with an average yield class

of 16 m^3 ha^{-1} yr^{-1} (COFORD 1994), stem concentrations of N = 0.05 % (based on spruce in Wales; Emmett and Reynolds 1996), and a wood density of 390 kg m^{-3}. The potential net uptake of 31.0 meq m^{-2} yr^{-1} was multiplied by the percent cover of forest in each catchment (based on TLC95).

17.2.6   Other European Applications of the FAB Model

There have been other large scale applications of the FAB model within Europe, including novel applications to alpine lakes in Switzerland (Posch et al. 2007) and remote lakes in several mountainous regions of Europe under the EU EMERGE Project (Curtis et al. 2005b). Both of these studies focused on small subsets of national lake datasets and are not explored further here, except to highlight the presence of (in some cases extremely) acidified lakes exceeding FAB critical loads in various alpine regions, including the Swiss and Italian Alps, the Tyrol of Austria and Italy, the Retezat Mountains of Romania, the French and Spanish Pyrenees, the Rila Mountains of Bulgaria and the Tatra Mountains of Poland and Slovakia (Curtis et al. 2005b).

17.3   Results: Critical Loads and Exceedances for European Surface Waters

17.3.1   Critical Loads of Sulphur and Nitrogen

Since the FAB model considers the effects of S and N deposition together, there is no unique critical load value for a given site. However, separate critical loads may be derived for both S and N when considered in isolation, i.e. assuming deposition of the other species to be zero (Posch et al. 1997, 2012). For example, the 5th percentile critical loads for S alone ($CL_{max}S$) are shown in Fig. 17.3. Since this model term does not include major sinks for acidity except for a minor in-lake retention term for S, it effectively provides a map of overall sensitivity to net acid inputs. Very low values of $CL_{max}S$ are found in southern Sweden, central and northern Finland, southern Norway and localised regions of northern England, north-west Scotland and Ireland. The corresponding map for N alone ($CL_{max}N$) is similar but moderated by site-specific modelled sinks for deposited N (not shown).

Cumulative distribution functions for $CL_{max}S$ in all five countries show marked differences in terms of the distribution of critical loads (Fig. 17.4). Sweden has around 10–15 % of sites with $CL_{max}S$ close to zero and a large proportion with low critical loads of < 1000 eq ha^{-1} yr^{-1}. Finland and Norway also have a large proportion of very acid-sensitive sites but fewer extreme critical loads close to zero. Ireland and the UK differ from the other countries in having a much smaller proportion of sensitive sites, and around 50 % with relatively high critical loads of > 1500 eq ha^{-1} yr^{-1}.
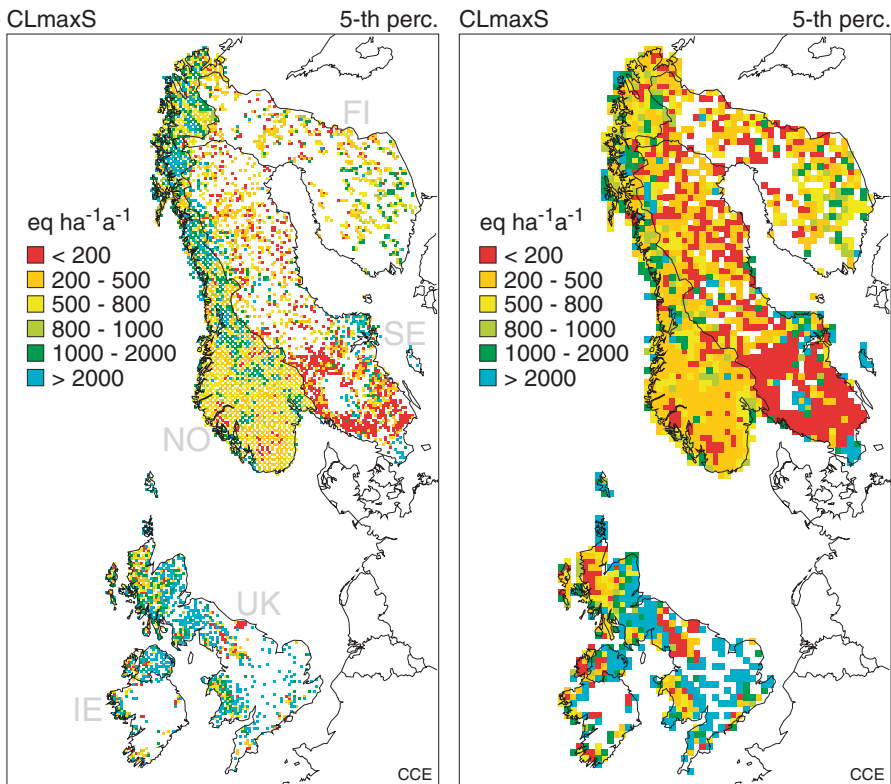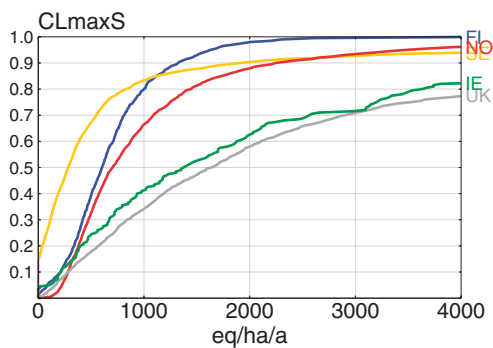
Fig. 17.3 5th percentile of $CL_{max}S$ on the 10 km × 10 km (*left*) and 25 km × 25 km (*right*) EMEP grid (the five countries are identified by their 2-letter codes)

Fig. 17.4 Cumulative distribution functions of $CL_{max}S$ for surface waters in the five countries (identified by their 2-letter codes)

17.3.2   Critical Load Exceedances

Exceedances of critical loads are computed as an excess flux of acidity comprising contributions from both sulphate and nitrate leaching for chosen S and N depositions (see Fig. 17.2 and Posch et al. 2001 for the calculation procedure). Here, results from
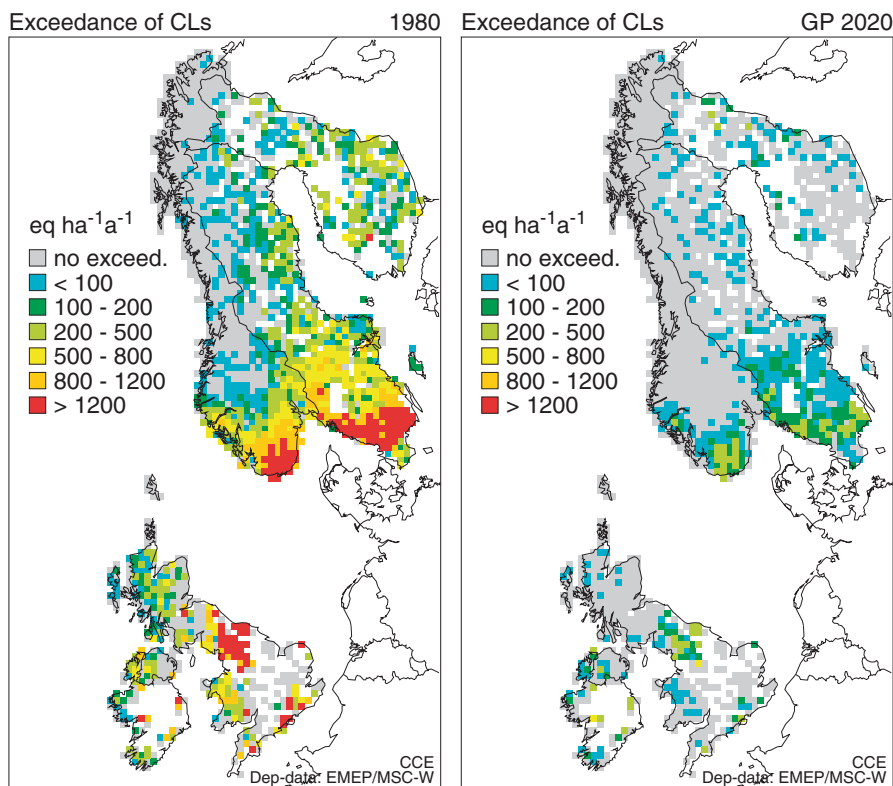
Fig. 17.5 Exceedance of acidity critical loads for surface waters in the five countries under S and N deposition for the years 1980 and 2020 (25 km × 25 km EMEP grid)

the multi-layer eulerian EMEP model (Simpson et al. 2012; Tarrasón et al. 2003) are used, consisting of relationships (source-receptor matrices) between European country emissions and depositions to specific ecosystems (forests, semi-natural vegetation, open land) in 50 km × 50 km grid cells covering Europe. Prior to 2010, historic country-specific emissions were used (see Schöpp et al. 2003 for data prior to 1990), whereas for 2020 emissions are those agreed in May 2012 in conclusion of the revision of the Gothenburg Protocol. It should be noted that there are differences between the critical load exceedances presented here for EMEP-scale deposition data, and estimates based on nationally derived depositions, which may be available at a finer spatial resolution and generated by different models.

Exceedance maps for the period of peak deposition in much of Europe (1980) and projected for 2020 on the 25 km × 25 km EMEP grid are provided in Fig. 17.5. The most severe exceedance of critical loads was observed in southern Norway, southern Sweden and parts of northern England in 1980, but exceedances were observed throughout most areas. Regions of greatest exceedance closely correspond with regions of greatest sensitivity as shown by $CL_{max}S$ (Fig. 17.3). The persistence of exceeded sites in 2020 suggests that currently planned emission reductions are insufficient to reach critical loads for all lakes, i.e. some lakes will remain acidified
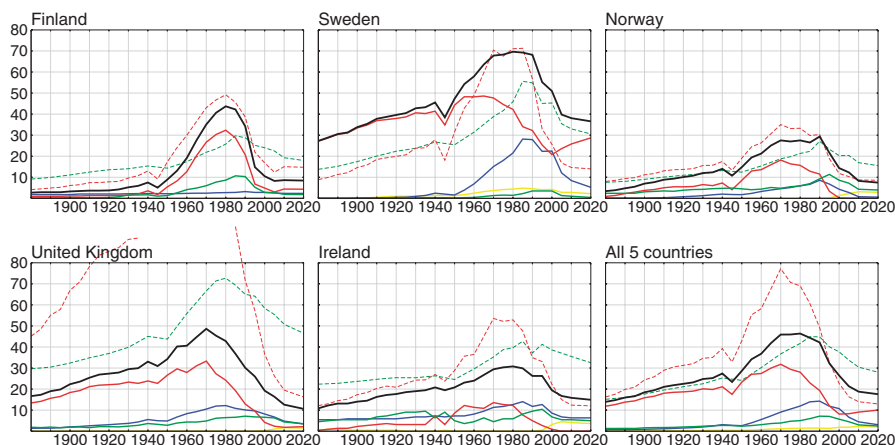
Fig. 17.6 Temporal development of percentages of lakes with critical loads exceeded in the five countries and the region as a whole (*black line*). Exceedance percentages split according to type (see coloured regions in Fig. 17.2): *red*: $S_{dep} > CL_{max}S$ but $N_{dep} < CL_{max}N$; *green*: $N_{dep} < CL_{max}N$ and $S_{dep} < CL_{max}S$; *blue*: $N_{dep} > CL_{max}N$ and $S_{dep} > CL_{max}S$; *yellow*: $N_{dep} > CL_{max}N$ and $S_{dep} < CL_{max}S$. The *dashed* lines show the total N (*green*) and S (*red*) deposition (in meq m^{-2} yr^{-1}; mean over all catchments; peaking at 165 in 1970 in the UK)

into the foreseeable future, although a partial chemical recovery and improvement in status will be achieved by 2020 as the magnitude of exceedances declines.

The temporal development of critical load exceedance from 1880 to 2020 for all countries is illustrated in Fig. 17.6, along with total deposition of excess S and N. Sulphur deposition peaked around 1970 in all countries except Sweden, where high levels persisted into the 1980s. Deposition in the UK reached a much greater and sharper peak than other countries, while Norway experienced the lowest peak in S deposition. Peaks in N deposition occurred later in all countries (1980–1990) and were much lower than peak S deposition. However, in all cases, N deposition exceeded S deposition by 2000 and this continues to 2020.

Exceedance of critical loads as a proportion of modelled sites shows a more variable peak between countries. The largest proportion of sites exceeding critical loads occurs in Sweden, peaking at about 70 % of modelled sites around 1980, which may be due to the higher level of protection selected in that country (see below). The next highest peak was found in the UK around 1970 with almost 49 % of modelled sites showing exceedance, then Finland in 1980 with 44 % exceedance. The proportion of exceeded sites reached only 31 % in Ireland (around 1980) and 29 % in Norway (around 1990). In all countries, the proportion of exceeded sites has decreased dramatically in response to declining S deposition, but all countries show exceeded sites persisting to 2020, varying from 8–11 % in Finland, Norway and the UK to 15 % in Ireland and almost 37 % in Sweden. The greatest uncertainty associated with these future exceedances is related to the long-term fate of N deposition as discussed below.

In 1980, around the time of peak exceedance for most countries, S was by far the dominant contributor to exceedance (Table 17.6). Sulphur deposition exceeded

Table 17.6 Proportion (%) of sites in exceedance categories of the Critical Load Function (see Fig. 17.2) and average deposition flux across all catchments (meq m^{-2} yr^{-1})

Year	Categories of exceedance according to CLF (%)								Deposition (meq m^{-2} yr^{-1})		
1980	Not exceeded	Total Exceeded	S or N (green)	S only (red)	N only (yellow)	S and N (blue)	Total $N_{dep} > CL_{max}N$ (blue+yellow)	Total $S_{dep} > CL_{max}S$ (blue+red)	N deposition	S deposition	N:S deposition ratio
Finland	56.3	43.7	8.6	32.4	0.0	2.7	2.7	35.1	25.8	49.2	0.52
Ireland	69.2	30.8	5.9	12.2	0.0	12.7	12.7	24.9	39.6	52.9	0.75
Norway	72.5	27.5	5.9	15.7	0.0	6.0	6.0	21.6	21.3	33.3	0.64
Sweden	30.4	69.6	1.5	42.2	4.4	21.5	25.9	63.8	45.9	71.1	0.65
UK	57.2	42.8	6.5	24.2	0.0	12.2	12.2	36.4	72.7	127.4	0.57
ALL	53.6	46.4	5.0	28.0	1.4	11.9	13.3	40.0	41.7	69.0	0.60
2020											
Finland	91.6	8.4	1.8	4.3	0.0	2.4	2.4	6.7	18.0	14.8	1.22
Ireland	85.1	14.9	5.0	0.0	3.6	6.3	10.0	6.3	32.5	12.0	2.70
Norway	92.5	7.5	4.0	0.1	2.9	0.6	3.4	0.7	15.5	8.6	1.81
Sweden	63.4	36.6	0.4	28.7	2.2	5.2	7.4	34.0	30.6	14.0	2.18
UK	89.4	10.6	3.5	2.2	1.5	3.4	4.9	5.6	46.5	16.2	2.86
ALL	82.4	17.6	2.5	10.0	2.0	3.1	5.0	13.1	28.0	12.9	2.16

$CL_{max}S$, indicating that critical loads were exceeded by S alone without reference to N deposition, in 22 (Norway) to 64 % (Sweden) of modelled sites. At that time, the relative contribution of N deposition to exceedance was much smaller than at present, as shown by N:S deposition ratios ranging from 0.52 (Finland) to 0.75 (Ireland), and taking into account that a proportion of N deposition is retained or removed in catchments before reaching surface waters. N deposition exceeded $CL_{max}N$ at only a small proportion of sites, from about 3 in Finland to 26 % in Sweden.

By 2020, major reductions in S deposition and much smaller reductions in total N deposition will mean that N deposition far outweighs S in terms of deposited acidity, with N:S deposition ratios ranging from 1.22 (Finland) to 2.86 (UK). However, the variability in modelled sinks for N deposition between countries results in a range of relative contributions to critical load exceedance. In Finland and Sweden, S deposition continues to dominate exceedance by 2020, but in Ireland and Norway N has overtaken S as the main agent of continued critical load exceedance. Relative contributions of S and N in the UK are very similar in 2020. The proportion of sites in which N deposition exceeds $CL_{max}N$ is low, ranging from about 2 % in Finland to almost 10 % in Ireland, but the future behaviour of deposited N is one of the key areas of uncertainty in the FAB model (see below).

17.4   Discussion: Limitations and Uncertainties in FAB Model Applications

17.4.1   National Approaches to Site Selection, Mapping and Setting Critical Limits

Submission of national critical load datasets to the Modelling & Mapping Programme under the LRTAP Convention allows significant flexibility in national approaches for deriving the requisite data. Some countries have used random surveys, often stratified regionally or by size class of lakes, to populate national water chemistry databases. Other countries have used additional datasets in acid-sensitive areas to supplement national random or grid-based surveys. Hence there is no consistency between countries in the degree to which national critical load datasets represent national populations of lakes. Furthermore, there are differences in the way that critical loads for lakes are represented in terms of ecosystem area.

Another difference between countries is in the choice of the critical limit, which for the SSWC and FAB models is defined as a critical ANC value. Except for Sweden, all the countries use brown trout as the indicator organism (endpoint) and a minimum ANC (critical limit) below which there is unacceptable damage. For Finland, Ireland and Norway, ANC_{oaa} takes into account the role of strong organic acids in determining this threshold, while the UK uses fixed ANC limits of 0 or 20 µeq l^{-1}. Sweden, on the other hand, uses ΔpH as the criterion for biological damage, in accordance with the Water Framework Directive whereby ecological status

should be assessed as a change from pre-industrial conditions. It is defined as the difference between pre-industrial pH and present-day pH. Both pH values are calculated from ANC and TOC; pre-industrial ANC comes from the MAGIC model. If ΔpH exceeds 0.4 units, then the lake is considered acidified, and the critical load is exceeded. This is independent of position on the pH scale and thus can indicate ecological changes in naturally acidic systems (where biota under pre-industrial conditions were already controlled by a low pH) and not just in naturally circumneutral lakes. The dose-response relationship between ΔpH and unacceptable biological change is in turn based on measured pH and relationships with fish, benthic fauna and periphyton for 74 lakes in southern Sweden, where pH showed a stronger predictive power than other chemical measures (Fölster et al. 2007).

The Swedish criterion of $\Delta pH = 0.4$ apparently gives lower critical loads than the ANC limit used in other countries, which also explains the drop in critical loads at the border between Norway and Sweden shown in Fig. 17.3. The implication is that Sweden has chosen to define critical loads with respect to more sensitive biota such as littoral invertebrate fauna and epiphyton in addition to fish, whereas the other four countries base their critical loads primarily on brown trout. It is also likely that a pH decline of 0.4 units in some Swedish sites would not result in breaching the critical ANC values used by other countries. Curtis and Simpson (2011) discussed the conceptual differences between the traditional critical loads approach where a fixed ANC limit allows differing degrees of change from reference conditions for different lakes, and the Water Framework Directive notion of defining ecological status relative to a fixed degree of change from reference conditions. The issue of aligning approaches under the LRTAP Convention and the EU Water Framework Directive has yet to be resolved.

17.4.2 National Approaches to the Nitrogen Mass Balance in FAB

There are four key N sink or removal terms used to calculate critical loads with the FAB model, and the national approaches for selecting them are compared below.

Net Uptake of N: All countries use similar assumptions about element concentrations in key tree species and projected harvest cycles to determine net removal of N in harvested biomass.

In-Lake Retention of N and S: All countries use similar mass-transfer coefficients to calculate in-lake retention of N and S. Previous studies using FAB found in-lake retention to be a minor sink for N and S in acid-sensitive lakes; for N potential terrestrial sinks are generally much greater (Curtis et al. 1998; Kaste et al. 2002; Kaste and Dillon 2003).

Immobilisation of N: Finland, Norway and Ireland all use a fixed value of 0.5 kg N ha^{-1} yr^{-1} for the immobilisation of N in catchment soils, which is the lower end of the suggested range in the Mapping Manual (www.icpmapping.org)

and constitutes a precautionary (worst-case nitrate leaching) approach. The UK employs a higher range of values from 1–3 kg N ha^{-1} yr^{-1} on the assumption that elevated rates of N immobilisation under anthropogenic N deposition loads may be sustainable over the long term. Sweden employs a unique approach, with a sliding scale for N immobilisation from 100% at very low deposition to zero at higher depositions. In all cases, long-term immobilisation in FAB provides a relatively minor sink for N when compared with the upper end of the deposition range, though it can be significant in lower deposition regions.

Denitrification Fluxes: Finland, Sweden and Ireland use a first-order term for denitrification, a linear interpolation between 10% for catchments with no peat and 80% for catchments with 100% peat cover. Norway uses a first-order term denitrifying a fixed 10% of net N inputs. The UK uses a constant (i.e. deposition-independent) term from a lookup table of denitrification fluxes for each soil type, so a catchment-weighted constant value is used for each site.

For peat catchments, denitrification within the FAB model therefore provides a major sink for N deposition of up to 80% of net inputs in Finland, Sweden and Ireland. However, it has been argued that the retention of large proportions of N deposition may be due to elevated, but possibly unsustainable, rates of N immobilisation in catchment soils (Curtis et al. 2006). One view might be that over the long term, a reduction in N immobilisation in soils could lead to an increase in denitrification fluxes as soilwater nitrate availability increases. However, for UK soils this hypothesis was tested experimentally by measuring denitrification fluxes from soils treated with ammonium nitrate solution in high and low N deposition settings. Neither untreated nor treated soils showed denitrification fluxes of the magnitude suggested by the peat fraction method (Curtis et al. 2006).

It may be concluded that the largest uncertainty within the N mass balance section of the FAB model lies in the relative importance of N immobilisation and denitrification fluxes, i.e. the future behaviour of N in terrestrial ecosystems. Nitrogen immobilisation in particular continues to present challenges at the catchment scale. The use of the FAB model to predict nitrate leaching fluxes at steady-state indicates much greater fluxes than currently observed (e.g. Curtis et al. 1998; Kaste et al. 2002). The question remains to what degree the FAB model adequately represents the real endpoint of the N saturation process, i.e. will N continue to be largely retained within terrestrial ecosystems, or will all N beyond long-term sinks specified in FAB ultimately leach as nitrate to surface waters? The latter case largely drives exceedance in Norway and Ireland. The same question has been addressed in recent modifications to the dynamic acidification model MAGIC (Oulehle et al. 2012).

17.4.3 F-Factor Issues and Lack of Steady-State

Under the original concept, a steady-state critical load should be a constant value regardless of when it is calculated. It is assumed that the calculation of critical load using

contemporary water chemistry is insensitive to the stage of acidification or indeed recovery from acidification, i.e. even though water chemistry changes in response to the acidification process, the calculated critical load should not. Hence some of the national datasets still employ water chemistry survey data from the early 1990s or even 1980s, when deposition loads were much higher than they are at present.

More recently, the time-independence of the F-factor in the SSWC and FAB models has been questioned by Rapp and Bishop (2009). They used the dynamic model SAFE to compare modelled hydrochemical changes in Swedish lakes with empirical F-factors and showed that while the F-factor approach may be adequate during the acidification phase, it does not work well during the recovery phase when base cation leaching may decrease below the pre-industrial value. Critical loads based on water chemistry sampled during the recovery phase would therefore be under-estimated. However, an earlier analysis by Henriksen (1995) suggested that variations in the F-factor were only likely to be significant for critical loads in regions of high deposition, and found little effect of varying F for Norway, Finland and Sweden. Aherne and Curtis (2003) also looked at the influence of different formulations of the F-factor on SSWC model exceedance and found little impact. One reason for these differing interpretations is that the later work of Rapp and Bishop (2009) considered largely recovered systems where minor variations in critical load would mean the difference between meeting critical loads or exceedance. As a result of reservations about the F-factor approach, Sweden and Norway have more recently used the dynamic model MAGIC to calculate critical loads for lakes in their national datasets (Tables 17.2, and 17.3), while other countries continue to use different forms of the F-factor approach. Posch et al. (2012) compared SSWC/ FAB and MAGIC based F-factors and found a strong correlation.

Conclusions

National scale applications of the FAB model provide maps of critical load exceedance which are highly consistent with areas of known acidification, as verified by independent chemical and biological monitoring studies, despite simplifications and uncertainties associated with steady-state models and critical limits (Chap. 2). Furthermore, many studies do show a strong relationship between increasing surface water ANC or pH and biological recovery (e.g. Hesthagen et al. 2011; Johnson and Angeler 2010; Kernan et al. 2010; Posch et al. 2012). In Finland, recovery in recruitment of perch populations matches well with reduced exceedance of critical loads (Posch et al. 2012). At long-term monitoring sites in the UK Acid Waters Monitoring Network, the re-appearance of acid-sensitive macrophytes and invertebrates has been linked to improvements in ANC with evidence that the critical ANC value of 20 μeq l^{-1} does indeed represent an important ecological threshold for some species (Kernan et al. 2010). Similar results were found in a Norwegian lake study where the achievement of critical load and ANC > 20 μeq^{-1} coincided with significant recovery in brown trout and invertebrate species (Hesthagen et al. 2011).

Swedish studies comparing acidified but recovering lakes with unimpacted reference lakes showed a movement of impacted phytoplankton and invertebrate assemblages towards those of reference lakes as pH of acidified lakes increased over 20 years (Johnson and Angeler 2010).

Hence the continued exceedance of FAB critical loads beyond 2020 in all five countries included here is a cause for concern as planned emission reductions under the recently revised Gothenburg Protocol do not go far enough. Critical load models like FAB evidently still have a role to play in shaping emissions policy for the protection of aquatic ecosystems. However, as emissions reductions become ever more expensive to achieve, the areas of uncertainty will face increasing scrutiny. The role of chemical and biological monitoring programmes will be critical in providing the supporting data against which model performance can be evaluated.

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