

# Critical loads of sulphur and nitrogen for freshwaters in Great Britain and assessment of deposition reduction requirements with the First-order Acidity Balance (FAB) model

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## Abstract

The critical loads approach is widely used within Europe to assess the impacts of acid deposition on terrestrial and freshwater ecosystems. Recent work in Great Britain has focused on the national application of the First-order Acidity Balance (FAB) model to a freshwaters dataset of 1470 lake and stream water chemistry samples from sites across Britain which were selected to represent the most sensitive water bodies in their corresponding 10 km grid square. A “Critical Load Function” generated for each site is compared with the deposition load of S and N at the time of water chemistry sampling. The model predicts that when catchment processes reach steady-state with these deposition levels, increases in nitrate leaching will depress acid neutralizing capacity (ANC) below the critical threshold of  $0 \mu\text{eq l}^{-1}$  at more than a quarter of the sites sampled, i.e. the critical load of acid deposition is exceeded at these sites. The critical load exceedances are generally found in upland regions of high deposition where acidification has been previously recognised, but critical loads in large areas of western Scotland are also exceeded where little biological evidence of acidification has yet been found. There is a regional variation in the deposition reduction requirements for protection of the sampled sites. The FAB model indicates that in Scotland, most of the sampled sites could be protected by sufficiently large reductions in S deposition alone. In the English and Welsh uplands, both S and N deposition must be reduced to protect the sites. Current international commitments to reduce S deposition throughout Europe will therefore be insufficient to protect the most sensitive freshwaters in England and Wales.

**Keywords:** critical loads; acidification; nitrate; FAB model; acid deposition

## Introduction

Acid deposition, in the past originating mainly from the oxidized sulphur compounds released by fossil fuel combustion, has long been recognised as an environmental problem on an international scale. The acidification of soils and waters, and subsequent damage to sensitive ecosystems, has been linked to atmospheric pollution in wide-ranging studies spanning many countries. As a result, international strategies have been negotiated for the control and abatement of sulphur emissions, culminating in the Second Sulphur Protocol under the Convention on Long-Range Transboundary Air Pollution, signed in Oslo in 1994 (UNECE, 1994).

The Oslo Protocol was underpinned scientifically by

critical loads models linking sulphur deposition fluxes to selected measures of environmental damage (Bull, 1995). These models were used to produce national and international maps of both critical loads, indicating the most acid sensitive areas, and critical load exceedances, showing the regions where acidification impacts on specified receptors were greatest (e.g. Kämäri *et al.*, 1992, 1993; Henriksen *et al.*, 1992; Langan and Wilson, 1994; Sverdrup *et al.*, 1994; CLAG, 1994). The maps introduced an “effects-based” method for spatial targeting of emission abatement measures, thereby providing a useful policy tool.

For freshwaters, the Steady-State Water Chemistry (SSWC) model was used to generate critical load and exceedance maps of sulphur for Scandinavia and the UK (Henriksen *et al.*, 1992; CLAG Freshwaters, 1995). The

SSWC model was employed because of its modest data requirements, needing only water chemistry data and an estimate of annual runoff to calculate the critical load, with an estimate of sulphur 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 (Seip, 1980; Henriksen, 1984), so that input fluxes of sulphate as acid 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. If a critical acid neutralizing capacity (ANC) concentration is selected from known relationships with biological response, it can be converted into a critical flux in catchment runoff and used to derive the long-term critical load (Henriksen *et al.*, 1992).

The Oslo Protocol ensured that measures would be adopted for the abatement of sulphur emissions across Europe, but current efforts are directed towards the negotiation of a similar protocol for nitrogen. It has been recognised in recent years that with a decline in sulphur deposition, the role of nitrogen in acidification will increase in relative and perhaps absolute terms, and many recent studies have noted the significance of nitrate in acid sensitive waters (Stoddard, 1994; Allott *et al.*, 1995; Dise and Wright, 1995; Henriksen *et al.*, 1997). The protons associated with both nitrate and ammonium ions (in the latter case via nitrification) in acid deposition will contribute to the acidification of surface waters in exactly the same way as sulphate-based acidity, with an equivalent reduction in ANC.

While the assumed mobility of the sulphate anion in catchments enables the conversion of deposition fluxes into surface water runoff fluxes, the same assumption cannot be made for nitrogen (N). The complex biological interactions affecting the transport of N (and hence its associated H<sup>+</sup>) through catchments, and the presence of mechanisms for the removal or long-term storage of N in catchment soils and vegetation, mean that it is not possible to use a single set of empirical relationships to link deposition and surface water chemistry. Instead, a process-oriented model is required which can be used to quantify the proportion of total N deposition which is transported through the terrestrial part of a catchment into the surface waters.

One such model which has been used for the derivation of linked critical loads for N and sulphur (S) is the First-order Acidity Balance (FAB) model (Posch *et al.*, 1997; UBA, 1996; Henriksen, 1998). The FAB model employs a simple charge balance for N and S, along with the original base cation leaching rate from the SSWC model, to construct a "critical load function" (CLF) which quantifies the deposition reduction requirements for either N or S.

This paper presents a full description of an application of the FAB model to British freshwaters to evaluate the long term impacts of current levels of acid deposition, and to assess the requirement for reductions in both sulphur and

nitrogen deposition to protect the most sensitive freshwater ecosystems across the country.

## Modelling methods and data requirements

### MODEL DESCRIPTION: CRITICAL LOADS

A key input to the FAB model is the pre-industrial leaching of non-marine base cations, obtained from the SSWC model (Henriksen *et al.*, 1992); its application to UK freshwaters has been described in reports by the Critical Loads Advisory Group (CLAG) to the UK government (CLAG, 1994; CLAG Freshwaters, 1995). The SSWC model is employed in the first instance to provide the pre-industrial base cation leaching, which is in turn required for the definition of critical acid anion flux and hence critical load.

Freshwater critical loads are based upon the maintenance of ANC above a critical concentration with respect to a specific target organism. ANC is defined as the sum of base cations minus the sum of strong acid anions (Henriksen *et al.*, 1992). It is assumed that all measured chloride is of marine origin and therefore delivered with an equivalent amount of base cations (although HCl deposition may be locally significant close to power stations). It is therefore possible to remove the neutral, marine component of atmospheric inputs from ANC calculations, using chloride as a tracer, by reference to the constant ionic composition of seawater. The definition of ANC is then given by:

$$\text{ANC} = [\text{BC}]^* - [\text{AN}]^* \quad (1)$$

where [BC] is the sum of base cation concentrations, [AN] is the sum of acid anion concentrations and "\*" denotes the non-marine component.

The critical flux of acid anions (i.e. critical S deposition with the SSWC model for S) is simply the difference between sustainable base cation leaching and the critical ANC flux. While the availability of exchangeable base cations in the soil complex may offer short-term buffering against acidification, in the long term steady-state considered by critical load models (decades and longer, e.g. Posch *et al.*, 1997; Jenkins *et al.*, 1997), only a supply of base cations from weathering can provide a permanent buffer (i.e. is not a transient response); it is this value, minus the required level of ANC protection, which determines the critical load (Posch *et al.*, 1997).

The basis of the SSWC model is the calculation of the flux of acid anions which, when combined with the long-term flux of base cations from weathering, will maintain ANC concentrations above the critical value:

$$L_{\text{crit}} = Q([\text{BC}]_0^* - \text{ANC}_{\text{crit}}) \quad (2)$$

where  $L_{\text{crit}}$  is the critical leaching flux of acid anions,  $Q$  is catchment runoff,  $[\text{BC}]_0^*$  is the pre-industrial concentration

of non-marine base cations and  $ANC_{crit}$  is the pre-selected critical ANC threshold.

$ANC_{crit}$  can be selected according to the required protection level for a given target organism, and the choice is therefore an arbitrary one. For freshwater critical loads modelling within Europe, the chosen target organism is usually brown trout (*Salmo trutta*), a fish of widespread distribution and economic importance in many upland areas of Europe. The correlation between the health of brown trout populations and ANC has been demonstrated through extensive studies in Scandinavia, and response curves have been derived to link the percentage of lakes holding healthy, reduced or extinct brown trout populations with ANC (Lien *et al.*, 1992). The requirement for an ANC value of 20–50  $\mu\text{eq l}^{-1}$ , as adopted in Scandinavian countries (Henriksen *et al.*, 1992), was deemed too stringent for British lakes where naturally occurring ANC values close to zero were found in some areas (CLAG, 1994): such ANC values could only be achieved by liming in these cases, even with zero acid deposition. For Britain a critical ANC value of 0  $\mu\text{eq l}^{-1}$  is therefore currently employed, which corresponds to a fifty percent probability of damage to brown trout populations (Lien *et al.*, 1992). Other methods have

Scotland where current N deposition levels are among the lowest in the country (RGAR, 1997). The change in base cation leaching per unit change in acid anion leaching is  $F$ , calculated according to the methodology of Brakke *et al.* (1990):

$$F = \sin\left(\frac{\pi [BC]_t^*}{S}\right) \quad (4)$$

where  $S$  is a constant which varies regionally according to geology, but from empirical studies is taken as 400  $\mu\text{eq l}^{-1}$  (Harriman and Christie, 1995). This means that in a surface water with  $[BC]_t^* = 400 \mu\text{eq l}^{-1}$ , the high base cation concentration indicates a weatherable catchment mineralogy likely to ensure the complete neutralization of incoming acidity, i.e. it is assumed that  $F = 1$ . Where measured base cations exceed the concentration  $S$ ,  $F$  has to be set to 1. It is now possible to calculate  $[BC]_0^*$  and hence  $L_{crit}$ , which becomes an input parameter to the FAB model.

With the FAB model, a charge balance incorporating the major processes affecting the acid anion budget is then invoked (Posch *et al.*, 1997):

$$N_{dep} + S_{dep} = \{fN_{upt} + (1 - r)(N_{imm} + N_{den}) + r(N_{ret} + S_{ret})\} + AN_{leach} \quad (5)$$

atmospheric inputs
“internal” catchment processes
leaching export

been developed for the derivation of a catchment dependent  $ANC_{crit}$  so that a single value does not have to be employed across the whole country for diverse lake types and conditions (Henriksen *et al.*, 1995; Posch *et al.*, 1999); these methods will be evaluated in future studies.

In the formulation of the SSWC model for sulphur (Henriksen *et al.*, 1992),  $L_{crit}$  is equivalent to the critical load for sulphur, because at steady-state all deposited sulphur will be present (it is assumed) in the runoff: the critical deposition flux is equal to the critical leaching flux of sulphate. In contrast, the FAB model employs a charge balance of acid anions to define those combinations of S and N deposition which will result in the critical leaching flux of acid anions (sulphate plus nitrate) over the longer term (decades).

Since  $ANC_{crit}$  is pre-selected, the unknown quantity is  $[BC]_0^*$ , which is estimated from the relationship between base cation leaching and the deposition inputs of strong acid anions, according to the “F-factor” (Brakke *et al.*, 1990):

$$[BC]_0^* = [BC]_t^* - F([AN]_t^* - [AN]_0^*) \quad (3)$$

where the subscript “t” indicates a current concentration and the subscript “0” indicates the pre-industrial concentration.  $[SO_4^{2-}]_0^*$  is estimated from an empirical relationship with  $[BC]_t^*$  derived from near-pristine lakes in northern Norway (Henriksen *et al.*, 1990). Pre-industrial nitrate concentrations are assumed to have been zero, which is supported by current observations in lakes in northern

$N_{dep}$	= total N deposition
$S_{dep}$	= total S deposition
$N_{upt}$	= net growth uptake of N by forest vegetation (removed by harvesting)
$N_{imm}$	= long term immobilisation of N in catchment soils
$N_{den}$	= N lost through denitrification in catchment soils
$N_{ret}$	= in-lake retention of N
$S_{ret}$	= in-lake retention of S
$AN_{leach}$	= acid anion leaching from catchment
$f$	= fraction of forested area in the catchment
$r$	= lake:catchment area ratio

The charge balance equates the deposition inputs of acid anions with the sum of processes which control their long term storage, removal and leaching exports. Several major assumptions are made in this formulation:

1. long term sinks of sulphur in the terrestrial part of the catchment (soils and vegetation) are negligible,
2. there are no significant N inputs from sources other than atmospheric deposition, i.e. no fertiliser application in the catchment,
3. ammonium leaching is negligible because any inputs are either taken up by the biota, adsorbed onto soils, or nitrified to nitrate.

The internal catchment processes affecting acid anion budgets introduced by the FAB model are linked to

vegetation cover, soil type and catchment morphology. For modelling applications in Britain, the calculation of these sink terms is described below.

#### *Net growth uptake of N (in forest)*

Over the long timescales (decades–centuries) considered by critical loads modelling, the only permanent sinks for N in the biomass occur when there is removal of biomass from the catchment. Short term, seasonal cycling of N does not affect the mean annual budget over these timescales. The main route for the removal of biomass from upland catchments is through the harvesting of forest. There are estimates available for the N content of various tree species, and if the time period of the harvesting cycle is known, a long-term figure for the export of nitrogen can be derived. For example, Emmett and Reynolds (1996) estimate the potential removal of nitrogen in thinnings and bolewood during harvesting of Sitka spruce stands of Yield Class 6 to 24 to be in the range 1.0–8.5 kgN ha<sup>-1</sup>yr<sup>-1</sup> for the UK. These figures assume harvesting at 40 years of age. It is recognised that the annual uptake of nitrogen will not be constant throughout the 40 years of forest growth, because there is a link between N uptake and tree age (Emmett *et al.*, 1993; Reynolds *et al.*, 1994).

Ideally, forestry data (species, age, yield class) should be used to determine N removal rates on a local basis. For national mapping, these data are not available so a default value in the mid-range of the published data (Emmett and Reynolds, 1996) is employed here ( $N_{\text{upt}} = 0.279$  keq ha<sup>-1</sup>yr<sup>-1</sup> or c. 4 kgN ha<sup>-1</sup>yr<sup>-1</sup>) for catchments containing areas of coniferous forest. Deciduous forest is ignored: it is assumed that there is little harvesting of deciduous trees, particularly in acid-sensitive areas.

Other possible routes for N export in biomass include grazing (weight gain of livestock) and burning (e.g. of heather on grouse moors) but these are assumed here to be negligible in the acid sensitive upland catchments of Britain.

#### *Long term immobilisation of N in catchment soils*

Estimates of the long term immobilisation of N in catchment soils have been derived by chronosequence studies, whereby the total N content of soil profiles is divided by the age of the profile (often assumed to be approximately 10,000 years since the last glaciation) to determine the annual immobilisation rate (UBA, 1996). These long-term estimates suggest a low immobilisation rate in the range 0.5–1.0 kgN ha<sup>-1</sup>yr<sup>-1</sup>. Under present conditions with increased N cycling due to elevated N deposition this range may extend to a much higher figure, and shorter term studies of the recent increases in N content in soil profiles indicate values of up to 10 kgN ha<sup>-1</sup>yr<sup>-1</sup> (UBA, 1996). For British soils, the working range adopted by the CLAG soils subgroup is 1–3 kgN ha<sup>-1</sup>yr<sup>-1</sup>, depending on soil type (Hall *et al.*, 1997).

#### *Denitrification*

It is assumed that N immobilisation and growth uptake are faster processes than denitrification (Posch *et al.*, 1997), and that denitrification can be linearly related to the net input of nitrogen into the soil system as follows:

$$N_{\text{den}} = f_{\text{de}}(N_{\text{dep}} - N_{\text{imm}} - fN_{\text{upt}}) \quad (6)$$

where  $f_{\text{de}}$  is the “denitrification fraction”, with a value between 0 and 1, ascribed to the catchment soils.

Posch *et al.* (1997) have argued that peat soils are likely to provide the highest rates of denitrification, and proposed a method for the calculation of a “denitrification fraction”. Denitrification is an anaerobic process and therefore a function of soil moisture content, which is very high in most peat soils. They assumed that, in peat soils, denitrification of net nitrogen inputs could be 80%, while the minimum denitrification fraction for other soil types would be around 10%. The denitrification rate for a catchment can therefore be estimated by an interpolation of these figures:

$$f_{\text{de}} = 0.1 + 0.7f_{\text{peat}} \quad (7)$$

where  $f_{\text{de}}$  is the denitrification fraction, and  $f_{\text{peat}}$  is the proportion of peat soils within the catchment. Denitrification within any catchment therefore varies between 10% and 80% of net inputs. This approach has been used in several countries around Europe, as recommended by the UNECE (UBA, 1996).

A problem with the above method for determining denitrification is that the resulting denitrification rate may be far higher than current values measured for British soils. For example, for peat soils  $f_{\text{de}} = 0.8$ , which implies that 80% of net N inputs (after N uptake and N immobilisation) are denitrified. With total N deposition exceeding 30 kgN ha<sup>-1</sup>yr<sup>-1</sup> across some parts of upland Britain (RGAR, 1997), the denitrification rates for unafforested peat catchments, after subtracting the component immobilised in catchment soils (a figure of 1–3 kgN ha<sup>-1</sup>yr<sup>-1</sup> is used for Britain), equate to 80% of at least 27 kgN ha<sup>-1</sup>yr<sup>-1</sup>, i.e. more than 21 kgN ha<sup>-1</sup>yr<sup>-1</sup>. This value is an order of magnitude higher than observed denitrification values for peat soils in Britain (Emmett and Reynolds, 1996) and five times greater than the recommended maximum value for UK soils (Hall *et al.*, 1997). It might, however, be argued that a possible reason for the low rates of denitrification observed in British soils is that current high immobilisation rates reduce the supply of N for denitrification. If N immobilisation rates decline to the low values used here, the supply of N for denitrifiers would increase and the rate of N removal via this route could rise. This issue is being addressed elsewhere through the measurement of denitrification potential in soil cores given very high rates of N supply.

For the British application of the FAB model, the denitrification rate is given a fixed value for certain soil types, independent of deposition. Each of the soil map units

on the 1:250,000 scale soil maps for Great Britain has been allocated one of three figures (1, 2 or 4 kgN ha<sup>-1</sup>yr<sup>-1</sup>, representing low, medium and high rates of denitrification), based largely on soil drainage characteristics (Hall *et al.*, 1997). It should be noted that in the British context, gley soils are assumed to yield a higher rate of denitrification than peat soils.

*In-lake retention of N and S*

The in-lake retention of acid anions is assumed to be a linear function of the net input of acidity. It is sequentially the final sink of acidity encountered by inputs which move through the catchment. The “in-lake retention fractions” for nitrogen ( $\rho_N$ ) and sulphur ( $\rho_S$ ) are calculated from a kinetic equation accounting for water retention time (Kelly *et al.*, 1987; Posch *et al.*, 1997) and their derivation for British lakes is described fully in Curtis *et al.* (1998). Both  $\rho_S$  and  $\rho_N$  are zero in stream catchments.

Using the above methods to derive the terms for “internal” catchment processes, the charge balance provides those combinations of S and N deposition for which  $AN_{leach} = L_{crit}$ , thereby defining the critical load function (Fig. 1). The constraints on the critical load function are

given by:

$$CL_{max}(S) = L_{crit}/(1 - \rho_S) \tag{8}$$

$$CL_{min}(N) = fN_{upt} + (1 - r)(N_{imm} + N_{den}) \tag{9}$$

$$CL_{max}(N) = CL_{min}(N) + L_{crit}/(1 - \rho_N) \tag{10}$$

It is not possible to define a single value to represent the critical load of total acidity using the FAB model, since the acid anions sulphate and nitrate behave differently in the way they are transported with hydrogen ions; one unit of deposition of S will not have the same net effect on surface water ANC as an equivalent unit of N deposition. Equations 8 and 10 effectively define the critical deposition loads for S and N individually.  $CL_{max}(S)$  defines the critical load for sulphur when total N deposition is less than  $CL_{min}(N)$ . This term is equivalent to the SSWC model critical load for sulphur, modified slightly for the in-lake retention of a small proportion of inputs. When S deposition exceeds  $CL_{max}(S)$ , the critical load is exceeded by S alone, regardless of the level of N deposition. The contribution of N deposition to an increase in exceedance over that resulting from S alone is determined by the FAB charge balance.  $CL_{min}(N)$  defines the deposition of total N ( $NH_x + NO_x$ ) at which terrestrial catchment processes effectively remove all N, so that deposition loads lower than  $CL_{min}(N)$  result in no net leaching of nitrate. The terrestrial sinks for N are fixed by soil type and coniferous forest cover. An important assumption here is that all N deposition is transported through the terrestrial part of the catchment, i.e. in lake catchments there is negligible deposition directly onto the lake surface.  $CL_{max}(N)$  defines the critical load for total N deposition when S deposition is zero. When total N deposition exceeds  $CL_{max}(N)$  the critical load is exceeded by N deposition alone, although critical load exceedance may be further increased by S deposition.

In practice, neither S nor N deposition will ever be zero, so the critical load for the deposition of one species is fixed by the deposition of the other, according to the line defining the unshaded area of the CLF in Fig. 1 (Posch *et al.*, 1997). For pairs of S and N deposition values which are located in the unshaded (white) area of the CLF, the site is protected. If the deposition values fall above the CLF, the critical load is exceeded. The options for protecting the site, in terms of deposition reductions, are then dictated by the location of the given deposition values in a particular segment of the CLF (Fig. 1). The colour coding of the CLF lends itself to mapping qualities of deposition reduction requirements.

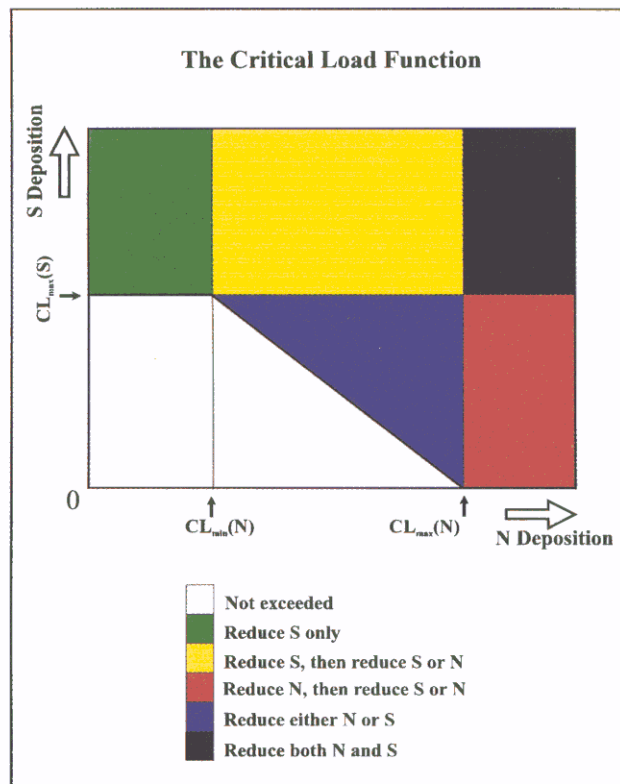


Fig. 1. The critical load function defined by the FAB model.

SOURCES OF INPUT DATA–CRITICAL LOADS MODEL

For the national FAB mapping exercise in Great Britain, datasets from various sources were collected. Their derivations are described below.

1. A national network of 1470 freshwater sites was selected

- to represent the most sensitive water body on a 10 km square grid basis (20 km in non-sensitive lowland areas), with lakes selected in preference to streams where possible (Kreiser *et al.*, 1993; Curtis *et al.*, 1996).
2. One-off dip samples were taken during autumn or early spring over the period 1992–94, and provided water chemistry which is assumed to approximate to flow-weighted mean chemistry (Forsius *et al.*, 1992). Chemical analysis was carried out according to the methodology of Harriman *et al.* (1990). These data were used to calculate  $L_{crit}$  with the SSWC model.
  3. Site catchments and lake outlines were digitized from topographical maps at 1:25,000 scale, from which lake to catchment area ratio ( $r$ ) was derived.
  4. Leaching fluxes from catchments were calculated using catchment area weighted runoff data from the 1 km grid annual mean values for the period 1992–94.
  5. Soil type and percentage cover were extracted for each catchment using digital catchment outlines and digital soils maps at 1:250,000 scale. The soils data were used to provide catchment weighted estimates of  $N_{imm}$  and  $N_{den}$  using the values suggested for each soil type in Hall *et al.* (1997).
  6. Coniferous forest cover ( $f$ ) was estimated for each catchment using digital catchment boundaries with the ITE land cover map (Fuller *et al.*, 1994) derived from satellite imagery at 25m pixel resolution.

The above datasets were used in the FAB charge balance to define the CLF for each of the 1470 freshwater sites, which can then be compared with sulphur and nitrogen deposition loads to assess spatial patterns of critical load exceedance and emission reduction requirements.

#### S AND N DEPOSITION DATA

The national S and N deposition data are generated at the 20km × 20km grid scale from measured and interpolated mean annual data in the UK (CLAG Deposition Fluxes, 1997; RGAR, 1997). For sulphur, total wet plus dry, seeder-feeder enhanced, non-marine values are used. For nitrogen, total N deposition is quantified as the sum of dry ( $NO_2 + NH_3$ ), cloud ( $NO_3^-$ ,  $NH_4^+$ ) and wet ( $NO_3^-$ ,  $NH_4^+$ ) deposition (CLAG Deposition Fluxes, 1997).

The deposition data are modelled from measurements made over the period 1992–1994, which corresponds with the period of freshwaters sampling. These are the most recently available data and are hereafter referred to as current deposition data.

The interpolated rainfall data used in the derivation of wet deposition are aggregated to the 20km × 20km scale, and the deposition data cannot be resolved to a finer scale than this (CLAG Deposition Fluxes, 1997). By contrast, the runoff data used to calculate catchment output fluxes from measured water chemistry are catchment-weighted 1km ×

1km data. The implications of this difference in data resolution are discussed below.

## Modelling results and predicted change

The charge balance which forms the basis of the FAB model and the “critical leaching of acid anions” ( $L_{crit}$ ) term calculated with the SSWC model are based on assumed long-term, steady-state conditions. The FAB model outputs therefore indicate the status of freshwater bodies under a steady-state condition between deposition and catchment processes. In this respect, national maps based on FAB outputs using current deposition data cannot necessarily reflect the current chemical status of those freshwaters, because a steady-state cannot be assumed at present with respect to nitrogen (Curtis *et al.*, 1998).

Since it is assumed that sulphate is a mobile anion (Seip, 1980) and that deposition inputs are in steady-state with leaching outputs, it might be expected that measured and predicted fluxes of non-marine sulphate are similar (while accepting the inherent uncertainties in the comparison of one-off spot measurements of chemistry with coarse-resolution, grid-based estimates of annual mean deposition and runoff). Although there is a deposition-dependent sink for sulphur in the FAB model through in-lake retention processes, this generally accounts for a very small percentage of the S budget and so the difference between measured and predicted fluxes under a given deposition load should be very small. For all chemical species, the outputs from the FAB model are effectively predictions of the chemical status of freshwaters under some future steady-state condition, and the most significant outputs in terms of anticipated changes in chemistry are nitrate and ANC.

The FAB model outputs under the current deposition load are described below with respect to the most acid sensitive areas as indicated by the two greatest sensitivity classes on the freshwater sensitivity map of Great Britain (Hornung *et al.*, 1995). The approximate locations of these regions are indicated in Fig. 2. Measured concentrations of non-marine sulphate, nitrate and ANC are compared with predicted concentrations by converting the FAB model output fluxes back into annual (flow-weighted) mean concentrations using runoff data (Figs. 3–5). Another significant model output in policy terms is the map of deposition reduction requirements from current load, whereby the FAB model indicates where current deposition of S and/or N will lead to future damage at steady-state (Fig. 6).

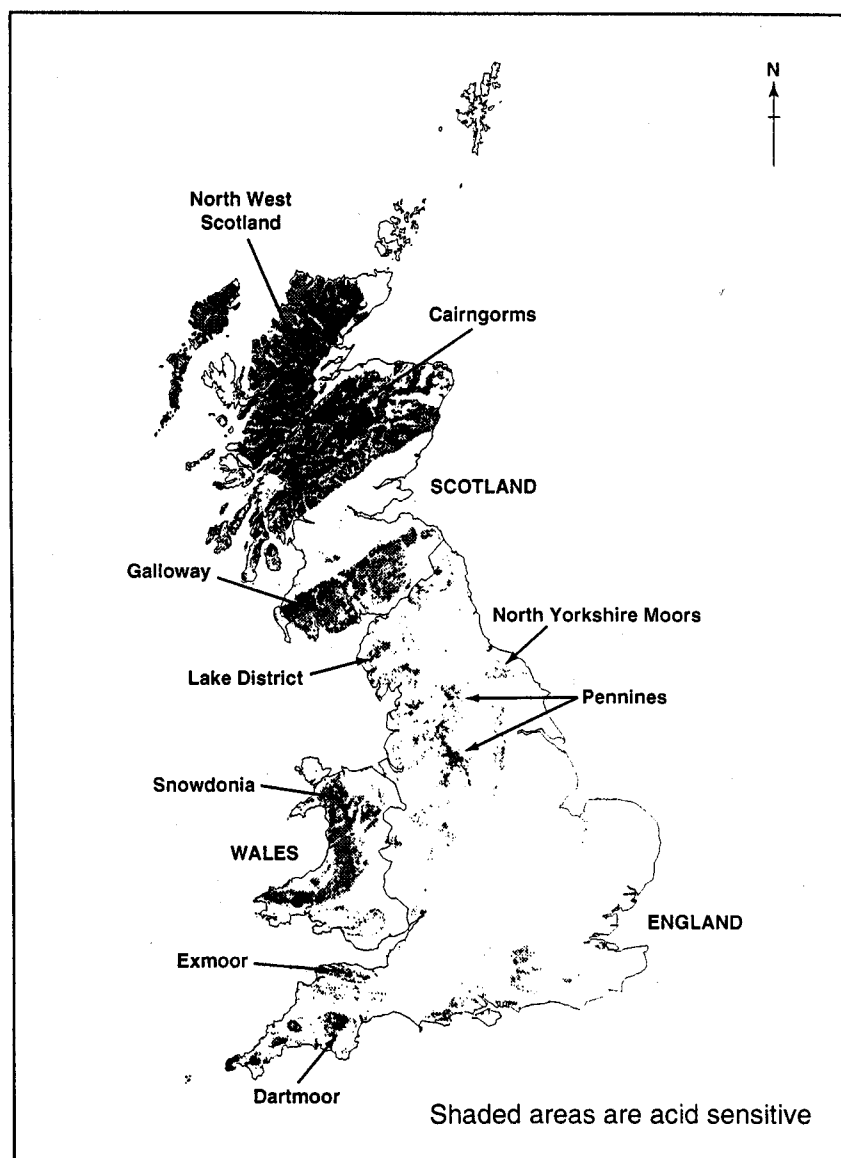


Fig. 2. Regions of acid-sensitive upland waters in Great Britain (from Curtis et al., 1999) as defined by the two greatest sensitivity classes on the national freshwater sensitivity map (Hornung et al., 1995).

#### PREDICTED CHANGES IN NON-MARINE SULPHATE LEACHING

The predicted, steady-state non-marine sulphate leaching flux ( $S_{leach}$ ) is derived by subtracting the in-lake retention component from deposition inputs:

$$S_{leach} = (1 - \rho_S) S_{dep} \quad (11)$$

For stream sites, where  $\rho_S = 0$ , the predicted leaching flux is therefore equal to the deposition flux. Division by the annual runoff converts the predicted leaching flux into a concentration, which can then be compared with measured values (Figs. 3a–b).

The national map of measured non-marine sulphate (Fig.

3a) shows a distinct gradient in concentrations, with the lowest values in the far north-western uplands of Scotland and the highest values in the east, and particularly the south-east of England. A similar pattern is observed in the map of modelled values (Fig. 3b) in which deposition data are used to predict concentrations, but lower values are shown in the south and east (very few  $>500 \mu\text{eq l}^{-1}$ ). This implies that there are direct terrestrial inputs in this region, where intensive agriculture and human population density are greatest. These non-atmospheric, urban and agricultural sources of S would cause problems in the application of the FAB model were it not for the fact that lowland regions most affected by this problem coincide almost entirely with areas in which surface waters are not sensitive to

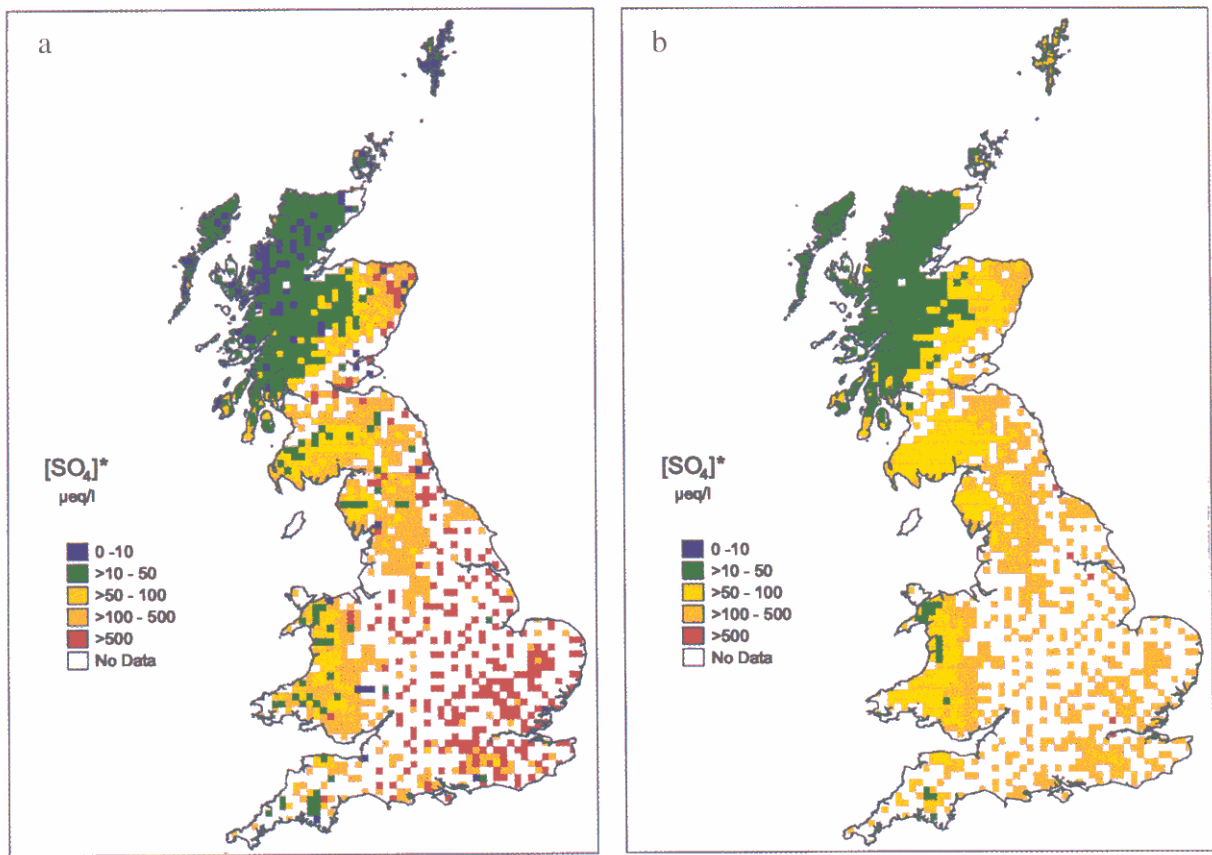


Fig. 3. Measured and modelled non-marine sulphate concentrations in the FAB mapping dataset. (a) Measured non-marine sulphate concentrations, (b) Predicted long-term non-marine sulphate concentrations under mean 1992–94 deposition loads of S and N.

acidification (Fig. 2). The sites which are located in these areas tend to have very high ANC values and are not exceeding their critical loads, thus avoiding the problem of spurious requirements to reduce S deposition.

In some localised groups of sites in the sensitive uplands, for example the far north-west of Scotland, northern Galloway, the central Lake District and the Black Mountains of South Wales (see Fig. 2), the prediction of slightly higher concentrations of sulphate than measured values could be due either to an overestimate of S deposition or an underestimate of S retention, whether in-lake or transient S adsorption within the soils of these areas.

#### PREDICTED INCREASES IN NITRATE LEACHING

Most of the processes included in the FAB model are selected to determine the sustainable removal or retention rates of nitrogen. It is recognised that current nitrogen dynamics within catchments may be dictated by short term elevated rates of certain processes, notably soil immobilisation of N (Dise and Wright, 1995), but these transient

processes must be excluded from the long term definition of critical loads (Curtis *et al.*, 1998).

Current nitrate measurements in British freshwaters show strong regional patterns (Fig. 4a). In northern Scotland, where S and N deposition levels are generally the lowest in Britain, most surface waters currently show negligible concentrations of nitrate ( $<10 \mu\text{eq l}^{-1}$ ), often below detection limits. The highest levels of nitrate ( $>100 \mu\text{eq l}^{-1}$ ) are found in lowland agricultural areas; eastern Scotland, south-west Wales and most of lowland (south-eastern) England. Water bodies in the acid-sensitive uplands of Britain experiencing moderate or high levels of N deposition tend to have nitrate concentrations which are intermediate between the near-zero levels of northern Scotland and the extreme levels in fertiliser impacted runoff in the lowlands (Allott *et al.*, 1995; INDITE, 1994). In these upland areas, the presence of high nitrate concentrations is generally attributable to atmospheric deposition, although simple input-output calculations in these areas still indicate a high level of nitrogen retention in catchments. An upland network of thirteen monitoring sites sampled monthly over two years indicated mean catchment retention levels of more than seventy-five percent for total inorganic nitrogen ( $\text{NH}_4^+ + \text{NO}_3^-$ ), but less than fifty percent for nitrate alone



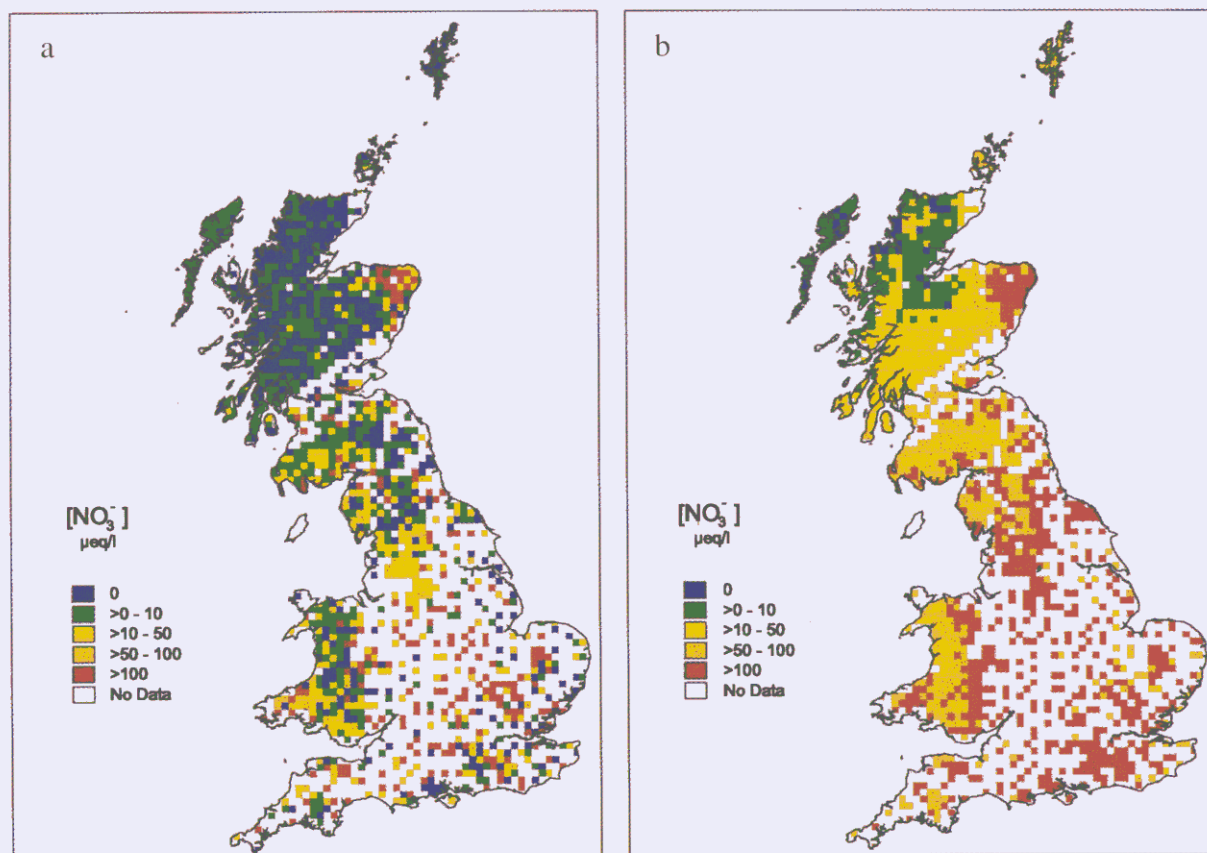


Fig. 4. Measured and modelled nitrate concentrations in the FAB mapping dataset. (a) Measured nitrate concentration. (b) Predicted long-term nitrate leaching under mean 1992–94 deposition load of N.

(Curtis *et al.*, 1998). This result implies the preferential retention of ammonium or its nitrification to nitrate.

The mass balance employed in the FAB model provides the potential, future nitrate leaching ( $N_{\text{leach}}$ ):

$$N_{\text{leach}} = (1 - \rho_N)(N_{\text{dep}} - fN_{\text{upt}} - (1 - r)(N_{\text{imm}} + N_{\text{den}})) \quad (12)$$

This leaching flux of nitrate can be converted into a concentration if no change in annual runoff is assumed.

The FAB model predicts that nitrate leaching will increase greatly in all areas of Britain, except for the extreme north west of Scotland, under current deposition loads (Fig. 4b). The sites in England are nearly all predicted to have nitrate concentrations of more than  $100 \mu\text{eq l}^{-1}$  upon reaching steady-state with current deposition levels, while most Welsh sites sampled would have nitrate concentrations in the range  $50\text{--}100 \mu\text{eq l}^{-1}$ . Nitrogen saturation and breakthrough is therefore predicted in most of the sampled upland catchments of England and Wales at some unspecified point in the future. In Scotland, a gradient of nitrate leaching is predicted from near zero concentrations

in the extreme north-west, to values in the range  $50\text{--}100 \mu\text{eq l}^{-1}$  across most of the south.

The national picture of severe future nitrate leaching (Fig. 4b) predicted by the FAB model results from the small size of available long-term nitrogen sinks in the model relative to current N deposition rates (Curtis *et al.*, 1998). This map therefore closely reflects the theoretical distribution of nitrate concentrations resulting from the conversion of total N deposition flux into concentrations using runoff data. The realisation of these large increases in acid anion leaching would have serious effects on the ANC of water bodies in sensitive areas (see below). In the non-sensitive lowlands, agricultural inputs of N are likely to be more significant than atmospheric inputs, but there will be no adverse impacts in terms of acidification in the surface waters draining these areas where, instead, eutrophication is more of a problem (INDITE, 1994).

#### CHANGES IN ANC ASSOCIATED WITH ACID ANION LEACHING RATES

If acid anion leaching does increase to a new steady-state level in the future, there will be an equivalent decrease in

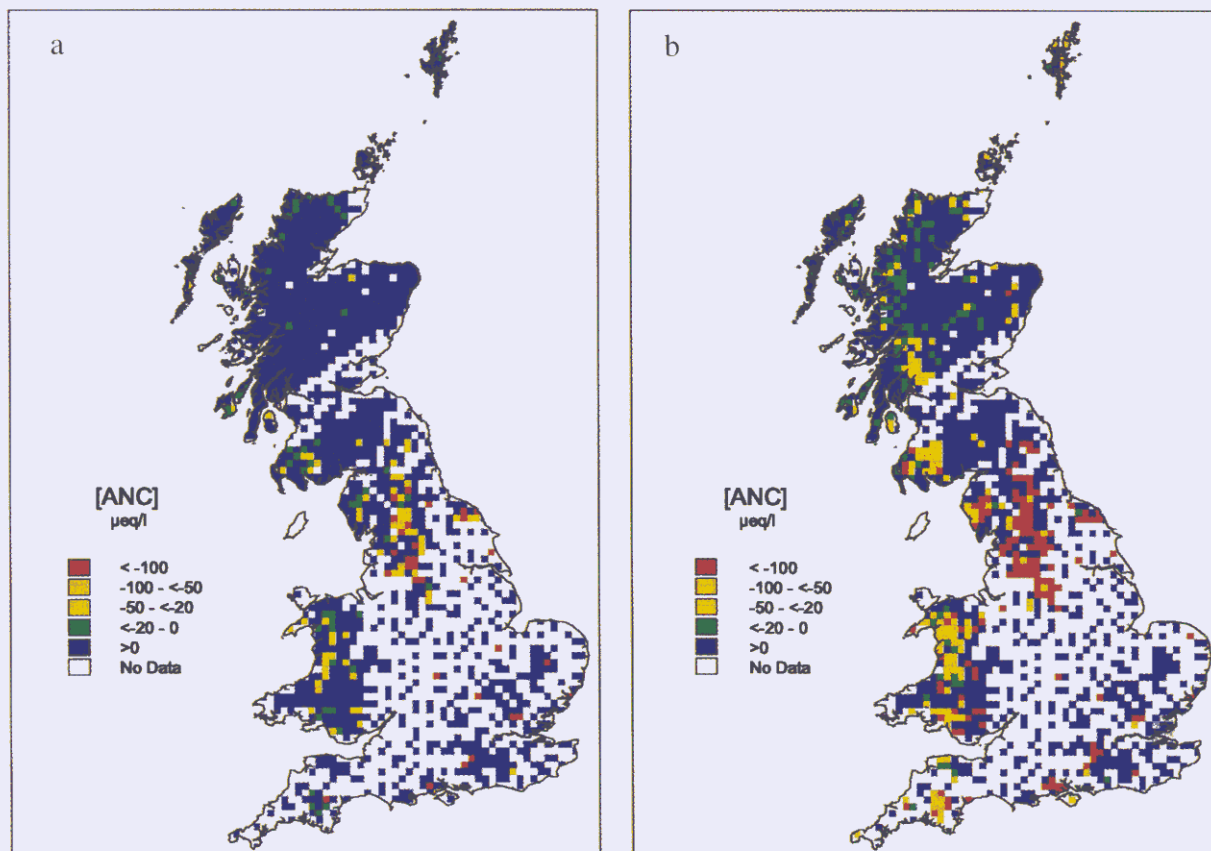


Fig. 5. Measured and modelled ANC in the FAB mapping dataset. (a) Measured ANC. (b) Predicted long-term ANC under mean 1992–94 deposition load of S and N.

ANC associated with this change, and this is ultimately what the freshwater critical load models use to define exceedance. A site with a predicted ANC of less than  $0 \mu\text{eq l}^{-1}$  induced by a given acid deposition load is exceeding its critical load, even though current measured ANC may be greater than zero until steady-state is achieved.

Figure 5a shows that apart from a few isolated sites scattered around the country, water bodies with negative ANC values generally occur in the recognised areas of acidification (see Fig. 2); Galloway in Scotland, the Lake District, Pennines and North Yorkshire Moors of northern England, and the uplands of Wales. The great majority of fresh waters have a positive ANC, although in north west Scotland there are very few sites with an ANC greater than  $20 \mu\text{eq l}^{-1}$ , indicating a region of extreme sensitivity to acidification. In sites with positive but low ANC, the impact of increased acid anion leaching is likely to be most significant in terms of causing critical load exceedance.

The charge balance of the FAB model can be employed to calculate the potential, future ANC at a site under a constant deposition load. Future sulphate and nitrate leaching are derived as above, while the base cation leaching from sites exceeding their critical load is provided by the SSWC

model. The SSWC model assumes that the only permanent, sustainable source of base cations is provided by weathering, so that despite current elevated rates of base cation leaching through cation exchange processes with acid inputs, base cation concentrations under a constant deposition load and runoff will decline to the value of  $[\text{BC}]_0^*$  where the SSWC critical load is exceeded (see above). In the FAB model, the potential, future ANC concentration ( $[\text{ANC}]_{\text{pot}}$ ) for exceeded sites is then provided by the difference between future base cation and acid anion concentrations:

$$[\text{ANC}]_{\text{pot}} = [\text{BC}]_0^* - (S_{\text{leach}} + N_{\text{leach}})/Q \quad (13)$$

where  $S_{\text{leach}}$  and  $N_{\text{leach}}$  are derived as in Equations 11–12. If the critical load is not exceeded in the FAB model, then the predicted increase in acid anion leaching will be partially offset by increased base cation leaching according to the “F-factor” (Brakke *et al.*, 1990). Such non-exceeded sites will, however, continue to maintain a positive ANC and are therefore not considered further.

The predicted increase in acid anion leaching under current deposition loads will lead to a decrease in ANC at many sites if no emissions abatement is implemented (Fig.

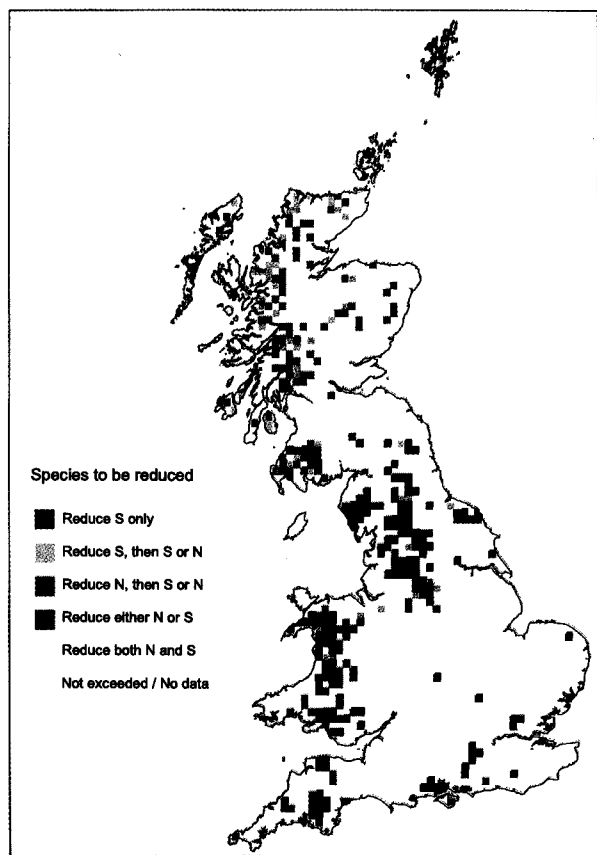


Fig. 6. Deposition reduction requirements modelled by FAB using ITE 1992–94 deposition data.

5b). In general, for England and Wales the reductions in ANC predicted by the FAB model are most apparent at sites with measured ANC values of less than zero (Fig. 5a), i.e. at sites which are already acidified. There is effectively a “consolidation” of acidified regions, with an increased spatial distribution of acidified sites as well as greater impacts at already acidified sites. In Wales, the region of acidification expands in area from parts of north and mid-Wales to most of the uplands in the whole country. In parts of central and northern Scotland, small increases in nitrate leaching, combined with extremely sensitive, dilute water bodies, will lead to a switching of ANC from positive to negative values, potentially with serious effects on the fisheries in these sensitive areas. Less significant is the further reduction in ANC throughout the Pennine region; fish are already absent from many water bodies there which have been severely acidified for many years.

Table 1. Deposition reduction requirements modelled by FAB under current deposition loads

Acid species requiring reduction	Number of sites			
	England	Scotland	Wales	Total
None (not exceeded)	412	586	86	1084
S only	0	6	0	6
S, then S or N	9	61	4	74
Both S and N	100	20	35	155
Either S or N	33	79	32	144
N, then S or N	4	2	1	7
<b>Total exceeded</b>	<b>146</b>	<b>173</b>	<b>72</b>	<b>386</b>
<b>Total number of sites</b>	<b>558</b>	<b>754</b>	<b>158</b>	<b>1470</b>
<b>Percent exceeded</b>	<b>26</b>	<b>22</b>	<b>46</b>	<b>26</b>

DEPOSITION REDUCTION REQUIREMENTS ACCORDING TO THE FAB MODEL

While the prediction of a negative ANC with the FAB model equates to a critical load exceedance, the utility of the FAB model lies also in the identification of the acid species which needs to be reduced to protect the site. If each site is colour coded according to its location on the critical load function (CLF—see Fig. 1) then a national map of deposition reduction requirements can be produced (Fig. 6).

Comparison of current deposition data with the critical load function indicates a regional pattern in the relative importance of S and N deposition. Many Scottish sites exceeding their critical loads require a mandatory reduction in S deposition to attain non-exceedance, i.e. sulphur alone is sufficient to cause critical load exceedance (Fig. 6). At most of the remaining exceeded sites in Scotland, reductions in S would provide sufficient protection, although N deposition could also be reduced to achieve the same outcome. It would therefore be possible to protect the majority of the Scottish sites through abatement of S emissions alone, without the need for recourse to N

Table 2. Relative contributions of S and N deposition to critical load exceedance

Acid species requiring reduction	Percentage of exceeded sites			
	England	Scotland	Wales	Total
Mandatory S	75	50	54	61
Mandatory N	71	13	50	42
Either S or N	23	46	44	37
Mandatory S and N	68	12	49	40

reductions. The only area of Scotland where N reductions are compulsory to protect freshwaters is the highly impacted Galloway region (Fig. 2).

In large areas of the acid sensitive regions of England and Wales defined in Fig. 2, both S and N deposition must be reduced if the sampled sites are to be protected in the long term. Nitrate is equally important an acid anion as sulphate in these areas, and the control of sulphur emissions alone, even with (unfeasible) reductions to zero, would be insufficient to protect these sites from acidification. The implication is that currently agreed international commitments to reduce S emissions under the Second Sulphur Protocol (UNECE, 1994) will be of limited benefit to the most sensitive freshwaters in Britain unless N deposition is also reduced.

The number of freshwater sites falling into each deposition reduction category identified on the CLF for the current deposition levels indicates the requirement for emissions reductions strategies (Table 1). Of the sampled population of 1470 lake and stream sites, 386 sites (26%) exceed their critical loads under current deposition levels. In terms of the proportion of water bodies with critical load exceedance, Wales is the most impacted, with 46% of sampled sites exceeded. Scotland and England have greater numbers of exceeded sites in absolute terms (Table 1), but smaller proportions with 22% and 26% exceeded, respectively. Note that these percentages refer to sampled sites, not the whole population of water bodies in these countries.

The proportions of sites affected by S or N deposition, or both, are given in Table 2, where the regional nature of deposition effects is exemplified. In England, 75% of exceeded sites require mandatory reductions in S deposition to be protected from acidification beyond the critical chemical value (below ANC = 0  $\mu\text{eq l}^{-1}$ ). However, most of these sites also require reductions in N deposition; 68% of exceeded sites would still be exceeded if S or N deposition were reduced to zero and the other remained constant. In England, therefore, nitrogen presents as great a problem as sulphur, at least in terms of numbers of sites affected. Expected reductions in S deposition under the Second Sulphur Protocol by 2010 (UNECE, 1994) may reduce the magnitude of exceedance at these sites, but will not suffice to protect them from acidification without reductions in N deposition.

In Scotland, a rather different situation is apparent. While the proportion of exceeded Scottish sample sites requiring mandatory S reductions is smaller than in England at 50%, the proportion requiring mandatory reductions in N deposition is negligible in relative terms, at just 13% (mostly in Galloway) compared with 71% in England (Table 2). In other words, sulphur is much more important in Scotland than nitrogen deposition and, with sufficiently large reductions in S deposition, up to 87% of currently exceeded sample sites could be protected. With no change in S deposition, reductions in N deposition could at best protect 50% of exceeded sites in the Scottish dataset.

Future reductions in S deposition under the Second Sulphur Protocol should therefore be much more successful at protecting surface waters in Scotland than in England.

The situation in Wales is closer to that of England than of Scotland. As in England, nitrogen and sulphur deposition are of almost equal importance, when assessed in terms of numbers of sites, in causing critical load exceedance. However, the proportion of exceeded sites at which reductions in both S and N deposition are required is smaller, at 49% compared with 68% in England (Table 2). Future reductions in S deposition could at best protect 50% of currently exceeded Welsh sample sites without any reduction in N deposition.

## Discussion

The results of the national model application have to be considered in the context of the uncertainties associated with the model structure, the catchment datasets and the deposition data used.

### UNCERTAINTIES ASSOCIATED WITH CRITICAL LOAD MODELS FOR FRESHWATERS

The input parameters to the FAB model, where limited empirical data have been used to specify various process rates at the national scale, will have associated errors which can only be reduced by further empirical study and revision of default values.

The nitrogen uptake term ( $N_{\text{upt}}$ ) incorporates two sources of uncertainty; the estimate of forest cover from satellite imagery and the single rate of N uptake selected for all coniferous forest areas. While these uncertainties are difficult to quantify, the affected proportion of the mapping dataset is small; only 12% of the 1470 catchments have an estimated coniferous forest cover of more than a tenth, and only two percent of catchments have more than half of their area afforested with conifers (CLAG, unpublished data).

The denitrification component of the FAB model is another source of uncertainty. The default terms used for British soil types are literature-based estimates (Hall *et al.*, 1997), and very few data are available for moorland sites in Britain. Current research efforts are therefore directed towards the measurement of denitrification rates from moorland soils in both the field and the laboratory.

Under current N deposition levels, N immobilisation in soil biomass may occur at elevated rates until N saturation occurs, i.e. until some other essential resource is limiting (Aber *et al.*, 1989), which may be reflected in decreased soil carbon:nitrogen ratios and increased nitrate leaching in the future (Dise and Wright, 1995). While this process could explain the very high rates of N retention currently

observed in many upland catchments, it poses a problem in the definition of critical loads, whereby a long-term steady-state is assumed. The approach adopted at the European level is to utilise the long-term estimates of soil N immobilisation obtained from chronosequence studies (UBA, 1996) which provide a much smaller figure ( $0.5\text{--}1.0\text{ kgN ha}^{-1}\text{ yr}^{-1}$ ) for immobilisation than current catchment N budgets would indicate. The dynamics of N immobilisation processes are a focus of current experimental research.

One specific source of error within the FAB model is the assumption that all acid deposition to a catchment passes through the terrestrial system and is thereby exposed to the various soil and vegetation sinks which act to remove nitrogen. Direct deposition to the lake surface is assumed to be negligible, but for certain water bodies in areas of very low relief, for example in the Flow Country of northern Scotland, the lake surface may form a major fraction of the total catchment area, in some cases as high as forty percent. While such cases are rare, the critical loads calculated with the FAB model will be significant overestimates for these sites.

#### UNCERTAINTIES ASSOCIATED WITH DEPOSITION DATA

There are several potential sources of uncertainty associated with the deposition data used, including interpolation (kriging) error, uncertainties in dry deposition velocity and precipitation amount and uncertainties associated with seeder-feeder enhancement of wet deposition at scales smaller than the  $20 \times 20\text{ km}$  grid employed (CLAG Deposition Fluxes, 1997). The uncertainty associated with deposition inputs into a  $20\text{ km}$  grid cell has been estimated as  $\pm 40\%$  for total sulphur over much of the country, rising to  $\pm 60\%$  towards the north and west of Britain (CLAG Deposition Fluxes, 1997). The uncertainties in N deposition fluxes to a  $20 \times 20\text{ km}$  grid square, and those associated with local topographical and land cover variations within a  $20 \times 20\text{ km}$  area, are unknown.

Another uncertainty relates to the issue of data resolution. While FAB is a catchment based model, with catchment specific estimates of soil and vegetation cover, the deposition and runoff data are not catchment specific. Mean deposition estimates are produced on a  $20 \times 20\text{ km}$  grid, while the mean total catchment area for sites in the mapping database is  $2.4\text{ km}^2$ . The uncertainty associated with the estimation of catchment fluxes from grid-based deposition and runoff data is unquantified.

#### INTERPRETATION OF FAB MODEL OUTPUTS

Interpretation of the chemical predictions and deposition reduction requirements generated by the FAB model requires an understanding of several key factors.

#### *The magnitude of critical load exceedance and deposition reduction requirements*

The national FAB map of deposition reduction requirements (Fig. 6) does not indicate the magnitude of critical load exceedance in terms of a deposition flux of acidity, because this notional figure cannot be interpreted directly as the size of the deposition reduction requirement. The reduction requirement for S or N deposition will inevitably be greater, in terms of equivalents of acidity, than the amount by which the critical load is exceeded, because of the catchment processes which retain a proportion of the inputs. Furthermore, because of the inter-dependence of S and N deposition in causing critical load exceedance, the FAB model cannot provide an absolute requirement for deposition reductions in either S or N beyond the upper deposition thresholds of  $CL_{\text{max}}(\text{S})$  and  $CL_{\text{max}}(\text{N})$  (Fig. 1), unless external constraints, for example costs, are imposed. Where the deposition load of either species lies below these thresholds, the reduction requirement for one cannot be specified without reference to the other. In other words, while the FAB model output indicates the options for reducing acid deposition in terms of S or N, it cannot provide independent figures for either. The utility of the model therefore lies in deposition scenario assessment, whereby the effect of given combinations of S and N deposition can be assessed to determine whether they provide adequate protection to the freshwater sites of concern. One such application of the model is described in Curtis *et al.* (1999) where the levels of protection afforded by three deposition scenarios (baseline, planned and potential) are compared.

#### *The timing of acidification damage*

A state of critical load exceedance does not necessarily mean that the affected water body has already been acidified or even significantly affected as yet by the excess acid deposition. If the critical load of a site is currently exceeded, then the critical chemical threshold will be crossed at some unspecified point in the future, i.e. *the current deposition load cannot be sustained if the site is to be protected*. In both the SSWC and FAB models, critical load exceedance is therefore not a function of current damage, but of *potential* future damage, which is explicit in the critical loads concept. The actual current state of a water body for which the critical load is exceeded is determined by such factors as deposition history (total cumulative acid load) and catchment characteristics like soil type and land cover, so that only dynamic models like MAGIC which incorporate such factors can predict the timing of acidification to a given chemical state (e.g. Jenkins *et al.*, 1997).

#### *Extrapolation of exceedance map to other freshwater sites*

Since the freshwaters sampling strategy focused on the most sensitive sites, rather than a randomly selected representative subsample, the results of the modelling exercise cannot

be extrapolated to the whole population of British water bodies. By definition, the selection of the most sensitive site in each  $10 \times 10$  km grid square should mean that, where the selected site is protected, the associated water bodies in the same grid square are also protected because they are less sensitive and will therefore have higher critical loads. This uncertainty was quantified by Curtis *et al.* (1995) who found that in SSWC model applications to the British freshwaters mapping dataset, the most sensitive site in a  $10 \times 10$  km grid square was correctly identified in one third of cases, while the selected site correctly represented the most sensitive critical load class of water bodies in the grid square in two thirds of cases. The implication here is that in around two thirds of mapped grid squares where critical loads are exceeded, there may be water bodies experiencing an even greater critical load exceedance than the mapped site.

## Conclusions

The national application of the FAB model to a dataset of 1470 British freshwaters indicates that more than a quarter of sampled sites might be damaged in the long term if the deposition of acidity is not sufficiently reduced. There is a distinct regional pattern in the relative importance of S or N deposition in causing acidification.

The uplands of northern England and Wales would experience the most severe acidification, mostly through increases in nitrate leaching. Ironically, many of the sites predicted to be most seriously impacted in the Pennine region are already acidified and fishless, so any further decrease in ANC cannot affect the (absent) target organism, brown trout. Potentially more serious is the decline in ANC predicted for many sites in upland Wales and the Lake District of northern England, where sites are currently less acidic and more likely to hold vulnerable fisheries. Perhaps most striking is the predicted decrease in ANC to less than  $0 \mu\text{eq l}^{-1}$  in sites across large parts of western Scotland, because these sites currently have positive ANC values and this region has not yet shown evidence of serious acidification damage to fisheries (Harriman *et al.*, 1995). Unlike England and Wales, the future acidification of Scottish freshwaters is determined largely by S deposition.

While previous (and ongoing) international efforts have been directed towards a strategy for the control of S emissions, the FAB model indicates that S emissions reduction alone will be insufficient to protect sites in much of upland England and Wales because of the magnitude of N deposition. Only the Scottish sample sites could be protected by S reductions alone, depending on the deposition levels attained. There is an evident need for reductions in N deposition across most of Great Britain if any benefits from future decreases in S deposition are to be realised. The potential effects of different possible scenarios for future reductions in S and N deposition have been

assessed using the FAB model in conjunction with modelled deposition data (Curtis *et al.*, 1999).

There is considerable uncertainty associated with the FAB model input data. The use of gridded data at various scales, from 25m square satellite derived landcover classes to 20km square interpolated deposition data, for catchment based estimates will have an associated error which is not quantified. In particular, the N sink terms are largely based on literature-derived values while very few data are available for the moorland systems which are typical of the more acid-sensitive British uplands. However, the resolution of national datasets is continually improving, providing more reliable estimates for catchment based parameters, while the processes responsible for the mismatch between measured and predicted nitrate concentrations are the focus of current intensive experimental work. Model parameterisation should therefore be subject to adaptation and improvement for application to British freshwaters.

The FAB model cannot predict the timescales over which acidification damage might occur, but this national application does provide a coherent, regional picture of the potential magnitude of change in sampled lakes and streams, and a graphic illustration of where sulphur or nitrogen deposition, individually or in combination, must be reduced from 1992–94 levels to protect these sensitive ecosystems. The signing of the Gothenburg Protocol in December 1999 has committed many European countries to reductions in S emissions beyond those required in the Oslo Protocol and also to substantial reductions in N emissions. The deposition levels and corresponding critical load exceedances which will result from these emissions reductions will become the focus of future modelling work.

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## References

- Aber, J.D., Nadelhoffer, K.D., Steudler, P. and Melillo, J.M., 1989. Nitrogen saturation in Northern forest ecosystems. *BioScience* 39, 378–386.
- Allott, T.E.H., Curtis, C.J., Hall, J., Harriman, R. and Battarbee,

- R.W., 1995. The impact of nitrogen deposition on upland freshwaters in Great Britain: a regional assessment. *Water Air Soil Pollut.* 85, 297–302.
- Brakke, D.F., Henriksen, A. and Norton, S.A., 1990. A variable F-factor to explain changes in base cation concentrations as a function of strong acid deposition. *Verh. int. Ver. Limnol.* 24, 146–149.
- Bull, K.R., 1995. Critical loads – possibilities and constraints. *Water Air Soil Pollut.* 85, 201–212.
- CLAG, 1994. *Critical Loads of Acidity in the United Kingdom*. UK Critical Loads Advisory Group, Summary Report to the Department of the Environment, February 1994. ITE, Penicuik. 61 pp.
- CLAG Deposition Fluxes, 1997. *Deposition fluxes of acidifying compounds in the United Kingdom*. Critical Loads Advisory Group, Sub-group report on Deposition Fluxes. ITE, Penicuik, 45 pp.
- CLAG Freshwaters, 1995. *Critical Loads of Acid Deposition for United Kingdom Freshwaters*. Critical Loads Advisory Group, Sub-group report on Freshwaters. ITE, Penicuik. 80 pp.
- Curtis, C.J., Allott, T.E.H., Battarbee, R.W. and Harriman, R., 1995. Validation of the UK critical loads for freshwaters: site selection and sensitivity. *Water Air Soil Pollut.* 85, 2467–2472.
- Curtis, C.J., Allott, T.E.H., Battarbee, R.W., Harriman, R. and Hall, J., 1996. *Screening and validation of the empirical maps for UK freshwater critical loads*. ECRC Research Papers No. 12, University College London, 120 pp.
- Curtis, C.J., Allott, T.E.H., Reynolds, B. and Harriman, R., 1998. The prediction of nitrate leaching with the first-order acidity balance (FAB) model for upland catchments in Great Britain. *Water Air Soil Pollut.* 105, 205–215.
- Curtis, C.J., Whyatt, J.D., Metcalfe, S.E., Allott, T.E.H. and Harriman, R., (1999) Assessing the impacts of international emissions reduction scenarios on the acidification of freshwaters in Great Britain with the First-order Acidity Balance (FAB) model and the Hull Acid Rain Model (HARM). *Energy Environ.* 10, 571–596.
- Dillon, P.J. and Molot, L.A., 1990. The role of ammonium and nitrate retention in the acidification of lakes and forested catchments. *Biogeochemistry* 11, 23–43.
- Dise, N.B. and Wright, R.F., 1995. Nitrogen leaching from European forests in relation to nitrogen deposition. *Forest Ecol. Manage.* 71, 153–161.
- Emmett, B.A., Reynolds, B., Stevens, P.A., Norris, D.A., Hughes, S., Görres, J. and Lubrecht, I., 1993. Nitrate leaching from afforested Welsh catchments – interactions between stand age and nitrogen deposition. *Ambio* 22, 386–394.
- Emmett, B.A. and Reynolds, B., 1996. Nitrogen critical loads for spruce plantations in Wales: is there too much nitrogen? *Forestry* 69, 205–214.
- Forsius, M., Kämäri, J. and Posch, M., 1992. Critical loads for Finnish lakes: comparison of three steady-state models. *Environ. Pollut.* 77, 185–193.
- Fuller, R.M., Groom, G.B. and Jones, A.R., 1994. The Land Cover Map of Great Britain: an automated classification of Landsat Thematic Mapper data. *Photogramm. Eng. Rem. Sens.* 60, 553–562.
- Hall, J., Hornung, M., Freer-Smith, P., Loveland, P., Bradley, I., Langan, S., Dyke, H., Gascoigne, J. and Bull, K., 1997. *Current status of UK critical loads data – December 1996*. ITE Monks Wood, UK, 22 pp. plus appendices.
- Harriman, R., Gillespie, E., King, D., Watt, A.W., Christie, A.E.G., Cowan, A.A. and Edwards, T., 1990. Short-term ionic responses as indicators of hydrochemical processes in the Allt a'Mharcaidh catchment, western Cairngorms, Scotland. *J. Hydrol.* 116, 267–285.
- Harriman, R. and Christie, A.E.G., 1995. Estimating critical loads for biota: the steady-state water chemistry (Henriksen) model. In: *Critical Loads of Acid Deposition for United Kingdom Freshwaters*. Critical Loads Advisory Group, Sub-group report on Freshwaters. ITE, Penicuik, U.K. 7–8.
- Harriman, R., Bridcut, E.E. and Anderson, H., 1995. The relationship between salmonid fish densities and critical ANC at exceeded and non-exceeded stream sites in Scotland. *Water Air Soil Pollut.* 85, 2455–2460.
- Henriksen, A., 1984. Changes in base cation concentration due to freshwater acidification. *Verh. int. Ver. Limnol.* 22, 692–698.
- Henriksen, A., 1998. *Application of the first-order acidity balance (FAB) model to Norwegian surface waters*. Report SNO 3809–98, Norwegian Institute for Water Research (NIVA), Oslo, Norway, 33 pp.
- Henriksen, A., Lien, L. and Traaen, T.S., 1990. *Critical loads for surface waters – Chemical criteria for inputs of strong acids*. Report 22/1990, Norwegian Institute for Water Research (NIVA), Oslo, Norway.
- Henriksen, A., Kämäri, J., Posch, M. and Wilander, A., 1992. Critical loads of acidity: Nordic surface waters. *Ambio* 21, 356–363.
- Henriksen, A., Posch, M., Hultberg, H. and Lien, L., 1995. Critical loads of acidity for Nordic surface waters – can the ANC<sub>limit</sub> be considered variable? *Water Air Soil Pollut.* 85, 2419–2424.
- Henriksen, A., Hindar, A., Hessen, D. and Kaste, Ø., 1997. Contribution of nitrogen to acidity in the Bjerkreim River in Southwestern Norway. *Ambio* 26, 304–311.
- Hornung, M., Bull, K.R., Cresser, M., Ullyett, J., Hall, J.R., Langan, S., Loveland, P.J. and Wilson, M.J., 1995. The sensitivity of surface waters of Great Britain to acidification predicted from catchment characteristics. *Environ. Pollut.* 87, 207–214.
- INDITE, 1994. *Impacts of nitrogen deposition in terrestrial ecosystems. Report of the United Kingdom Review Group on Impacts of Atmospheric Nitrogen*. UK Department of the Environment, London, 110 pp.
- Jenkins, A., Renshaw, M., Helliwell, R., Sefton, C., Ferrier, R. and Swingewood, P., 1997. *Modelling surface water acidification in the UK*. Report No. 131, Institute of Hydrology, Wallingford, UK, 54 pp.
- Kämäri, J., Amann, M., Brodin, Y.-W., Chadwick, M.J., Henriksen, A., Hettelingh, J.-P., Kuylenstierna, J., Posch, M. and Sverdrup, H., 1992. The use of critical loads for the assessment of future alternatives to acidification. *Ambio* 21, 377–386.
- Kämäri, J., Forsius, M. and Posch, M., 1993. Critical loads of sulfur and nitrogen for lakes II: regional extent and variability in Finland. *Water Air Soil Pollut.* 66, 77–96.
- Kelly, C.A., Rudd, J.W.M., Hesslein, R.H., Schindler, D.W., Dillon, P.J., Driscoll, C.T., Gherini, S.A. and Hecky, R.E., 1987. Prediction of biological acid neutralization in acid sensitive lakes. *Biogeochemistry* 3, 129–140.
- Kreiser, A.M., Patrick, S.T. and Battarbee, R.W., 1993. In *Critical Loads: Concept and Applications*. M. Hornung and R.A. Skeffington (Eds.). ITE Symposium No. 28, HMSO, London, 94–98.
- Langan, S.J. and Wilson, M.J., 1994. Critical loads of acid deposition on Scottish soils. *Water Air Soil Pollut.* 75, 177–191.
- Lien, L., Raddum, G.G. and Fjellheim, A., 1992. *Critical loads of acidity to freshwater-fish and invertebrates*. Fagrapport No. 23, NIVA, Oslo, Norway, 36 pp.
- Posch, M., Kämäri, J., Forsius, M., Henriksen, A. and Wilander, A., 1997. Exceedance of critical loads for lakes in Finland, Norway, and Sweden: reduction requirements for acidifying nitrogen and sulphur deposition. *Environ. Manage.* 21, 291–304.
- Posch, M., de Smet, P.A.M., Hettelingh, J.-P. and Downing, R.J., 1999. *Calculation and mapping of critical thresholds in Europe*. Status Report 1999, Co-ordination Centre for Effects. RIVM Report No. 259101009.
- Reynolds, B., Ormerod, S.J. and Gee, A.S., 1994. Spatial patterns in stream nitrate concentrations in upland Wales in relation to catchment forest cover and forest age. *Environ. Pollut.* 84, 27–33.
- RGAR, 1997. *Acid deposition in the United Kingdom 1992–1994*. Fourth Report of the Review Group on Acid Rain, June 1997. AEA Technology plc, Culham, UK, 176 pp.
- Seip, H.M., 1980. Acidification of freshwater – sources and mechanisms. In *Ecological impact of acid precipitation*, D. Drablos and A. Tollan (Eds.). Proceedings of an international con-

- ference, Sandefjord, Norway, March 11-14, 1980. SNSF Project, Oslo, Norway, 358-365.
- Stoddard, J.L., 1994. Long-term changes in watershed retention of nitrogen: its causes and aquatic consequences. In *Environmental Chemistry of Lakes and Reservoirs*, L.A. Baker (Ed.), ACS Advances in Chemistry Series No. 237, American Chemical Society, Washington, D.C., 223-284.
- Sverdrup, H., Warfvinge, P. and Nihlgård, B., 1994. Assessment of soil acidification effects on forest growth in Sweden. *Water Air Soil Pollut.* 78, 1-36.
- UBA, 1996. *Manual on methodologies and criteria for mapping critical levels/loads and geographical areas where they are exceeded.* Report for the UN ECE Convention on Long-Range Transboundary Air Pollution. Texte 71/96, Federal Environment Agency (Umweltbundesamt), Berlin, Germany, 142 pp. plus appendices.
- UNECE, 1994. *Protocol to the 1979 Convention on Long Range Transboundary Air Pollution on Further Reduction of Sulphur Emissions.* Document ECE/EB.AIR/40. UNECE, New York and Geneva, 106 pp.