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Appendix 1 – Overview of methods to calculate critical loads

Appendix 1A - Critical loads of acidity for soils and terrestrial habitats

Two methods are used in the UK for calculating acidity critical loads for terrestrial habitats: the empirical approach is used for non-woodland habitats and the simple mass balance (SMB) equation for woodland ecosystems. Both methods provide critical loads for systems at steady-state. These methods are described briefly below.

1A.1 Methods

Empirical critical loads of acidity for soils

Mineral weathering in soils provides the main long-term sink for deposited acidity. Using this principle, critical loads of acidity can be based on the amount of acid deposition which could be buffered by the annual production of base cations from mineral weathering (Nilsson & Grennfelt, 1988).

In the UK, empirical critical loads of acidity for soils have been assigned to each 1km grid square of the country based upon the mineralogy and chemistry of the dominant soil (series or map unit) present in the grid square (Hornung *et al.*, 1995). The data are mapped in five classes representing ranges of critical load values, with low critical loads for soils dominated by minerals such as quartz and high critical loads for soils containing free carbonates. Where a single critical load value is required, for example, when calculating the excess deposition above the critical load (ie, the exceedance), the mid-range values are applied, with the exception of those with the highest critical load, where the value at the top of the range is used (Hall *et al.*, 1998). However, this classification, based on weathering rates and mineralogy, is inappropriate for peat soils, which contain little mineral material. For these, the critical load was set based on a critical soil solution pH of 4.4 (Main Report, Section 4.2.2). Together, these empirical critical loads are assigned to the 1km grid squares of the country representing the sensitive non-woodland terrestrial BAP Broad Habitats. The distribution and mapping of these habitats are described in the Main Report (Main Report, Chapter 3).

The application of these methods in the UK represent a precautionary approach, setting the critical loads for mineral soils to prevent any further change in soil chemistry as a result of deposited acidity (Hornung *et al.*, 1997).

The simple mass balance (SMB) equation for calculating acidity critical loads for woodland ecosystems

The SMB equation is the most commonly used model in Europe for the calculation of acidity critical loads for woodland ecosystems. This model is based on balancing the acidic inputs to and outputs from a system, to derive a critical load that ensures a critical chemical limit (related to effects on the ecosystem) is not exceeded (Sverdrup *et al.*, 1990, Sverdrup & De Vries, 1994). The equation has been derived from a charge balance of ions in leaching fluxes from the soil compartment, combined with mass balance equations for the inputs, sinks, sources and outputs of sulphur and nitrogen (Posch *et al.*, 1995).

In the UK we apply the SMB equation to coniferous and broadleaved woodland habitats, except for wooded areas on peat soils, where the SMB is inappropriate; in such areas the acidity critical loads for peat soils are applied. The application of the SMB equation to non-woodland systems needs further development and testing because of uncertainties in the applicability of the critical chemical criteria to other ecosystems.

The SMB equation is parameterised according to the appropriate critical chemical criteria and critical limits that will protect the receptor from the adverse effects of acidification. A critical molar ratio of calcium to aluminium of one in soil solution is a common criterion applied in the SMB to protect the fine roots of trees. This criterion is more appropriate for mineral soils than organic soils; for the latter a critical pH is considered to be more suitable (Hall *et al.*, 2001a). Therefore, in the UK we have parameterised the model to use a Ca:Al=1 criterion for woodland on mineral soils and a critical pH of 4.0 as the criterion for woodland on organic soils. The equations currently being used are given below. In the case of peat soils, the empirical critical loads are still applied.

1A.2 SMB equations

1. SMB equation using Ca:Al ratio as chemical criterion (mineral soils).
NB. Base cation (BC) terms here relate to calcium only.

$CL(A) = ANC_w - ANC_{le(crit)}$
Where:

CL(A)	= *critical loads of acidity (calculated in eq ha^{-1} year^{-1})* *[using units given here divide* CL(A) *by 1000 to give keq ha^{-1} year^{-1}]*
ANC_w	= *Acid Neutralising Capacity produced by weathering (eq ha^{-1} year^{-1}) (base cation weathering)*
$ANC_{le(crit)}$	= *critical leaching of ANC (eq ha^{-1} year^{-1})* = $-Al_{le(crit)} - H_{le(crit)}$
$Al_{le(crit)}$	= *critical leaching of Aluminium (eq ha^{-1} year^{-1})* = $((1.5 * BC_{le}) / Ca{:}Al) * 1000$
BC_{le}	= *calcium leaching (keq ha^{-1} year^{-1})* = $BC_a - BC_u$
BC_u	= *net uptake of calcium (keq ha^{-1} year^{-1})* = *minimum (*u, BC_a*)*
u	= *calcium uptake (keq ha^{-1} year^{-1}), see values in Table 2.*
BC_a	= *calcium availability (keq ha^{-1} year^{-1})* = *maximum (*$Ca_w + Ca_{dep} - BC_{lemin}$, *0)*
Ca_w	= *calcium weathering (keq ha^{-1} year^{-1})*
Ca_{dep}	= *total (marine plus non-marine) calcium deposition for woodland 1995-97 (keq ha^{-1} year^{-1})*
BC_{lemin}	= *minimum calcium leaching (keq ha^{-1} year^{-1})* $Q * [BC_l] * 0.01$
Q	= *runoff (metres year^{-1})*
$[BC_l]$	= *limiting concentration for uptake of calcium (2μeq l^{-1})*
$H_{le(crit)}$	= *critical leaching of hydrogen ions (eq ha^{-1} year^{-1})* = $(1.5 * ((BC_{le} * 1000) / (K_{gibb} * Ca{:}Al)))^{1/3} * (Q * 10000)^{2/3}$
K_{gibb}	= *gibbsite equilibrium constant (mineral soils: 950 [m^6/eq^2])*
Ca:Al	= *Calcium:Aluminium ratio = 1*

2. SMB equation using critical pH as chemical criterion (organic soils).

$CL(A) = ANC_w - ANC_{le(crit)}$
Where:

CL(A)	= *critical loads of acidity (calculated in eq ha^{-1} $year^{-1}$)* *[using units given here divide* CL(A) *by 1000 to give keq ha^{-1} $year^{-1}$]*
ANC_w	= *Acid Neutralising Capacity produced by weathering (eq ha^{-1} $year^{-1}$) (base cation weathering)*
$ANC_{le(crit)}$	= *critical leaching of ANC (eq ha^{-1} $year^{-1}$)* = Q * ([H] + [Al])
Q	= *runoff (m^3 ha^{-1} = mm runoff * 10)*
[H]	= *hydrogen ion concentration (eq m^{-3})* = $10^{(-pH)}$ * 1000
pH	= *critical pH (4.0)*
[Al]	= *aluminium concentration (eq m^{-3})* = $K_{gibb} * H^3$
K_{gibb}	= *gibbsite equilibrium constant (organic soils: 9.5 [m^6/eq^2])*

1A.3 Critical Loads Function

In its simplest form, an acidity critical load can be defined graphically by a 45 degree diagonal line on a sulphur-nitrogen deposition plot (Appendix 4, Figure 5.1(a)). The line intercepts the x-axis (representing nitrogen deposition) and y-axis (representing sulphur deposition) at chemically equivalent points, each representing the nitrogen or sulphur deposition equal to the critical load for acidity. Each point along the diagonal line represents the critical load in terms of some combination of sulphur and nitrogen deposition.

To allow for the long-term nitrogen removal processes by the soil and through harvesting of vegetation, the simple diagonal line is shifted along the nitrogen axis to increase the nitrogen values across the entire acidity CLF (Appendix 4, Figure 5.1(b)). More nitrogen can then be deposited before the acidity critical load is exceeded. There are no similar removal processes that need to be considered for sulphur.

The intercepts of the CLF on the sulphur and nitrogen axes define the "maximum" critical loads of sulphur and nitrogen (Appendix 4, Figure 5.1(c)). The maximum critical load of sulphur (*CLmax(S)*) is the critical load for acidity expressed in terms of sulphur only, ie when nitrogen deposition is zero. Similarly, the maximum critical load of nitrogen (*CLmax(N)*) is the critical load of acidity expressed in terms of nitrogen only (when sulphur deposition is zero). The long-term nitrogen removal processes in the soil (for example, nitrogen uptake and immobilisation) define a "minimum" critical load for nitrogen (*CLmin(N)*, Appendix 4, Figure 5.1(c)).

The critical load values *CLmax(S), CLmax(N)* and *CLmin(N)* are calculated for each grid square, and together with the data used to calculate them, they form the data that the UK NFC is required to submit to the CCE.

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Appendix 1B - Critical loads of acidity for freshwaters

1B.1 Introduction

The First Order Acidity Balance (FAB) model continues to be used to calculate critical loads for freshwaters in the UK. It is applied to the set of lakes and streams described in the Main Report (Section 3.7.4). The FAB model is a charge balance model incorporating major processes affecting the acid anion budget for the lake/stream and catchment. It supersedes the Steady State Water Chemistry (SSWC) model, used until 1994 when sulphur was the main focus of attention for acidification of freshwaters. The main advantage of the FAB model is that it can be used to derive a steady state mass balance for nitrogen, taking account of the several key nitrogen processes in catchments, such as denitrification, nitrogen immobilisation, removal in vegetation and nitrogen retention in lakes.

While the SSWC model is no longer used directly for the calculation of critical loads for freshwaters, the FAB model that supersedes it employs identical methods for the calculation of pre-industrial base cation leaching, which is still fundamental to the application of FAB. Hence a description of the SSWC model and the key principles is included here.

The critical chemical criterion used to indicate the threshold for damage, and thus determine the critical load for freshwaters, is Acid Neutralising Capacity (ANC) (see 1B.2 and Main Report, Section 6.1). The most wide-ranging studies linking ANC to biological damage have been carried out in Norway, where hundreds of lakes have been surveyed for fish population data and water chemistry. These surveys provided the data for a widely used dose-response function linking ANC to probability of damage to brown trout populations (Lien *et al.*, 1992; 1996), where damage is defined as a reduction in fish populations. Since brown trout is a widespread and economically important species in UK fresh waters, it provides an ideal indicator species for national critical loads applications.

In Norway, the critical ANC selected for critical loads work is 20 μeql^{-1}, which represents only a 10% probability of damage to brown trout populations. In the UK, uncertainties regarding the occurrence of waters with natural, pre-industrial ANC values of less than 20 μeql^{-1} have meant that a lower critical ANC of zero has been used, providing less stringent critical loads but a lower probability of protection to brown trout. According to the Norwegian work, zero ANC represents a 50% probability of reduced brown trout populations, including a 10% probability of extinction. Ideally, fish and water chemistry survey data from the UK would be used to derive a dose-response function that is appropriate for UK waters, but these data are currently lacking at the national scale. Therefore debate is still ongoing over the applicability of the Norwegian dose-response function for UK waters, but it continues to be used in the absence of an alternative (see Section 6.3).

1B.2 ***Acid neutralising capacity (ANC): the critical chemical parameter***

ANC is operationally defined as the sum of base cations minus the sum of acid anions (Henriksen *et al.*, 1992):

$$ANC = \sum BC - \sum AA$$
$$= [Ca^{2+}] + [Mg^{2+}] + [K^{+}] + [Na^{+}] - [SO_4^{2-}] - [NO_3^{-}] - [Cl^{-}] \quad \textbf{(Equation 1)}$$

which is, when expressed in equivalents (μeql^{-1}), numerically equivalent to

$$ANC = [HCO_3^{-}] + [OA^{-}] - [H^{+}] - [Al^{n+}] \quad \textbf{(Equation 2)}$$

where $[HCO_3^{-}]$ = bicarbonate ion
$[OA^{-}]$ = organic anions
$[H^{+}]$ = hydrogen ion
$[Al^{n+}]$ = Σ positively charged Al species

This definition excludes ions which are generally present in relatively small concentrations (e.g. NH_4^{+}, Fe^{n+}, F^{-}), but in waters where these other ions are thought to be important, they should be included in the calculation of ANC to avoid a charge balance error (i.e. where the sum of all positively charged species does not equal the sum of all negatively charged species).

Since critical loads relate to acid deposition inputs only, the next stage is to quantify and remove the proportion of ions deriving from neutral sea-spray inputs so that the definition of ANC becomes:

$$ANC = [BC_t^{*}] - [AA_t^{*}] \quad \textbf{(Equation 3)}$$

where $[BC_t^{*}]$ is the current, measured sum of non-marine base cations (= $Ca^{*} + Mg^{*} + K^{*} + Na^{*}$) and $[AA_t^{*}]$ is the sum of non-marine acid anions (= $SO_4^{*} + NO_3$); * denotes the non-marine component. It is assumed that all chloride is derived from marine sources, so that the marine contribution of each ion may be subtracted as a proportion of measured chloride concentration from the known ratios of these ions in seawater, using "seasalt correction factors" for each ion.

In the UNECE Mapping Manual (UBA, 1996), seasalt correction factors are given as:

$$[Ca^{*}] = [Ca] - 0.037 \times [Cl]$$
$$[Mg^{*}] = [Mg] - 0.198 \times [Cl]$$
$$[Na^{*}] = [Na] - 0.858 \times [Cl]$$
$$[K^{*}] = [K] - 0.018 \times [Cl]$$
$$[SO_4^{*}] = [SO_4] - 0.103 \times [Cl]$$

(all concentrations in μeql^{-1})

The concentration of NO_3^{-} in sea spray is assumed to be negligible.

1B.3 The SSWC model formulation

The equation defining ANC forms the basis of the freshwater critical load modelling approach. The key to the SSWC model is the calculation of the sustainable supply of ANC, or the inherent buffering capacity of the system.

Empirical relationships are invoked to determine the pre-industrial concentration of base cations ($[BC_0^*]$) from weathering, and this effectively sets the long-term critical load because it represents the only source of base cations over the long term. The base cations released by weathering processes are numerically equivalent to the bicarbonate produced, and therefore indicate the sustainable rate of production of ANC that defines the critical load.

Given a pre-selected critical ANC value, the freshwater critical load is simply the input flux of acid anions from atmospheric deposition that gives the critical ANC when subtracted from the pre-industrial flux of base cations (Henriksen *et al.*, 1992):

$$\text{Critical load} = ([BC_0^*] - [ANC_{crit}]).Q \qquad \textbf{(Equation 4)}$$

Concentrations are multiplied by runoff (Q) from the site to convert them into fluxes. The critical load is therefore a critical flux of acid anions.

The steady-state water chemistry (SSWC) model employs certain assumptions and empirical relationships in order to determine the "permanent" buffering provided by the pre-industrial base cation concentration ($[BC]_0^*$) which is the sum of weathering supply ($[BC_w]$) plus base cation deposition ($[BC^*_{dep}]$) if it is assumed that base cation deposition has not significantly changed since pre-industrial times.

The first step is to quantify the proportion of measured base cation leaching which is derived from transient ion-exchange processes (BC_{ex}) and is proportional to the load of acid anions. This proportion of base cations per unit input of acid anions (as a surrogate for H^+) derived from ion exchange in the soil complex is represented in the SSWC model by the term "F", calculated according to the methodology of Brakke *et al.* (1990):

$$F = \sin\left(\frac{\Pi}{2}\frac{[BC]_t^*}{S}\right)$$

where $[BC]_t^*$ is measured non-marine base cation concentration and *S* is a constant which varies regionally according to geology, but from empirical studies is taken as 400 μeql^{-1} (Harriman and Christie, 1995). This constant determines the current non-marine base cation concentration which represents a catchment likely to be unaffected by acid deposition; when $[BC]_t^* = S$, F=1 and base cation leaching is increased by exactly the value of the acid anion load, resulting in no change in ANC in runoff. For values of $[BC]_t^*$ greater than S (400 μeql^{-1}) F is set to 1, since it would otherwise decrease again with $[BC]_t^*$ according to the sine function. F can be interpreted as an index of the "exchangeability" of base cations in the soil exchange complex of a catchment.

F is used to calculate the pre-industrial base cation concentration according to the following equation (Henriksen *et al.*, 1992):

$$[BC]_0^* = [BC]_t^* - F([AA]_t^* - [AA]_0^*) \qquad \textbf{(Equation 5)}$$

where $[AA]_0^*$ is the pre-acidification concentration of non-marine acid anions from weathering and natural atmospheric sources and the measured leaching rate of non-marine base cations ($[BC]_t^*$) represents the sum of weathering, non-marine deposition and ion-exchange sources (BC_{leach}). Data from near-pristine lakes in northern Scotland indicate that "background" concentrations of NO_3^- are close to zero, while "background" concentrations of SO_4^{2-} are determined from empirical relationships between base cations and sulphate in near-pristine lakes (see Henriksen *et al.*, 1990, 1992).

Since $[BC]_0^*$ is now known, the SSWC critical load can be defined by Equation 4. If the critical load of acid deposition is exceeded, BC_{leach} will decline from its current value $[BC]_t^*$ as the base cation pool in the soil exchange complex is depleted, until it reaches the concentration $[BC]_0^*$ when the ANC of the water body will cross the threshold concentration $[ANC_{crit}]$. The magnitude of critical load exceedance, expressed as a flux of acid anions, provides the theoretical ANC of the water body when BC_{leach} has declined to $[BC]_0^*$.

The critical load term derived using the SSWC model forms a key input to the FAB model. The major difference between the two models lies in their treatment of N retention and leaching processes.

1B.4 Treatment of nitrogen processes in critical loads models

With critical loads models for sulphur (SSWC) it was assumed that the SO_4^{2-} anion is mobile in catchments, and therefore S deposition would quickly reach steady-state with leaching into surface waters (Seip, 1980). For N, as a major nutrient, the situation is much more complex, since deposition inputs enter the terrestrial N cycle. Terrestrial processes can remove or immobilise N deposition over very long timescales or even permanently. For example, denitrification returns N to the atmosphere as N_2O, NO or N_2 and permanently neutralises the associated acid inputs. Microbial cycling of N can lead to immobilisation in refractory organic matter, again retaining the associated protons.

Catchment input-output budgets generally indicate that only a small proportion of N deposition is leached into surface waters with its associated protons (e.g. Curtis *et al.*, 1998). If the retention of a known proportion of N deposition represented a steady-state situation then there would be no problem modelling the effects of N deposition and setting a critical load for N. For example, if the current retention rate for N is 90%, it might be assumed that as the N deposition load changes, 10% of the new load will still be leached into surface waters, and this would be very easy to model. The SSWC model was adapted to take account of NO_3^- leaching by using measured NO_3^-, converted into a flux using runoff, as a measure of the contribution of N deposition to critical load exceedance (Kämäri *et al.*, 1992). This method makes no reference to actual N deposition, and hence cannot take account of possible changes in NO_3^- leaching under a different N deposition scenario.

Studies of terrestrial N dynamics indicate that a process of N saturation can occur, whereby N accumulates in the soil-vegetation system until it is present in excess of biological demand and leaching increases, until potentially almost all N deposition is leached. It cannot therefore be assumed that N leaching under a fixed deposition load will remain constant. Furthermore, it cannot be assumed that if the N deposition load changes, the proportion of N leaching will remain constant. This process of N saturation takes place over a timescale that is largely independent of the processes which affect S leaching and its associated cation exchange processes. N therefore adds a further dimension to the problem of defining a steady-state situation.

A steady-state critical loads model for total acidity that can be used for scenario testing therefore requires the quantification of N retention processes over the very long term. If the *sustainable* (as opposed to short-term) rates of N retention or removal can be quantified, then the critical load can be determined. In the formulation of process-oriented models such as FAB, best available knowledge (in terms of published data) has been used to derive a steady-state mass balance for N. Several key retention processes have been identified: denitrification, N immobilisation, N removal in vegetation (grazing, forestry, burning) and in-lake retention. The long-term, steady-state rates at which all these key processes operate for a given deposition load of N (and the equivalent terms for S) must be quantified if a critical load of total acidity is to be defined.

1B.5 General description of the FAB Model

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

$$\underbrace{\mathbf{N_{dep} + S_{dep}}}_{\textit{atmospheric inputs}} = \underbrace{\{\boldsymbol{f}\mathbf{N_{upt}} + \mathbf{(1-}\boldsymbol{r}\mathbf{)(N_{imm} + N_{den})} + \boldsymbol{r}\mathbf{(N_{ret} + S_{ret})}\}}_{\textit{“internal” catchment processes}} + \underbrace{\mathbf{AN_{leach}}}_{\textit{leaching export}}$$

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

All units are expressed in equivalents (moles of charge) per unit area and time. Braces enclose “internal” catchment processes, i.e. those terrestrial and in-lake processes which operate on acid anion inputs to control the net export in catchment runoff.

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

1. long term sinks of S 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. NH_4^+ leaching is negligible because any inputs are either taken up by the biota, adsorbed onto soils, or nitrified to NO_3^-.

The acid anion balance of the FAB model can provide the critical leaching rate of acid anions (critical $AN_{leach} = L_{crit}$) which will depress ANC below the pre-selected critical value (ANC_{crit}) as in the SSWC model (Henriksen *et al.*, 1992; see above). Therefore, at critical load, AN_{leach} can be substituted as:

$$L_{crit} = BC_{crit} - ANC_{crit}$$

(UBA, 1996).

The internal catchment processes affecting acid anion budgets introduced by the FAB model are linked to vegetation cover, soil type and catchment morphology. In order to calculate critical loads for S and N, the sink terms for acid anions have to be quantified, as described below.

1B.5.1 N_{upt} : net growth uptake (in forest)

The only permanent sinks for N in the biomass occur when there is some form of harvesting or removal of biomass from the catchment. Short term, seasonal processes which lead to biological uptake of N during the growing season do not form a net sink for N over the long term, since the N content of biomass is subject to biological cycling. There is no net assimilation of N within catchment biomass over the long term, because it is released on the death of plants through decomposition and mineralisation.

The main route for the removal of biomass from upland catchments is through the harvesting of forest and the removal of wood from the catchment. If there are estimates available for the N content of the relevant tree species in a catchment, and if the time period of the harvesting cycle is known, a long-term figure for the export of N can be derived. Ideally, forestry data (species, age, Yield Class) should be used to determine N removal rates on a local basis.

There are other possible routes for the removal of biomass from catchments; in grazed or burned moorland catchments there may be small net losses of N from other types of catchment vegetation in terms of weight gain of removed livestock and losses in smoke. If figures are available for N losses through these processes, they can be incorporated into the long-term N mass balance (e.g. INDITE, 1994).

Since N uptake and biomass removal processes are specific to certain vegetation types, the derivation of the mass balance requires both the N removal/loss rate per unit area, and the area of each specific vegetation type within the catchment. In the standard formulation of the FAB model, only forest harvesting is considered so the proportion of the catchment covered in forest (*f*) is required.

1B.5.2 *N_{imm} : long term immobilisation of N in catchment soils*

Estimates of the long term immobilisation of N in different soil types have been derived by the analysis of total N content of soil profiles, which 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. A suggested long-term range of net immobilisation rates (including N-fixation) is 2-5 kgN ha^{-1} yr^{-1} (Downing *et al.*, 1993, UBA, 1996), although it is noted that under present conditions with high growth due to elevated N deposition this range may extend to a much higher figure. Since N immobilisation rate varies with soil type, the relative proportion of the catchment covered by each soil type is required to determine the mean value for soils in the whole (terrestrial) catchment. Note that the term '*1-r*' is used to weight the mean immobilisation rate by the terrestrial part of the catchment only, because in-lake retention processes are considered separately.

1B.5.3 *Input-dependent 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. Net in-lake retention of N is calculated as:

$$rN_{ret} = \rho_N [N_{dep} - fN_{upt} - (1-r)(N_{imm} + N_{den})]$$

The "in-lake retention fraction" for N (ρ_N) is calculated from a kinetic equation accounting for water retention time:

$$\rho_N = \frac{S_N}{S_N + \zeta/\tau} = \frac{S_N}{S_N + Q/r}$$

where S_N is the mass transfer coefficient for N, ζ is mean lake depth, τ is water residence time in the lake, Q is runoff and r is the lake to catchment area ratio (Kelly *et al.*, 1987). A similar equation is used to calculate ρ_S, the "in-lake retention fraction" for S.

Literature default values for the mass transfer coefficients of N and S are recommended by the Mapping Manual in the absence of site-specific data, with S_N = 5.0 m yr^{-1} and S_S = 0.5 m yr^{-1} (Kelly *et al.*, 1987, Dillon and Molot, 1990). The appropriateness of these literature default values was explored in a literature review by Curtis (2001a).

It should be noted that for stream sites, the lake:catchment ratio r is zero (the surface area of catchment streams is ignored in the calculation of r, since only in-lake processes are assumed to contribute to net retention of S and N). Hence both ρ_S and ρ_N are effectively zero.

Most published formulations of the FAB model do not take into account direct deposition onto the lake surface in lake catchments. Instead, it is assumed that all deposition falls onto the terrestrial catchment and is subject to terrestrial retention

processes, so that only the non-retained portion of inputs reaches the lake. The most recently published version (Henriksen & Posch, 2001) does take into account direct deposition to the lake surface, but this more complex formulation is not discussed further since it has not been applied in the UK. It would, however, result in slightly smaller critical loads and greater exceedance values for lake sites.

1B.5.4 *Critical load function for fixed denitrification rates*

The Mapping Manual (UBA, 1996) recommends a first order denitrification term (f_{de}) based on the percentage cover of peat soils. The problem with the 'f_{de}' method for determining denitrification is that the resulting denitrification rate may be far higher than measured values, for example in UK soils (Curtis *et al.*, 2000). For peat soils f_{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^{-1} yr^{-1} 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^{-1} yr^{-1} is used for Britain), equate to 80% of at least 27 kgN ha^{-1} yr^{-1}, i.e. more than 21 kgN ha^{-1} yr^{-1}. 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.*, 1998).

For the UK the FAB model is therefore modified to include the denitrification component as a fixed value for certain soil types, independent of deposition (Curtis *et al.*, 2000). If a fixed value of N_{den} is used, then each soil type must be allocated a denitrification value and its proportional cover within the catchment must be quantified, in an identical way to the calculation of mean N immobilisation for the catchment.

It might, however, be argued that a possible reason for the low rates of denitrification observed in certain soils is that current high immobilisation rates reduce the supply of N for denitrification. If N immobilisation rates decline to the low values recommended by the Mapping Manual (UBA, 1996), the supply of N for denitrifiers would increase and the rate of N removal via this route could rise. This issue provided one of the key questions addressed in the Defra contract 'Freshwater Umbrella: Critical Loads for Acidity and Metals', where it was demonstrated that current rates of denitrification are negligible in the four upland systems studied, while potential maximum rates under excess N availability were in a similar range to those suggested in Hall *et al.* (1998) (see Table 1B.1 below).

The new critical load constraints on the critical load function (Fig. 1B.1) with a fixed denitrification term are:

$$\mathbf{CL_{max}(S) = L_{crit}/1\text{-}\rho_S}$$
$$\mathbf{CL_{min}(N) = \mathit{f}N_{upt} + (1\text{-}r)(N_{imm} + N_{den})}$$
$$\mathbf{CL_{max}(N) = \mathit{f}N_{upt} + (1\text{-}r)(N_{imm} + N_{den}) + (L_{crit}/1\text{-}\rho_N)}$$

This formulation effectively adds the denitrification component onto the $CL_{min}(N)$ part of the CLF, so that the difference between $CL_{min}(N)$ and $CL_{max}(N)$ is dictated solely by the in-lake retention of N, the only input-dependent parameter for N.

1B.6 The CLF and the nature of critical load exceedance

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 SO_4^{2-} and NO_3^- 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. The above sets of equations effectively define the critical deposition loads for S and N individually. $CL_{max}(S)$ defines the critical load for S when total N deposition is less than $CL_{min}(N)$. In stream catchments this term is equivalent to the SSWC model critical load for S, and for lakes is 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_y) at which terrestrial catchment processes effectively remove all N, so that deposition loads lower than $CL_{min}(N)$ result in no net leaching of NO_3^-. The terrestrial sinks for N are fixed by soil type and 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 (see above). $CL_{max}(N)$ 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.

While the precise definition of the various CLF thresholds will vary depending on whether lakes or streams, or fixed or input-dependent denitrification rates, are used, the interpretation of the critical load function is identical. The nature of critical load exceedance for any given pair of S and N deposition values is illustrated in Figure 1B.1.

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 Figure 1B.1 (Posch *et al.*, 1997). For pairs of S and N deposition values that 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 (Figure1B.1). The colour coding of the CLF in this way lends itself to the mapping of deposition reduction requirements. However, the use of the CLF in this way can only provide qualitative information, in terms of whether reductions in either N or S deposition, or both, are optional or compulsory to attain non-exceedance. There is no quantitative indication of the amount by which the deposition of either species must be reduced, and indeed this is not possible because of the inter-dependence of the two species in jointly causing critical load exceedance. The requirement to reduce deposition of either species depends on the status of the other species, i.e. whether it remains constant or changes to a different, known value.

There are only two specific cases where the deposition reduction requirement for a species can be quantified. If the deposition of S is zero, the critical load exceedance

and reduction requirement for N can be quantified. Such a situation is, however, extremely unlikely to occur because of the ubiquitous nature of atmospheric S pollution. A more feasible case in this formulation of the FAB model is that N deposition may be less than $CL_{min}(N)$, implying that all deposited N is retained within the catchment. In such a case, the FAB model would provide a critical load exceedance value for S. This particular case only holds true for the assumption that N deposition directly onto the lake surface is negligible; otherwise some N leaching is inevitable and there is again a situation where both S and N are contributing to exceedance.

Despite the impossibility of quantifying the deposition reduction requirement for either N or S independently, except in the special circumstances above, the FAB model can provide a numerical value for critical load exceedance (see below). This value can only be interpreted as the amount by which the total acid flux (from both S and N) exceeds the critical load, and provides no indication of how site protection (non-exceedance) might be attained.

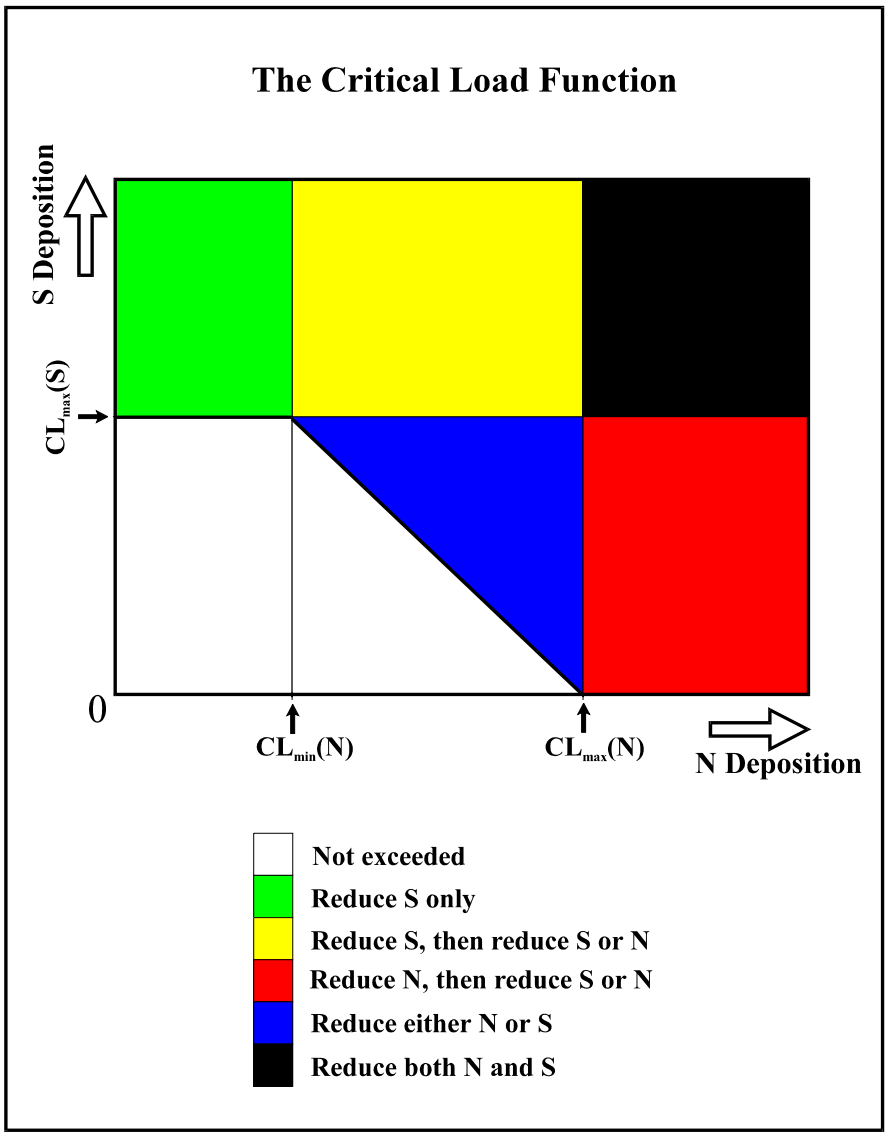
While the CLF diagram provides a visual indication of the nature of critical load exceedance for a site given any pair of deposition values for S and N, the FAB model actually provides a prediction of steady-state leaching of base cations, acid anions and hence ANC, which determines whether the critical load is exceeded and by how much.

1B.7 FAB model predictions and quantifying critical load exceedance

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, FAB outputs using current deposition data cannot necessarily be expected to reflect the current chemical status of freshwaters, because a steady-state cannot be assumed at present with respect to N (Curtis *et al.*, 1998).

While the FAB model is used to derive a critical load function for both S and N deposition, an exceedance of the critical load cannot be expressed in terms of a required reduction in both S and N deposition. It can, however, be calculated as the amount by which the acid anion leaching flux resulting from the specified deposition flux exceeds the critical load for a site.

Figure1B.1: Schematic interpretation of exceedance on the CLF



Critical load exceedance is simply the difference between the predicted leaching fluxes of S+N and the critical leaching flux of acid anions 'L_{crit}'. Exceedance can be calculated as:

$$\textbf{Total exceedance} = A_S S_{dep} + (1-\rho_N)\,(N_{dep} - fN_{upt} - (1-r)(N_{imm} + N_{den})) - L_{crit}$$

An alternative interpretation is that the critical load exceedance calculated in this way indicates the amount by which the leaching flux of acid anions must be reduced, which could potentially be achieved by many different combinations of reductions in S and N deposition.

Table 1B.1: Default soil N sinks

Mapping Soil code	General Soil description	Immobilisation		Denitrification	
		kgN ha^{-1} yr^{-1}	keq ha^{-1} yr^{-1}	kgN ha^{-1} yr^{-1}	keq ha^{-1} yr^{-1}
1	Terrestrial raw soil	3	0.2143	1	0.0714
2	Raw gley soils	1	0.0714	1	0.0714
3	Lithomorphic soils	1	0.0714	1	0.0714
3.1	Rankers	1	0.0714	1	0.0714
3.2	Sand rankers	1	0.0714	1	0.0714
3.4	Rendzinas	1	0.0714	1	0.0714
3.6	Sand parendzinas	1	0.0714	1	0.0714
3.7	Rendzina-like alluvial soils	1	0.0714	1	0.0714
4.1	Calcareous pelosols	1	0.0714	2	0.1429
4.2	Non-calcareous pelosols	1	0.0714	2	0.1429
4.3	Argillic pelosols	1	0.0714	2	0.1429
5.1	Brown calcareous earths	1	0.0714	1	0.0714
5.2	Brown calcareous sands	1	0.0714	1	0.0714
5.3	Brown calcareous alluvial soil	1	0.0714	1	0.0714
5.4	Brown earths	1	0.0714	1	0.0714
5.5	Brown sands	1	0.0714	1	0.0714
5.6	Brown alluvial soils	1	0.0714	1	0.0714
5.7	Argillic brown earths	1	0.0714	1	0.0714
5.8	Paleo-argillic brown earths	1	0.0714	1	0.0714
6.1	Brown podsolic soils	3	0.2143	1	0.0714
6.3	Podsols	3	0.2143	1	0.0714
6.4	Gley podsols	3	0.2143	1	0.0714
6.5	Stagnopodsols	3	0.2143	1	0.0714
7.1	Stagnogley soils	1	0.0714	4	0.2857
7.2	Stagnohumic gley soils	3	0.2143	4	0.2857
8.1	Alluvial gley soils	1	0.0714	4	0.2857
8.2	Sandy gley soils	1	0.0714	4	0.2857
8.3	Cambic gley soils	1	0.0714	4	0.2857
8.4	Argillic gley soils	1	0.0714	4	0.2857
8.5	Humic-alluvial gley soils	1	0.0714	4	0.2857
8.6	Humic-sandy gley soils	1	0.0714	4	0.2857
8.7	Humic gley soils	1	0.0714	4	0.2857
9	Man-made soils	1	0.0714	1	0.0714
9.2	Disturbed soils	1	0.0714	1	0.0714
10.1	Raw peat soils	3	0.2143	1	0.0714
10.2	Earthy peat soils	3	0.2143	1	0.0714
2.2	Unripened gley soils	1	0.0714	4	0.2857
1.1	Raw sands	1	0.0714	1	0.0714
4	Pelosols	1	0.0714	2	0.1429
5	Brown soils	1	0.0714	1	0.0714
6	Podzolic soils	3	0.2143	1	0.0714
7	Surface-water gley soils	1	0.0714	4	0.2857
8	Ground-water gley soils	1	0.0714	4	0.2857
10	Peat soils	3	0.2143	1	0.0714

(Source: Hall *et al.*, 1998)

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Appendix 2 BAP Broad Habitats and LCM2000 classes

Land Cover Map 2000 records terrestrial and inshore examples of widespread Broad Habitats. The Habitats are elements in a classification designed for monitoring and maintaining biodiversity under the UK Biodiversity Action Plan. In practice, LCM2000 identified 16 Target classes (Level-1) to be mapped with 90% accuracy; it subdivided these into 27 Subclasses (Level-2), allowing the construction of the widespread Broad Habitats. A summary of the Broad Habitats and relevant LCM classes used in the mapping of critical loads in this update, is provided in the table below.

Table A2.1

Broad Habitat	**LCM Target (Level –1)**	**Code**	**LCM subclass (Level-2)**
Bog	Bog	12.1	Bog
Dwarf Shrub Heath	Dwarf Shrub Heath	10.1 10.2	Dwarf Shrub Heath Open Shrub Heath
Montane	Montane	15.1	Montane
Broad-leaved, mixed and yew woodland	Broad-leaved /mixed woodland	1.1	Broad-leaved /mixed woodland
Coniferous woodland	Coniferous woodland	2.1	Coniferous woodland
Calcareous grassland	Semi-natural & natural grasslands and bracken	7.1	Calcareous grass
Acid grassland	Semi-natural & natural grasslands and bracken	8.1	Acid grass
Supra-littoral sediment	Supra-littoral rock and sediment	19.1	Supra-littoral sediment
Littoral sediment	Littoral rock and sediment	21.1	Littoral sediment
Inland rock	Inland bare ground	16.1	Inland bare ground

The mapping of these Broad Habitats for this critical loads update, used additional data sets such as species distributions, soil information etc. The methods are described in detail in Chapter 3 of the Main Report.

Appendix 3. Relationships between the UK BAP Broad Habitats, EUNIS classes and UK Broad Habitat critical load maps

Table A3.1

BAP Broad Habitat (BH)	EUNIS class(es) assigned to each Broad Habitat	Relationship between BAP BH and EUNIS class[1]	UK Broad Habitat critical loads maps[2]
1. Broadleaved, mixed & yew woodland	G1 Broadleaved woodland	#	Broadleaved woodland (managed)
	G1-LA Broadleaved woodland (lichens & algae)	None	Atlantic oak woods
	G1&G3 Broadleaved woodland & coniferous woodland	#	Coniferous & broadleaved woodland (unmanaged)
	G1&G3-GF Broadleaved woodland & coniferous woodland (ground flora)	None	Coniferous and broadleaved woodland (unmanaged) – excluding Atlantic oak woods
2. Coniferous woodland	G3 Coniferous woodland	#	Coniferous woodland (managed)
7. Calcareous grassland	E1.26 Sub-Atlantic semi-dry calcareous grassland	None	Calcareous grassland
8. Acid grassland	E1.7 Non-Mediterranean dry acid & neutral closed grassland	<	Dry acid grassland[3]
	E3.5 Moist or wet oligotrophic grassland	>	Wet acid grassland[3]
10. Dwarf shrub heath	F4.2 Dry heaths	None	Dry heathland[4]
	F4.11 Northern wet heaths	None	Wet heathland[4]
12. Bogs	D1 Raised & blanket bogs	=	Bogs
13. Standing open water & canals	C1 Surface standing waters	#	Catchment areas as defined by UCL – lake and stream sites not mapped separately
14. Rivers & streams	C2 Surface running waters	#	
15. Montane	E4.2 Moss & lichen dominated mountain summits	>	Montane/*Racomitrium* heath
19. Supralittoral sediment	B1.3 Shifting coastal dunes B1.4 Coastal stable dune grassland	None	Dune grasslands

[1] These relationships have been taken from EUNIS web site (http://mrw.wallonie.be/dgrne/sibw/EUNIS/home.html), with the exception of that for acid grassland which differs from the published relationship on the web site.
[2] Habitats mapped within each BAP Broad Habitat and representing the EUNIS classes.
[3] Wet and dry acid grassland mapped as a single UK map (each 1km square is either mapped as wet or dry acid grassland)
[4] Wet and dry heathland mapped as a single UK map (each 1km square is either mapped as wet or dry heathland)

Relationship key:
= exact equivalence between classifications
partial overlap between classification units
> habitat in other system includes that in EUNIS
< habitat in EUNIS includes that in other system