

### 5.4 Critical Loads for Aquatic Ecosystems

The purpose of critical loads for aquatic ecosystems is to estimate the maximum deposition(s) below which 'significant harmful effects' on biological species do not occur. Similar to terrestrial ecosystems, the links between water chemistry and biological impacts cannot be modelled adequately at present (see also Wright and Lie 2002) as such, water quality criteria are generally used to derive critical loads for aquatic ecosystems.

In this Section we deal only with the modelling of critical loads of acidity for aquatic ecosystems. The models are restricted to freshwater systems, since models for marine ecosystems do not seem to exist. Empirical critical loads of nitrogen for fresh waters, as well as coastal and marine habitats, can be found in Section 5.2.

The following description is largely based on the review by Henriksen and Posch (2001), but amended with new or additional information where available. Three models for calculating critical loads of acidifying *N* and *S* deposition are described. Models of critical loads for surface waters also include their terrestrial catchment to a greater or lesser extent. Therefore, it is advised to consult Section 5.3 for some of the terminology and variables used in the context of critical loads for soils.

#### 5.4.1 The Steady-State Water Chemistry (SSWC) model

##### 5.4.1.1 Model derivation

The critical load of a lake or stream can be derived from present day water chemistry using the SSWC model, if weighted annual mean values, or estimates thereof, are available. It assumes that all sulphate ( $SO_4^{2-}$ ) in runoff originates from sea salt spray and anthropogenic deposition (no adsorption or retention). The model uses Acid Neutralising Capacity (*ANC*) as the variable linking water

chemistry to sensitive indicator organisms in freshwaters.

In the SSWC model (Sverdrup et al. 1990, Henriksen et al. 1992, Henriksen and Posch 2001) a critical load of acidity,  $CL(A)$ , is calculated from the principle that the acid load should not exceed the non-marine, non-anthropogenic base cation input and sources and sinks in the catchment minus a buffer to protect selected biota from being damaged, i.e.:

(5.48)

$$CL(A) = BC_{dep}^* + BC_w - Bc_u - ANC_{limit}$$

where  $BC_{dep}^*$  ( $BC = Ca + Mg + K + Na$ ) is the sea-salt corrected (with *Cl* as a tracer; see Chapter 2) non-anthropogenic deposition of base cations,  $BC_w$  is the average weathering flux,  $Bc_u$  ( $Bc = Ca + Mg + K$ ) is the net long-term average uptake of base cations in the biomass (i.e., the annual average removal of base cations due to harvesting), and  $ANC_{limit}$  the lowest *ANC*-flux that does not damage the selected biota. Since the average flux of base cations weathered in a catchment and reaching the lake is difficult to measure or to compute from available information, a critical load equation that uses water quality data alone has been derived.

In pre-acidification times the non-marine flux of base cations from the lake,  $BC_0^*$ , is given by (all parameters are expressed as annual fluxes, e.g. in  $eq/m^2/yr$ ):

(5.49)

$$BC_0^* = BC_{dep}^* + BC_w - Bc_u$$

Thus we have for the critical load from eq. 5.48:

(5.50)

$$CL(A) = BC_0^* - ANC_{limit} \\ = Q \cdot (BC_0^* - ANC_{limit})$$

where the second identity expresses the critical load in terms of the catchment runoff *Q* (in  $m/yr$ ) and concentrations ( $[X] = X/Q$ ).

To estimate the pre-acidification flux of base cations we start with the present flux of base cations,  $BC_t^*$ , given by:

(5.51)

$$BC_t^* = BC_{dep}^* + BC_w - BC_u + BC_{exc}$$

where  $BC_{exc}$  is the release of base cations due to ion-exchange processes. Assuming that deposition, weathering rate and net uptake have not changed over time, we obtain  $BC_{exc}$  by subtracting eq. 5.49 from eq. 5.51:

(5.52)

$$BC_{exc} = BC_t^* - BC_0^*$$

This present-day excess production of base cations in the catchment is related to the long-term changes in inputs of non-marine acid anions by the so-called  $F$ -factor (see below):

(5.53)

$$BC_{exc} = F \cdot (\Delta SO_4^* + \Delta NO_3^*)$$

For the pre-acidification base cation flux we thus get from eq. 5.52 ( $\Delta X = X_t - X_0$ ):

(5.54)

$$BC_0^* = BC_t^* - F \cdot (SO_{4,t}^* - SO_{4,0}^* + NO_{3,t}^* - NO_{3,0}^*)$$

The pre-acidification nitrate concentration,  $NO_{3,0}$ , is generally assumed zero.

### 5.4.1.2 The $F$ -factor

According to eqs. 5.52 and 5.53, and using concentrations instead of fluxes, the  $F$ -factor is defined as the ratio of change in non-marine base cation concentrations due to changes in strong acid anion concentrations (Henriksen 1984, Brakke et al. 1990):

(5.55)

$$F = \frac{[BC_t^*] - [BC_0^*]}{[SO_{4,t}^*] - [SO_{4,0}^*] + [NO_{3,t}^*] - [NO_{3,0}^*]}$$

where the subscripts  $t$  and  $0$  refer to present and pre-acidification concentrations, respectively. If  $F=1$ , all incoming protons are neutralised in the catchment (only soil acidification), at  $F=0$  none of the incoming protons are neutralised in the catchment (only water acidification). The  $F$ -factor was estimated empirically to be in the range 0.2–0.4, based on the analysis of historical data from Norway, Sweden, U.S.A. and Canada (Henriksen 1984). Brakke et al. (1990) later suggested that the  $F$ -factor should be a function of the base cation concentration:

(5.56)

$$F = \sin\left(\frac{\pi}{2} [BC_t^*] / [S]\right)$$

where  $[S]$  is the base cation concentration at which  $F=1$ ; and for  $[BC_t^*] > [S]$   $F$  is set to 1. For Norway  $[S]$  has been set to 400 meq/m<sup>3</sup> (ca. 8 mgCa/L) (Brakke et al. 1990).

In eq. 5.56 the present base cation concentration is used for practical reasons. To render the  $F$ -factor independent from the present base cation concentration (and to simplify the functional form), Posch et al. (1993) suggested the following relationship between  $F$  and the pre-acidification base cation concentration  $[BC_0^*]$ :

(5.57)

$$F = 1 - \exp\left(-[BC_0^*] / [B]\right)$$

where  $[B]$  is a scaling concentration estimated to be 131 meq/m<sup>3</sup> from paleolimnological data from Finland (Posch et al. 1993). Inserting this expression into eq. 5.55 gives a non-linear equation for  $[BC_0^*]$  which has to be solved by an iterative procedure. The two expressions for the  $F$ -factor give similar results when used to calculate critical loads for surface waters in Norway (see Henriksen and Posch 2001).

The use of the  $F$ -factor, defined as a function of the base cation concentration (Henriksen 1984) was originally derived from Norwegian lake data. In Norway the range of runoff is wide (0.3–5 m/yr), with an average of about

1 m/yr. In other Nordic countries, such as Sweden and Finland, runoff is low compared to most of Norway. The weathering rate of a catchment is largely dependent on the bedrock and overburden. Thus, catchments with similar bedrock and overburden characteristics should have similar weathering rates. If one catchment has a high runoff, e.g., 2 m/yr, and another one has a low runoff, e.g., 0.3 m/yr, their base cation fluxes will be similar, but their concentrations will differ considerably. Thus, in the  $F$ -factor (eq. 5.56) the  $BC$ -flux should be used instead of the concentration (Henriksen and Posch 2001):

$$(5.58) \quad F = \sin\left(\frac{\pi}{2}Q \cdot [BC^*]_t / S\right)$$

where  $S$  is the base cation flux at which  $F=1$ . For Norway,  $S$  has been estimated at 400 meq/m<sup>2</sup>/yr. Again, if  $Q \cdot [BC^*]_t > S$ ,  $F$  is set to 1. Similarly, fluxes could be introduced for the formulation in eq. 5.57.

### 5.4.1.3 The non-anthropogenic sulphate concentration

The pre-acidification sulphate concentration in lakes,  $[SO_4^*]_0$ , is assumed to consist of a constant atmospheric contribution and a geologic contribution proportional to the concentration of base cations (Brakke et al. 1989):

$$(5.59) \quad [SO_4^*]_0 = a + b \cdot [BC^*]_t$$

The coefficients in this equation, estimated

for different areas and by different authors, are summarised in Table 5.16.

Details on the procedures and data sources for estimating these coefficients can be found in the references given. In Henriksen and Posch (2001) it is shown that the exceeded area for Norwegian lakes (in 1994) is influenced very little by the choice of coefficients for calculating non-anthropogenic sulphate. Similar results have been reported for Irish lakes (Aherne and Curtis 2003).

Larssen and Høgåsen (2003) suggested that the atmospheric contribution in eq. 5.59 be derived from background  $S$  deposition, as estimated by atmospheric transport models:

$$(5.60) \quad [SO_4^*]_0 = S_{dep,0} / Q + b \cdot [BC^*]_t$$

For southern Norway,  $S_{dep,0}$  is about 50 mgS/m<sup>2</sup>/yr from the EMEP long-range transport model, i.e., about 3 meq/m<sup>2</sup>/yr. With  $Q$  varying between 0.5 and 1 m/yr this results in an atmospheric contribution to  $[SO_4^*]_0$  of about 3–6 meq/m<sup>3</sup>.

The SSWC model has been developed for and is particularly applicable to dilute oligotrophic waters located on granitic and gneissic bedrock with thin overburden, such as large parts of Fennoscandia, Scotland, Canada and Ireland. In such areas, surface waters are generally more sensitive to acid inputs than soils. The model assumes that all sulphate in runoff originates from deposition alone, except for a small geologic contribution. In areas where the geological conditions lead to more alkaline waters, the SSWC model has to be modified, since significant

**Table 5.16:** Constants to estimate the non-anthropogenic sulphate concentration with eq. 5.59, derived from empirical data ( $N$  is the number of samples and  $r$  is the correlation coefficient).

$a$ (meq/m <sup>3</sup> )	$b$	$N$	$r$	Reference
15	0.16	143	0.38	Lakes, Norway (Brakke et al 1989)
8	0.17	289	0.78	Lakes, Norway (Henriksen and Posch 2001)
5	0.05	n.g.	n.g.	Groundwater, Sweden (Wilander 1994)
14	0.10	61	0.29	Lakes, Finland (Posch et al. 1993)
19	0.08	251	0.66	Lakes, N. Norway+Finland+Sweden (Posch et al. 1997)
9.5	0.08	60	0.66	Lakes, Ireland (Aherne et al. 2002)

amounts of sulphate from geological sources can be present in the runoff water. A modification for this kind of conditions has been developed for Slovakia (Závodský et al. 1995).

### 5.4.1.4 The ANC-limit

Lien et al. (1996) analysed the status of fish and invertebrate populations in the context of surface water acidification and loss of *ANC* in Norwegian lakes and streams. The data for fish came from populations in 1095 lakes, mostly from the regional lake survey carried out in 1986 (Henriksen et al. 1988, 1989). The critical level of *ANC* varied among fish species, with Atlantic salmon being the most sensitive, followed by brown trout. They concluded that Atlantic salmon appeared to be a good indicator of acidification of rivers, and trout seemed to be a useful indicator for acidification of lakes. Based on an evaluation of fish and invertebrate populations, a critical lower limit of  $[\text{ANC}] = 20 \text{ meq/m}^3$  was suggested as the tolerance level for Norwegian surface waters (Lien et al. 1996; see Figure 5.6). This limit has been widely used (Kola, northern Russia: Moiseenko 1994; southern central Alps: Boggero et al. 1998; China: Duan et al. 2000); however, it has been set to zero in the United Kingdom (CLAG 1995) and to  $40 \text{ meq/m}^3$  in south-central Ontario, Canada (Henriksen et al. 2002).

Lydersen et al. (2004) argued that the *ANC*-limit should be corrected with the amount of

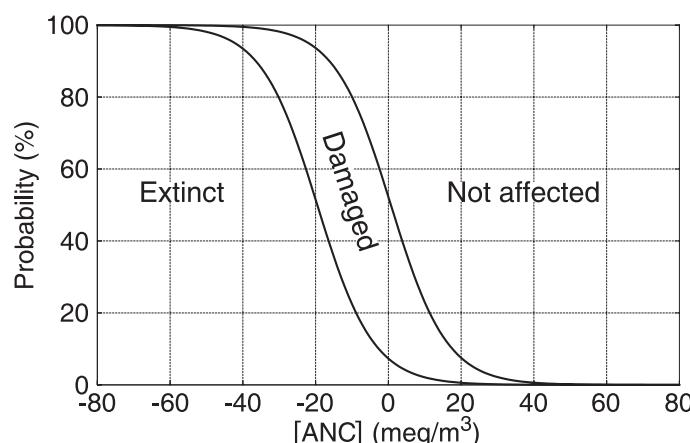
organic acids present in the lake. And they showed that the fit between observed fish status and *ANC* can be (slightly) improved, if an 'organic acid adjusted' *ANC*,  $[\text{ANC}]_{oaa}$ , is used (instead of the 'standard' *ANC*). They define this quantity as:

(5.61)

$$[\text{ANC}]_{oaa} = [\text{ANC}] - \frac{1}{3} \cdot m \cdot \text{TOC}$$

where  $m \cdot \text{TOC}$  is the total organic carbon expressed in  $\text{meq/m}^3$  ( $m$  being the charge density). Such a correction leads to a lower *ANC*-limit, i.e., higher critical loads.

Figure 5.6 indicates that in the *ANC* range 0–50  $\text{meq/m}^3$  there is a decreasing probability from about 50 to 0% of damage to fish populations. The lakes studied receive very low to very high (for Norway) levels of deposition, thus including a wide range of affected lakes. This implies that for a given *ANC*-value lakes of varying sensitivity exist, receiving varying amounts of deposition. This could reflect that fish have responded to the same *ANC* differently in different lakes, indicating that a catchment-dependent *ANC*-limit would be more appropriate than a fixed value for all lakes. In other words, every lake has its own characteristic *ANC*-limit (in the range shown in Figure 5.6). Less sensitive lakes, i.e., lakes with higher critical loads, should have a higher *ANC*-limit, since less sensitive ecosystems will have a higher biological variety/diversity and thus require a higher *ANC*-limit to keep that diversity intact.



**Figure 5.6:** Relationship between the *ANC* concentration in lake water and the probability for damage and extinction of fish (brown trout) populations in lakes, derived from Norwegian data (after Lien et al. 1996).

The simplest functional relationship with this feature is a linear relationship between  $[ANC]_{limit}$  and the critical load  $CL$ :

(5.62)

$$[ANC]_{limit} = k \cdot CL$$

This gives the following implicit equation for the critical load (see eq. 5.50):

(5.63)

$$CL = Q \cdot ([BC^*]_0 - k \cdot CL)$$

which yields after re-arranging for  $CL$ :

(5.64)

$$CL = Q \cdot [BC^*]_0 / (1 + k \cdot Q)$$

and thus from eq. 5.62:

(5.65)

$$[ANC]_{limit} = k \cdot Q \cdot [BC^*]_0 / (1 + k \cdot Q)$$

This is a special case of a more general expression derived earlier using somewhat different arguments (Henriksen et al. 1995). As for the constant  $[ANC]_{limit}$  used earlier, the proportionality constant  $k$  should be derived from data. If we assume that for  $CL=0$ , the  $[ANC]_{limit}=0$ ; if we further assume that for a critical load of 200 meq/m<sup>2</sup>/yr the ANC-limit should not exceed 50 meq/m<sup>3</sup>, as has been assumed in Sweden, we arrive at a  $k$ -value of  $50/200 = 0.25$  yr/m. In addition, for  $CL$ -values above 200 meq/m<sup>2</sup>/yr we set the  $[ANC]_{limit}$  to the constant value of 50 meq/m<sup>3</sup>. The value of  $k$  is derived from experience in the Nordic countries and, as such, reflects the geology, deposition history and biological diversity (fish species) of that region. For different regions other  $k$ -values may be more appropriate.

### 5.4.2 The empirical diatom model

The empirical diatom model is an alternative approach to the SSWC model and is devel-

oped from paleolimnological data (Battarbee et al. 1995). Diatom assemblages in cores from acidified lakes usually show that prior to acidification the diatom flora, and therefore water chemistry, changed little over time. The point of acidification is indicated by a shift towards a more acidophilous diatom flora. Diatoms are amongst the most sensitive indicators of acidification in freshwater ecosystems, hence it can be argued that the point of change in the diatom record indicates the time at which the critical load for the site was exceeded.

The acidification status (as defined by diatom analyses) of 41 sites in the United Kingdom (UK) was compared to site sensitivity (defined by lake-water calcium concentrations) and current deposition loading. The optimal separation of acidified and non-acidified sites is given by a  $[Ca]_0:S_{dep}$  ratio of 94:1 (Battarbee et al. 1995), acidified sites having a ratio less than 94:1. This critical ratio, determined by logistic regression, can be used to define critical sulphur loads for any site, including streams. Critical load values are calculated from pre-acidification calcium concentrations using the  $F$ -factor (Brakke et al. 1990). For example, the critical sulphur load for a lake with a  $[Ca]_0$ -value of 40 meq/m<sup>3</sup> is approximately 0.43 keq/ha/yr.

The diatom model has been adapted to provide critical loads, and critical load exceedances, for total acidity (sulphur and nitrogen). Exceedance values for total acidity require a measure of the fraction of deposited nitrogen leached to the surface waters. This is calculated from the differences between the ratios of sulphate/nitrate in the water and in the deposition at the site. In this way the fraction of the nitrogen deposition contributing to acidification,  $f_N$ , is added to the value of sulphur deposition to provide a total 'effective' acid deposition:

(5.66)

$$f_N = \frac{S_{dep}^*}{N_{dep}} \left/ \frac{[SO_4^*]}{[NO_3^-]} \right.$$

This model assumes equilibrium between sulphur deposition and sulphate in water, and only applies to sites with no additional catchment nitrogen inputs. The diatom model has been re-calibrated for total acidity loads by substituting total effective acid deposition for sulphur deposition. The resulting critical ratio is 89:1, slightly lower than when considering sulphur alone. The basic equation for the critical load of total acidity in the empirical diatom model is therefore as follows:

(5.67)

$$CL(A) = \frac{[Ca^*]_0}{89}$$

where  $CL(A)$  is in  $\text{keq/ha/yr}$  and  $[Ca^*]_0$  in  $\text{meq/m}^3$ . The pre-acidification  $Ca$ -concentration is calculated as:

(5.68)

$$[Ca^*]_0 = [Ca^*]_t - F_{Ca} \cdot ([SO_4^*]_t - [SO_4^*]_0 + [NO_3]_t - [NO_3]_0)$$

with

(5.69)

$$F_{Ca} = \sin\left(\frac{\pi}{2} [Ca^*]_t / [S_{Ca}]\right)$$

and  $[S_{Ca}]$  is the  $Ca$ -concentration at which  $F_{Ca}=1$ . It can vary between 200 and 400  $\text{meq/m}^3$ , depending on location. In the UK critical loads mapping exercise a value of  $[S_{Ca}]=400 \text{ meq/m}^3$  has been used, and in waters with  $[Ca^*]_t > [S_{Ca}]$ ,  $F_{Ca}$  was set to 1. The pre-acidification nitrate concentration,  $[NO_3]_0$ , is assumed zero. The pre-acidification sea-salt corrected sulphate concentration,  $[SO_4^*]_0$ , is estimated according to eq. 5.59 (Brakke et al. 1989).

The diatom model has been calibrated using sites and data from the UK. However, a major advantage of the approach is that predictions for any lake can be validated by analysing diatoms in a sediment core. In this way the applicability of the model to sites outside the UK can be tested.

### 5.4.3 The First-order Acidity Balance (FAB) model

The First-order Acidity Balance (FAB) model for calculating critical loads of sulphur ( $S$ ) and nitrogen ( $N$ ) for a lake takes into account sources and sinks within the lake and its terrestrial catchment. The original version of the FAB model has been developed and applied to Finland, Norway and Sweden in Henriksen et al. (1993) and further described in Posch et al. (1997). A modified version was first reported in Hindar et al. (2000) and is described in Henriksen and Posch (2001). The FAB model is designed to be equivalent to the Simple Mass Balance model for a catchment, and it largely follows its derivation (see Section 5.3), the main difference being that the leaching of  $ANC$  is modelled according to the SSWC model (see section 5.4.1).

#### 5.4.3.1 Model derivation

The lake and its catchment are assumed small enough to be properly characterised by average soil and lake water properties. With  $A$  we denote the total catchment area (lake + terrestrial catchment),  $A_l$  is the lake area,  $A_f$  the forested area and  $A_g$  the area covered with grass/heath land. We have  $A_l + A_f + A_g \leq A$ , and a non-zero difference represents a land area on which no transformations of the deposited ions take place ('bare rock').

Starting point for the derivation of the FAB model is the charge balance ('acidity balance') in the lake water running off the catchment:

(5.70)

$$S_{runoff} + N_{runoff} = BC_{runoff}^* - ANC_{runoff}$$

where  $BC^*$  stands for the sum of (non-marine) base cations and  $ANC$  is the acid neutralising capacity. In the above equation we assume that the quantities are total amounts per time (e.g. eq/yr). In order to derive critical loads we have to link the ions in the lake water to their depositions, taking into account their sources and sinks

in the terrestrial catchment and in the lake.

For  $X = S, N$  and  $BC$  the mass balance in the lake is given by:

$$(5.71) \quad X_{runoff} = X_{in} - X_{ret}, \quad X = S, N, BC$$

where  $X_{in}$  is the total amount of ion  $X$  entering the lake and  $X_{ret}$  the amount of  $X$  retained in the lake. The in-lake retention of  $S$  and  $N$  is assumed to be proportional to the input of the respective ion into the lake:

$$(5.72) \quad X_{ret} = \rho_X \cdot X_{in}, \quad X = S, N$$

where  $0 \leq \rho_X \leq 1$  is a dimensionless retention factor. Thus the mass balances for the lake become:

$$(5.73) \quad X_{runoff} = (1 - \rho_X) \cdot X_{in}, \quad X = S, N$$

The total amount of sulphur entering the lake is given by:

$$(5.74) \quad S_{in} = A \cdot S_{dep}$$

where  $S_{dep}$  is the total deposition of  $S$  per unit area. Immobilisation, reduction and uptake of sulphate in the terrestrial catchment are assumed negligible, and sulphate ad/desorption is not considered since we model steady-state processes only. eq. 5.74 states that all sulphur deposited onto the catchment enters the lake, and no sources or sinks are considered in the terrestrial catchment.

In the case of nitrogen we assume that immobilisation and denitrification occur both in forest and grass/heath land soils, whereas net uptake occurs in forests only (equalling the annual average amount of  $N$  removed by harvesting); the deposition onto the remaining area (lake + 'bare rocks') enters the lake unchanged. Thus the amount of  $N$  entering

the lake is:

$$(5.75) \quad N_{in} = (A - A_f - A_g) \cdot N_{dep} + A_f \cdot (N_{dep} - N_i - N_u - N_{de})_+ + A_g \cdot (N_{dep} - N_i - N_{de})_+$$

where  $N_{dep}$  is the total  $N$  deposition,  $N_i$  is the long-term net immobilisation of  $N$  (which may include other long-term steady-state sources and sinks; see Chapter 5.3),  $N_{de}$  is  $N$  lost by denitrification, and  $N_u$  the net growth uptake of  $N$ , all per unit area. The symbol  $(x)_+$  or  $x_+$  is a short-hand notation for  $\max\{x, 0\}$ , i.e.,  $x_+ = x$  for  $x > 0$  and  $x_+ = 0$  for  $x \leq 0$ . The effects of nutrient cycling are ignored and the leaching of ammonium is considered negligible, implying its complete uptake and/or nitrification in the terrestrial catchment.

While immobilisation and net growth uptake are assumed independent of the  $N$  deposition, denitrification is modelled as fraction of the available  $N$ :

$$(5.76) \quad N_{de} = \begin{cases} f_{de} \cdot (N_{dep} - N_i - N_u)_+ & \text{on } A_f \\ f_{de} \cdot (N_{dep} - N_i)_+ & \text{on } A_g \end{cases}$$

where  $0 \leq f_{de} < 1$  is the (soil-dependent) denitrification fraction. The above equation is based on the assumption that denitrification is a slower process than immobilisation and growth uptake. Inserting eq. 5.76 into eq. 5.75 one obtains:

$$(5.77) \quad N_{in} = (A - A_f - A_g) \cdot N_{dep} + A_f \cdot (1 - f_{de}) \cdot (N_{dep} - N_i - N_u)_+ + A_g \cdot (1 - f_{de}) \cdot (N_{dep} - N_i)_+$$

If sufficient data for quantifying the sources and sinks of base cations in the catchment, such as deposition, weathering and uptake,

are available, the runoff of base cations ( $BC^*_{runoff}$ ) could be described in the same way as  $S$  and  $N$ . This would be in analogy to the derivation of the SMB model for (forest) soils. Alternatively, water quality data can be used to quantify the runoff of base cations and  $ANC$ , as is done in the SSWC model (see section 5.4.1).

To arrive at an equation for critical loads, a link has to be established between a chemical variable and effects on aquatic biota. The most commonly used criterion is the so-called  $ANC$ -limit (see above), i.e. a minimum concentration of  $ANC$  derived to avoid 'harmful effects' on fish:  $ANC_{runoff,crit} = A \cdot Q \cdot [ANC]_{limit}$ .

Defining  $L_{crit} = (BC^*_{runoff} - ANC_{runoff,crit})/A$ , inserting eq. 5.74 and 5.77 into eq. 5.73 and eq. 5.70 and dividing by  $A$  yields the following equation to be fulfilled by critical depositions (loads) of  $S$  and  $N$ :

(5.78)

$$(1 - \rho_S) \cdot S_{dep} + (1 - \rho_N) \cdot \{(1 - f - g) \cdot N_{dep}$$

where we have defined:

(5.79)

$$f = A_f/A, \quad g = A_g/A \\ \Rightarrow 1 - f - g \geq r \quad \text{with} \quad r = A_l/A$$

Eq. 5.78 defines a function in the  $(N_{dep}, S_{dep})$ -plane, the so-called critical load function (see Figure 5.7), and in the following we will look at this function in more detail. The general form of the critical load function is:

(5.80)

$$a_S \cdot S_{dep} + a_N \cdot N_{dep} = L_N + L_{crit}$$

with

(5.81)

$$a_S = 1 - \rho_S, \quad a_N = (1 - \rho_N) \cdot b_N, \\ L_N = (1 - \rho_N) \cdot M_N$$

The quantity  $M_N$  and the dimensionless coefficient  $b_N$  depend on  $N_{dep}$ :

(a)  $N_{dep} \leq N_i$ : In this case  $(N_{dep} - N_i)_+ = 0$  and  $(N_{dep} - N_i - N_u)_+ = 0$ , which means that all  $N$  falling onto forests and grassland is immobilised and only the  $N$  deposition falling directly onto the lake and 'bare rocks' contributes to the leaching of  $N$ :

(5.82)

$$b_N = b_1 = 1 - f - g, \quad M_N = M_1 = 0$$

(b)  $N_i < N_{dep} \leq N_i + N_u$ : In this case  $(N_{dep} - N_i)_+ = N_{dep} - N_i$ , but  $(N_{dep} - N_i - N_u)_+ = 0$ , meaning that all  $N$  deposition falling onto forests is immobilised or taken up, but  $N$  falling onto the other areas is (partially) leached:

(5.83)

$$b_N = b_2 = 1 - f - g \cdot f_{de},$$

$$M_N = M_2 = (1 - f_{de}) \cdot g \cdot N_i$$

(c)  $N_{dep} > N_i + N_u$ : Some  $N$  deposition is leached from all areas:

(5.84)

$$b_N = b_3 = 1 - (f + g) \cdot f_{de},$$

$$M_N = M_3 = (1 - f_{de}) \cdot [(f + g) \cdot N_i + f \cdot N_u]$$

The maximum critical load of sulphur is obtained by setting  $N_{dep} = 0$  in eq. 5.78:

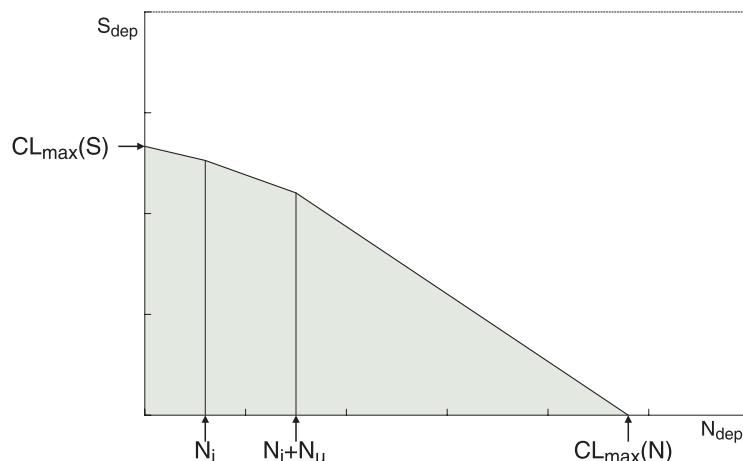
(5.85)

$$CL_{max}(S) = L_{crit}/a_S$$

Setting  $S_{dep} = 0$  and considering the three different cases for  $N_{dep}$ , gives the following expression for the maximum critical load for nitrogen:

(5.86)

$$CL_{max}(N) = \min \{ (L_{crit}/(1 - \rho_N) + M_i) / b_i, \\ i = 1, 2, 3 \}$$



**Figure 5.7:** Piece-wise linear critical load function of  $S$  and acidifying  $N$  for a lake as defined by catchment properties. Note the difference with the critical load function for soils (see Figure 5.1). The grey area below the  $CL$  function denotes deposition pairs resulting in an  $ANC$  leaching greater than  $Q \cdot [ANC]_{limit}$  (non-exceedance of critical loads; see Chapter 7).

### 5.4.3.2 Systems of lakes

The above derivation of the FAB model is for (small) headwater lakes only. Critical loads are generally calculated for such lakes, since lakes with (many) upstream lakes tend to have larger catchments, and several (implicit) assumptions of the FAB model, e.g. uniform depositions, will be violated. Nevertheless, in some areas systems of lakes can be found on a small scale, and therefore a model for such systems is desirable.

When computing the critical load of acidity with the SSWC model (which uses annual average lake water chemistry) for a lake receiving runoff from upstream lakes, one implicitly computes *the critical load for that lake including all its upstream lakes*, since water samples taken from (the outlet of) the lowest lake is a mixture of the water of that lake and all its upstream lakes. Consequently, when applying the FAB model to such a lake, one has to be aware that one also computes the critical load for the whole system of lakes and thus must take into account the catchment and lake characteristics of all lakes in the system. To do this in a more explicit way, two methods for computing the critical load of a system of lakes have been developed (Hindar et al. 2000). Both require the same input data, but they differ in the complexity of the calculations

involved. The formulae will not be derived here, and the interested reader is referred to the literature (see also Hindar et al. 2001), where also the differences between the methods are demonstrated, using data from lake systems in the Killarney Provincial Park in Ontario, Canada. An application to lakes in the Muskoka river catchment (Ontario, Canada) can be found in Aherne et al. (2004).

### 5.4.4 Input data

In addition to the data required for the SSWC and diatom model (runoff and concentrations of major ions in the lake runoff water), the FAB model needs also information on (a) the area of lake, catchment and different land cover classes, (b) terrestrial nitrogen sinks, and (c) parameters for in-lake retention of  $N$  and  $S$ .

#### Runoff:

The runoff  $Q$  is the amount of water leaving the catchment at the lake outlet, expressed in m/yr. It is derived from measurements or can be calculated as the difference between precipitation and actual evapotranspiration, averaged over the catchment area. A long-term climatic mean annual value should be taken. Sources for data and models for evapotranspiration can be found in Section 5.3.

### Ion concentrations:

In addition to runoff, the concentrations of major ions in the runoff water, i.e. sulphate, nitrate and base cations, are needed to calculate SSWC critical loads, and these come from the analysis of representative water samples.

The critical load for a site should be calculated with yearly flow-weighted average chemistry and yearly average runoff. Since such values are not available for a large number of lakes, critical loads are mostly calculated on the basis of a single sample considered representative of yearly flow-weighted averages. A sample collected shortly after the fall circulation of a lake is generally assumed to fulfil this purpose. To check this claim, Henriksen and Posch (2001) compared critical load values calculated from yearly flow-weighted average concentrations with critical loads calculated from a single fall value for sites for which long-term data series are available. Results for seven Norwegian catchments show that the single fall value is fairly representative for the annual average chemistry. Similarly, results from eight Canadian catchments show that a single spring sample is fairly representative of the annual average chemistry (Henriksen and Dillon 2001).

### Lake and catchment characteristics:

The area parameters  $A$ ,  $A_l$ ,  $A_f$  and  $A_g$ , which are needed in the FAB model, can generally be derived from (digital or paper) maps.

### Terrestrial N sinks:

The uptake of  $N$  can be computed from the annual average amount of  $N$  in the harvested biomass. If there is no removal of trees from the catchment,  $N_u=0$ .

$N_i$  is the long-term annual immobilisation (accumulation) rate of  $N$  in the catchment soil. Note that at present immobilisation may be substantially higher due to elevated  $N$  deposition.

The denitrification fraction  $f_{de}$  depends on the soil type and its moisture status. In earlier FAB applications it has been estimated from the fraction of peat-lands,  $f_{peat}$ , in the catchment by  $f_{de}=0.1+0.7f_{peat}$

(Posch et al. 1997).

For more details on these parameters see Section 5.3.

### In-lake retention of $N$ and $S$ :

Concerning in-lake processes, the retention factor for nitrogen  $r_N$  (see eq. 5.72) is modelled by a kinetic equation (Kelly et al. 1987):

(5.87)

$$\rho_N = \frac{s_N}{s_N + z/\tau} = \frac{s_N}{s_N + Q/r}$$

where  $z$  is the mean lake depth,  $\tau$  is the lake's residence time,  $r$  is the lake:catchment ratio ( $=A_l/A$ ) and  $s_N$  is the net mass transfer coefficient. There is a lack of observational data for the mass transfer coefficients, especially from European catchments, but Dillon and Molot (1990) give a range of 2–8 m/yr for  $s_N$ . Values for Canadian and Norwegian catchments are given in Kaste and Dillon (2003).

An equation analogous eq. 5.87 for  $r_S$  with a mass transfer coefficient  $s_S$  is used to model the in-lake retention of sulphur. Baker and Brezonik (1988) give a range of 0.2–0.8 m/yr for  $s_S$ .