APPENDIX 1

Draft manuscript:

A Review of Uncertainties in Inputs for UK Acid and Nutrient Nitrogen Critical Loads Calculations

Elizabeth Heywood, Jane Hall, Brian Reynolds, Geoff Smith, Bridget Emmett, Mike Hornung, Chris Curtis and Mike Ashmore

Running head: Uncertainties in inputs for UK critical loads

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Authors: Elizabeth Heywood¹, Jane Hall¹, Brian Reynolds, Geoff Smith¹, Bridget Emmet, Mike Hornung⁴, Chris Curtis³, Mike Ashmore²

Affiliation: Centre for Ecology and Hydrology, Monks Wood, Cambridgeshire, PE28 2LS, United Kingdom ²Department of Environmental Science, University of Bradford ³Environmental Change Research Centre, University College London ⁴Centre for Ecology and Hydrology, Merlewood

Address: Centre for Ecology and Hydrology,

Monks Wood,

Abbots Ripton,

Huntingdon,

Cambridgeshire PE28 2LS,

United Kingdom

E-mail; elhe@ceh.ac.uk,

Telephone: +44(0) 1487 772506

Fax: +44(0) 1487 773467

Abstract. The calculation of critical loads for acidity and nutrient nitrogen uses data from a wide range of sources. For many of these inputs, no estimate of uncertainty has been calculated by the UK. This information is required at both the national and international scale for policy negotiations. Within Europe, protocols controlling the emissions of sulphur and nitrogen are being implemented and further reductions in these pollutants have an associated high cost. Consequently, it is imperative that uncertainties in the calculation of exceedances are quantified before further reductions in pollutants can be justified. The UK uses empirical and mass balance modelling techniques to calculate critical loads. The uncertainties in the inputs to these calculations are therefore required. The aim of this paper is to estimate these uncertainties. A comparison of the uncertainties derived for UK data and those calculated by other European countries reveals a wide range of uncertainty estimates. **Keywords**: acidity, critical loads, empirical, nutrient nitrogen, simple mass balance, uncertainties

1 Introduction

Air pollutants can adversely affect sensitive habitats, as highlighted in the report of the National Expert Group on Transboundary Air Pollution (NEGTAP, 2001). The critical load forms the basis of the effects-based approach, used to guide national and international policy on reducing the environmental impacts of transboundary air pollutants, such as sulphur dioxide, nitrogen oxide and ammonia. A critical load is defined as "A quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to present knowledge" (Nilsson *et al.* 1988 pp.8).

The amount of deposited pollutant above the critical load is termed the exceedance. Reducing the exceedance of critical loads is one of the main aims of international agreements to curb transboundary air pollution, such as the UNECE Protocol to Abate Acidification, Eutrophication and Ground-level ozone (1999), and the EC National Emission Ceilings Directive (2001). Syri *et al.* (1994) state that 'For many countries achieving emission ceilings will require costly abatement installation with high unit reduction costs'. Emission reductions and their costs can be weighted against the risk of critical load exceedance, thus providing additional information for the decision-making process. The inclusion of uncertainties in critical load and exceedance calculations is going to become a key activity in the UK and the rest of Europe in preparation for reviews of international protocols.

Each party to the UNECE's Convention on Long Range Transboundary Air Pollution (CLRTAP) is required to submit national input data and critical load calculations on a regular basis to the Coordination Centre on Effects (CCE) in the Netherlands. The critical loads for UK habitats are calculated and mapped by the National Focal Centre (NFC) for Critical Loads, at CEH Monks Wood. The development of methods to assess the uncertainties in critical load calculations is one of the priority tasks identified by the CLRTAP Working Group for Effects. This information is also required by the Department of Food and Rural Affairs (DEFRA) for national policy making decisions.

The UK NFC provided a short summary of a preliminary analysis with the CCE call for critical loads data in March 2003. This paper describes this work in more detail. The methods used to calculate the uncertainty estimates are described and a preliminary mathematical analysis carried out. However, it should be noted that this is preliminary work and the uncertainty estimates presented here are not being used for any policy negotiations, as further work is still ongoing.

A number of methods exist to determine the critical loads of acidity or nutrient nitrogen, which fall into two broad categories (i) mass balance and (ii) empirical approaches. The mass balance approach is based on balancing the long term chemical inputs and outputs (affecting acidity or nitrogen), to derive a critical load that ensures a critical chemical limit is not exceeded. Empirical approaches assign an acidity critical load to soils on the basis of soil mineralogy and/or chemistry. For example, at the Critical Loads Workshop at Skokloster (Nilsson and Grennfelt, 1988) soil forming materials were divided into five classes on the basis of the dominant weatherable minerals. Empirical nutrient nitrogen critical loads are evaluated based on observed changes in the structure and function of ecosystems, based on experimental and field evidence (Bobbink *et al.*, 1996, Bobbink *et al.*, 2002).

The UNECE's Mapping Manual (UBA, 1996) provides guidance on the calculation of critical loads, so that transparent common methods are applied across Europe. However, there remain variations in methods within Europe for the calculation of critical loads for different receptor systems. In this paper each method used for estimating critical loads in the UK is reviewed. It is not within the scope of this paper to discuss all the different methods used to calculate critical loads within Europe.

For each method, the theory is outlined; the estimated uncertainties for the UK described and comparisons made with other European countries. The uncertainties in each of the input parameters have either to be assessed "directly" (e.g. for empirical critical loads) or derived from the uncertainties in the variables used to calculate them, e.g. the variables and parameters needed in the mass balance models. Since the SMB model describes steady-state conditions, it uses long-term averages for the fluxes. Short-term variations - e.g. episodic, seasonal, inter-annual, due to harvest and as a result of short-term natural perturbations - are not considered, but are assumed to be included in the calculation of the long-term mean. Parameters may show variability spatially. This variability has been used to calculate uncertainty ranges for some input parameters. Where values have been taken from default ranges given in the literature, these ranges have been used to calculate the uncertainty around the value used. For a few parameters no estimates of uncertainties in the UK is available at the present time.

2 Critical Loads for Acidity

Two methods are used in the UK for calculating acidity critical loads for terrestrial habitats: the empirical approach used to provide estimates for non-woodland habitats and a simple mass balance equation for woodland habitats. Both methods make use of the empirical critical loads for soils.

2.1 Empirical Critical Loads for Acidity for non-woodland habitats

2.1.1 BACKGROUND TO UK EMPIRICAL ACIDITY CRITICAL LOADS

Empirical critical loads of acidity for soils have been assigned to the dominant soil in each 1km grid square of the UK. The critical loads are calculated using two methods. Hornung *et al.*, (1995), developed the method for calculating and mapping acidity critical loads for mineral and organic soils. These have been determined by linking the mineralogical classification of soils by Sverdrup and Warfvinge (1988) with the classification resulting from the Skokloster workshop (Nilsson and Grennfelt, 1988). The approaches were adapted to be made appropriate to UK conditions.

The calculation of acidity critical loads for peat soils sets the critical load to the amount of acid deposition that would give rise to an effective rain pH of 4.4. Yesmin *et al.* (1996) showed that the best correlation between transformed mycorrhizal infection of *Calluna* roots and deposition parameters was with effective rain pH; Dawod (1996), Procter and Maltby (1998) and Parveen (2001) have shown that peat soil solution pH equals effective rain pH. This method is specific to the UK and is not used by any other country that submits data to the CCE.

The estimated uncertainties associated with both empirical methods are described below.

2.1.2 UNCERTAINTIES IN EMPIRICAL ACIDITY CRITICAL LOADS

2.1.2.1 Empirical critical loads for non-peat soils

The data are mapped in five critical load classes (in keq ha⁻¹ yr⁻¹) <0.2; 0.2-0.5; 0.5-1.0; 1.0-2.0; and >2.0. For the current UK maps the mid-range empirical critical load values are used i.e. 0.1, 0.35, 0.75, 1.5 and 4.0 for the highest critical load. These values are consistent with work on soil weathering rates by Langan *et al.* (1995). He reported that the results of the PROFILE model (Warfvinge and Sverdrup, 1992), which is used widely in Europe to calculate weathering rates, compare very favourably with those suggested for the calculation of critical loads using the empirical approach; he showed that the calculated weathering rate falls mostly within the class range (Langan *et al.* 1995). The range was adopted to represent uncertainties in the weathering rate. Due to the limited data it was decided to apply a uniform distribution between the ranges in Table 1.

[Table 1 about here]

The median uncertainty from the table is $\pm 50\%$, which corresponds with the expected uncertainty using the "Skokloster" method of assigning empirical critical loads given in Sverdrup *et al.* (1990). The Austrian NFC bases the weathering rate classes on the Skokloster ranges. They estimate an uncertainty range of $\pm 40\%$ by using professional judgment (Schneider, 2001).

2.2.2 ACIDITY CRITICAL LOADS FOR PEAT SOILS

The UK is using a critical hydrogen ion concentration equivalent to an effective rain pH of 4.4 to calculate acidity critical loads for peat soils. This was derived from published regression analyses of rainwater to surface water pH based on UK data. The intercept of the regression was at pH 4.40 and the standard error of the intercept (i.e. the standard deviation of the sampling distribution) has been calculated from these data as 0.09. This method has been applied in the calculation of acidity critical loads for all peat-dominated 1km squares in the UK using the equation:

 $CLA = q * [H^+]$

where q = runoff in metres $\left[H^{+}\right] = 10^{pH}$ =critical hydrogen concentration equivalent to pH 4.4

The critical hydrogen concentration is the antilog of the pH. Using error analysis this gives us an uncertainty in H^+ of $s_{H^+} = 2.303 \cdot H^+ \cdot s_{pH}$. Taking the s_{pH} as 0.09 we have a coefficient of variation for the critical hydrogen concentration of 20%.

The run-off data are the mean 1km values for 1941-1970; the same data set as used in the Simple Mass Balance equation for acidity critical loads for forest soils. The

uncertainty of this parameter has been given a coefficient of variation of 23% and its derivation is discussed fully below. Error propagation techniques are used to calculate the total uncertainty of the acidity critical loads for peat soils giving a final coefficient of variation \pm -30%.

2.2 Simple Mass Balance Method for Acidity Critical Loads for woodland habitats

2.2.1 BACKGROUND TO UK MASS BALANCE ACIDITY CRITICAL LOADS

The Simple Mass Balance (SMB) model is a method widely used in Europe for predicting critical loads for acidity. The 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 and De Vries, 1994). In its simplest form, the SMB critical load for acidity can be expressed as:

 $CL(Ac) = BC_{w} - ANC_{le(crit)}$

where BC_w base cations produced by weathering and $ANC_{le(crit)}$ = critical leaching of Acid Neutralising Capacity (ANC).

The empirically derived critical loads described in the above section (i.e. assignment according to the proceedings of the Skokloster workshop) are used as weathering rates in the calculation of the SMB equation in the UK. However other countries within Europe use different methods. A full description of the alternative methods can be found in the UNECE Mapping Manual (UBA, 2004).

 $ANC_{le(crit)}$ can be calculated by either setting the critical aluminium and hydrogen ion concentrations and converting them to critical fluxes, or by defining the fluxes via a critical molar ratio of calcium or base cations to aluminium Ca:Al or BC:Al. Selecting the most appropriate criteria depends on whether the plant response is more sensitive to aluminium or pH toxicity. Two methods for calculating $ANC_{le(crit)}$ are described: one for mineral soils (more sensitive to aluminium) and organic soils (more sensitive to pH).

In the following section the equation is described in detail and the uncertainty associated with each input estimated for the UK and compared with other European countries.

2.2.2 UNCERTAINTIES IN MASS BALANCE ACIDITY CRITICAL LOADS

2.2.2.1 Chemical criteria

Work by Hall *et al.* (2001a & b) highlighted that the critical molar Ca:Al ratio in the soil solution is more appropriate for mineral soils than organic soils. The UNECE Workshop on Chemical Criteria and Critical Limits (Hall *et al.*, 2001c) recommended critical soil solution pH as the preferred criterion for organic soils.

To calculate critical ANC leaching the following equation is used:

 $ANC_{le(crit)} = -Al_{le(crit)} - H_{le(crit)}$

Using Ca:Al ratio as chemical criterion (mineral soils)

Critical leaching of aluminium (eq ha⁻¹ year⁻¹) is given by:

$$Al_{le(crit)} = \left(1.5 \times \frac{Ca_{le}}{Ca/Al}\right) \times 1000$$

and the critical leaching of hydrogen ions (eq ha⁻¹ year⁻¹) as: 1/3

$$H_{le(crit)} = \left(1.5 \times \frac{Ca_{le} \times 1000}{K_{gibb} \times Ca/Al}\right)^{1/2} \times (Q \times 10000)^{2/3}$$

where K_{gibb} is set to 950 m⁶/eq² and Ca_{le} (keq ha⁻¹ year⁻¹) is calcium leaching which is defined in terms of total (marine plus non-marine) calcium deposition for woodland, Ca_{dep} (keq ha⁻¹ year⁻¹) ,calcium weathering, Ca_w (keq ha⁻¹ year⁻¹) and net uptake of calcium, Ca_u (keq ha⁻¹ year⁻¹):

$$Ca_{le} = Ca_{dep} + Ca_{w} - Ca_{le\min} - Ca_{le}$$

where minimum calcium leaching (keq ha⁻¹ year⁻¹) is:

 $Ca_{lemin} = Q \times [Ca_l] \times 0.01$

There is a limiting concentration for uptake of calcium (Ca_l) of 2 μ eq l⁻¹.

The UK uses the critical ratio Ca/Al in the critical load calculations. Many countries apply the ratio Bc/Al. In most countries a value of one is applied, as suggested in the Mapping Manual (UBA, 2004) regardless of whether the Ca:Al or Bc:Al ratio is applied. This value has been questioned by Løkke et al. (1996) and Skeffington (1999). Cronan and Grigal (1995) reviewed the use of the Ca:Al molar ratios used as stress indicators in forest ecosystems. They estimated that there was a 50% risk of impacts on tree growth or nutrition with a Ca:Al ratio in soil solution as low as one, with this risk increasing as the ratio decreases. They estimated the overall uncertainty of the Ca:Al molar ratio, to be approximately \pm 50%. Alveteg (2001) who investigated the uncertainty in the Swedish national assessment of critical loads and exceedance for forest soils assumed a uniform distribution with a range of \pm 50%. The German NFC use the Bc:Al ratio in their acidity critical load calculations. Becker (pers. comm.) has carried out an uncertainty analysis for all input parameters to the critical load calculation. An uncertainty range of $\pm 10\%$ was used for the Bc:Al ratio and a uniform distribution was assumed. Compared to the other uncertainty estimates this would seem to be an optimistic assumption. Zak (2000) carried out Monte Carlo simulations of the SMB equation for Alice Holt in the United Kingdom using a most likely value for Bc:Al of 0.6, a minimum value of 0.3, and a maximum value of 1.0 (a triangular distribution is assumed). This is equivalent to a percentage uncertainty range of between -50 and +67%, close to the Cronan and Grigal estimate. For the UK national uncertainty assessment it was decided to adopt a uniform distribution with a range of \pm 50%.

Critical leaching of ANC is defined in this case as: $ANC_{le(crit)} = Q \times ([H] + K_{gibb} [H^3])$ where K_{gibb} is set at 9.5 m⁶/eq² for organic soils and [H] is the hydrogen ion concentration (eq m⁻³) calculated in terms of a critical pH: $[H] = 10^{(-pH)} \times 1000$

The pH value of 4.0 is recommended in the UNECE Mapping Manual (UBA, 2004) and is used by the UK. Ulrich (1987) proposed a critical pH of 4.2. There is an obvious paucity of literature on limits for organic soils. Hence a uniform distribution was assumed with a minimum value of 3.8 and a maximum value of 4.2.

2.2.2.2 Gibbsite equilibrium constant

The gibbsite equilibrium constant (K_{gibb}) , simulate the relationship between aluminium and hydrogen ions in soil solution:

 $[Al] = K_{gibb} \times [H]^3$

The value applied to mineral soils is 950 m⁶/eq² and the value for organic soils is 9.5 m⁶/eq². These values are based on the percentage of organic matter in the soil and are recommended in the UNECE Mapping Manual (UBA, 2004). A review of the current literature gives uncertainty ranges for K_{gibb} and pK_{gibb} (pK_{gibb} = -log₁₀(K_{gibb} in (mol/L)⁻²). NB. 1 m⁶/eq² = 10⁶/3 mol/L⁻².

Zak (2000) estimates the most likely value of K_{gibb} to be 300, but could be between 200 and 500 m⁶ eq⁻² at the Alice Holt field site in the UK. These numbers imply that a triangular distribution should be used for K_{gibb} . The maximum value from this range is well below those used for mineral soils for national critical loads mapping, as knowledge about the soil organic matter content would have been used.

Zak *et al.* (1997) estimated pK_{gibb} at the Phynlimon research area in Wales to be uniformly distributed from 8.0 to 9.0. This is equivalent to the range in the mapping manual for soils with low organic matter.

Abbott *et al.* (2003) used the ranges defined in the mapping manual (UBA, 2004) for the Liphook soil type (soils with low organic matter; B/C-layers: 300-3000 m⁶/eq²) to define the uncertainty ranges dependent on soil type. They used a log-normal distribution to weight towards low values (mean = 1025, standard deviation = 410 m⁶ eq⁻²).

The above uncertainty ranges were all derived for site specific studies. For the UK national scale analysis it was decided to use the CCE recommendation of $\pm 20\%$ and a uniform distribution (Suutari *et al.* 2001).

2.2.2.3 Calcium deposition

The calculation of the acidity critical load for forested mineral soils, based on the Ca:Al criterion, requires total calcium deposition (wet plus dry, marine plus nonmarine) values. To date no estimation of calcium deposition uncertainty has been calculated for the UK specifically. Other European countries mostly use the Bc:Al ratio and hence have made estimates of the uncertainties in base cation deposition uncertainty. Uncertainties in both base cation and calcium deposition estimates have been reviewed.

Draaijers *et al.* (1996) used error propagation, to estimate the random and systematic errors in total (wet and dry) base cation deposition for an average grid cell of 10x20 km in Europe. He quoted worst case coefficient of variation of between 90-150% and best case of 60-90% for random and systematic errors. A normal distribution was assumed.

The Netherlands used uncertainties derived by de Vries *et al.* (2000) for the deposition of base cations. By comparing model inputs at the European scale and value derived in various individual countries he estimated the uncertainty was generally less than 50%, although it could be more than 100%. This has been interpreted as a normal distribution with a CV of 50%.

Austria estimate base cation deposition from an empirical interpolation model. They estimate an average variance of $\pm 30\%$ for each Austrian grid square of 2.75x2.75 km² (Schneider J, 2001). Suutari *et al.* 2001 assumed a uniform distribution for the Austrian estimate. Germany (pers. comm.) chose a uniform uncertainty range of $\pm 20\%$ as a percentage of the mean. These two national estimates of uncertainty are in good agreement with one another.

Base cation deposition is estimated in Finland using interpolated data from a nationwide network of stations measuring monthly bulk deposition. An intercomparison of measurement methods and laboratory analysis carried out by Johansson and Janssen (1994) resulted in errors of less than $\pm 20\%$ of the base cation deposition values for the measurement network used. The coefficient of variation for the base cation deposition was increased to $\pm 30\%$ to account for measurement errors. Finland's estimate however assumes a normal distribution so does not correspond to the uncertainty estimates given by Austrian and Germany.

A base cation uncertainty of approximately \pm 70% was estimated using error propagation by Barkman *et al.* 1999 using a combination of triangular and rectangular distributions to account for the correlations between parameters. He was carrying out an uncertainty analysis for 67 forest site with Sweden. Abbott *et al.* 2003 carried out a site-specific Monte Carlo analysis at Liphook in Hampshire. They estimated a variation of \pm 50% and a log-normal distribution.

Varied estimates of uncertainty have been derived for this parameter and these vary widely depending on the whether they are attempting to represent uncertainty at the European, national or site specific scale.

As there is no UK specific uncertainty estimate for this parameter a subjective decision had to be made with discussion with atmospheric deposition experts. It was decided to use a normal distribution with a CV of $\pm 50\%$ for both base cation and

calcium deposition uncertainty for every 5km grid square in the UK. It should be realized when using these estimates in an uncertainty analysis that correlations between deposition parameters must be accounted for.

2.2.2.4 Base cation and calcium weathering

Base cation weathering

The empirical critical loads of acidity for soils (section 3.1.1), are based on the mineralogy and weathering rate characteristics of the dominant soils, and can therefore be used to provide ANC_w inputs to the SMB. The weathering rate uncertainty is hence the same as that used in section 3.1.2.

The methods used for calculating weathering rate in the UK differ from those of other European countries, but then these all differ from each other. It is hence almost impossible to compare uncertainty estimates. Discussed below are some of the different methods used by other European countries to calculate weathering rate and the associated uncertainty estimates.

Following a study of weathering rate calculation by PROFILE, Hodson *et al.* (1997) estimated the uncertainty range as high as $\pm 250\%$. However Barkman *et al.* (1997), again using the PROFILE model, derived uncertainty ranges of between $\pm 20-100\%$, dependent on mineral type.

The German NFC uses the soil type and general bedrock geology approximation. They estimated the uncertainty range using this method to be $\pm 20\%$ (Suutari *et al.*, 2001). He used a uniform distribution to define the uncertainty distribution. Becker (pers. comm.) state that "These uncertainties are derived from the experiences concerning the uncertainties of investigated parameters at the German Level II plots, which contains of uncertainties in measurement, laboratory analysis and further methods." The Netherlands, using the same method, considered that the overall uncertainty associated with the weathering rate would be somewhat larger than $\pm 50\%$ (de Vries *et al.*1994).

Finland estimated BC_w (= $Ca_w + Mg_w$ in Finnish calculations) using the total base cation content correlation. The uncertainty in the parameter values was expressed in terms of the coefficient of variation. The estimated uncertainty in weathering rate was separately calculated by varying the effective temperature sum by 10% (CV) and total soil contents of calcium and magnesium by 5% (CV). A triangular distribution was assumed. The overall uncertainty for BC_w was estimated to have a CV of 24 % (Johansson and Janssen, 1994).

Zak *et al.* (2000), who carried out a site specific analysis, gives a minimum value of 300, maximum value of 350 and a most likely value of 375 eq ha⁻¹ year⁻¹. This is equivalent to a triangular distribution with a percentage range -14% to +7%. *Calcium weathering*

The formulation of the SMB adopted in the UK for woodland on mineral soils uses a critical molar Ca:Al ratio of one in the soil soultion as the chemical effects criteria.

This means that the base cation terms in the calculation of $ANC_{le(crit)}$ need to be considered in terms of calcium only. As calcium weathering is a fraction of the total base cation weathering, estimates are obtained by applying "calcium correction" values to the base cation values:

$Ca_w = ANC_w *$ calcium correction value

The correction factors were derived from 'expert judgement' by the National Soils Resources Institute (NSRI), MLURI soil surveys and CEH Merlewood. An estimate was made of the proportion of calcium bearing minerals with differing weathering rates in the total weatherable minerals of the soil parent materials. Thus, for example, in soils over limestone or chalk, the weatherable minerals would be almost entirely rapidly weathering carbonates and in most cases calcium carbonates so the calcium correction factor would be one. In soils from granites, the main weatherable minerals releasing base cations would be relatively slowly weathering aluminium silicates which contain low to moderate amounts of calcium and hence were assigned values of 0.2 or 0.3. In terms of uncertainty, the uncertainties decrease from the low to high factors. Table 2 gives estimated uncertainties for each of the correction factors.

[Table 2 about here]

Hence the calcium weathering term will incorporate two sources of uncertainy; the estimate of base cation weathering and the calcium correction values selected for each 1km square of the United Kingdom.

2.2.2.5 Base cation, calcium and nitrogen uptake values

Uptake values are based on site-specific measurements from the ten Level II Intensive Forest Health monitoring sites in the UK. The map below shows the locations of these sites; there are three oak sites, three Scots pine sites and four Sitka spruce sites. [Figure 1 about here]

The mean of the three oak sites is applied to all broadleaved woodland, and the mean of the seven conifer sites applied to all coniferous woodland areas. The coefficient of variation of the three broadleaf and seven conifer plots are then taken as representative uncertainty estimates for their respective forest categories and the uncertainty ranges are given in Table 3. A normal distribution is assumed. [Table 3 about here]

These uptake values give us an estimate of the variation in the data sets used to calculate the uptake values on a country-wide basis and have been used in the national assessment of critical load uncertainty. Forest research plan to increase the number of points in this data set. It may then be possible to use geostatistical interpolation techniques to predict uncertainty spatially.

Two UK site specific studies estimated uncertainty in uptake values. Abbott *et al.* (2003) investigated a coniferous woodland site and used $\pm 50\%$ to represent the variation in both base cation and nitrogen uptake, although it was not clear what distribution type had been chose. Alice Holt, the site studied by Zak (2000), is an oak site and base cation uptake was given a most likely value of 433, minimum value of 300 and a maximum value of 610 eq ha⁻¹ year⁻¹ (-44% to +41%). Site specific

uncertainties in Sweden were estimated by Barkman *et al.* 1999. Owing to the use of the measure of the long-term biomass growth the range in annual uptake was assess to be approximately \pm 50% and a triangular distribution was assumed. If the percentage deviations of the triangular distribution were considered to represent the tails of the assumed normal distribution of the national estimate we would have almost direct correspondence between the national estimate and the Alice Holt and Swedish estimates.

All European countries use the same basic equation to calculate uptake values:

Loss from site	=	average volume	*	basic wood *	^c concentration
		increment		density	in wood
(keq ha ⁻¹ year ⁻¹)		$(m^3 ha^{-1} year^{-1})$		$(g m^{-3})$	(keq g^{-1})

However the methods used to estimate the uncertainty vary widely from country to country.

Germany used error propagation techniques to determine the uncertainty of the uptake value from the combination of uncertainties in volume, density and concentration. In this case the relative standard error of uptake values is calculated as

$$r_{uptake} = \sqrt{\left(r_V^2 + r_D^2 + r_N^2 + r_{BM}^2\right)}$$

where r_v = relative standard error in volume r_D = relative standard error in wood density r_N = relative standard error in nutrient content r_{BM} = relative standard error in conversion factor from stem biomass into whole tree biomass

The final uncertainty for both base cation and nitrogen content was estimated to be $\pm 15\%$ and a uniform distribution was assumed.

Austria uses expert judgement to estimate the average variance for both nitrogen and base cation uptake as $\pm 20\%$ for a 2.5x2.5 km grid square. Suutari *et al.* (2001) assumed a uniform distribution for these parameters.

The Netherlands compare model inputs from previous studies to estimate the uncertainty in both base cation and nitrogen uptake to be generally less than \pm 50%, although possibly more than \pm 100%. Hence we can assume a CV of 50% and normal distribution. Finland also assumes a normal distribution for the uptake uncertainties. Johansson and Janssen (1994) derive coefficients of variation for base cation uptake to be 37, 48 and 40% for birch, spruce and pine, and for nitrogen 37, 55 and 44% respectively. Both of these countries are predicting much wider bounds of uncertainty than have been assumed by the UK.

2.2.2.6 Runoff data

The UK national critical loads maps are produced using a mean annual runoff value for each 1 km square, calculated using a simple water balance model (Gustard *et al.* 1992). This model calculates the annual average runoff depth as the difference between the rainfall and actual evaporation losses. Arnell *et al.* (1990) evaluated a median CV for annual runoff of 23% for UK catchments and this is the range used for the UK national uncertainty analysis (a normal distribution has been assumed). Abbot *et al.* (2003) and Zak (2000) both assumed triangular distributions for their UK site specific studies. However Zak (2000) used a range of -30% to +33% and Abbott *et al.* (2003) a range of $\pm 10\%$.

Austria bases their calculation of runoff on the following empirical relationship:

$$Q = \left(P - \left[\left(12 - H \times 0.005\right) \times \frac{P}{100}\right] + \left(420 - H \times 0.005\right)\right) \times \left(1 - NK \times a\right)$$

where: P = precipitation (mm) H = altitude (m) NK = slope correction factor a = constant factor

Hence the accuracy of the runoff data will be influenced by the accuracy of all these data sets. The long-term uncertainty of the precipitation data is estimated at $\pm 30\%$ (Schneider J, 2001). However it is stated that "A problem for the calculation of Q is the lack of survey data on soil physical characteristics, so a variance of $\pm 50\%$ is expected at the spatial scale of the grids." Suutari *et al.* 2001 assumed a uniform distribution for this parameter in their uncertainty analysis. Suutari *et al.* 2001 reports that Germany assigns an estimate of $\pm 15\%$ to the uncertainty in run off. They assume a uniform distribution.

The Netherlands use interpolated data from weather stations to obtain values for precipitation. Interception fractions, relating interception to precipitation, have been derived from the literature for pine, spruce and deciduous forest. The uncertainty in runoff has been estimated by De Vries *et al.* (1994) at approximately \pm 50%, although he states that it might be as low as \pm 25%. To account for this variation a normal distribution with a coefficient of variation of \pm 50% is assumed. Finland assumes measurement errors are very small for run-off and they assign a coefficient of variation of only 5% to the runoff (Johansson and Janssen, 1994). These UK estimate lies between these two extremes.

3 Critical Loads of acidity for Freshwaters

3.1 Background to the FAB model

The FAB model applied to UK freshwater (Curtis *et al.*, 2000) is based on previous work by Henriksen *et al.* (1998) and Posch *et al.* (1997a). It has recently been reformulated to takes account of direct deposition to the lake surface (Henriksen & Posch (2001)). Critical loads are calculated for a number of freshwater sites in

acidified regions for which survey information exists. The FAB model is an aquatic charge balance equation and takes into account in-lake processes. While it supersedes the Steady State Water Chemistry (SSWC) model, 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 SSEC model and the key principles is included here.

3.1.1 THE SSWC MODEL FORMULATION

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):

$$L_{crit} = \left(\left[BC_0^* \right] - \left[ANC_{crit} \right] \right) \times Q$$

where $L_{crit} = critical$ leaching flux of acid anions

Q = catchment runoff $[BC]_{o}^{*}$ = pre-industrial concentration of non-marine base cation ANC_{crit} = pre-selected critical ANC threshold

The proportion of base cations per unit inpt of acid anions derived from ion exchange in the soil complex is represented in the SSEC model by the term "F", calculated according to the methodology of Brakke *et al.* (1990):

$$F = \sin\left(\frac{\pi}{2} \frac{\left[BC\right]_{r}^{*}}{S}\right)$$

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 μ eql⁻¹ (Harriman and Christie, 1995). F is used to calculate the pre-industrial base cation concentration according to the following equation (Henriksen *et al.*, 1992):

$$[BC]_{0}^{*} = [BC]_{t}^{*} - F([AA]_{t}^{*} - [AA]_{0}^{*})$$

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 nonmaringe base cations ($[BC]_{t}^{*}$) represents the sum of weathering, non-marine deposition and ion-exchange sources (BC_{leach}). Background concentrations of SO_{4}^{2-} are determined from empirical relationships between base cations and sulphate in near-pristine lakes (Henriksen *et al.*, 1990, 1992).

3.1.2 GENERAL DESCRIPTION OF THE FAB MODEL

The acidity balance for the FAB model is as follows:

$$N_{dep} + S_{dep} = fN_{u} + (1 - r)(N_{i} + N_{de}) + rN_{ret} + rS_{ret} + BC_{l} - ANC_{l}$$

Where:

$$\begin{split} N_{dep} &= nitrogen \ deposition \\ S_{dep} &= sulphur \ deposition \\ f &= fraction \ of \ forest \ in \ the \ catchment \\ r &= lake \ to \ catchment \ ratio \\ N_u &= net \ growth \ uptake \ of \ N \ by \ coniferous \ forests \ in \ the \ catchment \\ N_i &= nitrogen \ immobilization \ in \ the \ catchment \ soils \\ N_{de} &= nitrogen \ lost \ by \ denitrification \\ N_{ret} &= nitrogen \ retention \ in \ the \ lake \\ S_{ret} &= sulphur \ retention \ in \ the \ lake \\ BC_l &= base \ cation \ leaching \ from \ the \ catchment \\ ANC_l &= ANC \ leaching \ from \ the \ catchment \end{split}$$

3.2 Uncertainties in the FAB model

Curtis *et al.* (2000) discuss some of the uncertainties of the FAB model when applied to the UK national datasets. Posch *et al.* (1993) assessed the uncertainty of the SSWC Model for lakes in Finland.

3.2.1 CRITICAL LEACHING FLUX OF ACID ANIONS, Lcrit

The L_{crit} term from FAB is calculated using the equation:

$$L_{crit} = \left(\begin{bmatrix} BC_o^* \end{bmatrix} - \begin{bmatrix} ANC_{crit} \end{bmatrix} * Q \right)$$

Where BC_0^* is the pre-industrial concentration of non-marine base cations, ANC_{crit} is the pre-selected critical ANC threshold and Q is the annual run-off. Uncertainties are associated with each variable.

The steady-state FAB model requires data inputs that are based on long-term averages. However, such data are not always available. In the UK, the FAB model currently uses data from a single water sample collected at each site mapped. Therefore, a major source of uncertainty is how representative the water chemistry from this single sample is of the annual mean chemistry.

Pre-industrial concentration of non-marine base cations are derived from time series data for base cation concentrations for each of the sites contained within the freshwater dataset. The standard deviation of the time series is calculated to give an estimate of how far the single sample may vary from the mean chemistry. The coefficient of variation of pre-industrial base cations has been calculated for 13 N budget sites. For lakes the median coefficient of variation is $\pm 20\%$ and for rivers it is $\pm 40\%$. The derivation of run-off uncertainty has been recorded already in section 2.2.2.6

3.2.1.2 Choice of ANCcrit

For freshwaters ANC is chosen as the critical chemical value and fish as the biological indicator. ANC_{crit} is hence the critical concentration for fish. A stakeholder workshop was held prior to the 2004 data submission to discuss and agree the most appropriate value(s) of ANC_{crit} to be applied in the calculation of acidity critical loads for UK freshwaters. The workshop presented evidence from palaeolimnological, static and dynamic models that suggests that the great majority of surface waters in the UK had a pre-industrial ANC of >20 μ eql⁻¹. Biological data suggest that a number of organisms may be adversely affected when mean ANC declines to 0 μ eql⁻¹ but an increase from 0 to 20 μ eql⁻¹ represents a major improvement in biological status. Hence ANC_{crit} = 20 μ eql⁻¹ may reasonably be considered to provide a defensible threshold for acidity.

From this evidence a triangular distribution with a minimum at 0, maximum at 40 and a mode at 20 μ eql⁻¹ would seem appropriate. This also agrees with the Posch *et al.* (1993) study where ANC_{crit} was varied around 20 μ eq L⁻¹ following a triangular distribution with a range of $\pm 100\%$.

3.2.2 FOREST UPTAKE DATA

The freshwater critical load data set incorporates N_{uptake} data based on managed forest cover data, which provides a long-term sink for N. Hence the nitrogen uptake term incorporates two sources of uncertainty; the estimate of forest cover and the single rate of N uptake selected for coniferous and deciduous areas.

The managed coniferous woodland areas were calculated as:

Managed conifers = (ratio of FR managed coniferous woodland are to FR total woodland area) * LCM coniferous woodland area

Where FR total woodland area = sum of managed and unmanaged coniferous and broadleaved Forest Research woodland data

And LCM coniferous woodland area = Land Cover Map 2000 managed and unmanaged coniferous woodland area

The managed broadleaved woodland areas were calculated as:

Managed broadleaved = (ratio of FR managed broadleaved woodland area to FR total woodland area) * LCM broadleaved/mixed woodland area

LCM broadleaved woodland area = Land Cover Map 2000 managed and unmanaged broadleaved woodland area

The accuracy of the land cover data for coniferous forests in the UK has been estimated as 95% Fuller *et al.* 2002. The uncertainty associated with N uptake is discussed in section 2.2.2.5. Since the accuracy of the landcover data is considered to be so much better than the uptake data the uncertainty in N uptake is expected to dominate this parameter.

3.2.3 DENITRIFICATION DATA

For the UK the FAB model is 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, Denitrification uncertainty is discussed in section 4.2.2.2.

For freshwater calculations the denitrification data incorporates two sources of uncertainty; the derived lake to catchment ratio and the rates of denitrification selected depending on soil type. MLURI have assessed uncertainty in catchment boundary delimination (pers comm.). Their method assessed water catchment sensitivity to errors in digital elevation data for boundary delimination. They have applied their model to a national database of 700 standing waters in Scotland. They found that the catchment area error varied between 0 and 15%.

3.2.4 LONG TERM NITROGEN IMMOBILISATION

Estimates of the long term immobilization 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 immobilization rate. In the UK a value of 1 or 3 kg ha⁻¹ yr⁻¹ is chosen depending on soil type and organic matter content (Hornung *et al.*1995a), 1 being chosen for more mineral soils and 3 for more organic soils. Nitrogen immibilisation uncertainty is discussed in section 4.2.2.2. 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. The uncertainty in the lake:catchment ratio has already been discussed in section 3.2.3. Note 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.

3.2.5 INLAKE RETENTION COMPONENT

The in-lake retention of acid anions is assumed to be proportional to the net input of acidity i.e.

$$rN_{ret} = \rho_N [N_{dep} - fN_u - (1 - r)(N_i + N_{de})]$$

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

$$\rho_{N} = \frac{S_{N}}{S_{N} + Q/r}$$

where S_N is the mass transfer coefficient for nitrogen, Q is runoff and r is the lake to catchment area ratio.

The range of mass transfer coefficients recommended in the UNECE Mapping Manual (Ss = 0.2 to 0.8 m yr⁻¹ and S_N = 2 to 8 m yr⁻¹) has been found to be appropriate for lakes in European countries. Posch *et al.* (1993) used mid-range values (S_s=0.5 m yr⁻¹, S_N = 5 m yr⁻¹) for the most probable value and appointed triangular distributions with a range of \pm 60%. The UK will use the same uncertainty range.

3.2.5.1 Runoff data

Whitehead *et al.* (2002) note that runoff data in the UK has to be interpolated onto a catchment area from grid-based model outputs that are themselves uncertain (as discussed in section 2.2.6 of this report). The uncertainties in the interpolation procedure have already been discussed.

Posch *et al.* (1993) assumed the runoff in each catchment to vary 20% around its interpolated value i.e. \pm 10%, following a triangular distribution.

4 Critical Loads for Nutrient Nitrogen

The UNECE recommend two main approaches to calculating critical loads for nutrient nitrogen. The empirical approach (section 4.1), in which critical loads are estimated for different ecosystems based on experimental or field evidence of thresholds for change in species composition, plant vitality or soil processes.

The second is a steady state mass balance approach (section 4.2) in which long-term inputs and outputs of nitrogen from the system are calculated, with the critical load being exceeded when any excess nitrogen input is calculated to lead to exceedance of a critical rate of nitrogen leaching. This mass balance approach is applied to managed woodland. The mass balance approach is best suited to managed ecosystems of low biodiversity, in which inputs and outputs can be quantified with some confidence and the key concern is nitrate leaching. The empirical approach is better suited to semi-natural communities for which the long-term protection of biodiversity and/or system function is the key concern. For these reasons, the UK chooses to use both mass balance and empirical approaches in mapping critical loads across the country, applying them to different types of ecosystem.

4.1 Empirical Critical Loads for Nutrient Nitrogen

4.1.1 BACKGROUND TO THE UK EMPIRICAL CRITICAL LOADS OF NUTRIENT NITROGEN

A review of the empirical critical loads recommended for application across Europe has resulted in the adoption of new recommended empirical critical values at a formal UNECE workshop held in Berne in November 2002. The proposed mapping values for non-forest ecosystems in the UK are summarised in Table 4, alongside the recommended ranges of critical loads adopted at the Berne workshop. The reasons for the choice of the mapping values are explained in detail in Hall et al. (2003).

4.1.2 UNCERTAINTIES IN EMPIRICAL NUTRIENT NITROGEN CRITICAL LOADS

It is important to note in interpreting Table 4 that the critical loads are expressed as a range rather than a single value. This range indicates the real variation in sensitivity within a particular ecosystem, for example, because of differences in nutrient status, management etc.

Table 4 provides estimates of the reliability of each critical load. There are three rated as "reliable", six rated as "quite reliable" and three are derived from "expert judgement" or best guess. It is important to note that this represents a judgement of the extent and quality of the scientific evidence available from which critical loads might be estimated. Even where the evidence is classed as reliable, there may be different views on its interpretation and therefore on the appropriate critical load range.

So far in Europe no uncertainty ranges have been reported on nutrient nitrogen empirical critical loads. Abbott et al. 2003 discusses some of the uncertainties in empirical nitrogen critical loads. Experts in the UK believe an estimate of uncertainty needs to combine both the range of empirical critical loads and the estimate of reliability. Hence in the UK, uncertainty has been estimated using triangular functions, with the selected UK mapping value as the maxima of the distribution, and the ends of the ranges representing the tails of the distributions. Additionally, the critical load ranges have been extended for each reliability category to incorporate an element of uncertainty, as follows:

range as published ##

 $\pm 5 \text{ kg N ha}^{-1} \text{ year}^{-1}$ beyond the range $\pm 10 \text{ kg N ha}^{-1} \text{ year}^{-1}$ beyond the range #

(#)

The only exception to this rule, was the critical load for bogs (EUNIS class D1), where the UK is using the upper limit of the range as its mapping value (ie, 10 kg N ha⁻¹ year⁻¹); to deal with this the maximum of the range was increased to 12 kg N ha⁻¹ year⁻¹ to provide a reasonable estimate of uncertainty.

4.2 Steady State Mass Balance Critical Loads for Nutrient Nitrogen

4.2.1 BACKGROUND TO THE UK MASS BALANCE NUTRIENT NITROGEN CRITICAL LOADS

The steady-state mass balance (SSMB) method is used to provide critical loads to protect the tress rather than the ground flora. In the UK the SSMB is applied to managed (coniferous and broadleaved) woodlands. The critical load is calculated as: $CL_{nut}N = N_u + N_i + N_{le(acc)} + N_{de}$

where N_u = nitrogen uptake

 N_i = nitrogen immobilisation $N_{le(acc)}$ = acceptable level of nitrogen leaching N_{de} = denitrification

Nitrogen immobilization and denitrification values are dependent on soil type. Acceptable nitrogen leaching is ecosystem specific.

4.2.2 UNCERTAINTIES IN MASS BALANCE NUTRIENT NITROGEN CRITICAL LOADS

4.2.2.1 Nitrogen Immobilisation

The net amount of nitrogen that can be sustainably stored (accumulated) in forest soils is a highly uncertain parameter. Sverdrup *et al.* 1990 (page 55) gives a range of nitrogen immobilization of 1-3 kg ha⁻¹ yr⁻¹. In the UK a value of 1 or 3 kg ha⁻¹ yr⁻¹ is chosen depending on soil type and organic matter content (Hornung *et al.*1995a), 1 being chosen for more mineral soils and 3 for more organic soils. The higher value is chosen for more organic soils since the nitrogen content of the soil profiles tend to be higher.

Emmett and Reynolds (1996) calculated a long-term average of nitrogen accumulation in the soil of 2.1 kg ha⁻¹ yr⁻¹ for a productive spruce forest using the accepted method of dividing the total nitrogen content of soil profiles by the age of the profile i.e. the number of years since glaciation. The mapping manual (2004) recommends values between 0.2 and 0.5 kg ha⁻¹ yr⁻¹. According to previous mapping guidelines N_i values of between 0.5 and 1 kg ha⁻¹ yr⁻¹ (UBA, 1996) and 2-5 kg ha⁻¹ yr⁻¹ Posch *et al.* (1993) have been recommended. These results illustrate that there is no consensus yet on long-term sustainable immobilization rates.

A uniform uncertainty range of 0.5 - 1.5 kg ha⁻¹ yr⁻¹ was chosen for more mineral soils for the UK national uncertainty range. The lower bound was chosen to correspond to the mapping manual recommended values of between 0.5 and 1 kg ha⁻¹ yr⁻¹. An uncertainty range of 1.5-4.5 kg ha⁻¹ yr⁻¹ was chosen for more organic soils.

Abbott *et al* (2003) chose to vary the N_i value for the site specific analysis between the lower bound used by the UK (1 kg ha⁻¹ yr⁻¹) and an extended upper bound of 4.9 kg ha⁻¹ yr⁻¹. They chose a uniform distribution to represent the uncertainty.

Germany rank nitrogen immobilization rates according to temperature. A uniform uncertainty range of $\pm 5\%$ is used by Suutari *et al.* 2001. The Netherlands chose an uncertainty range of $\pm 50\%$ following De Vries *et al.* 1994,2000.

The UK employs default values for denitrification (Hall *et al.*, 1998, see Appendix 1B). Recent work under Defra's Freshwater Umbrella (Curtis, 2001; Curtis, 2003), suggests that the default values are much more appropriate than the UNECE Mapping Manual (UBA, 1996) suggested method of 10-80% denitrification, determined by percentage peat cover. The assumption of very high denitrification rates in peat soils disguises the fact that most retained N in mass balance models is probably immobilised in soils rather than denitrified.

Grennfelt & Thornelof, 1992 (Appendix 1) give default range values of between 0 and 5 kg ha⁻¹ yr⁻¹, with 4-5 used for sites with waterlogged soils and high deposition, 2-3 for sites with waterlogged soils and low deposition and 0-1 for aerated soils. In the UK values of 1, 2 and 4 kg ha⁻¹ yr⁻¹ are chosen depending on soil type (Hornung *et al.* 1995a) and defined in the Status Report 1998 appendix V. Uncertainty ranges were chosen to correspond with those ranges given by Grennfelt & Thornelof (1992) and to attempt to create symmetrical range around the default values already being used. Hence for those soils using a default value of 1 an uncertainty range of 0.5-1.5 kg N ha⁻¹ yr⁻¹ was chosen, for those using a default value of 2 an uncertainty range of 1.5 - 2.5 kg N ha⁻¹ yr⁻¹ was used. A uniform distribution was chosen to reflect the high level of uncertainty in the choice of Nde.

Abbott *et al.* (2003) applies an uncertainty of \pm 50% centred on the lowest value used in UK calculations. If we assume a uniform distribution then this range is comparable to the one chosen for a national estimate if uncertainty.

The Netherlands derives denitrification fractions for each soil type based on data given in Breeuwsma (1991) and corrected for the more acidic forest soils. De Vries *et al.* 1994, 2000) estimated the uncertainty as $\pm 50\%$. Suutari *et al.* (2001) give Germany's estimated uncertainty of the denitrification fraction as $\pm 20\%$.

4.2.2.3 Nitrogen Uptake

Nitrogen uptake values and uncertainties for forest systems have already been discussed in section 3.2.2.5.

4.2.2.4 Nitrogen leaching

For managed conifers, Emmett *et al.* (1993) and Emmett & Reynolds (1996) suggest a range of 1-5 kg N ha⁻¹ year⁻¹. The UK has set and a single value of 4 kg N ha⁻¹ year⁻¹ as this is similar to annual leaching losses from upland acid grassland in Wales (Emmett *et al.* 1993) and is also similar to leaching losses observed in coniferous forests in natural conditions. The range suggested by Emmett *et al.* (1993) and Emmett & Reynolds (1996) has been used as a measure of the uncertainty associated with this parameter. A triangular distribution with the mode set at 4, minimum at 1 and maximum at 5 kg N ha⁻¹ year⁻¹ has been used.

For managed broadleaved woodland, a range of 1-3 kg N ha⁻¹ year⁻¹ (Emmett, pers. Comm.) was considered, and a precautionary value of 3 kg N ha⁻¹ year⁻¹ has been

used in the calculation of $CL_{nut}N$. The mapping manual (UBA, 1996) recommends a range of 2 – 4 kg ha⁻¹ yr⁻¹ for nitrogen leaching for temperate deciduous. Using the lower bound of the recommendation by Emmett and the upper bound of the mapping manual we have an uncertainty range of between 1 and 4 kg N ha⁻¹ year⁻¹. A triangular distribution was again chosen with the mode at 3 kg N ha⁻¹ year⁻¹.

There is very little literature within Europe on the uncertainty range that could be associated with this parameter. De Vries *et al.* (2000) calculates the natural N leaching rate by multiplying the precipitation excess by a natural background NO₃ concentration in drainage water of 0.02 mol_c m⁻³ and assumed an uncertainty in the critical N leaching rate of \pm 50%.

5 Conclusions and Future Development

This study has highlighted the high degree of variation in the reporting of uncertainty in critical load model parameters throughout Europe. There are conflicting views on the uncertainties that should be attached to critical load input parameters. Table 1 summarises the estimates of uncertainties derived by other European countries and those derived by the UK.

Weathering rate uncertainty demonstrates well the disparity between uncertainty estimates and yet sensitivity analyses (Hodson and Langan, 1999, Final defra report) have demonstrated that it is one of the most influential parameters when attempting to calculate critical loads nationally. The German NFC estimated the smallest range in weathering rates as $\pm 20\%$ and assumed a uniform distribution. The Austrian NFC assumed an uncertainty range of $\pm 40\%$, also assuming a uniform distribution. Finland used a coefficient of variation of 24%, but a normal distribution was assumed, which would give a 95% confidence range of 50%. Both Sverdrup *et al.* (1990) and De Vries *et al.* (2000) assumed a range of $\pm 50\%$ but no information about the distribution was given. Barkman *et al.* (1997) gave a CV of roughly $\pm 30\%$ although this was variable depending on tree type. Hodson *et al.* (1997) estimated by far the largest uncertainty ranges as $\pm 250\%$. The UK estimate of between $\pm 33\%$ and $\pm 100\%$ depending on soil type is generally comparable with the ranges used elsewhere in Europe

There are several problems that must be considered when interpreting the uncertainty ranges given in the literature:

- 1. Inconsistency in the way the uncertainty ranges are quoted, sometimes without the underlying distribution being explicitly stated. It is not always clear whether the percentage being quoted is a range or a coefficient of variations. Also a range quoted as $\pm 20\%$ has been assumed to mean a variation of between -20% of the mean to +20% of the mean, i.e. a **total** range of 40% about the mean. This assumption may however be incorrect at times.
- 2. Part of the variation between different countries' estimates of uncertainty is due to the different methods of calculating parameters. This is particularly the case for the calculation of weathering rate where there are many different methods used across Europe.
- 3. The methods used to assess the uncertainty ranges vary. Some analyse the parameter values over time, some the uncertainties in the input values to the

parameter being calculated and some compare field data to laboratory data. Where expert judgement has been used a large degree of subjectivity will be included in the assessment.

- 4. An important question which was noted in Abbott *et al.* (2003) addresses the spatial scale in the assessment of an uncertainty range. When estimating an uncertainty range for a parameter, e.g. base cation deposition, within a grid square are we accounting for the variability within the grid square? If so the estimates of uncertainty for base cation deposition would be expected to vary depending on the resolution of the data.
- 5. A further discrepancy, recognised by Abbott *et al.* (2003) is that ecosystem values are not always explicitly defined, eg uptake values.

Plausible ranges and distributions are required for Monte Carlo analysis to assess the uncertainty in critical loads and their exceedances. To get the most out of such an analysis, good-quality data characterising the uncertainties is needed. This paper is useful because it highlights arbitrariness of many of the assumptions made so far and attempts to objectively define input ranges for the UK uncertainty analysis. It involved a hierarchy of error assessment including:

- Expert judgement by single expert
- Consensus judgement by group of experts
- Uncertainty range estimated from observations at different sites
- Uncertainty estimated from model calculations

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Table 1. Uncertainties of the base cation weathering rates

Mid-range valueRange (\pm percentage of
mid-range value)

0.1	100	
0.35	43	
0.75	33	
1.5	33	
4.0	50	

Table 2. Uncertainties of the calcium correction factors.

Calcium correction factor	Range (\pm percentage of factor)
0.02 (NI and IOM only)	100
0.05 (Scotland only)	100
0.1	100 (0.0 – 0.2)
0.15 (Scotland only)	100
0.2	80 (0.04 - 0.36)
0.3	60 (0.12 – 0.48)
0.4	40 (0.24 – 0.56)
0.6	20 (0.48 – 0.72)
1.0	10 (0.8 – 1.0)

Table 3	The input parameter values and their coefficients of variation (CV) for
the uncertaint	analysis on the net uptake

Woodland Type	Uptake Values (keq ha ⁻¹ year ⁻¹)			CV		
	base cations	calcium	nitrogen	base cations	calcium	nitrogen
Conifers	0.27	0.16	0.21	23 %	27 %	27 %
Broadleaved Ca-rich soils	0.41	0.29	0.42	14 %	12 %	7 %
Broadleaved Ca-poor soils	0.315	0.195	0.42	14 %	12 %	7 %

Table 4

ClnutN EUNIS classes (UK only)	Critical Load Range (kg N ha ⁻¹ yr ⁻¹)	UK Mapping Value (kg N ha ⁻¹ yr ⁻¹)	Reliability score
Native Scots Pine in	10-15	12	#
UK			
Atlantic oak woods in	10-15	10	(#)
UK			
Calcareous grassland	15-25	20	##
Acid grassland	10-20	15	#
E3.5 Moist or wet	10-20	15	#
oligotrophic grassland			
E3.52 Nardus stricta	10-20	15	#
swards			
F4.11 "U" Calluna	10-20	15	(#)
dominated wet heath			
F4.11 "L" Erica	10-25	15	(#)
dominated wet heath			
F4.2 Dry heaths	10-20	12	##
D1 Raised and blanket	5-10	10 (lower for low	##
bogs		rainfall)	
Moss/lichen mountain	5-10	7	#
summits (montane)			
B1.4 Coastal stable	10-20	15	#
dune grassland			
Unmanaged woodlands	10-15	12	#

reliable, # quite reliable, (#) expert judgement

Critical	Country	Range	Distribution	Justification
loads				
parameter	-			
BC_{dep} and	Europe	90-150%(10x20km)	Normal	Draaijers et al. (1996)
Ca _{dep}	Sweden	30%+40% (site specific)	Uniform+ Triangular	Barkman (1999)
	Austria	30% (2.5x2.5 km)	Uniform	Schneider (2001)
	Germany	20%	Uniform	Suutari et al. (2001)
	Netherlands	50% (site specific)	Normal	De Vries <i>et al.</i> (2000)
	Finland	30%	Normal	Johansson and Janssen (1994)
	UK (National)	50% (site specific)	Normal	Abbott <i>et al.</i> (2003)
BC _u	Sweden	Coniferous 50% Deciduous 50%	Triangular	Barkman (1999)
	Austria	Forests 20%	Uniform	Schneider (2001)
	Netherlands	50%	Normal	De Vries et al. (2000
	Germany	15%	Uniform	Suutari et al. (2001)
	Finland	Birch 37% Spruce 48% Pine 40%	Normal	Johansson and Janssen (1994)
	UK (Alice Holt)	Broadleaved –44% to + 41%	Triangular	Zak (2000)
	UK (Liphook)	Coniferous 50%	-	Abbott <i>et al.</i> (2003)
	UK (National)	Conifers 23% Broadleaved 14%	Normal	Experimental data; this paper
ANC _w	Europe	50% (Skokloster method)	-	Sverdrup et al. (1990)
	Scotland	250% (Profile)	-	Hodson <i>et al.</i> (1997)
	Sweden	25-80% (Profile) dependent on tree type	Normal	Barkman (1999)
	Germany	20% (soil type and general bedrock geology approximation)	Uniform	Suutari <i>et al.</i> (2001)
	Netherlands	50% (soil type and general bedrock geology approximation)	-	De Vries <i>et al.</i> (2000)
	Finland	24% (total base cation content correlation)	Normal	Johansson and Janssen (1994)
	Austria	40% (Skokloster)	Uniform	Schneider (2001)
	UK (Alice Holt)	-14% to + 7% (Profile)	Triangular	Zak (2000)

Table 5 Summary of UK critical load values and justification for their use

	UK (National)	$100 \text{ eq } ha^{-1} yr^{-1} = \pm 100\%$ $350 \text{ eq } ha^{-1} yr^{-1} = \pm 43\%$ $750 \text{ eq } ha^{-1} yr^{-1} = \pm 33\%$ $1500 \text{ eq } ha^{-1} yr^{-1} = \pm 33\%$ $4000 \text{ eq } ha^{-1} yr^{-1} = \pm 33\%$	Uniform	Langan <i>et al.</i> (1995)
Nu	Sweden	± 50%Coniferous 50%Deciduous 50%	Triangular Triangular	Barkman (1999)
	Austria	20%	Normal/uniform	Schneider (2001)
	Netherlands	50%	Normal	De Vries <i>et al.</i> (2001)
	Germany	15%	Uniform	Suutari <i>et al.</i> (2001)
	Finland	Birch 37% Spruce 55% Pine 44%	Normal	Johansson and Janssen (1994)
	UK (Liphook)	Coniferous 50%	-	Abbott <i>et al.</i> (2003)
	UK (National)	Coniferous 27% Broadleaved 7%	Normal	Experimental data, this paper
Ni	Germany UK	5% Minimum: 71 eq	Uniform Uniform	Suutari <i>et al.</i> (2001) Abbott <i>et al.</i> (2003)
	(Liphook)	$ \begin{array}{c} ha^{-1} yr^{-1} \ (1 \ kg \ N \ ha^{-1} yr^{-1}) \\ Maximum: 350 \ eq \\ ha^{-1} yr^{-1} \\ Mean: 210.5 \ eq \ ha^{-1} yr^{-1} \\ \end{array} $		
	UK (National)	Uncertainty range: 0.5-1.5 kg N ha ⁻¹ yr ⁻¹ for more mineral soils Uncertainty range: 1.5-4.5 kg N ha ⁻¹ yr ⁻¹ for more organic soils	Uniform	Sverdrup <i>et al.</i> (1990) Emmet and Reynolds (1996)
N _{le(acc)}	Europe	0.5-1 kg N ha ⁻¹ yr ⁻ for managed conifers 2-4 kg N ha ⁻¹ yr ⁻ for broadleaved	-	UBA (1996)
	Finland	50%	Normal	De Vries <i>et al.</i> (2001)
	UK (National)	$\begin{array}{c} 1-5 \text{ kg N ha}^{-1} \text{ yr}^{-1} \\ \text{for managed} \\ \text{conifers} \\ (0.0715 - 0.3575 \\ \text{keq ha}^{-1} \text{ yr}^{-1} \\ 1-3 \text{ kg N ha}^{-1} \text{ yr}^{-1} \\ 1-3 \text{ kg N ha}^{-1} \text{ yr}^{-1} \\ \text{for managed} \\ \text{broadleaved} \\ (0.0715 - 0.2145 \\ \text{keq ha}^{-1} \text{ yr}^{-1} \\ \end{array}$	Triangular Mode = 4 kg N ha ⁻¹ yr ⁻¹ for conifers Mode = 3 kg N ha ⁻¹ yr ⁻¹ for broadleaved	Emmett <i>et al.</i> (1993) and Emmett & Reynolds (1996)
N _{de}	UK (Liphook)	50%	Normal	Abbott <i>et al.</i> (2003)

	UK (National)	0.5 - 1.5 kg N ha ⁻¹ yr ⁻¹ for aerated soils 1.5-2.5 kg N ha ⁻¹ yr ⁻¹ for sites with waterlogged soils and low deposition 2.5-5.5 kg N ha ⁻¹ yr ⁻¹ for sites with waterlogged soils and high deposition.	Uniform	Grennfelt & Thornelof, 1992 (Appendix 1)
Q	Austria	50%	-	Schneider (2001)
	Netherlands	50%	Normal	De Vries <i>et al.</i> (2001)
	Finland	5%	Normal	Johansson and Janssen (1994)
	Germany	15%	Uniform	Suutari <i>et al.</i> (2001)
	UK (Alice Holt)	-30% to + 33%	Triangular	Zak (2000)
	UK (Liphook)	10%	Triangular	Abbott <i>et al.</i> (2003)
	UK (National)	23%	Normal	Arnell et al. (1990)
Kgibb	Europe	20%	Uniform	Suutari et al. (2001)
0	UK (Alice Holt)	-33% to + 50%	Triangular	Zak (2000)
	UK (site specific)	300 – 3000 dependent on soil type	Log-normal Mean = 1025 SD = 410	Abbott <i>et al.</i> (2003)
	UK (National)	20%	Uniform	Suutari et al. (2001)
Bc:Al	Sweden	50%	Uniform	Alveteg (2001)
ratio	Germany	10%	Uniform	Suutari et al. (2001)
	UK (Alice Holt)	-50% to + 67%	Triangular	Zak et al. (2000)
	UK (National)	50%	Uniform	Cronan and Griegal (1995)