Assessing the impact of international emissions reduction scenarios to combat the acidification of freshwaters in Great Britain with the First-order Acidity Balance (FAB) model and the Hull Acid Rain Model (HARM)

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Abstract
Critical loads models for acidity underpin the negotiation of international strategies for the abatement of acid deposition through emissions controls. In Great Britain and Scandinavia, critical loads for freshwater ecosystems are calculated with the First-order Acidity Balance (FAB) model, which provides a catchment based estimate of deposition reduction requirements of both sulphur and nitrogen species in order to protect a selected aquatic target organism. The FAB model is here applied to a national freshwaters database for Great Britain using three deposition scenarios generated with the Hull Acid Rain Model (HARM). Critical load exceedance and changes in two important chemical indicators (nitrate and acid neutralizing capacity) are assessed for 1990 baseline deposition levels, planned emissions reductions under existing international commitments (REF scenario), and a potential stringent deposition reduction scenario under a multi-pollutant, multi-effect strategy (EI0 scenario). Model outputs indicate that the proportion of sampled sites exceeding their critical load will be reduced by sixty-four and seventy-eight percent respectively under the two future deposition scenarios. Planned reductions in emissions under the REF scenario will protect most Scottish freshwaters, but substantial areas of the English and Welsh uplands will remain exceeded. Most of the required reductions in acid deposition for both Scotland and Wales would be met by the more stringent EI0 scenario. In the most sensitive areas of northern England, even greater reductions in both S and N emissions than those described under the EI0 scenario are required. While S remains the most important source of acid deposition even after future reductions, freshwaters in sensitive areas cannot be protected by abatement of S emissions alone. There is a clear need for a strategy to reduce N deposition if British freshwaters in sensitive areas are to be protected.

Introduction
Critical loads maps have for several years been used to inform policy makers at both national and international levels of the spatial distribution of sensitive ecosystems and the extent of damage caused by acid deposition (e.g. CLAG, 1994, Posch et al., 1997a). Critical loads modelling for various ecosystems provided a major part of the scientific input into the negotiation of the Second Sulphur Protocol under the Convention on Long Range Transboundary Air Pollution, which was signed in Oslo in 1994 (UNECE, 1994).

For freshwater ecosystems, relatively simple steady-state models were used to determine critical loads for sulphur deposition, for example the Steady-State Water Chemistry (SSWC) model (Henriksen et al., 1992) and the diatom model (Battarbee...
et al., 1996), and national maps of critical loads of sulphur to freshwaters have been published (e.g. CLAG Freshwaters, 1995, Henriksen et al., 1998).

After the signing of the Second Sulphur Protocol, which committed the signatory nations to major reductions in sulphur deposition, international attention turned towards the increasingly evident problem of nitrogen deposition. Surface waters had been showing signs of elevated nitrate concentrations which were attributed to atmospheric sources in sensitive upland areas (e.g. Brown, 1988, Aber et al., 1989, Aber. 1992, Stoddard, 1994, Allott et al., 1995a, Lepistö, 1995). There was therefore an obvious demand for a critical load model for both S and N deposition, since the previous freshwaters models could not account for the relative complexity of N cycling processes in catchments compared with sulphur.

A model developed for just such a purpose is the First-order Acidity Balance (FAB) model (Posch et al., 1997b). This model provides a “critical load function” (CLF) which quantifies the deposition reduction requirements for either N or S, given the value of the other. Deposition loads of S and N can be compared with the CLF in order to determine whether the critical load is exceeded, and if so, whether S or N deposition or some combination of both must be reduced in order to protect a water body.

Since the FAB model is process based, it can be used to determine the likely effects of different potential deposition scenarios, i.e. it is not limited to the calculation of critical load exceedance for current loads of total acid deposition (S+N) only, as were previous freshwaters models (Allott et al., 1995b, Harriman et al., 1995). It therefore provides a useful tool for assessing the long term effects of future reductions in acid deposition, such as those planned or implemented for S under the Second Sulphur Protocol. With a new protocol for nitrogen soon to be negotiated, and with further reductions in S deposition proposed under new European Union initiatives, the FAB model provides a means of testing the combined effects of future loads of S and N deposition.

Two scenarios based on planned and potential reductions in acid emissions have been used as inputs into the Hull Acid Rain Model (HARM: Metcalfe et al., 1995a), which provides estimates of S and N deposition based on the anticipated changes in emissions. Here we use the FAB model in combination with HARM modelled deposition data to assess the potential impacts of two future deposition scenarios, plus a selected baseline for comparison, on a national dataset of British freshwaters.

2. Methods: model descriptions and input data requirements

For critical loads work in Great Britain, the selected critical chemical parameter for freshwaters is acid neutralising capacity (ANC), with a critical value of 0 µeq l⁻¹ (CLAG Freshwaters, 1995) which has been shown to represent a 50% probability of damage to brown trout populations in extensive studies in Norway (Lein et al., 1992). The FAB model must therefore determine the deposition loads of S and N which will depress ANC below this value in the long term (Curtis et al., 1998a).
Two distinct modelling tasks are necessary in order to provide the required data. First is the derivation of the critical load function for each of the freshwater sites in the dataset. The second aspect is the calculation of appropriate S and N deposition data for these sites which can be related to the CLF to determine whether the critical load is exceeded, and by which species.

2.1 Model description and input data: the FAB model

The formulation of the FAB model and its application to the British freshwaters dataset is described in detail in Curtis et al. (1998a), and only a brief description is given here.

The basis of the FAB model is a charge balance incorporating inputs, catchment retention or removal processes, and leaching outputs of acid anions (Posch et al., 1997b):

\[
N_{\text{dep}} + S_{\text{dep}} = f N_{\text{upt}} + (1-r)(N_{\text{imm}} + N_{\text{den}}) + r(N_{\text{ret}} + S_{\text{ret}}) + AN_{\text{leach}}
\]

(Equation 1)

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

All units are expressed in equivalents (moles of charge) per unit area and time.

It is assumed that there are no other inputs of S or N (e.g. from agriculture) and that there are no other significant processes in operation which affect the catchment budget of acid anions. The proportion of total N deposition which is delivered as \(NH_4^+\) is assumed to be either retained within the terrestrial catchment or nitrified.

Various sources are used to provide the necessary input data; their derivation is described fully by Curtis et al. (1998a, 1998b), and summarised below. Deposition inputs are discussed separately in Section 2.2.

Water chemistry was based on the analysis of one-off dip samples, according to the methodology described in Harriman et al. (1990), from a national network of 1470 freshwater sites across Great Britain (excluding the Isle of Man, for which soils data were unavailable). The lake and stream sites were selected to represent the most sensitive water body on a 10km square grid basis (20km in non-sensitive lowland areas), with lakes selected in preference to streams where possible (Kreiser et al.,...
1993). For each of the study sites, a catchment outline was defined on 1:25,000 scale topographical maps and digitized using a Geographical Information System. Lake to catchment ratio \( r \) was obtained from the digital catchment estimates of lake and catchment area. Surface water runoff was calculated for each catchment by the overlay of digital catchment boundaries onto a 1km grid resolution dataset of annual mean runoff for the period 1992-94. Leaching fluxes of major ions from each catchment were calculated from water chemistry and catchment weighted runoff estimates.

Net \( N_{\text{upt}} \) was based on a single uptake rate of 0.279 keq ha\(^{-1}\) yr\(^{-1}\) (or c. 4 kgN ha\(^{-1}\) yr\(^{-1}\)) for coniferous forest only (Emmett and Reynolds, 1996, Curtis et al., 1998a). The proportion of coniferous forest cover \( f \) was derived from the ITE Land Cover Map of Great Britain (Fuller et al., 1994) by overlaying digital catchment boundaries.

Catchment weighted soils data were obtained by the overlay of digital catchment boundaries onto digital soils maps at a scale of 1:250,000 to obtain the proportion of each major soil type for every catchment (Curtis et al., 1998a). Rates of \( N \) immobilisation and denitrification were estimated for each catchment using literature based default values for each soil type (Hall et al., 1997).

The in-lake retention of \( S \) and \( N \) was calculated with a kinetic equation (described fully in Curtis et al., 1998) using literature based mass-transfer coefficients \( (S_s = 0.5 \text{ m yr}^{-1} \) for sulphur, \( S_N = 5 \text{ m yr}^{-1} \) for nitrogen), estimated runoff, and lake to catchment ratio (Kelly et al., 1987, Dillon and Molot, 1990). Note that for stream catchments, the lake to catchment ratio is zero and there is no in-lake retention of \( S \) or \( N \).

With all the components of the mass balance of \( S \) and \( N \) now in place, it is possible to define the critical load function by calculating a critical value of acid anion leaching \( (\text{AN}_{\text{leach}}) \). The SSWC model is employed to determine the pre-industrial base cation leaching rate, which is deemed to represent the long-term, sustainable supply of base cations from weathering and excludes any shorter term provision of base cations from the soil exchange complex (Henriksen et al., 1992). Given the sustainable supply of base cations and a selected critical value of ANC, the critical leaching flux of acid anions \( (L_{\text{crit}}) \) can be calculated:

\[
L_{\text{crit}} = Q ([BC]_0^{\text{pre}} - [\text{ANC}_{\text{crit}}]) \tag{Equation 2}
\]

where \([BC]_0^{\text{pre}}\) is the pre-industrial concentration of non-marine base cations from the SSWC model, \( Q \) is runoff, and \([\text{ANC}_{\text{crit}}]\) is selected to be 0 \( \mu \text{eq l}^{-1} \).

Substitution of Equation 2 into the charge balance of the FAB model (Equation 1) provides those combinations of \( S \) and \( N \) deposition for which \( \text{AN}_{\text{leach}} = L_{\text{crit}} \). If \( \text{AN}_{\text{leach}} \) is greater than \( L_{\text{crit}} \) for a given combination of \( S \) and \( N \) deposition, the critical load is exceeded. There is clearly no unique combination of \( S \) and \( N \) deposition which defines the critical load of acidity, because a reduction in one species would permit an increase in deposition of the other to achieve the same net flux of acid anions (and therefore acidity). The critical load function defines the range of paired values of \( S \)
and N deposition which equate to the critical load of acidity (Bull, 1995, Posch et al., 1997b, Curtis et al., 1998a).

There are three key points on the CLF which signify individual thresholds for S or N deposition. The maximum allowable deposition of sulphur, called CL\(_{\text{max}(S)}\), at which critical load is not exceeded, can only occur when N deposition is zero. Similarly, the maximum allowable deposition of total N, CL\(_{\text{max}(N)}\), is only possible when S deposition is zero if the site is to be protected. There is an additional threshold for N deposition, CL\(_{\text{min}(N)}\), which indicates the minimum level of N deposition which leads to nitrate leaching and can therefore contribute to critical load exceedance. The derivation of these terms is explained fully by Posch et al. (1997b) and Curtis et al. (1998a).

For any combination of S and N deposition, the FAB model will predict the potential leaching of sulphate and nitrate, and hence of ANC, thereby determining whether the critical load is exceeded. If S deposition exceeds CL\(_{\text{max}(S)}\) then it must be reduced in order to protect the site, regardless of N deposition. Similarly, N deposition must be reduced to less than CL\(_{\text{max}(N)}\) regardless of S deposition. For situations where neither threshold is exceeded but the critical load is exceeded, there is a choice between reducing S deposition or N deposition, or some combination of both, in order to prevent critical load exceedance.

2.2 HARM modelled deposition scenarios

The national S and N deposition data used here were generated at the 10 km x 10 km grid scale by the Hull Acid Rain Model (HARM: Metcalfe et al., 1995a), a receptor-oriented, Lagrangian statistical model which can operate at the regional scale. HARM is driven by emissions of SO\(_2\), NO\(_x\), and NH\(_3\) across Europe at a spatial scale of 50 km (Barrett and Berge, 1996) and emissions of SO\(_2\), NO\(_x\), NH\(_3\) and HCl across the UK at a spatial scale of 10 km (Salway et al., 1997). Emissions may be varied on a country by country basis across Europe to reflect planned or proposed emission reduction requirements and on a source or sector specific basis across the UK due to the more detailed nature of the UK inventories (Metcalfe et al., 1995b). HARM has undergone considerable validation. HARM performs particularly well for S and oxidised N, but when compared with a reduced N dry deposition field derived from concentrations modelled using FRAME (RGAR, 1997) underpredicts substantially. As a result, the HARM modelled current deposition underestimates the relative importance of total N deposition compared with the RGAR estimates. Overall, however, HARM compares favourably with the outputs of other deposition models when tested against measured and interpolated data in the UK (RGAR, 1997). Modelled deposition data may therefore be used in conjunction with the FAB model to assess the extent and magnitude of exceedance across Great Britain under a range of possible future scenarios.

Two emission reduction scenarios, relating to planned and potential measures for emissions reductions within Europe, are considered here and their effects on national critical load maps for freshwaters are compared. These scenarios are based on those
described in a recent report to the European Commission and UNECE (Amann et al., 1998) in which current legislation, nationally planned emission reductions and a range of acidification, eutrophication and ozone scenarios were all taken into account.

The "Reference" Scenario for the year 2010

The officially adopted or internationally announced ceilings on national emissions resulting from the Second Sulphur Protocol of the UNECE form the basis of the "Current Reduction Plan" (CRP) Scenario. In addition, the Current Legislation (CLE) Scenario considers the impacts of adopted national or international legislation for emission control (as opposed to national emission ceilings), against a background of projected future energy consumption. These two emissions scenarios have been combined to assess the most likely outcome in terms of emissions reductions; the most stringent of the two for each individual country is used in the Reference (REF) scenario (Amann et al., 1998). The REF scenario was constructed for the purpose of assessing the likely environmental impacts of the current emission strategies through its application in critical loads modelling exercises.

Under the REF scenario SO₂, NOₓ and NH₃ emissions across Europe are reduced by 61%, 40% and 14%, relative to 1990 levels, by the year 2010. Under this scenario EU countries are expected to achieve greater reductions than non-EU countries. In HARM these reductions are achieved through scaling current (1994) emissions on a cell by cell basis for each country within the UNECE European Monitoring and Evaluation Programme (EMEP) grid domain. Emissions from natural oceanic sources and international shipping are not abated in this scenario.

UK emission targets under the REF Scenario are listed in Table 1. These targets were represented in HARM as a series of spatially disaggregated emission grids with each emission grid relating to a specific source category. Emission estimates for each source category were provided by the National Atmospheric Emissions Inventory (Culham, Oxfordshire) after consultation with government departments (the Department of Trade and Industry, the Department of Transport) and the Environment Agency, and are largely based on energy projections listed in Energy Paper 65 (DTI, 1995). Spatially disaggregated totals for SO₂ and NOₓ emissions are listed in Table 2.

The "E10" Scenario

The stated ultimate target in the Fifth Environmental Action Programme of the European Union is to achieve no exceedance of critical loads throughout Europe. However, this is deemed unattainable in the short term, given economic and technological constraints, hence an interim target is under consideration. Integrated assessment modelling on a European scale with the Regional Air Pollution Information System (RAINS) model has been directed towards identifying the most cost-effective gap closure strategy (Amann et al., 1998). "Gap closure" implies a closing of the gap between current levels of ecosystem protection and ultimate target levels (Bull. 1995).
Many different methods of attaining gap closure targets have been assessed through integrated assessment modelling, resulting in the definition of several scenarios at the European level, with names like “b1” or “b5” (Amann et al., 1996). These scenarios are essentially “moving targets” because even at the national level, there are many different factors to be accounted for, such as national controls on the sulphur content of fuels or legislation to control emissions from shipping; there is as yet no single definitive emissions scenario.

Table 1: UK emission ceilings used in HARM deposition scenarios (kilo-tonnes)

<table>
<thead>
<tr>
<th>Species</th>
<th>1990 Base</th>
<th>REF Scenario</th>
<th>E10 Scenario</th>
</tr>
</thead>
<tbody>
<tr>
<td>SO₂</td>
<td>3754</td>
<td>980</td>
<td>385</td>
</tr>
<tr>
<td>NOₓ</td>
<td>2800</td>
<td>1161</td>
<td>1123</td>
</tr>
<tr>
<td>NH₃</td>
<td>329</td>
<td>298</td>
<td>277</td>
</tr>
</tbody>
</table>

Table 2: UK emissions of SO₂ and NOₓ under the REF Scenario (kilo-tonnes)

<table>
<thead>
<tr>
<th>Source Type</th>
<th>SO₂</th>
<th>NOₓ</th>
</tr>
</thead>
<tbody>
<tr>
<td>Point Sources (Power Stations, Refineries, Industry)</td>
<td>551.5</td>
<td>451.6</td>
</tr>
<tr>
<td>Buses</td>
<td>1.6</td>
<td>37.2</td>
</tr>
<tr>
<td>Diesel Cars</td>
<td>5.1</td>
<td>39.6</td>
</tr>
<tr>
<td>Petrol Cars</td>
<td>22.2</td>
<td>125.0</td>
</tr>
<tr>
<td>Heavy Goods Vehicles</td>
<td>11.1</td>
<td>163.5</td>
</tr>
<tr>
<td>Light Goods Vehicles – Diesel</td>
<td>2.8</td>
<td>14.9</td>
</tr>
<tr>
<td>Light Goods Vehicles – Petrol</td>
<td>0.9</td>
<td>1.1</td>
</tr>
<tr>
<td>Motorcycles</td>
<td>0.1</td>
<td>0.9</td>
</tr>
<tr>
<td>Railways</td>
<td>1.7</td>
<td>19.8</td>
</tr>
<tr>
<td>Other Stationary Sources</td>
<td>340.7</td>
<td>196.3</td>
</tr>
<tr>
<td>Ships in Port</td>
<td>42.3</td>
<td>111.1</td>
</tr>
<tr>
<td><strong>Total</strong></td>
<td><strong>980.0</strong></td>
<td><strong>1161.0</strong></td>
</tr>
</tbody>
</table>

Despite the minor changes and adjustments which are frequently made to these potential scenarios, it is still instructive to select a particular example in order to assess the utility of the FAB model and the general spatial pattern of impacts in terms of critical loads exceedance at the national level. Here we adopt the “E10” scenario (Amann et al., 1998). E10 represents a joint optimisation to address problems
resulting from acidification, eutrophication and ground level ozone. For acidification the scenario aims to achieve 90% gap closure of accumulated excess acidity. Accumulated excess acidity is a function of critical load exceedance, expressed as a flux of acidity to a site, and the area of the site, summed for each site within a 150 × 150 km EMEP grid square. All ecosystems are included in its derivation, not just freshwaters. This joint optimisation approach increases NO\textsubscript{x} controls, but relaxes the requirement to reduce SO\textsubscript{2} emissions compared with a scenario to address acidification alone (Amann \textit{et al.}, 1998).

Under the E10 scenario SO\textsubscript{2}, NO\textsubscript{x} and NH\textsubscript{3} emissions across Europe are reduced by 73%, 53% and 26% relative to the 1990 base year making this scenario much more stringent than the REF scenario. These reductions were achieved through scaling current (1994) national EMEP emissions to meet the required targets. Emission targets for the UK are listed in Table 1. Under this scenario, SO\textsubscript{2} emissions are reduced by 87% relative to 1990 levels. NO\textsubscript{x} and NH\textsubscript{3} reductions are less severe in comparison (60% and 16% respectively).

Creating a reasonable distribution of UK emissions under this scenario is not a trivial task given the severity of the SO\textsubscript{2} emission reductions. We therefore assumed that all coal-fired power stations would be closed apart from Drax, Ratcliffe-on-Soar and Longannet, which would be operating with FGD or equivalent. Remaining point and area emission sources used in the REF scenario were then scaled down to meet the specified national targets for SO\textsubscript{2} and NO\textsubscript{x} under the E10 scenario. NH\textsubscript{3} emissions were also scaled on a flat rate basis.

**Base Year (1990) Deposition**

Deposition associated with the two future emissions reduction scenarios (for 2010) is compared with HARM modelled estimates of deposition for the base year 1990. Base year deposition was determined from a model run in which current (1994) European (EMEP) and UK emissions were scaled to the 1990 totals listed in the Fifth Interim Report (Amann \textit{et al.}, 1998). For comparability, all three HARM deposition data sets were derived using 1961-1990 rainfall field in the calculation of wet deposition (Barrow \textit{et al.}, 1993). It is therefore assumed that there are no changes in total deposition levels due to variations in mean rainfall for the assessment years of 1990 and 2010.

**3 Modelling results and inter-comparison of HARM deposition scenarios**

The most significant outputs of the FAB model are predictions of nitrate, ANC and deposition reduction requirements for S and N. Changes in sulphate leaching under a fixed deposition load are expected to be less significant because of the relatively minor role of S in biological cycling within catchments, compared with N.

The FAB model outputs under the three HARM deposition scenarios are described below with respect to the most impacted areas as indicated by published critical load
exceedance maps using the SSWC and diatom models (CLAG Freshwaters, 1995). The approximate locations of these regions are indicated in Figure 1.

3.1 Predicted differences in nitrate leaching

The discrepancy between measured rates of nitrate leaching and those predicted by the FAB model have been discussed in previous studies (Curtis et al., 1998a, 1998b), but in general relate to the longer term, sustainable basis of critical loads definitions compared with transient catchment processes currently in operation. Measured nitrate levels are shown for comparison in Figure 2a.

The FAB model predicts that nitrate leaching would greatly increase from measured values (Fig. 2a) in sensitive upland areas across almost the whole country (except for north west Scotland) under a constant deposition load (HARM 1990 data - Figure 2b). The upland waterbodies of England and Wales are nearly all predicted to have nitrate concentrations of at least 20-50 µeq l⁻¹ upon reaching steady-state with 1990 deposition levels. Even in central areas of northern Scotland, where low deposition levels coincide with high denitrification and immobilisation rates (due to the predominance of waterlogged soils), there are still small predicted increases in nitrate leaching. It should be noted, however, that UK NOₓ emissions fell between 1990 and 1995 from 2897 to 2295 k tonnes.

In contrast, the nitrate concentrations predicted by FAB in some lowland areas (e.g. Eastern Scotland) appear to decrease in the future. This is an artefact of the model assumptions, whereby only atmospheric N sources are included as net inputs to catchments: in those areas where measured nitrate levels indicate agricultural runoff, the predicted concentrations may be lower, depending on the relative contributions of agriculture and total N deposition at each site. In lowland England, measured and predicted nitrate leaching are both very high, but any apparent links between the two are probably spurious. Measured nitrates in south east England are often high because of the predominance of intensive agriculture (and therefore fertilizer inputs) and urban areas in this region (INDITE, 1994). These sources of N are not included in the FAB model formulation, and the high nitrate concentrations predicted by FAB are due to relatively high deposition inputs in regional terms. Atmospheric inputs are, however, likely to be small relative to agricultural and urban inputs (INDITE, 1994). This shortfall of the model rarely presents a problem in practice because those water bodies subjected to unquantified agricultural inputs of nitrogen are almost exclusively insensitive to acidification. In such cases, it is likely that base cation rich soils were a prerequisite for agriculture and so the surface waters will have a naturally high ANC (Allott et al., 1995).

Comparison of measured nitrate (Fig. 2a) and predicted nitrate under 1990 deposition levels (Fig. 2b) which remained fairly constant over the freshwaters sampling period (RGAR, 1997) shows that nitrate leaching had not reached predicted levels by the time of sampling. This observation is consistent with the FAB model’s basis in a long term steady-state situation, which had evidently not been reached at the time of
sampling. Other studies have indicated that predicted nitrate levels using the FAB model are far greater than those currently observed, reflecting transient, elevated rates of N retention in catchments (Curtis et al., 1998b).

Figure 1: Selected regions of acid sensitive freshwaters in Great Britain
(from Curtis et al., 1998a)

Under the HARM REF scenario, the reduction in N deposition associated with greater use of catalytic converters on motor vehicles leads to much lower predictions of nitrate in all freshwaters than under 1990 deposition levels (Fig. 2c). The change relative to measured values is more complex (Fig. 2a). In Scotland, nitrate concentrations are predicted to decline from measured values across most of the country apart from areas centred on the Trossachs and the English border (Fig. 1),
where small increases are found. In Wales, the northern Pennines and the North Yorkshire Moors, increases are predicted, while other sensitive areas experience little change or a slight decrease. The predictions of lower nitrates across lowland England are spurious for the reasons discussed earlier, i.e. because agricultural inputs dominate the N budget.

If the ultimate deposition reductions associated with the HARM E10 scenario were achieved, then the FAB model indicates a slight reduction in nitrate leaching relative to the REF scenario (Fig. 2d), with similar regional changes from measured chemistry. The main difference from the REF scenario is a reduction in nitrate leaching from the 20-50 µeq l⁻¹ range to the 10-20 µeq l⁻¹ range in the northern Pennines and Lake District, while nitrate from atmospheric sources would virtually disappear from freshwaters in a greater proportion of Scotland. The deposition of N would be so low that the catchment sinks for nitrogen would effectively intercept all N, thereby preventing any from being leached in runoff. Current nitrogen retention rates are very large but significant leaching indicates a degree of nitrogen saturation (Stoddard, 1994), so the prediction of zero nitrate leaching under the E10 scenario indicates a return to N limitation in some areas, which may have biological consequences in catchments where atmospheric inputs of nitrogen may for years have acted as an important nutrient source.

3.2 Predicted differences in ANC

The predicted changes in acid anion leaching would result in an equivalent decrease in ANC, and where predicted ANC is less than 0 µeq l⁻¹ the critical load is exceeded. Patterns of measured (Fig. 3a) and predicted ANC under contemporary deposition loads are discussed by Curtis et al. (1998a).

If HARM 1990 deposition loads remained constant, there would be a decrease in ANC at many sites (Figure 3b), most significantly in sensitive upland areas. In northern Scotland and upland Wales there is a greater distribution of sites with negative ANC values compared with current measured ANC, while the Pennine region is affected mainly by a decrease in ANC at sites which have already acidified to below zero ANC.

The reductions in deposition associated with the HARM REF scenario are sufficient to lead to major reductions in acid anion leaching which elevate ANC to positive levels in all but the most acidified areas (Figure 3c). Under the REF scenario, the only strongly acidified regions are the Pennines, Lake District and North Yorkshire Moors, with a few other negative ANC values at scattered locations in other sensitive parts of the country but few in Scotland.

Further reductions in deposition are associated with the HARM E10 scenario and have an even greater effect on predicted surface water chemistry (Figure 3d). Very few negative ANC values are predicted for Scotland and Wales, but clusters of acidified sites still persist in the ultra-sensitive Lake District, North Yorkshire Moors and especially Pennine regions.
3.3 Further deposition reduction requirements indicated by the FAB model

One of the major benefits of the FAB model is that it can be used to determine which acid species (S or N) needs to be reduced in order to protect impacted sites. Using a colour coding system based on the requirement to reduce S or N deposition, it is possible to map the options for deposition reduction requirements (Figure 4). This approach has previously been described by Curtis et al. (1998a).

The HARM 1990 deposition data indicate that the majority of sites exceeding their critical loads require a mandatory reduction in S deposition in order to attain non-exceedance, i.e. the critical loads are currently exceeded by sulphur alone (Fig. 4a). In much of the far north and north-west of Scotland, N deposition is irrelevant in terms of acidity critical loads; only sulphur reductions are required, because no nitrate leaching is predicted (or observed in the measured data – see Figs. 2a-2b). At the majority of sites outside this region of north-west Scotland, initial reductions in S deposition could be followed by further reductions in either S or N deposition in order to protect the sites, i.e. some nitrate leaching is predicted and so critical load exceedance could be reduced with N deposition reductions. Significantly, the most impacted areas of the Pennines, Lake District and North Yorkshire Moors, plus a few sites in south-east England, require mandatory reductions in both S and N deposition, i.e. either sulphur or nitrogen deposition alone is sufficient to cause critical load exceedance. This indicates that measures to reduce sulphur alone, as in the Second Sulphur Protocol, will be insufficient to protect the most sensitive freshwaters in these areas unless N emissions are also reduced.

The FAB model indicates that by 2010, under existing commitments to reduce S deposition and with associated decreases in N deposition under the REF scenario, large areas of Britain will be protected from acidification, including most of Scotland and Wales (Figure 4b). A few sites in upland Wales, Dartmoor and south-east England are still exceeded, but in general a further reduction in either S or N deposition would suffice to protect them. Since the reductions in N deposition under the HARM REF scenario are small relative to the reductions in S, the regions most impacted under the HARM 1990 deposition levels are still experiencing widespread exceedance of critical loads under the HARM REF scenario (Fig. 4a-4b). However, it is not just N deposition which requires further reduction to protect these sites; further reductions in S deposition are also mandatory across much of the Pennines.

The level of protection afforded under the HARM E10 scenario is more dramatic, although the benefits relative to the HARM REF scenario are small compared with the transition from HARM 1990 to HARM REF deposition levels (Figure 4). Only a very small number of isolated sites are still exceeded in Scotland or Wales, and only one Scottish site, in Galloway, is still affected by N deposition. However, even under the very stringent reductions of the E10 scenario, many of the exceeded sites in England still remain unprotected. The Pennines, central Lake District and North Yorkshire Moors regions would still be highly impacted, and further reductions in both S and N are mandatory across much of the Pennines and at the few remaining exceeded sites in south east England.
Figure 2: Measured and modelled nitrate concentrations in the British freshwaters mapping dataset

2a: Current measured nitrate

2b: Predicted nitrate concentrations under HARM 1990 deposition load

2c: Predicted nitrate concentrations under HARM RHF scenario deposition load

2d: Predicted nitrate concentrations under HARM e10 scenario deposition load
Figure 3: Measured and modelled ANC concentrations in the British freshwaters mapping dataset

3a: Current measured ANC

3b: Predicted ANC under HARM 1990 deposition load

3c: Predicted ANC under HARM REF scenario deposition load

3d: Predicted ANC under HARM e10 scenario deposition load
Figure 4: Deposition (emissions) reduction requirements according to the FAB model under HARM deposition scenarios

4a: Further reductions required under HARM 1990 deposition loads
4b: Further reductions required after implementation of HARM REF scenario emissions reductions
4c: Further reductions required after implementation of HARM e10 scenario emissions reductions
Intercomparison of the number of freshwater sites falling into each of the mapped deposition reduction categories for the various deposition scenarios provides a measure of the effectiveness of the emissions reductions strategies adopted (Table 3).

Of the sampled population of 1470 lake and stream sites, 295 sites (20%) exceed their critical loads under the HARM 1990 deposition load. Of these, more than 80 percent require mandatory reductions in S deposition while a quarter would be exceeded by S or N alone.

The deposition reductions associated with the HARM REF scenario will protect nearly two thirds (64%) of the sites which were exceeded in 1990, leaving only 106 sites (7% of the sampling population) unprotected. Around one third of the exceeded sites require further mandatory reductions in N deposition in order to be protected, while two thirds require further mandatory reductions in S deposition.

Additional deposition reductions under the HARM E10 scenario would increase the number of protected sites to nearly four fifths (78%) of those exceeded at 1990 levels, with only 4.4% of the total sampling population remaining unprotected. While the implementation of the HARM REF scenario emissions reductions would leave only a third of sites unprotected relative to 1990, the HARM E10 scenario would leave more than sixty percent of sites unprotected relative to the HARM REF scenario. In terms of the number of sample sites protected, the additional benefits provided by the more stringent E10 scenario are relatively small, compared with the initial gains of implementing the REF scenario.

<table>
<thead>
<tr>
<th>Acid species required reduction</th>
<th>Number of sites in scenario</th>
<th>1990</th>
<th>REF</th>
<th>E10</th>
</tr>
</thead>
<tbody>
<tr>
<td>None (not exceeded)</td>
<td>1175</td>
<td>1364</td>
<td>1405</td>
<td></td>
</tr>
<tr>
<td>S only</td>
<td>37</td>
<td>14</td>
<td>10</td>
<td></td>
</tr>
<tr>
<td>S, then S or N</td>
<td>129</td>
<td>24</td>
<td>7</td>
<td></td>
</tr>
<tr>
<td>Both S and N</td>
<td>75</td>
<td>32</td>
<td>27</td>
<td></td>
</tr>
<tr>
<td>Either S or N</td>
<td>54</td>
<td>36</td>
<td>20</td>
<td></td>
</tr>
<tr>
<td>N, then S or N</td>
<td>0</td>
<td>0</td>
<td>1</td>
<td></td>
</tr>
<tr>
<td>Total exceeded</td>
<td>295</td>
<td>106</td>
<td>65</td>
<td></td>
</tr>
</tbody>
</table>

4 Discussion

The predictions of the modelling exercise have to be considered in the light of the limitations of the critical load models, the HARM deposition model, and the data used as inputs to HARM to describe future emissions reductions.
4.1 Uncertainties associated with critical load models

There are several sources of uncertainty within the critical load models. For example, a single chemical sample, used to calculate the sustainable leaching rate of base cations which can buffer acid inputs in the long-term, is assumed to represent mean annual chemistry despite the short-term chemical variability of freshwaters (Curtis et al., 1998a).

In a national model application, general assumptions have to be made regarding the rates at which various N retention processes operate in different soil and vegetation types. A single value for N uptake in forest is used, despite known regional variations in tree species, productivity, harvesting practice etc. which will significantly affect the N budget over a harvesting cycle. Denitrification is calculated from catchment soil cover estimates, with low, medium and high rates allocated to soils on the basis of their general drainage characteristics, but there is likely to be very significant spatial and temporal variation at much smaller scales than can be accounted for in such a large number of catchments. Very few data are available for denitrification rates in British moorland soils so the uncertainty in these figures is not known. Perhaps the greatest single source of uncertainty lies in the determination of sustainable N immobilisation rates in soils, since the values used are estimated from long-term N accumulation rates in the soil profile over the period of soil formation, which extends back several millennia before the relatively recent phenomenon of acid deposition and elevated N loads from the atmosphere (Curtis et al., 1998a).

Despite the various uncertainties in the definition of the critical load function and associated with the catchment data inputs, an important consideration is that the national freshwaters critical loads dataset provides a consistent method for evaluating the potential effects of different emissions abatement (and therefore deposition reduction) scenarios. While there is uncertainty associated with the critical loads calculated for individual sites, on a regional and national basis the critical loads data provide a uniform measure of sensitivity to acidification for comparison with any given load of atmospheric deposition of acidity. However, if the scenarios are to be compared directly, it is necessary to consider the uncertainties associated with the deposition data in some detail.

4.2 Uncertainties associated with HARM deposition data

One problem with HARM deposition data is that the model does not perform well in predicting dry deposition of reduced nitrogen, a shortfall common to most deposition models in the UK (RGAR, 1997). Although HARM performs well relative to other models in reproducing wet ammonium deposition, it nevertheless tends to underestimate. More significant is the tendency of HARM to underestimate dry reduced N deposition, with a much greater difference from measured values relative to wet deposition. Since the FAB model assumes that reduced N which is not retained within the terrestrial catchment will be nitrified to nitrate, there will be an associated underprediction of nitrate leaching and therefore of critical load exceedance.
In a previous application of the FAB model to the British freshwaters dataset using interpolated deposition data from measurements at a national monitoring network of sites, the relative importance of N deposition was found to be much greater than with the most comparable dataset used here, the HARM 1990 data (Curtis et al., 1998a). This difference is largely due to the apparent underestimate of reduced N deposition from the HARM model. Any reliable quantification of these underestimates, however, requires a high quality map of reduced N dry deposition based on measurement data. Such a map is not available at present, but should be possible in the near future using data from the DETR’s new rural ammonia monitoring network. The effects of model underprediction of N inputs are likely to be greatest in Wales and the south west and least in Scotland north of the central valley.

With respect to the known underestimation of N deposition with the HARM model, the predictions of increased nitrate leaching, ANC decline and critical load exceedance must be considered as “best case” scenarios. The regional picture of deposition reduction requirements for nitrogen could look much worse if N deposition were increased. For example, Curtis et al. (1998a) found that on the basis of deposition estimates for N interpolated from measurements at a national network of sites (RGAR, 1997) the role of N in causing acidification relative to S deposition was much more significant.

4.3 Uncertainties in emissions estimates

In addition to the uncertainties associated with model structures described above, there are also uncertainties in the emissions data used to drive the HARM deposition model. There are uncertainties with current emissions inventories because the sources of each pollutant cannot all be known, and emissions estimates are often based on assumptions that cannot be quantified (RGAR, 1997).

Emissions of SO₂ and HCl are predominantly governed by fuel content and consumption. Fuel consumption for large combustion sources is well quantified on an annual basis, but the S content of fuels is very variable and smaller combustion sources are more difficult to quantify. The overall uncertainty in emission estimates of SO₂ has been stated as 5-10% (RGAR, 1997).

The uncertainty associated with emissions of N oxides is less well known, because of the importance of diffuse sources (e.g. road vehicles), but has been estimated with Monte-Carlo methods to lie in the range 20-30% (RGAR, 1997). For reduced nitrogen the uncertainties are much greater (INDITE, 1994).

Since future deposition scenarios are based on expected reductions in emissions which have not yet been realised, it is impossible to estimate the uncertainty in predicted emissions. The uncertainty associated with current emissions inventories are described above, but for future scenarios there is the additional question of whether targets will ever be met. However, the purpose of this modelling exercise it to ascertain the potential effects of possible emissions and deposition reductions, and it is beyond the scope of this paper to discuss the likelihood of their implementation.
The REF scenario represents a “best estimate” of current commitments to reduce emissions in Britain and Europe, and may be regarded as the most likely outcome in terms of deposition for the target year of 2010. The inputs to the E10 scenario are less certain, but it is considered here as a typical example of the major reduction scenarios which are currently under discussion at the international level within Europe. In this sense, the E10 scenario indicates the order of magnitude of possible outcomes under the forthcoming Second Nitrogen Protocol of the UNECE.

5 Conclusions

The FAB model runs using the two future HARM deposition scenarios indicate that major reductions in critical load exceedance relative to a 1990 baseline would result from both of them by 2010. While more than 20% of sampled sites were exceeding their critical loads under HARM 1990 deposition levels, only 7% are left unprotected under the REF scenario, and this proportion is reduced to 4.4% under the more stringent E10 scenario. Since these sites were selected to represent the most sensitive freshwaters (Kreiser et al., 1993) in Britain, these figures should be over-estimates of the proportion of the total population of British freshwaters for which critical loads are exceeded.

Although there may be uncertainties in the exact numbers, it is evident that the potential magnitude of the deposition reductions considered would be sufficient to afford long-term protection to a great majority of the sampled freshwater sites in Britain. It is also apparent that the additional benefits provided under the E10 scenario compared with current commitments to emissions reductions (REF scenario) will be relatively small.

The HARM deposition data indicate that while N may be a significant contributor to acidification in freshwaters, S is still the main cause for concern, even after the full implementation of the Second Sulphur Protocol. At the most impacted sites, both S and N deposition will have to be reduced, even after the drastic decreases associated with the E10 scenario. Not only are the planned reductions in S deposition insufficient to protect all freshwater sites in Britain, but regional problems of acidification will persist unless N deposition is also reduced in sensitive areas. Given the known underestimation of ammonium deposition with the HARM model, it is likely that the maps presented here are “best case” outcomes.

The utility of the FAB model for the assessment of the likely impacts of different deposition scenarios has been demonstrated, and the potential contribution of such modelling work to the negotiations of a future nitrogen protocol is clear. Model outputs can be fed into integrated assessment models at the European scale in order to perform cost-benefit type analyses on the various possible abatement measures and strategies. At the regional scale, the FAB model provides a coherent picture of future chemical change and damage assessed as critical load exceedance.
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References


