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Quantifying the scale dependence in estimates of wet and dry deposition and the implications for critical load exceedances

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Abstract

Two sources of uncertainty in the deposition estimates used to calculate critical load exceedances are investigated. An analysis of the uncertainties in the deposition models for the UK suggests predictions of total sulphur deposition on a 20 km scale are within ±40% to ±80% depending on the region, with greater uncertainty over the higher rainfall regions, but catchment studies indicate that predictions within ±30% can be achieved. Critical loads for soils are calculated at a 1 km scale and a simulation study shows that deposition estimates calculated either at a 20 km (UK) or a 150 km (European) scale both underestimate critical load exceedances in complex terrain using the current mapping and modelling procedures. Models for deposition at a 1 km scale are not available for complex terrain but an alternative approach is proposed which gives a probability distribution of critical load exceedances within an area.

INTRODUCTION

The procedures developed recently within Europe to regulate emissions of acidifying gases are based on an assessment of the deposition of the various chemical species and their effects on vegetation, soil and freshwaters. In the first instance, emissions of sulphur dioxide (SO_2) will be reduced within the Sulphur Protocol agreed within the United Nations Economic Commission for Europe (UNECE, 1994). In developing such protocols the distributions of sources and sensitive receptors throughout Europe are of major importance so that emission reductions can be targeted for maximum benefit.

A critical loads approach has been adopted to identify the geographical distribution of sensitive receptors and to assess the magnitude of current effects. In conjunction with long-range transport models, the critical loads approach can also be used to assess the effect of different strategies to control the sources of acidifying gases.

The critical load exceedance, or the amount by which the estimated deposition exceeds the estimated critical load for an area, is calculated assuming no uncertainty in either estimate. The aim of this paper is to explore uncertainties in the method associated with the current deposition estimates. Of particular interest is the problem of scale, since critical loads are often derived at different spatial scales from those used in deposition modelling, and these approaches may lead to systematic bias in estimating the magnitude of exceedance and its spatial distribution.

CRITICAL LOADS

A critical load is defined by UNECE as 'a quantitative estimate of exposure to one or more pollutants below which significant harmful effects on sensitive elements of the environment do not occur according to present knowledge' (CLAG, 1994). The aim of emission reduction policy is to minimise the area of Europe which has a critical load exceedance. Many of the areas with large critical load exceedances are remote from sources and are often remote from any gas or rainfall monitoring stations, so concentration and deposition fields must be modelled.

Maps for critical load exceedances for soils in Britain have been produced with a resolution of 1 km x 1 km. These show the squares where the estimated deposition input, derived from 20 km x 20 km square values (assumed to be constant for all the 400 1 km x 1 km squares within the 20 km x 20 km square), exceeds the calculated critical load value for the 1 km x 1 km square (CLAG, 1994). On a European scale, deposition values are taken from the output of the EMEP models which operate at a 150 km x 150 km scale (Sandnes, 1993). For policy purposes, similar maps are also produced with deposition estimated from various emission reduction scenarios using long-range transport models.

The critical load considered in this paper is for acidification of soils with no modification for land use or base cation deposition.

DEPOSITION MODELS

The deposition models considered in this paper are those used to provide the deposition of non-marine sulphur to 20 km squares in the UK. Total deposition is calculated as the sum of deposition through 3 deposition pathways: wet, cloud droplet and dry. The importance of the 3 deposition pathways varies considerably across the UK, depending on region and landscape type, and the accuracy of the predicted total deposition will depend both on the accuracy of the inputs and on the particular mix of pathways at a specified location.

Wet deposition is modelled as a product of rainfall and SO_4^{2-} concentration in rain. When the landscape has the rapid altitude variations which are typical of many areas of the UK, both the rainfall amount and rain ion concentrations, enhanced by the presence of polluted orographic cloud at higher elevations, must be adjusted for altitude effects. These effects are included in the model in a simplified form developed for average topography rather than for detailed topographic and meteorological inputs (Dore *et al*, 1992).

Both cloud droplet deposition and dry deposition are modelled using the standard resistance analogy 'big-leaf' model (Hicks *et al,* 1987). Cloud droplet deposition is modelled to high elevation land uses, assumed to be either moorland or forest, at deposition rates close to those for momentum (Fowler *et al,* 1993). The surface resistances are set to zero and only an aerodynamic resistance, r_a, is used to calculate the cloud droplet deposition velocity. For dry deposition the 3 resistances, aerodynamic resistance, r_a, a quasi-laminar boundary layer resistance, r_b, and a canopy resistance, r_c are all required for calculating the gas deposition velocity (Cape *et al,* 1991). Deposition is a product of deposition velocity and concentration.

Assuming all the above models are correct in their formulation, the effect of the accuracy of the input variables on the predicted total deposition at the 20 km scale will now be explored.

Model inputs

The wet deposition model is dependent on three inputs. The rainfall amount and rain ion concentrations are taken from interpolated maps derived from a network of monitoring sites (RGAR, 1990). There are approximately 4000 rainfall collectors and there were almost 60 sites recording concentrations in rain of the major ions, although this has now been reduced to about 30 sites. Estimates of rainfall and rain ion concentration for each 20 km square are derived from a kriging interpolation (Webster *et al,* 1991). The orographic enhancement factor was calculated from 30 year average rainfall records and is assumed to be constant over time.

For cloud droplet deposition the aerodynamic resistance, r_a, is derived from a land use data base (Bunce *et al,* 1983) which is used to determine the proportion of each square which is expected to be covered by forest. The roughness length is fixed for the two upland land use categories, forest and moorland, and the wind speeds are 30 year averages (Thompson *et al,* 1982) for 40 km x 40 km squares interpolated to 20 km values and modified to increase mean wind speeds over high ground. The time for which vegetation is covered by cloud is estimated from long-term cloud cover observations over the UK (Weston, 1992). The cloud ion concentration is derived from the rain ion concentration field.

The dry deposition model requires greater detail in land use categorisation and more meteorological information. The land use data base (Bunce *et al,* 1983) is used to determine the proportions of each square expected to be covered by arable crops, grassland, forest, moorland and urban areas. Climatological data, in the form of 30 year averages for 40 km x 40 km squares interpolated to 20 km values, of wind speed (modified for altitude), temperature, sunshine hours and rainfall (Thompson *et al,* 1982) are input. The latitude and longitude of the square are used to generate hourly values of incoming solar radiation for one day in each month for clear sky, overcast sky and wet conditions. A stomatal resistance, r_s, is generated for each hour of the day from typical vegetation characteristics. Lower optimum temperatures, 25°C, are used than appear elsewhere in the literature because there is no explicit constraint on stomatal opening when a plant is water stressed. Soil and leaf surface resistance are combined and set to constant values depending on expected surface wetness, derived from the rainfall and cloud frequency data and estimates of hours of dew. The total canopy resistance with wet surfaces is set to 20 s m^{-1}, a value which reflects average conditions in the UK. The combined canopy resistance, r_c, is used with vegetation and wind speed dependent resistances, r_a and r_b, to calculate daily deposition velocities for clear sky, overcast sky and wet conditions. The annual sulphur deposition is calculated from the deposition velocities using the estimated occurrence of each weather type in each square and the interpolated SO_2 concentration field derived from data from about 30 monitoring sites.

Model sensitivity

The wet deposition model is linear in both inputs, rainfall and rain ion concentration. The ratio of cloud ion to rain ion concentrations used in determining both the orographic enhancement factor for wet deposition and the cloud ion concentrations for cloud droplet deposition was set to 2. Recent experimental work (Fowler *et al*, in press) indicates this value is conservative for the high rainfall areas of the UK where ratios of 4 to 8 are common. However, the observational data are biased, since they typically sample the base of the cloud where ion concentrations are highest, and a value of 2 gives an estimate over a wide variety of upland landscapes consistent with more complex modelling approaches (Choularton *et al*, 1988).

The cloud droplet deposition model is linear both in cloud ion concentration and in the time vegetation is covered by cloud and is non-linear in wind speed. In this case, a higher ratio of cloud ion to rain ion concentrations than currently used would be justifiable. The proportion of forest within the square has a substantial effect on modelled cloud droplet deposition at higher altitudes.

The dry deposition model is relatively insensitive to several of its inputs, as long as the values are varied within reasonable limits for the UK. Plausible modifications of temperature would alter dry deposition by less than 5%. Wind speed has a non-linear effect on dry deposition and a 40% increase in wind speed would increase dry deposition to forests by 6% and to other land uses by 15%. Doubling the time with wet leaf surfaces would increase deposition to forests by 30% and to other land uses by 10% to 15%. The model is linear with respect to gas concentration. Misspecification of land use has its greatest effect through the presence or absence of forests in remote areas when it can substantially affect deposition.

The values for wind speed adjustment, orographic enhancement of rainfall and cloud frequency are averages over large areas and would not necessarily be appropriate for finer scale modelling.

Possible uncertainty in the estimates of total sulphur deposition

The deposition models are linear with respect to the input concentrations of SO_4^{2-} or SO_2 and errors in these interpolated concentration fields are transmitted directly into the predicted deposition fields.

The Review Group on Acid Rain presented data for 1986 to 1988 including error maps for both precipitation-weighted mean concentration of non-marine sulphate and for mean annual wet deposited sulphate (calculated as a product of rainfall and concentration without inclusion of any altitude effect) (RGAR, 1990). Incorporating an orographic enhancement, which was not used for the original maps, the uncertainty, measured as twice the Kriging error, was approximately 2.5 kg S ha^{-1} y^{-1} over most of England, 4 kg S ha^{-1} y^{-1} over south-west England, Wales, and most of Scotland increasing to 10 kg S ha^{-1} y^{-1} over the high rainfall areas in the west of Scotland. Assuming an error in the rainfall estimate to a 20 km square of ±10%, these values indicate a combined uncertainty (i.e. an approximate 95% confidence interval assuming a Normal distribution of residuals) in wet deposition estimates of about ±40% rising to ±80% in high altitude, high rainfall areas.

Kriging interpolation has been used to derive the SO_2 concentration field, but there are a relatively small number of sites for its successful implementation. There is evidence that the assumptions underlying the derivation of a Kriging error map are not satisfied so the spatial error analysis must be considered an approximation. The error map gave values of 1-2 ppbV over most of the UK, ranging from about 25% of the annual mean concentration in central and eastern England to over 100% of the annual mean concentration in the north-west of Scotland (Vincent, K.J. and Campbell, G.W., *pers. com.)*. Assuming an uncertainty in deposition velocity of ±20%, the overall uncertainty in dry deposition estimates would range from ±40% in central England to well over ±100% in many areas of Scotland.

The major pathway for deposition varies in different areas of the UK with 70% as dry deposition in central England to 80% as wet deposition in north-west Scotland. Ignoring any uncertainty in cloud droplet deposition, the estimates of total sulphur input to a 20 km square could have an uncertainty of ±40% in central England increasing to ±80% on the west of Scotland and Wales.

CATCHMENT STUDIES

A number of studies have required more detailed measurements and modelling of specific areas and extra information has become available. These provide valuable support for the interpretation of deposition estimates.

In studies of the Plynlimon and Llyn Briane catchments in west Wales deposition inputs were modelled using detailed land use and altitude data and local concentration measurements (Reynolds *et al*, 1993). Inputs of sulphur to the catchment modelled from the detailed measurements in 1987 and 1988 averaged 26 kg S ha^{-1} y^{-1} while measured averaged output in the stream water for the same period was 27 kg S ha^{-1} y^{-1}. The inputs to the area estimated from the 20 km squares used in the UK map were in the range 20 to 30 kg S ha^{-1} y^{-1} giving reasonable agreement between the national and the more detailed local model.

A similar exercise based on a study in east England at Beacon Hill, an area dominated by dry deposition, between 1985 and 1988 gave an estimated total sulphur input of 37 kg S ha^{-1} y^{-1} compared against an estimated outflow in the streams of 37 kg S ha^{-1} y^{-1} (Vitkovic and Black, 1994). If the dry year in 1984 is included, the estimated outflow was 31 kg S ha^{-1} y^{-1} and the input increased to 40 kg S ha^{-1} y^{-1}. The estimated input from the 20 km national map squares were approximately 30 kg S ha^{-1} y^{-1}, somewhat lower than the more detailed model estimates.

In a study in the north of Scotland a specific requirement was to model the current input of sulphur to an area of moorland and to estimate the effect of afforestation. The area lay within

2 UK 20 km squares with estimated total sulphur deposition of 5.9 kg S ha^{-1} y^{1}. Detailed modelling of 4 areas gave values of 5.7, 6.1, 6.7 and 6.9 kg S ha^{-1} y^{1}.

These 3 studies indicate that the 20 km deposition estimates were within ±30% of more detailed model estimates, with little evidence of gross systematic error in the national maps.

SIMULATING DEPOSITION DATA AT DIFFERENT SPATIAL SCALES

Deposition estimates are produced for Europe at the 150 km scale and 50 km scale by EMEP. For the UK, deposition is estimated at a scale of 20 km while in the Netherlands deposition estimates will in time be provided at the 5 km scale. Assuming that the deposition models give reliable estimates at their own scale, do they provide good estimates for critical load exceedances when the critical loads are calculated at a 1 km scale?

For this exercise, a method was devised to produce deposition estimates to 1 km squares using data at the 20 km scale. For comparison purposes, deposition was also estimated using exactly the same procedures but implemented at the 20 km scale and the 100 km scale.

Each 20 km square was divided into the number of 1 km squares within 7 altitude bands which were assumed to be at 31, 92, 184, 458, 519, 762 and a notional 1066 m above sea level.

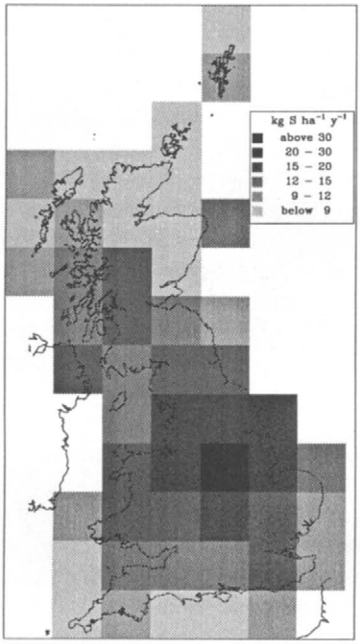

Figure 1 Total deposition of Sulphur modelled at the 100 km scale.

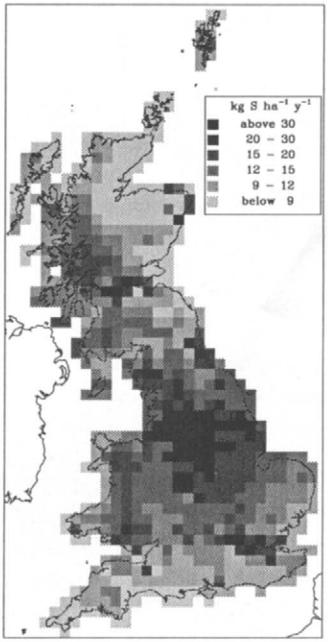

Figure 2 Total deposition of Sulphur modelled at the 20 km scale.

The rainfall to the 1 km squares were estimated from a linear function assuming 600 mm rain at 50 m and 2000 mm rain at 600 m, and then adjusted to match the 20 km square rainfall amounts. The orographic enhancement of wet deposition was estimated by assuming all rainfall amounts in excess of 600 mm had twice the rain ion concentration. Gas and rain ion concentrations were assumed constant over the 20 km square but wind speed was adjusted to the altitude band values. Deposition was calculated for each altitude band and land uses were assigned to the 1 km squares assuming moorland at the highest altitudes, then forest, grass, arable and urban in descending order. This method would not give accurate estimates to any specific 1 km square but does give a distribution of possible deposition values.

Deposition maps

The map of total sulphur deposition at the 100 km scale provides an impression of relatively smooth variation (Figure 1). The 20 km map, although not identical to the 'standard' modelled map, gave very similar structure over the UK (Figure 2).

At the 1 km scale, the information was in distributional form and could not be accurately located spatially. Therefore, a pair of maps were produced: one giving the value for the whole 20 km square as the minimum deposition from the 400 1 km squares (Figure 3) and the other giving it the maximum deposition from the 400 1 km squares (Figure 4). This indicated the range of possible deposition values at the 1 km scale. The minimum 1 km deposition gave a structure

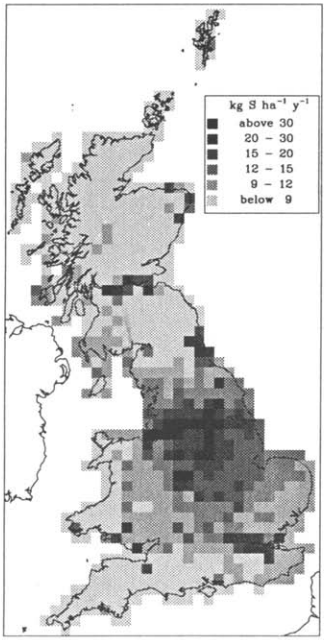

Figure 3 Minimum Sulphur deposition to a 1 km square within the 20 km square.

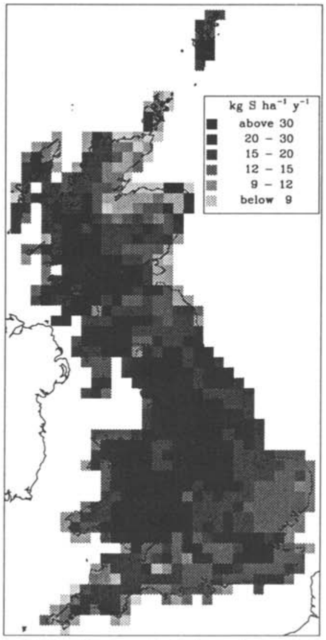

Figure 4 Maximum Sulphur deposition to a 1 km square within the 20 km square.

quite similar to the 20 km map but without showing the areas associated with high wet deposition in west Wales and west Scotland. These areas have rapid changes in altitude and the low level ground did not receive high total deposition according to these models. The maximum 1 km deposition gave values above 20 kg S ha^{-1} y^{1} for large areas of the UK indicating that variations in altitude and land use could give high depositions to local areas practically everywhere.

Distribution of 1 km deposition estimates

The categorisation of altitude and land use only allows typically 7 different deposition values for the 400 1 km squares within the 20 km square. This gives little information on the distribution of values. For 5 areas of the UK, ten 20 km squares covering an area of 40 km E-W x 100 km N-S were grouped together to compare distributions in different landscapes.

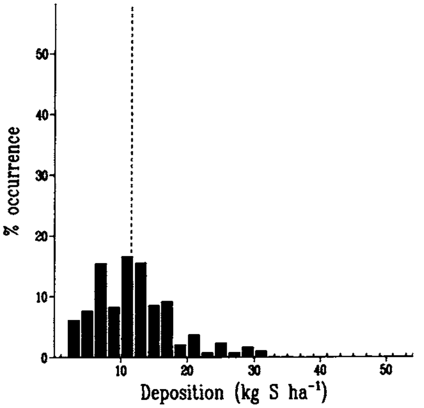

Figure 5 North-West Scotland : Distribution of 1 km deposition in a 40 km x 100 km area, dotted line indicates average 20 km deposition.

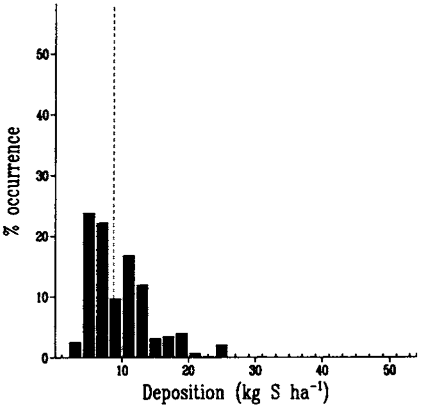

Figure 6 North-East Scotland : Distribution of 1 km deposition in a 40 km x 100 km area, dotted line indicates average 20 km deposition.

In a high rainfall area in the north-west of Scotland with an average 20 km deposition of 12 kg S ha^{-1} y^{-1} dominated by wet deposition (over 80% of total deposition), the 1 km deposition values ranged from 2 to 36 kg S ha^{-1} y^{1} (Figure 5). In a drier area in the north-east of Scotland wet deposition was 60% of total deposition, the average 20 km deposition was 9 kg S ha^{-1} y^{-1} and the range of 1 km deposition values was 2 to 26 kg S ha^{-1} y^{1} (Figure 6). 7% and 9% of the two areas respectively received over twice the 20 km average deposition.

In more polluted areas further south, in a high rainfall area of Wales with wet deposition at 50% of total deposition the average 20 km value was 21 kg S ha^{1} y^{1} and the range was 8 to 52 kg S ha^{-1} y^{-1} (Figure 7). To the east in a drier area in east England where wet deposition is 30% of total deposition the average 20 km value was 25 kg S ha^{1} y^{1} with a range of 16 to 48 kg S ha^{-1} y^{-1} (Figure 8). In Wales about 2% of the area exceeded twice and 14% of the area

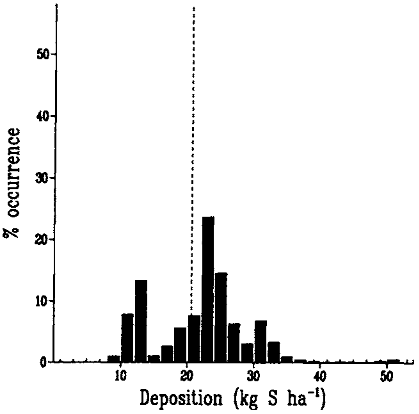

Figure 7 West Wales : Distribution of 1 km deposition in a 40 km x 100 km area, dotted line indicates average 20 km deposition.

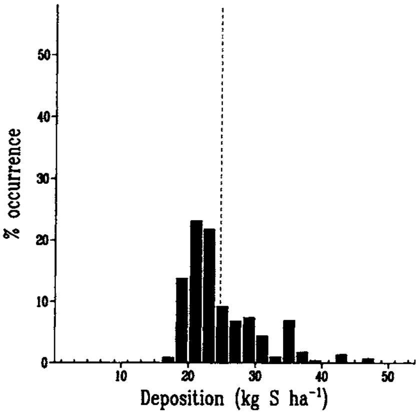

Figure 8 East England : Distribution of 1 km deposition in a 40 km x 100 km area, dotted line indicates average 20 km deposition.

exceeded 1.5 times the average 20 km deposition. For east England 4% exceeded 1.5 times the average 20 km deposition.

In East Anglia, an area more similar to the Netherlands in landscape, the average 20 km deposition was 14 kg S ha^{-1} y^{-1} (with 50% from wet deposition) with a much shorter range from 10 to 22 kg S ha^{-1} y^{-1} (Figure 9).

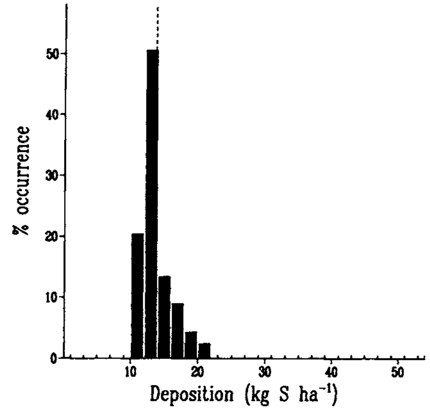

Figure 9 East Anglia : Distribution of 1 km deposition in a 40 km x 100 km area, dotted line indicates average 20 km deposition.

In all cases the distribution of 1 km values was positively skewed. Only in East Anglia, with an altitude and land use pattern similar to the Netherlands, was the range of 1 km estimates relatively short. In the UK context, where sensitive ecosystems are often on higher, poorer land, these simulations indicate that in most areas where critical loads are currently exceeded, the 20 km deposition value will be conservative. For substantial areas of land the 1 km deposition would be 1.5 to 2 times greater than the 20 km deposition.

CRITICAL LOAD EXCEEDANCES AT DIFFERENT SPATIAL SCALES

The critical load exceedances were calculated for the different deposition maps. The results (Table 1) indicate that the difference between the total areas exceeded at the 100 km scale and the 20 km scale, using the same model, was small. Use of the minimum 1 km estimate for the whole square reduced the number of exceeded squares by 15% but the maximum 1 km estimates increased the number of exceeded squares by 75%. These figures give some bounds to the likely impact of deposition scale problems on critical load exceedances.

Table 1
Critical load exceedances for different spatial scales of deposition

Exceedance ($keq\ ha^{-1}\ y^{-1}$)	Number of 1 km x 1 km squares exceeded (in thousands)			
	deposition scale			
	100 km	20 km	Min 1 km	Max 1 km
not exceeded	120	124	140	50
0.0 - 0.2	30	25	24	6
0.2 - 0.5	35	30	24	14
0.5 - 1.0	29	29	18	25
> 1.0	12	17	20	130
total exceeded	106	101	85	176

CONCLUSIONS

The analysis of the accuracy of the current UK national 20 km deposition models, although not a full error analysis of the system, indicated that the uncertainty in predicted total sulphur deposition was about ±40% in central England rising to ±80% on the west of Scotland and Wales. The catchment studies with more detailed models and measurements gave improved deposition values which agreed well with measured stream flow and chemistry in the area. The national estimates were within ±30% of the detailed model values, considerably better than the uncertainty analysis suggested. There was an indication of underestimation by using the national estimates but no definite evidence of bias.

The simulation study of 1 km deposition values from 20 km data, with extra information on altitude and altitude dependencies, showed that the distribution of deposition to 1 km squares within an area is positively skewed for typical landscapes in the UK. Even for a flat landscape in East Anglia the skew distribution was apparent. Therefore the 20 km deposition estimates will be biased and will underestimate 1 km deposition in substantial areas. The importance of the bias for critical load exceedance calculations depends on which 1 km squares have low critical load values. In the UK the high deposition, high altitude regions are also often the areas with very sensitive receptors. Therefore the smallest critical loads tend to have depositions towards the upper tail of the distribution of 1 km deposition values. The simulations indicate that 1 km values of 1.5 to 2 times the 20 km values would be appropriate for such sensitive ecosystems.

The application of minimum and maximum 1 km estimates to the whole 20 km square indicates bounds to the problems that deposition scale poses for critical load exceedances. The area predicted to exceed its critical load estimated from the 20 km deposition estimate is considerably nearer the minimum 1 km value than the maximum 1 km value. Transferring from a 100 km estimate to a 20 km estimate of deposition makes little difference in the total areas with critical load exceedances. Although using the maximum 1 km deposition value is unrealistic, these results indicate a potential substantial underestimation of areas of critical load exceedance, both at the UK national 20 km scale and at the European 150 km scale (EMEP).

The current deposition models are not directly applicable at a scale of 1 km. In the absence of accurate fine scale modelling, the alternative approach is to provide statistical distributional information for deposition to an area. This information would not only incorporate the scale dependency problem but also include the effects of the uncertainties in all the inputs to the models. The critical loads themselves are estimates which have a quantifiable uncertainty. The critical load exceedance for an area would then be provided as a probability distribution of exceedances formed by combining the distributions for deposition and for critical load. This approach would have the further advantage of a direct indication of the uncertainty in the estimates of critical load exceedances.

The large scale deposition estimates (150 km) used throughout Europe and the nationally derived estimates at other scales (20 km or 5 km) lead to significant underestimates of the critical load exceedance at the 1 km resolution. The systematic bias results from fine scale variability in deposition not being represented in the coarse scale modelling. In the absence of deposition models for complex terrain at the 1 km scale, a statistical approach is suggested which will provide critical load exceedances in distributional form and give a clearer indication of the uncertainty in the estimates.

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