UNCERTAINTY ANALYSIS OF DEPOSITION DATA USED IN THE CALCULATION OF EXCEEDANCE OF ACIDITY CRITICAL LOADS FOR UK ECOSYSTEMS

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Abstract.

The critical load concept is based on the maximum load of pollutant deposition an element of the environment can tolerate without harmful effects occurring. The amount of excess deposition above the critical load is called the exceedance. Within Europe, protocols controlling the emissions of sulphur and nitrogen are being implemented and further reductions have an associated high cost. Therefore as the exceedance gets smaller, there is a greater necessity to determine the accuracy of the values.

An uncertainty analysis of the exceedance calculations was performed by Monte Carlo and fixed value analysis, varying deposition errors in the range ± 40%. The uncertainty in calculation of exceedances was studied for 1995-97 and 2010 deposition scenarios.

The analysis to date indicates that taking into account the uncertainties in deposition estimates and using a 95% confidence suggests we may be currently underestimating the area of ecosystems exceeded. More work is needed to attain better knowledge of the size and shape of the deposition distributions in order to draw firmer conclusions from the analysis.

Key words: uncertainty, Monte Carlo simulation, acidity critical load exceedance

1. Introduction

Acidification can lead to deleterious environmental effects, including depletion of fish stocks from lakes and streams, soil degredation and deforestation due to phytotoxicity (Rodhe *et al.* 1995). Acidification is caused by emissions of sulphur dioxide, nitrogen oxides and ammonia resulting from the combustion of fossil fuels. Sensitivity to acidification varies, depending on the type of ecosystem (eg woodland, grassland etc.). A ‘critical loads’ approach has been developed and adopted to identify the spatial distribution of ecosystems at risk from acidification and eutrophication. This paper will focus on acidification only. A critical load has been 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). Critical loads have formed the basis for national and international deliberations on the control of atmospheric emissions and depositions of sulphur and nitrogen. The excess deposition above the critical load, the critical load exceedance, is used to quantify the potential harmful effects.

It should be noted that the empirical and mass balance methods on which national critical loads are based, define long-term critical loads for systems at steady-state. Therefore exceedance is an indication of the *potential* for harmful effects to systems at steady-state. This means that current exceedance of critical loads does not necessarily equate with damage to the ecosystem.

Previously critical load exceedances have been calculated as deterministic estimates. However each parameter in the calculation of critical load and exceedance has some uncertainty associated with it. Until now exceedance calculations have not accounted for these inherent uncertainties in input parameter values. Exceedance estimates could be produced along with confidence limits on model predictions. As the cost of further reducing emissions increases, it becomes more important to take uncertainties into account when estimating exceedances. Syri *et al.* (2000) state that "the total costs for a country can vary considerably with small changes in environmental targets".

This paper outlines the results of a preliminary study undertaken to quantify the variation in acidity critical load exceedance caused by the uncertainty in sulphur and nitrogen deposition values alone. This study presents the initial results, obtained by 1) fixed value uncertainty and 2) Monte Carlo analyses on two deposition scenarios. This paper is concerned with the effect of deposition uncertainty on exceedance values at a national level. The uncertainty of exceedance values will involve a product of many factors. Those contributing significantly to the uncertainty of the final product are those with the largest errors. Hence deposition alone has been investigated as an initial attempt to gain some knowledge of exceedance uncertainty. The uncertainty range would be widened by uncertainty in critical loads calculations, but these additional effects are not considered here.

The response of the exceedance calculation to variations in sulphur alone, nitrogen alone and sulphur and nitrogen together was investigated, i.e. a sensitivity analysis. Nitrogen and sulphur were varied together to investigate any synergistic effects. Deposition was varied 1) by fixed amounts and 2) randomly. Exceedance predictions have been measured in 2 ways:

1) the total area of sensitive ecosystems exceeded in a grid square
2) the Accumulated Exceedance (AE). This takes account of both the exceeded ecosystem area and the magnitude of exceedance and is defined as:

$$\underset{(\text{keq year}^{-1})}{\text{AE}} = \underset{(\text{keq ha}^{-1}\ \text{year}^{-1})}{\text{exceedance}} * \underset{(\text{ha})}{\text{exceeded ecosystem area}}$$

The aims of this study were to:

- Quantify the effect of uncertainty in nitrogen and sulphur deposition on percentage area of UK sensitive ecosystems exceeded.
- Quantify the effect of uncertainty in nitrogen and sulphur deposition on accumulated exceedance.
- Investigate whether there is a linear relationship between deposition uncertainty and area of critical load exceedance or accumulated exceedance.
- Consider whether critical load exceedance is more sensitive to sulphur or nitrogen deposition.
- Produce exceedance estimates along with confidence limits on exceedance prediction.

2. Methods

This section will describe the input data required, the exceedance calculation and uncertainty analysis methods used. The critical load information contains a number of records for 1km squares of the UK. Currently critical loads data are calculated for six ecosystems. The deposition data is total nitrogen and sulphur values mapped for every 5 km grid square of the UK. The exceedance values are then calculated for every 1km grid square. These data can then be aggregated to map parameters at 5km resolution or summed across grid-squares to give national statistics.

2.1 INPUT DATA

Critical loads data

To examine the acidifying effects of both sulphur and nitrogen deposition simultaneously, the Critical Loads Function (CLF) was developed in Europe (Posch *et al.*, 1999; Posch & Hettelingh, 1997; Posch *et al.*, 1995; Hettelingh *et al.*, 1995).

The maximum critical load of sulphur (*CLmax(S)*) is the critical load for acidity expressed in terms of sulphur only, ie when nitrogen deposition is zero. Similarly, the maximum critical load of nitrogen (*CLmax(N)*) is the critical load of acidity expressed in terms of nitrogen only (when sulphur deposition is zero). The long-term nitrogen removal processes in the soil (nitrogen uptake and immobilisation) define a “minimum” critical load for nitrogen (*CLmin(N)*). The UK currently calculates these “minimum” and “maximum” critical loads for areas of acid grassland, calcareous grassland, heathland, coniferous woodland, deciduous woodland and freshwaters (Hall *et al.*, 1998). It is these critical load values for each ecosystem considered nationally that are used for this study.

Deposition data

Input data are a combination of wet and dry nitrogen, sulphur and ammonia deposition. Deposition estimates for moorland were applied in the calculations for acid grassland, calcareous grassland and heathland; woodland deposition estimates were used for coniferous and deciduous woodland and mean deposition estimates for freshwaters.

Two different deposition scenarios have been used to investigate the effect of uncertainty on exceedance within the United Kingdom:

Scenario 1) is based on 5 km resolution mean deposition for 1995-97. Maps of total (ie the sum of wet, dry and cloud) deposition for each 5km grid square of the UK have been provided by CEH Edinburgh for this work (Smith *et al.* 2001, Smith *et al.* 2000). These are annual mean values are based on three years data. Figure 1(a) shows non-marine sulphur and Figure 1 (c) shows the total nitrogen deposition for this scenario.

Scenario 2) is the Gothenburg Protocol deposition scenario modelled from the Hull Acid Rain Model (HARM) (Metcalfe *et al.*, in press) and the Fine Resolution Ammonia model (FRAME) (Singles *et al.*, 1998; Sutton *et al.*, 1995) atmospheric deposition models. This is a deposition scenario for 2010 based on assuming the implementation of the recent United Nations Economic Committee for Europe Gothenburg Protocol (UNECE, 1999). HARM outputs are used for sulphur and nitrogen oxide deposition while FRAME is used for ammonia deposition. The deposition scenario is illustrated in figures 1(b) for sulphur and 1 (d) for nitrogen.

These two scenarios are referred to elsewhere in this paper as the 1995-97 and 2010 baseline scenarios.

2.2 THE EXCEEDANCE CALCULATION

A suite of programs in ARC/INFO Macro Language (AML) have been developed and linked via a C program to calculate critical loads exceedances within a GIS framework. This program, EXCEED, calculates the area of ecosystem exceeding the acidity critical load within a grid-square, and also the accumulated exceedance for each grid square. The program produces summary statistics for each ecosystem type and for England, Northern Ireland, Scotland, Wales, Great Britain and the United Kingdom separately.

Exceedance outputs can be produced at 1km, 5km, regional and national scales. For this study, statistics for the whole UK were used to give an estimate of the effect of deposition uncertainty at the national level. Maps at 5 km resolution of the ecosystem areas exceeded and accumulated exceedance for the 1995-97 and 2010 baseline deposition scenarios are shown in Figure 2. The existing national methodology (used to produce this figure) is referred to as the deterministic approach throughout this paper.

2.3 UNCERTAINTY ANALYSES

Uncertainty analysis involves the computation of the total uncertainty in the output induced by quantified uncertainty in data inputs and model parameters. Two methods for examining uncertainty in deposition values were used; fixed value analysis and Monte Carlo analysis. These techniques are described below.

The Fixed Value Analysis

For the purposes of this study, the estimated total sulphur and nitrogen deposition to any 5 km square in the United Kingdom is assumed to have an uncertainty of $\pm 40\%$. Smith *et al.* 1995 stated that the basis on which these estimates are made is quite fragile, as there are many uncertainties in the system that cannot be quantified directly without extensive investigation.

Six simulations were investigated, for this fixed values analysis, D1 to D6:

(D1) $N_{\text{gen}} = N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}} + 0.4 \cdot S_{\text{base}}$

(D2) $N_{\text{gen}} = N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}} - 0.4 \cdot S_{\text{base}}$

(D3) $N_{\text{gen}} = N_{\text{base}} + 0.4 \cdot N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}}$

(D4) $N_{\text{gen}} = N_{\text{base}} - 0.4 \cdot N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}}$

(D5) $N_{\text{gen}} = N_{\text{base}} + 0.4 \cdot N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}} + 0.4 \cdot S_{\text{base}}$

(D6) $N_{\text{gen}} = N_{\text{base}} - 0.4 \cdot N_{\text{base}}$

$S_{\text{gen}} = S_{\text{base}} - 0.4 \cdot S_{\text{base}}$

where

N_{base} is the total (oxidised + reduced) nitrogen deposition value as estimated by 1995-97 or 2010 baseline deposition scenario
S_{base} is the non-marine sulphur deposition value as estimated by 1995-97 or 2010 baseline deposition scenario
N_{gen} is the generated nitrogen deposition scenario used in the simulation
S_{gen} is the generated sulphur deposition scenario used in the simulation

The last two scenarios were run as worst/best case scenarios calculated by increasing (worst case) and reducing (best case) both nitrogen and sulphur deposition.

The Monte Carlo Analysis

Monte Carlo simulation (Mowrer, 1997) is a well-established technique for assessing the effects of uncertainty in inputs and model parameters. The majority of previous studies in this area have recognised that the underlying distribution of deposition values is largely unknown due to limited data availability (Jonsson *et al.*, 1995, Barkman *et al.* 1995). Hence, previous studies have assumed a uniform distribution between the upper and lower quoted deposition values. Therefore, a uniform distribution was selected for use within the present study. The following Monte Carlo simulations were used:

(D7) $N_{\text{gen}} = N_{\text{base}}$

$S_{\text{gen}} = S_0 + r \cdot (S_1 - S_0)$

(D8) $N_{\text{gen}} = N_0 + r \cdot (N_1 - N_0)$

$S_{\text{gen}} = S_{\text{base}}$

(D9) $N_{\text{gen}} = N_0 + r \cdot (N_1 - N_0)$

$S_{\text{gen}} = S_0 + r \cdot (S_1 - S_0)$

where
N_{gen} is the generated nitrogen deposition scenario used in the simulation
S_{gen} is the generated sulphur deposition scenario used in the simulation
N_0 is the lower limit for nitrogen deposition ($N_0 = N_{\text{base}} - 0.4 \cdot N_{base}$)

S_0 is the lower limit for sulphur deposition ($S_0 = S_{\mathrm{base}} - 0.4 \cdot S_{base}$)
N_1 is the higher limit for nitrogen deposition ($N_1 = N_{\mathrm{base}} - 0.4 \cdot N_{base}$)
S_1 is the higher limit for nitrogen deposition ($S_1 = S_{\mathrm{base}} - 0.4 \cdot S_{base}$)
r is a random value between 0 and 1

Monte Carlo simulations involve strong assumptions on the mutual independence among the different input variables. It has therefore been assumed that the nitrogen and sulphur depositions are independent, i.e. *r* in D9 is different for sulphur and nitrogen.

The major technical difficulty when carrying out the Monte Carlo analysis was dealing with the large amount of data associated with simulating 1km squares of the entire United Kingdom. The software developed was coded in C and run on a Sun Unix workstation. Data analysis was carried out using IDL 5.0 and Excel 97. Exceedance data were mapped using ARC Version 8.0.1 (Environmental Systems Research Institute).

3. Results

The following section is divided into three main sections giving the results for: 1) the deterministic approach using the baseline scenarios previously defined 2) the fixed value analysis in which fixed value uncertainty has been applied to the baseline deposition scenarios (simulations D1 to D6), 3) the Monte Carlo analysis in which random uncertainty has been applied to the baseline deposition scenarios (simulations D7 to D9).

3.1 DETERMINISTIC

The total area of each ecosystem exceeded and the accumulated exceedance across the United Kingdom for the 1995-97 and 2010 baseline deposition scenarios, not accounting for uncertainty, have previously been calculated at the UK National Focal Centre (Hall *et al.* 2001). The ecosystem area exceeded in the UK for 1995-97 has been calculated as 68 265 km^2 and for 2010 as 29 330 km^2. The accumulated exceedance in the UK for 1995-97 has been calculated as 7 236 915 keq year^{-1} and for 2010 as 1 436 411 keq year^{-1}. The exceeded areas and accumulated exceedance have been mapped at 5km resolution and are shown in figure 2.

The frequency distribution of exceedance values for all 1 km grid squares was formed for both 1995-97 and 2010 baseline deposition scenarios (figure 3). This value is either positive or negative depending on whether the deposition did or did not exceed the critical load. The peak of the 1995-97 frequency distribution (figure 3(a)) falls into the positive range. The peak of the frequency distribution for 2010 is in the negative range (figure 3(b)). This is to be expected, as the deposition values for 2010 are less than those for 1995-97 (figure 1), so the critical loads are less likely to be exceeded, resulting in smaller areas of ecosystems exceeded.

The distributions of critical load values are shown in Figure 4. The variables *Clmax(S)* (Fig 4(a)) and *Clmax(N)* (Fig 4(b)) showed a bimodal pattern of variation. This reflects the two types of ecosystem present in the UK, namely woodland and non-woodland. *Clmin(N)* values (Fig 4(c)) are low and the mode falls at 0.25 keq. The bimodal distribution of the critical load parameters may be justification for using the Monte Carlo method of uncertainty analysis. Using other types of uncertainty analysis would involve complex mathematics when calculating exceedance using the combination of deposition and critical loads data.

3.2 FIXED VALUE UNCERTAINTY ANALYSIS

The effects of fixed value uncertainty in deposition values on the total area of ecosystems exceeded and accumulated exceedance are displayed graphically in Figure 5 (scenarios D1 to D6). Table 1 gives the changes for all six of these simulations from the deterministic values.

Area Exceeded

The worst case scenario for the 1995-97 causes an increase in the area of critical load exceedance of 12 676 km^2, whereas the best case scenario causes a decrease of 30 448 km^2.

For the 2010 deposition the worst case scenario causes an increase in exceeded area of 14 065 km^2 and the best case scenario a decrease in area exceeded of 20 767 km^2.

Across the range of deposition uncertainty the (–40% to +40% of the baseline deposition estimate) there is a non-linear relationship between deposition and area of critical load exceedance for both the 1995-97 and 2010 deposition scenarios. This behaviour is explained in the discussion section.

Table 1 shows that nitrogen deposition causes a larger change in exceeded area than sulphur deposition.

Accumulated exceedance

The 1995-97 worst case scenario causes an increase in accumulated exceedance of 6 660 613 keq year^{-1}. The best case scenario causes a decrease in accumulated exceedance of 5 299 371 keq year^{-1}.

The 2010 deposition worst case scenario causes an increase of accumulated exceedance of 2 277 535 keq year^{-1}. The best case scenario causes a decrease in accumulated exceedance of 1 264 338 keq year^{-1}.

Across the range of deposition uncertainty the (–40% to +40% of the baseline deposition estimate) there is a non-linear relationship between deposition and accumulated exceedance for both the 1995-97 and 2010 deposition scenarios. This behaviour is explained in the discussion section.

Table 1 shows that nitrogen deposition causes a larger change in accumulated exceedance than sulphur deposition.

3.3 MONTE CARLO UNCERTAINTY ANALYSIS

The above simulations represent the worst/best case scenarios; these are unrealistic in that they assume that *all* of the deposition values have been under or overestimated. A much more sensible simulation is one in which the deposition value is varied randomly, by the same amount for every grid square, between the two extremes for every trial.

A uniform distribution was selected for use within the present study so that *r* in scenarios *D7*, *D8* and *D9* is drawn from a uniform distribution between 0 and 1.

It should be noted that the results of the Monte Carlo simulations are stochastic in nature so a result obtained from one run of the simulation will not produce the same result as the following run. 250 trials for each nitrogen and sulphur deposition value were used; this value was chosen through testing sets of 100, 250 and 1000 runs, which showed that 250 runs achieved reproducible results. To check whether the number of calculations for the United Kingdom was sufficient, the cumulative standard deviations were calculated according to:

$$s_x = \sqrt{\frac{n\Sigma x^2 - (\Sigma x)^2}{n(n-1)}}$$

where n = number of trials and x = result of Monte Carlo simulation. Figure 6 displays the cumulative standard deviations for the 250 trials of each of the deposition scenarios. Each graph, through its stabilisation, shows that enough trials were undertaken. Note, the difference in scales of the Y coordinate axes in Figure 6.

Table 2 gives the range of outputs from the Monte Carlo simulations D7, D8, D9, i.e the difference between the smallest and largest values of the Monte Carlo trials.

Area Exceeded

The results given in Table 2 demonstrate that 1) Sulphur uncertainty shows low variation (D7). Variation in exceeded area is around 25% of the deterministic value in both the 1995-97 and 2010 simulations 2) Nitrogen uncertainty shows that critical load exceedance is strongly driven by nitrogen deposition (D8). Variation in exceeded area is around 25 000 km^2 for the 1995-97 and 2010 simulations. 3) Sulphur and nitrogen uncertainty show that variation in exceeded area is approximately 32 500 km^2 for the 2010 simulation and over 40000 km^2 for the 1995-97 simulation (D9).

Accumulated Exceedance

The results in Table 2 show that 1) Sulphur uncertainty shows low variation. Variation in accumulated exceedance is around 50% of the deterministic value in both the 1995-97 and 2010 simulations. 2) Nitrogen uncertainty shows that accumulated exceedance is strongly driven by nitrogen deposition. Variation in exceeded area is over 100% of the deterministic value for the 1995-97 and nearly 200% of the deterministic value for the 2010 simulations. 3) Sulphur and nitrogen uncertainty show that variation in accumulated exceedance is approximately 1.5 times the deterministic value for the 1995-97 simulation and over twice the deterministic value for the 2010 simulation.

Frequency Distributions

Figure 7 gives the frequency distributions of ecosystem areas exceeded generated in the trial runs. The distribution for the 1995-97 data (figure 7(a)) does not show an optima and has a median value of 67 941 km^2. The 2010 frequency distribution (figure 7(b)) shows an almost uniform distribution, the median value is 27 157 km^2.

The frequency distribution of the 1995-97 accumulated exceedance (figure 7(c)) is not obviously skewed but an Anderson-Darling normality shows that the distribution is not normal. The median value is 7 147 424 keq year^{-1}. The 2010 accumulated exceedance frequency distribution (figure 7(d)) is slightly skewed with a median value of 1 223 183 keq year^{-1}.

The probabilistic exceedance plots shown in figure 8 are formed from the Monte Carlos outputs of the 250 trials. The results for area exceeded and accumulated exceedance were ranked from largest to smallest. Each was assigned a probability according to the equation:

$$P = \frac{r}{N+1}$$

where r = rank and N = number of trials. For example in graph 7(a) the value 78 673 km^2 is the 13th largest out of 250 values. The probability of this being exceeded is therefore 13 / 250 x 100 = 5.2%. Put another way, the probability of having protected all ecosystems at risk using the value 78 673 km^2 is 94.8%. These plots are discussed fully in the next section.

4. Discussion

The focus of this discussion is the 2010 results. A comparison is made between these results and the 1995-97 simulations.

4.1 FIXED VALUE ANALYSIS

Area exceeded

The deterministic estimate of areas of critical load exceedance for 2010 was 29 330 km^2 (Table 1) of sensitive ecosystems. By comparison, the worst case scenario resulted in 43 496 km^2 and the best case scenario 8 564 km^2. Note that this range is not equally spread around the deterministic value, i.e. it is non linear (figure 5). This was also seen to be the case for the 1995-97 simulation. Both the 1995-97 and 2010 simulations show a larger decrease in exceeded area than increase when fixed value deposition uncertainty is taken into account. To explain this non-linear effect it was observed that the areas where ecosystem were just exceeded received larger deposition than those which were just non-exceeded. It would be expected that areas with high deposition would be more likely to be exceeded. When a percentage change in deposition is made the *absolute* value of this change will be larger than the same percentage change applied to a smaller deposition. The outcome of this is that it is more likely for an area which was just exceeded (due to a large deposition) to become unexceeded when the deposition is decreased than it is for an area which was just unexceeded (due to a small deposition) to become exceeded when the deposition is increased.

Accumulated exceedance

The range of accumulated exceedance from the ‘best case’ to the ‘worst case’ scenario was 172 073 to 3 713 946 keq year^{-1} for the 2010 deposition scenario.

Both the 1995-97 and 2010 simulations show a larger increase in accumulated exceedance than decrease when fixed value deposition uncertainty is taken into account. This is further explained below:

Consider an increase in deposition. This may lead to exceedance of critical loads that were previously not exceeded, in which case the area of ecosystems exceeded increases and the new exceeded ecosystem area contributes to the accumulated exceedance. Alternatively it will enhance the exceedance of critical loads already exceeded. This will not alter the area of ecosystems exceeded (since once the area is exceeded it is only counted once), however, it will increase the accumulated exceedance value since the magnitude of exceedance is greater.

Now consider a decrease in deposition. This may lead to non-exceedance of critical loads that were previously exceeded, in which case the area of ecosystems exceeded is decreased and the value of accumulated exceedance is also reduced. For ecosystems where the critical load was previously non-exceeded, the non-exceedance (i.e. negative) value will decrease further. This will have no impact on the area of ecosystem exceeded or the accumulated exceedance - since both are only calculated for exceeded areas.

Sensitivity to nitrogen and sulphur deposition

Nitrogen deposition uncertainty was shown to cause wider output variation than sulphur deposition uncertainty (figure 5) for both area exceeded and accumulated exceedance. This is because the nitrogen deposition is larger than sulphur deposition (figure 1) for both the 1995-97 and 2010 scenarios. Hence perturbing the deposition values by $\pm 40\%$ produces a larger uncertainty on the (higher) nitrogen deposition values than it does on the (smaller) sulphur deposition values.

4.2 MONTE CARLO UNCERTAINTY ANALYSIS

Monte Carlo analysis was used as an alternative method for assessing the uncertainty of deposition on UK acidity critical load exceedances.

The probabilities for the areas of ecosystem exceeded in 2010 are shown in Figure 8. The area in the UK remaining at risk of acidification in 2010 was calculated to be 29 330 km^2, i.e. 31% of the total ecosystem area (Hall *et al.* 2001). Figure 8(a) summarizes the estimates of protected areas of the deterministic approach and the Monte Carlo analysis. The estimated range of values the area exceeded takes is from approximately 10 000 km^2 to approximately 40 000 km^2. A conservative interpretation (i.e. risk of exceedance less than 5%) gives approximately 35% increase

(39 540 km^2) in area exceeded above the deterministic approach. If the deposition uncertainty is taken into account, there is a 40% probability of exceeding the critical loads for the 29 330 km^2 estimated to be exceeded by the deterministic approach. If a 50% probability of exceeding were used as an indicator, the estimate of area at risk would be slightly more optimistic (27 200 km^2) than the deterministic approach.

The probabilities of ecosystem exceedance decrease by 2010 because the deposition levels decrease and thus the critical load are less likely to be exceeded.

Figure 8(b) shows the probabilistic graph for accumulated exceedance. The accumulated exceedance for 2010 was calculated to be 1 436 411 keq $year^{-1}$. It summarizes the estimates of accumulated exceedance of that deterministic approach and the Monte Carlo approach. The values range from about 300 000 keq $year^{-1}$ to approximately 3 500 000 keq $year^{-1}$. A conservative interpretation (risk of exceedance less than 5%) give over 100% increase (3050 000 keq $year^{-1}$) compared to the deterministic approach. When deposition uncertainty is taken into account, there is a 42% probability of the accumulated exceedance being greater than the 1 436 411 keq $year^{-1}$ estimated by the deterministic approach. If a 50% probability of exceeding were used as an indicator, the accumulated exceedance would be slightly more optimistic (1 230 000 keq $year^{-1}$) than the deterministic approach.

4.3 COMPARISON WITH PREVIOUS WORK

The Monte Carlo results for area exceeded are in agreement with Barkman *et al.* (1995). Barkman *et al.* 1995 investigated the effects of uncertainties in both input data and model parameters in calculations of the critical load of acidity and its exceedance, for forest ecosystems in two studies using Monte Carlo simulations of the regional PROFILE model (Warfvinge and Sverdrup 1992; Sverdrup and Warfvinge 1991). He found for southern Sweden that taking into account the uncertainties in critical load and deposition estimates and allowing a 5% exceedance probability would result in more pessimistic estimates of protected areas than according to the deterministic approach.

In this paper the variation in deposition gives a non-linear relationship with area exceeded. This is in disagreement with the results of Smith *et al.* 1995. He established that in the range of –40% to +40% of the current 20 km deposition estimate there was an almost linear relationship between deposition and area of critical load exceedance. There are a number of possible reasons for these contradictions. Smith *et al.* used the simple empirical approach of dividing soil materials into five classes defined on the basis of their dominant weatherable minerals (Nilssone and Grennfelt, 1988). Each 1 km grid square had one critical load value only. More recently the calculation of critical loads has changed considerably and become more complex. More parameters have been incorporated into the empirical calculation of critical loads and the simple mass balance equation is now used for woodland.

5. Conclusions

Uncertainties in deposition estimates have a large effect on exceedance calculations.

The fixed value analysis suggests that by including uncertainty in 2010 deposition in the calculation of exceedance, the uncertainty in areas exceeeded could be between +50 and -70% of the deterministic value. The Monte Carlo analysis narrows this range to +35 and -50% and gives a most likely estimate of 27 200 km^2, a decrease of 7% of the deterministic value.

The fixed value analysis suggests that by including uncertainty in 2010 deposition in the calculation of accumulated exceedance, the uncertainty in maybe even greater e.g. between +160 and -90% of the deterministic value. The Monte Carlo analysis narrows this range to +100 and -75% and gives a most likely estimate of 1 230 000 keq $year^{-1}$, a decrease of 14% of the deterministic value.

The Monte Carlo analysis was more useful than the fixed value analysis for estimating the impacts of uncertainties in the calculation of exceedances in this study. It was possible with this method to associate exceedance probabilities with a whole range of exceeded areas and accumulated exceedances. The fixed value analysis only allowed you to look at the worst and best case situations.

In the Monte Carlo analysis a uniform distribution was used to model the uncertainty of the deposition distributions. The decision to use a uniform distribution was justified by a lack of information about the form of the underlying nitrogen and sulphur deposition distribution. In reality a uniform distribution may not be the most appropriate probability function for this variable. Seiler et al. (1996) suggests that the triangular distribution may be more appropriate in this case.

It was interesting to look at both the areas exceeded and accumulated exceedance as they both represent different ways of looking at potential ‘harm’ to the environment. However, accumulated exceedance was more useful as it takes account of both the area of ecosystem exceeded *and* the magnitude of the exceedance.

The Gothenburg Protocol is an important step towards ensuring protection of UK ecosystems from acidification. After its implementation in 2010, the area of sensitive ecosystems exceeded should only be 41%, base on the 5% probability of exceedance calculations. This equates to a 50% reduction in the area of ecosystems exceeded in 1995-97.

Current international emission abatement strategies are based on exceedance calculations without considering uncertainties. Syri *et al.* (2000) has already stated that “For many countries achieving emission ceilings will require costly abatement installations with high unit reduction costs.” The Monte Carlo approach provides information about the confidence limits for the environmental impact estimates of emission reduction efforts. 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 be a key activity in the UK and Europe over the next two to three years, in preparation for reviewing international protocols.

The analyses in this paper are based on looking at uncertainty in one parameter only, input deposition. Other parameters in the calculation affect the exceedance estimates and these parameters will also have uncertainties, which are important to analyse. It should be remembered that the uncertainty in critical loads has not yet been taken into account and this may change these results significantly. This should be a major focus of future work.

The decrease in deposition has been applied uniformly across all grid squares. In reality changes in emissions would result in a heterogeneous change in deposition. Smith *et al*. (1995) states that the uncertainty value of 40% may rise to 80% in the west of Scotland and in Wales. Hence, in future, uncertainties in deposition should be applied in different amounts in different areas according to more realistic criteria, derived from further empirical work. Uncertainties due to spatial resolution must also be further investigated.

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References

Barkman, A., Warfinge, P., Sverdrup, H., 1995. Regionalization of critical loads under uncertainty. Water, Air and Soil Pollution 85, 2515-2520.

Hall, J., Broughton, R., Bull, K., Curtis, C., Fowler, D., Heywood E., Hornung, M., Metcalfe, S., Reynold, B., Ullyett, J. Whyatt, D., 2001. Status of UK Critical Loads and Exceedances, January 1998. Part 2 – Exceedances. In preparation.

Jonsson, C., Warfvinge P., Sverdrup, H., 1995. Uncertainty in predicting weathering rate and environmental stress factors with the PROFILE model. Water, Air and Soil Pollution 81, 1-23.

Nilsson, J., Grennfelt, P., (ed). Critical loads for sulphur and nitrogen (Report 1988:15). Nordic Council of Ministers, Copenhagen, Denmark, 1988.

Posch, M. 1995. Percentiles and protection isolines. In: Posch, M., de Smet, P.A.M., Hettelingh, J.-P. & Downing, R.J. (Eds), Calculation and Mapping of Critical Thresholds in Europe: Status Report 1995. Coordination Centre for Effects, National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands. pp 43-47.

Posch, M. & Hettelingh, J.-P. 1997. Remarks on critical load calculations. In: Posch, M., de Smet, P.A.M., Hettelingh, J.-P. & Downing, R.J. (Eds), Calculation and Mapping of Critical Thresholds in Europe: Status Report 1997. Coordination Centre for Effects, National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands. pp 25-28.

Posch, M., de Smet, P.A.M. & Hettelingh, J.-P. 1999. Critical loads and their exceedances in Europe: an overview. In: Posch, M., de Smet, P.A.M., Hettelingh, J.-P. & Downing, R.J. (Eds), Calculation and Mapping of Critical Thresholds in Europe: Status Report 1999. Coordination Centre for Effects, National Institute of Public Health and the Environment (RIVM), Bilthoven, The Netherlands. pp 3-11.

Seiler, F., Alvarez, J., 1996. On the Selection of Distributions for Stochastic Variables. Risk Analysis 16, 5-18.

Smith, R., Fowler D., 2001. Uncertainty is estimation of wet deposition of sulphur. Water, Air and Soil Pollution (in press).

Smith, R., Fowler, D., Sutton, M., Flechard, C., Coyle, M., 2000. Regional esimtation of pollutant gas dry deposition in the UK: model description, sensitivity analyses and outputs. Atmospheric Environment 34, 3757-3777.

Smith, R., Hall, J., Howard, D.C., 1995. Estimating uncertainty in the current critical loads exceedance models. Water, Air and Soil Pollution 85, 2503-2508.

Sverdrup, H., Warfvinge, P., 1991. On the geochemistry of chemical weathering, in K. Rosen (ed.), Chemical Weathering under Field Condition. Reports on Forest Ecology and Forest Soils 63, Swedish University of Agricultural Science, Uppsala, Sweden, 79.

Syri, S., Suutari, R., Posch, M., 2000. From emission in Europe to critical load exceedances in Finland – uncertainty analysis of acidification integrated assessment. Environmental Science & Policy 3, 263-276.

Warfvinge, P. and Sverdrup, H., 1992. Water, Air and Soil Pollution 63, 119.

FIGURES

Figure 1	Baseline deposition scenarios at a 5 km resolution. a) 1995-97 non-marine sulphur b) 2010 non-marine sulphur c) 1995-97 total nitrogen (nitrogen oxide + ammonia) d) 2010 total nitrogen (nitrogen oxide + ammonia). Sources CEH Edinburgh, Edinburgh University and Lancaster University
Figure 2	5 km maps where acidity loads are exceeded by acid deposition for a) area of ecosystems exceeded by 1995-97 baseline deposition b) area of ecosystems exceeded by 2010 baseline deposition c) accumulated exceedances for 1995-97 baseline scenario deposition d) accumulated exceedances for 2010 baseline scenario. Source: CEH Monks Wood
Figure 3	Exceedance frequency distributions a) 1995-97 simulation b) 2010 simulation
Figure 4	5-percentile critical loads (bases on 6 ecosystems) a) Clmaxs b) ClmaxN, CLminN
Figure 5	Critical load exceedance for the UK (total area of sensitive ecosystems 95782.4 km^2) Sulphur and nitrogen variation (short dash), nitrogen variation only (solid line) , sulphur variation only (long dash) a) area critical load exceedance for 1995-97 simulation b) area of critical load exceedance for 2010 acid simulation c) accumulated exceedance for 1995-97 simulation d) accumulated exceedance for 2010 simulation
Figure 6	Change in cumulative standard deviation. 1995-97 acid deposition (solid line), 2010 acid deposition (short dash). a) Sulphur variation b) Nitrogen variation c) Sulphur and nitrogen variation
Figure 7	Frequency distribution a) Area exceeded 1995-97 simulation b) area exceeded 2010 simulation c) accumulated exceedance 1995-97 scenario b) accumulated exceedance 2010 simulation

Figure 8 The estimates of a) ecosystem areas exceeded and b) accumulated exceedance in UK for 2010 with the Monte Carlo approach.

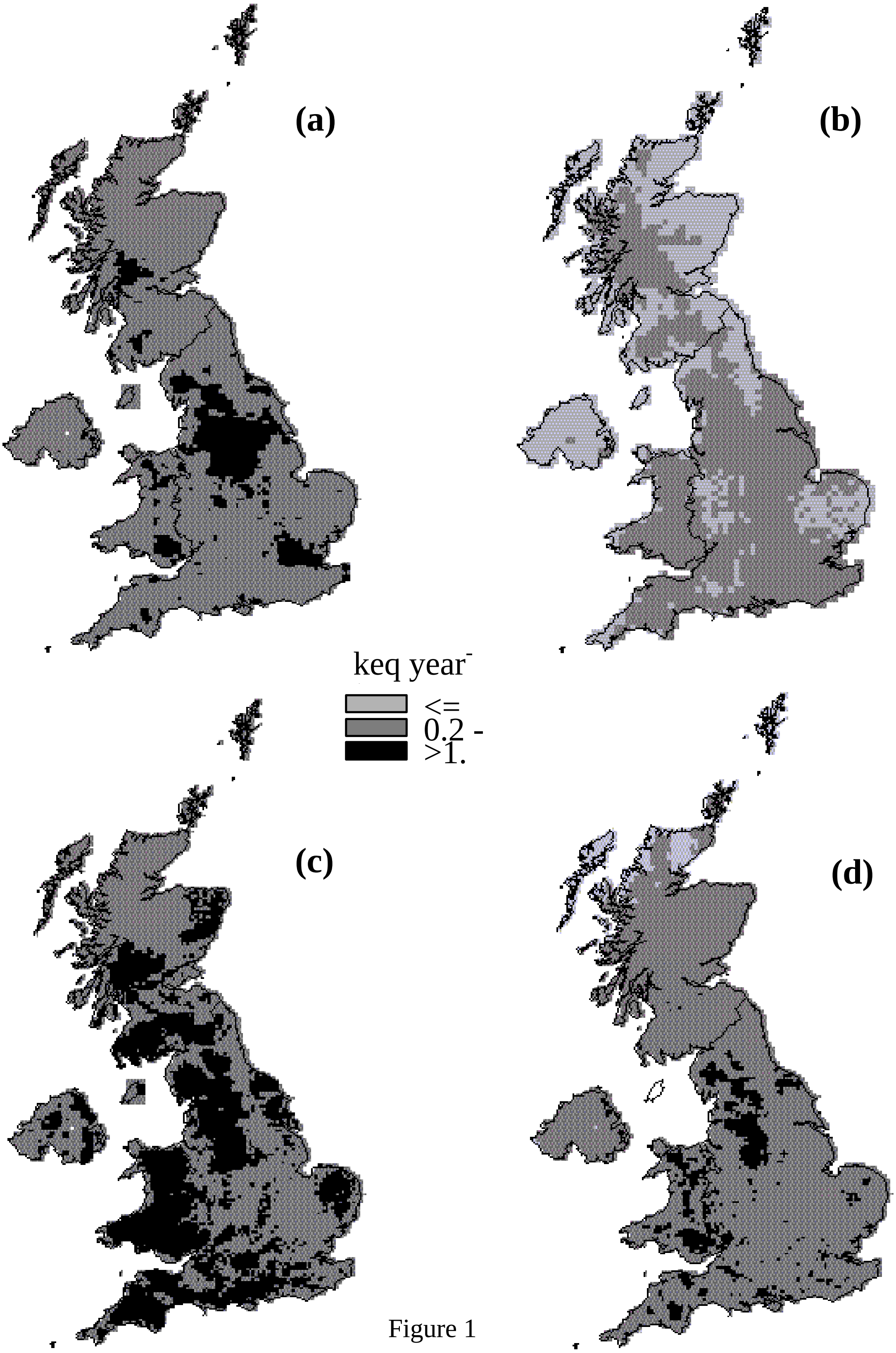


Figure 1

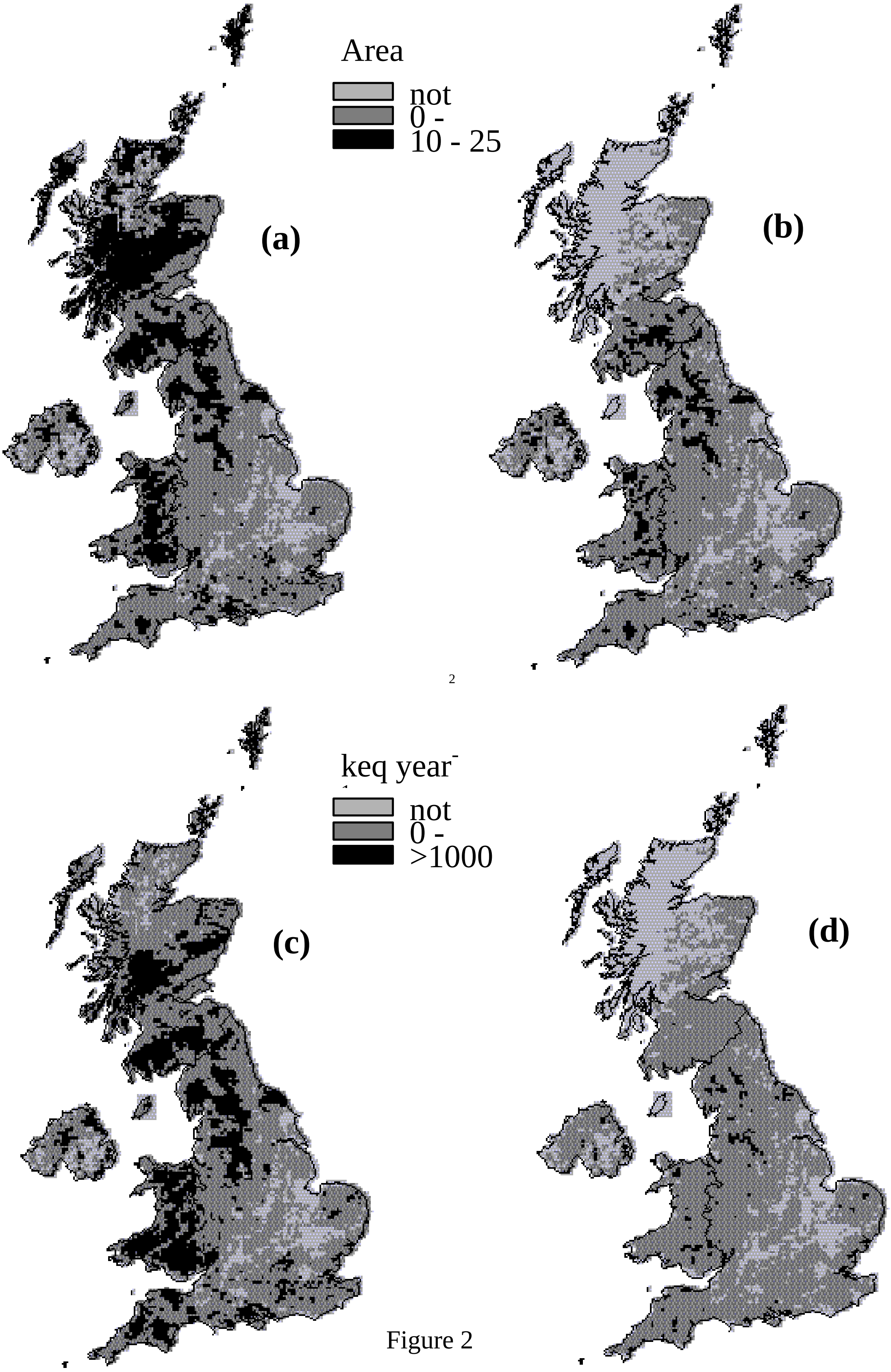


Figure 2

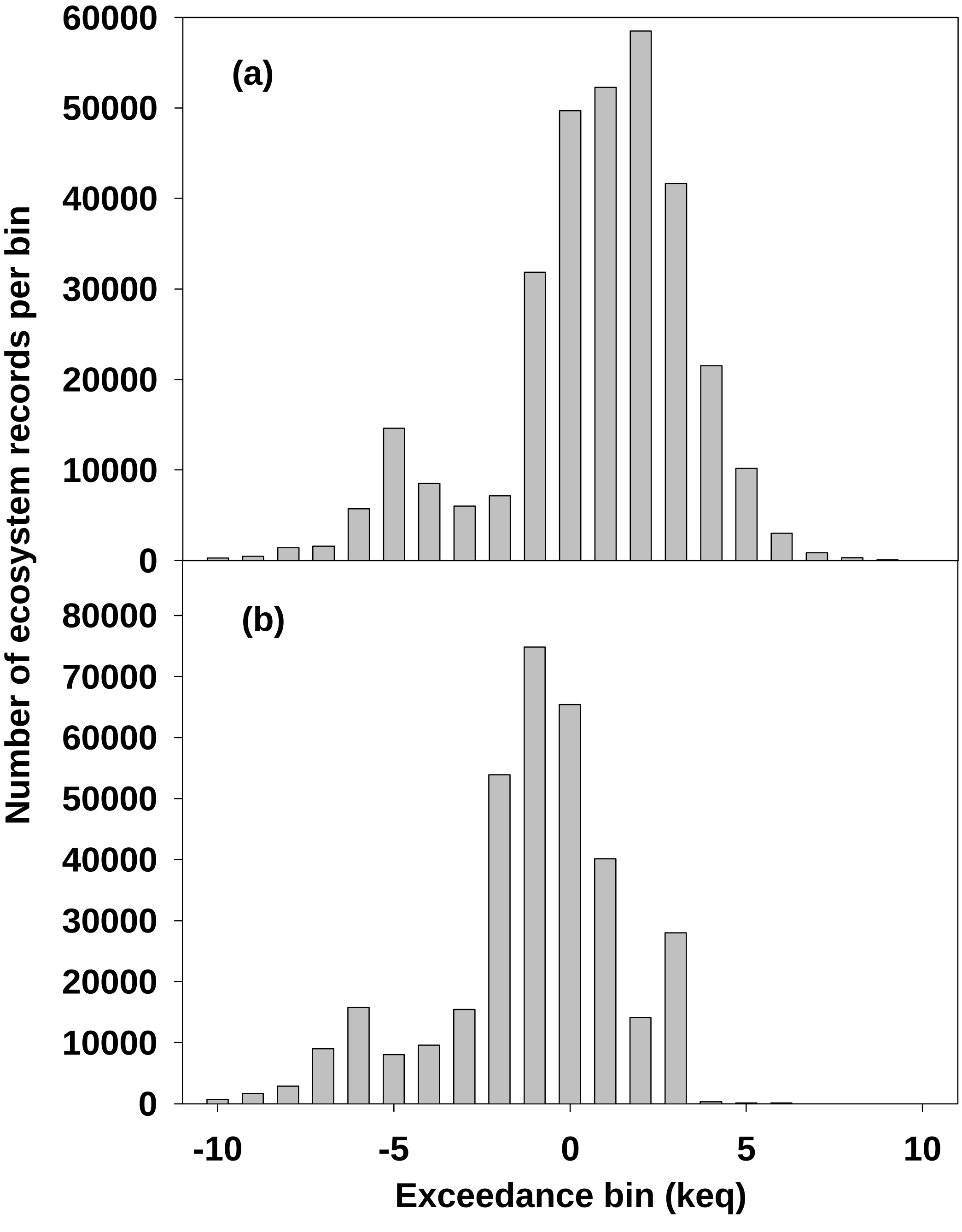

(a)
(b)
60000
50000
40000
30000
20000
10000
0
80000
70000
60000
50000
40000
30000
20000
10000
0
-10
-5
0
5
10
Number of ecosystem records per bin
Exceedance bin (keq)

Figure 3

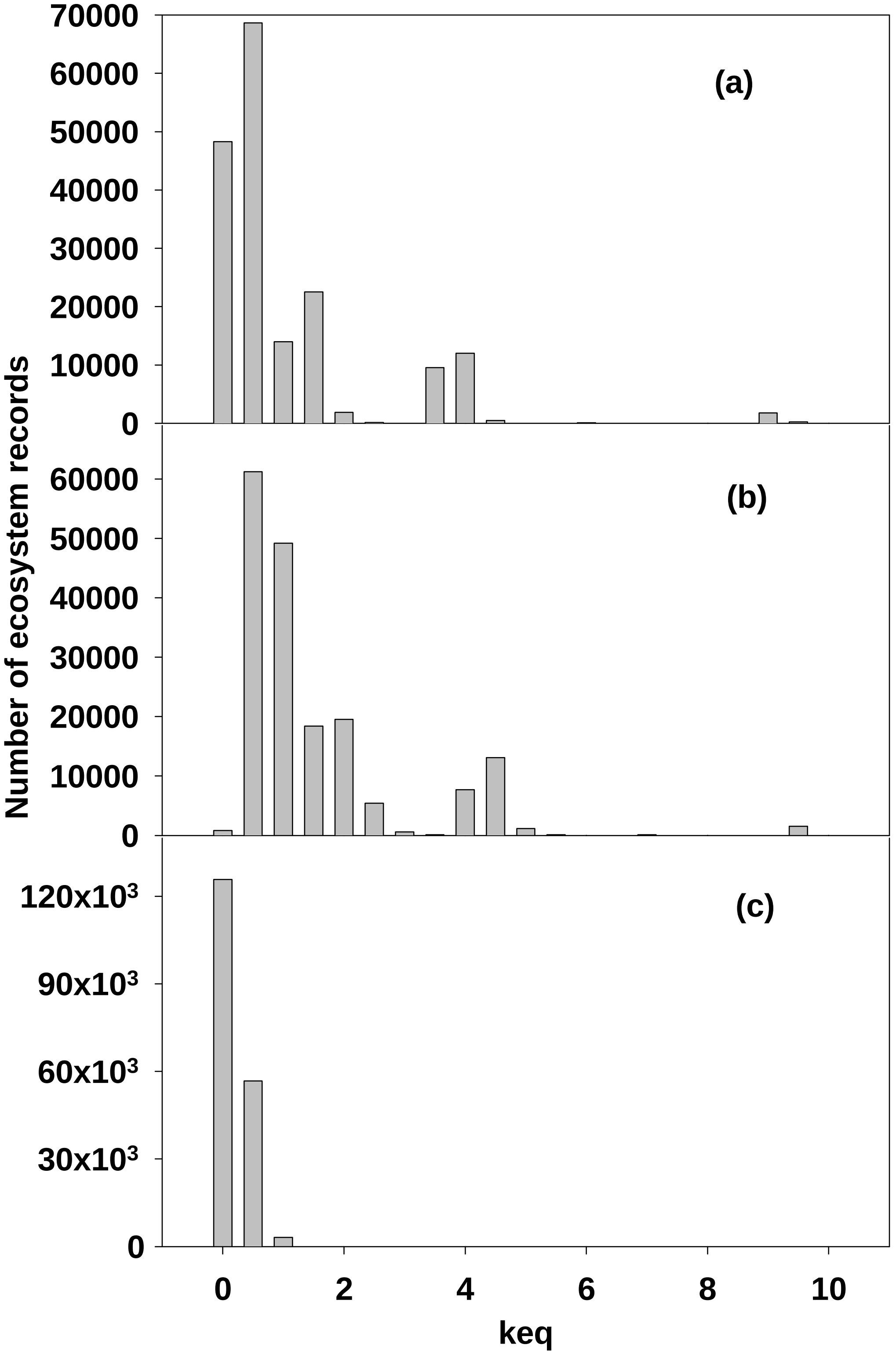

70000
60000
50000
40000
30000
20000
10000
0
(a)
60000
50000
40000
30000
20000
10000
0
(b)
120x10³
90x10³
60x10³
30x10³
0
(c)
0
2
4
6
8
10
keq
Number of ecosystem records

Figure 4

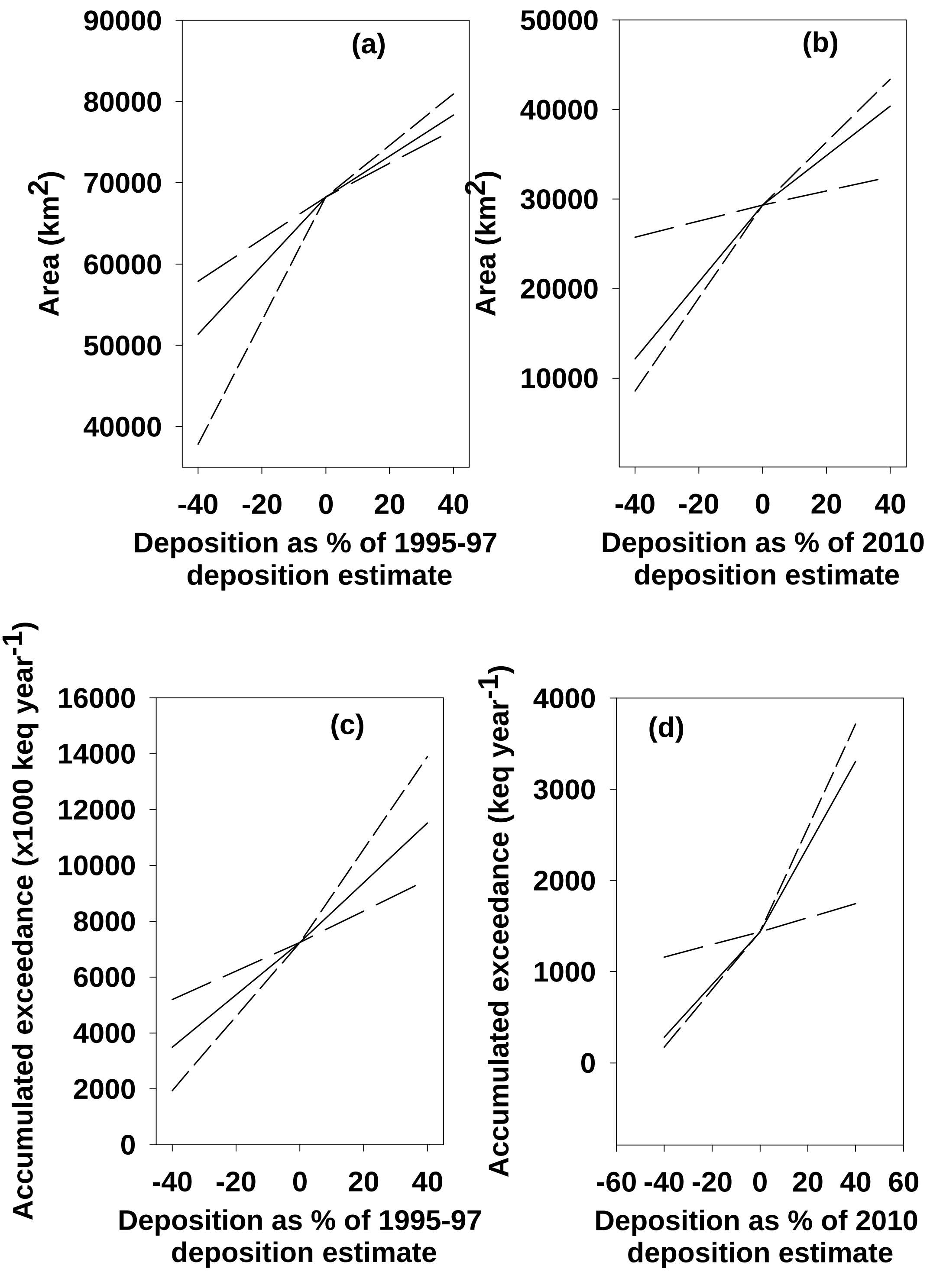
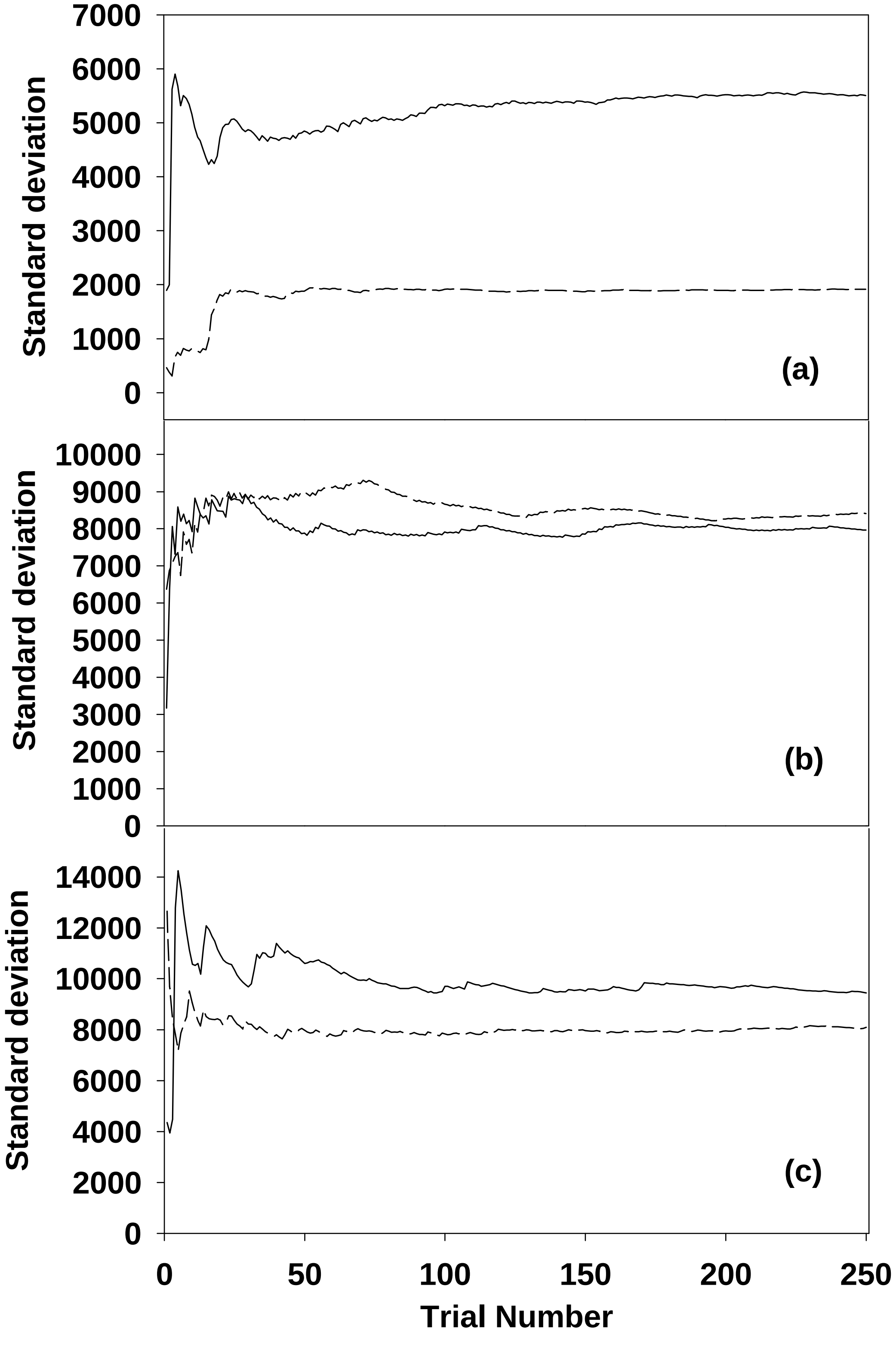
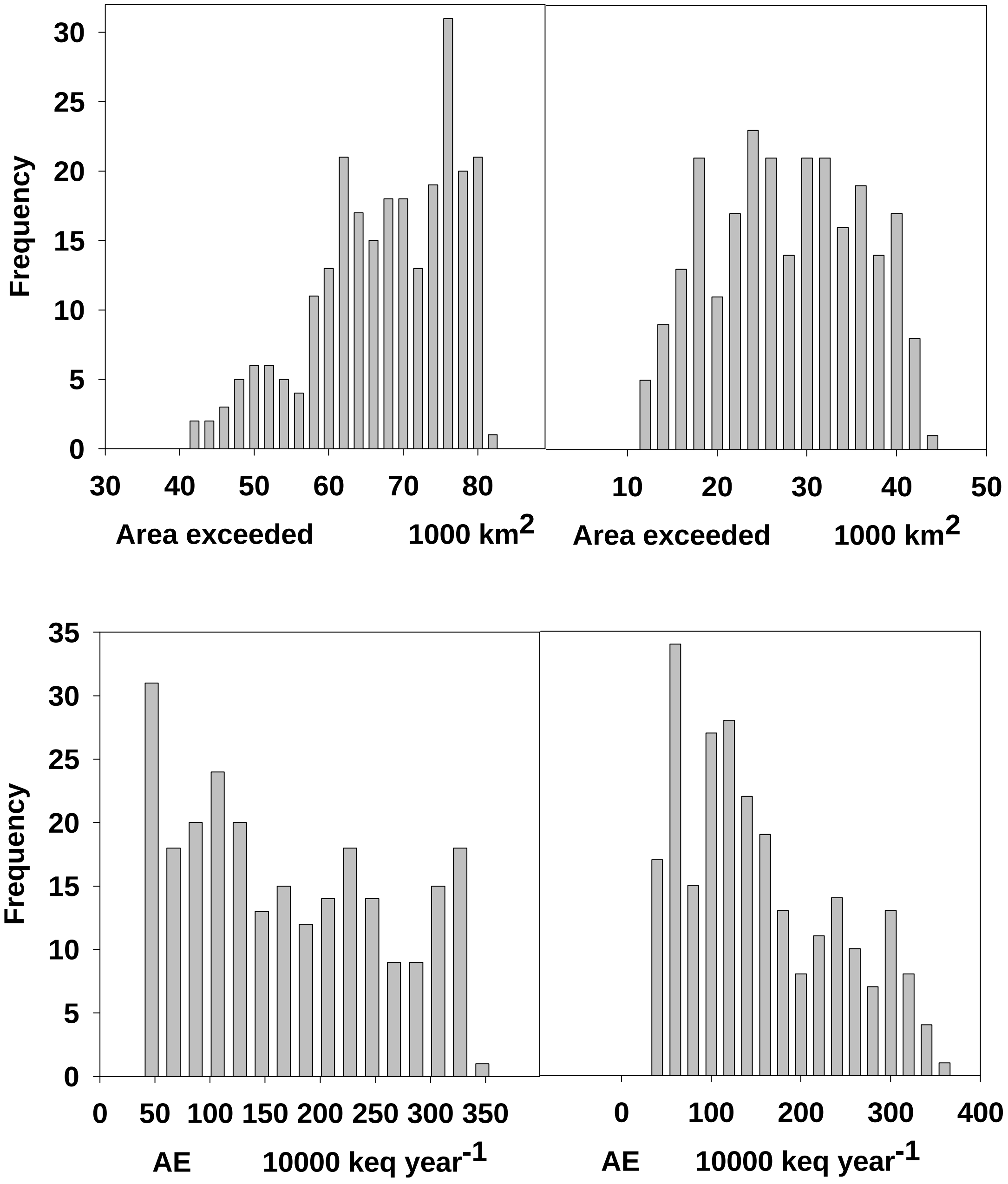

(a)
Area (km^2)
90000
80000
70000
60000
50000
40000
-40 -20 0 20 40
Deposition as % of 1995-97 deposition estimate
(b)
Area (km^2)
50000
40000
30000
20000
10000
-40 -20 0 20 40
Deposition as % of 2010 deposition estimate
(c)
Accumulated exceedance (x1000 keq year^{-1})
16000
14000
12000
10000
8000
6000
4000
2000
0
-40 -20 0 20 40
Deposition as % of 1995-97 deposition estimate
(d)
Accumulated exceedance (keq year^{-1})
4000
3000
2000
1000
0
-60 -40 -20 0 20 40 60
Deposition as % of 2010 deposition estimate

Figure 5



Standard deviation
7000
6000
5000
4000
3000
2000
1000
0
(a)
Standard deviation
10000
9000
8000
7000
6000
5000
4000
3000
2000
1000
0
(b)
Standard deviation
14000
12000
10000
8000
6000
4000
2000
0
(c)
0
50
100
150
200
250
Trial Number

Figure 6



Frequency
0
5
10
15
20
25
30
30
40
50
60
70
80
Area exceeded
1000 km^2
10
20
30
40
50
Area exceeded
1000 km^2
Frequency
0
5
10
15
20
25
30
35
0
50
100
150
200
250
300
350
AE
10000 keq year^{-1}
0
100
200
300
400
AE
10000 keq year^{-1}

Figure 7

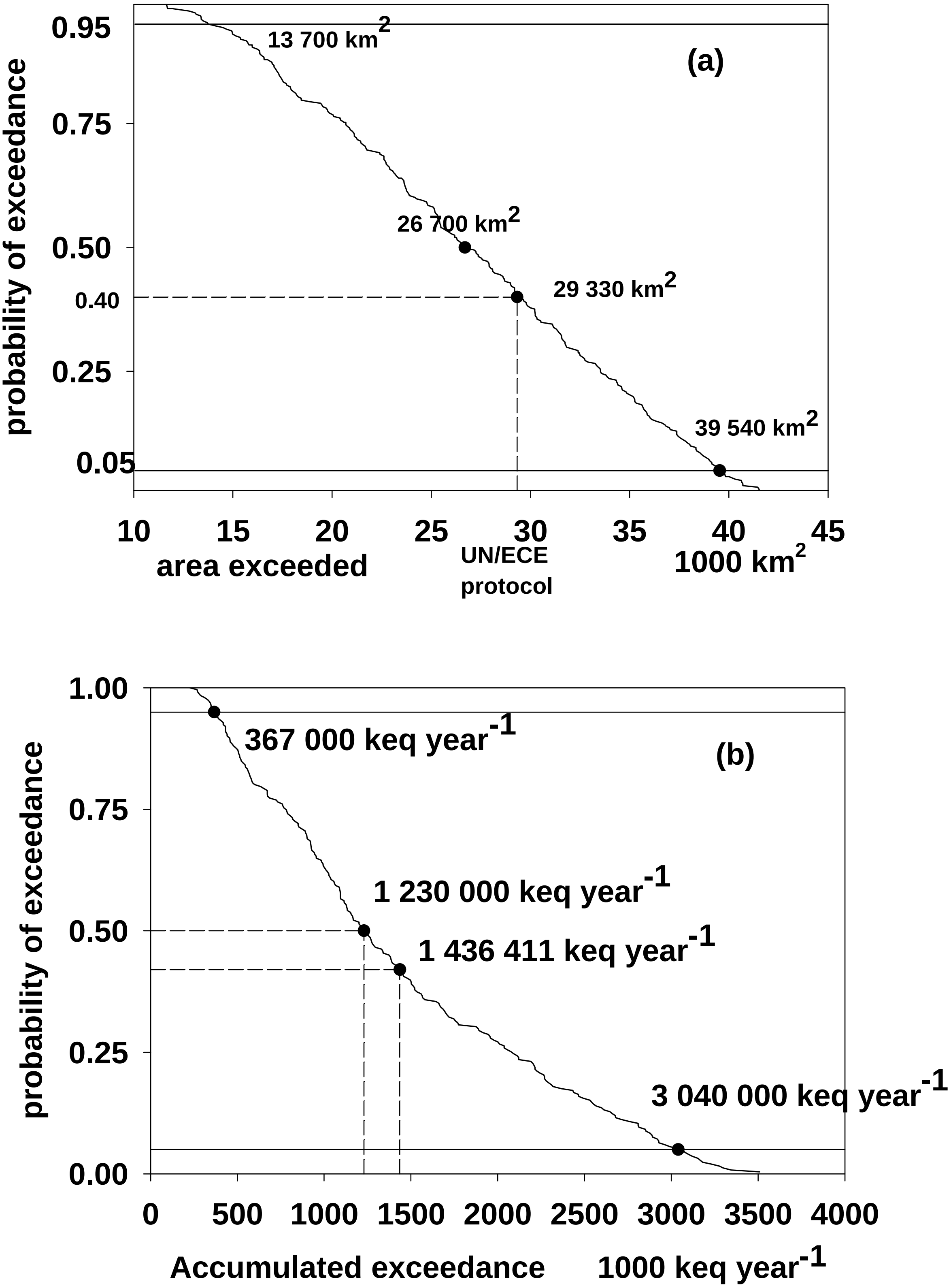

0.95
0.75
0.50
0.40
0.25
0.05
probability of exceedance
13 700 km^2
(a)
26 700 km^2
29 330 km^2
39 540 km^2
10
15
20
25
30
35
40
45
area exceeded
UN/ECE
protocol
1000 km^2
1.00
0.75
0.50
0.25
0.00
probability of exceedance
367 000 keq year^{-1}
(b)
1 230 000 keq year^{-1}
1 436 411 keq year^{-1}
3 040 000 keq year^{-1}
0
500
1000
1500
2000
2500
3000
3500
4000
Accumulated exceedance
1000 keq year^{-1}

Figure 8

TABLES

Table 1 Change in area of critical load exceedance and accumulated exceedance from the deterministic case for 1995-97 and 2010.

Table 2 Range of Monte Carlo area exceeded and accumulated exceedance results from 250 trials for 1995-97 and 2010.

Table 1

Simulation	Change in ecosystem area exceeded (km ^{2})		Change in AE (keq year $^{-1}$)	
	1995-97	2010	1995-97	2010
D1	8 224	3 165	2 246 559	309 284
D2	10 388	3 572	2 044 765	277 427
D3	10 080	11 071	4 276 278	1 867 068
D4	16 897	17 170	3 745 202	1 157 104
D5	12 676	14 065	6 660 613	2 277 535
D6	30 448	20 767	5 299 371	1 264 338

Table 2

Simulation	Range of area exceeded		Range of accumulated area exceeded	
	1995-97	2010	1995-97	2010
D7	17769	6614	4049159	578592
D8	26849	25673	7984705	2787779
D9	41859	32497	11253889	3281056