

1 An analysis of the likely success of policy actions under uncertainty:  
2 recovery from acidification across Great Britain

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14 *Environmental Science and Policy* (in press)

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17 ABSTRACT

18 In the context of wider debates about the role of uncertainty in environmental science and the  
19 development of environmental policy, we use a Generalised Likelihood Uncertainty Estimate (GLUE)  
20 approach to address the uncertainty in both acid deposition model predictions and in the sensitivity  
21 of the soils to assess the likely success of policy actions to reduce acid deposition damage across  
22 Great Britain. A subset of 11, 699 acid deposition model runs that adequately represented observed  
23 deposition data were used to provide acid deposition distributions for 2005 and 2020, following a  
24 substantial reduction in SO<sub>2</sub> and NO<sub>x</sub> emissions. Uncertain critical loads data for soils were then  
25 combined with these deposition data to derive estimates of the accumulated exceedance (AE) of  
26 critical loads for 2005 and 2020. For the more sensitive soils, the differences in accumulated  
27 exceedance between 2005 and 2020 were such that we could be sure that they were significant and  
28 a meaningful environmental improvement would result. For the least sensitive soils, critical loads  
29 were largely met by 2020, hence uncertainties in the differences in accumulated exceedance were of  
30 little policy relevance. Our approach of combining estimates of uncertainty in both a pollution model  
31 and an effects model, shows that even taking these combined uncertainties into account, policy-  
32 makers can be sure that the substantial planned reduction in acidic emissions will reduce critical  
33 loads exceedances. The use of accumulated exceedance as a relative measure of environmental  
34 protection provides additional information to policy makers in tackling this 'wicked problem'.

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36 *Keywords:* HARM, GLUE, uncertainty, critical loads, soil acidification

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## 40 **1. Introduction**

41 The many types of uncertainty that can affect policy making and how these can be presented to and  
42 then handled by policy makers, have become topics of increasing interest. Schneider and Kuntz-  
43 Duriseti (2002) considered uncertainty in climate change policy. They suggested that whilst one  
44 approach is to reduce (bound) the uncertainty by collecting more data, more understanding and  
45 building better models, the other approach is to reduce the effects of (manage) any uncertainty in  
46 understanding by taking it into account in policy making. This second approach can be traced back  
47 to ideas about ecosystem resilience and recovery after disturbance developed in the 1970s.

48 Refsgaard et al. (2007) in a review of uncertainty in the context of water management, suggested  
49 that uncertainty in its widest sense can usefully be regarded as the degree of confidence a decision  
50 maker has about possible outcomes and/or the probabilities of these outcomes. Uusitalo et al.  
51 (2015) suggested that uncertainty analysis can provide decision makers with a realistic picture of  
52 possible outcomes, in a context where results are going to be better or worse, not true or false, i.e.  
53 that environmental problems are 'wicked problems'. Whilst some types of uncertainty are  
54 unquantifiable, other types can be quantified through approaches such as sensitivity analysis, the  
55 use of multiple models and exploring the impact of parameter uncertainty. Here we take a  
56 quantitative approach to uncertainty in the context of recovery from the problem of acidification in  
57 Great Britain. We quantify and then combine the uncertainties in outputs from one acid deposition  
58 model and one measure of ecosystem health to assess whether current emissions reduction policies  
59 are likely to deliver ecosystem protection. We believe that this is the first effort to combine the  
60 uncertainties in both these elements in a single assessment.

61 European policymakers have been concerned about the acidification of sensitive soils and terrestrial  
62 ecosystems, driven by emissions of acidic species, sulphur dioxide (SO<sub>2</sub>) and nitrogen oxides (NO<sub>x</sub>)  
63 since the 1970s. These concerns have led to concerted policy actions within the United Nations  
64 Economic Commission for Europe (UN ECE) and the European Union (EU), designed to reduce

65 emissions and hence, the damaging deposition. The UN ECE agreed the Convention on Long-Range  
66 Transboundary Air Pollution (CLRTAP) in 1979 and has since promulgated a series of Protocols to the  
67 Convention, initially involving SO<sub>2</sub> and NO<sub>x</sub> separately and then combined with ammonia (NH<sub>3</sub>) under  
68 the Gothenburg Protocol (1999), referred to as the 'Multi-pollutant, Multi-effect Protocol'. A revision  
69 of the Gothenburg Protocol was agreed in 2012 (referred to here as RGP, see Amann et al., 2012;  
70 Reis et al., 2012). The EU has tackled the need to reduce emissions through a series of directives  
71 focussing initially on Large Combustion Plant (1988 and 2001), giving rise to the National Emission  
72 Ceilings Directive (NECD). In 2005, the EU put forward its Thematic Strategy on Air Pollution, Clean  
73 Air for Europe (CAFÉ) and under this framework is renegotiating the NECD with current  
74 commitments extending to 2029, with new commitments after 2030 (for an assessment of the NECD  
75 see Hettelingh et al., 2013a). Within these policy contexts, the chosen measure of ecosystem  
76 sensitivity was the critical load (CL) (Hettelingh et al., 1995), where the CL is the amount of  
77 deposition the chosen receptor can apparently tolerate without damage being likely (Bull, 1992).  
78 Where deposition was greater than (exceeded) the CL, damage was assumed to occur. CLs have  
79 been developed for a range of receptors (soils, freshwaters and a variety of terrestrial ecosystems)  
80 using a number of different methodologies (for the latest UK information see  
81 <http://www.cldm.ceh.ac.uk/>, for details of the most recent changes in methodology across Europe  
82 see Slootweg et al. 2015). It has been long recognised that there is variability between  
83 representations of CLs and that there are uncertainties in their calculation (see Zak et al., 1997), but  
84 CLs remain central to policymaking in this area and are an accepted risk assessment tool (Hettelingh  
85 et al., 2013b; Holmberg et al., 2013). The success of any emissions reduction policy is gauged by the  
86 resulting reduction in CL exceedance and system recovery (chemical and biological) (Posch et al.,  
87 2012), recognising that any system is unlikely to recover to exactly its pre-acidification state  
88 (Helliwell et al., 2014).

89 As it soon became evident that CLs would not be achievable across the whole of Europe in the  
90 foreseeable future, the concept of 'gap-closure' was adopted to formulate acid deposition policies

91 (see Amann et al., 2012 and the references therein). Gap closure implies reducing CL exceedance by  
92 a given fraction, say 50%, and then using integrated assessment modelling to find an equitable and  
93 fair distribution of emission reductions across the European countries to achieve the gap-closure  
94 target. Whilst this is a pragmatic approach, the approach cannot use meeting CLs as its optimisation  
95 target (and hence cannot guarantee complete ecosystem protection) and so a new index of  
96 environmental protection has been defined in terms of reducing 'accumulated exceedance' (AE)  
97 which captures both the magnitude and areal extent of exceedance. This index requires the  
98 combination of both CL and acid deposition data, both of which are uncertain.

99 The historical reductions in emissions across the EU-28 countries (by 87% for SO<sub>2</sub>, 54% for NO<sub>x</sub> and  
100 27% for NH<sub>3</sub> since 1990) (European Environment Agency (EEA), 2015) and measured decreases in  
101 deposition, have been reflected by measurable recovery in pH and acid neutralising capacity in many  
102 surface waters (Battarbee et al., 2014; Kernan et al., 2010) and reductions in CL exceedance (De Wit  
103 et al., 2015; RoTAP, 2012). Forward projections of current emission reduction commitments and the  
104 agreement of any additional reductions, however, depend on the application of atmospheric  
105 transport and deposition models, whose outputs can then be compared with CLs to assess the likely  
106 resulting environmental improvement (gains). Acid deposition models are uncertain because the  
107 parameterisations on which they are based and the input parameters that are fed into them, both  
108 contain simplifications and assumptions. CL are also uncertain, as described above. It is important,  
109 therefore, that policymakers have confidence in the outcomes of this modelling procedure  
110 (deposition and CL exceedance) given all the uncertainties inherent in both the atmospheric  
111 transport and CL models and can be assured that the higher costs of additional future emission  
112 reductions (assuming that the cheaper options have already been adopted) will actually increase  
113 protection of sensitive ecosystems and that recovery from acidification will continue. Two questions  
114 therefore arise: 1) can we really be sure that the emissions reductions proposed to reduce AE  
115 will produce discernible environmental improvement or will they be lost in uncertainty? and 2) does  
116 the change of approach from an absolute target (CL exceeded or not) to a relative one (based on

117 accumulated exceedance), change our perception of environmental improvement? Here we address  
118 both these questions. The concerns around the implications of scientific and model uncertainty for  
119 policy making that we address here in relation to acidification are relevant across a range of  
120 environmental issues.

121 We address our two questions about the impact of scientific uncertainty on achieving environmental  
122 protection, by exploring the impact of uncertainties in one atmospheric transport and deposition  
123 model, the Hull Acid Rain Model (HARM, Metcalfe et al., 2005) and one representation of CL (for  
124 soils), based on the Skokloster classification, by comparing estimates of accumulated exceedance of  
125 CL in 2005 and 2020 and assessing the likelihood of environmental protection across Great Britain  
126 (GB). This builds on an initial assessment of the impacts of uncertainty in HARM on CL exceedance  
127 across Wales reported by Heywood et al. (2006a). We provide a brief description of HARM and set  
128 out our approach to representing uncertainty in HARM and the CL for soils data set. We describe  
129 how we have combined estimates of deposition and sensitivity to acidification (CLs) to yield  
130 estimates of accumulated exceedance (AE) and how we have assessed the significance of the  
131 modelled changes. Our method is illustrated with reference to one 10 km x 10 km grid square in the  
132 Peak District in northern England, before going on to present and discuss the results for the whole of  
133 GB and consider the wider implications of this more rigorous approach for policy making.

## 134 **2. Methodology**

### 135 2.1 HARM and the GLUE framework

136 HARM is a receptor-orientated Lagrangian statistical model which is driven by emissions of SO<sub>2</sub>, NO<sub>x</sub>  
137 and NH<sub>3</sub> across the UK and the wider European area. Over a number of years, the model has been  
138 used to help in the formulation of acidification control policies in the UK. It provides estimates of  
139 wet and dry sulphur and nitrogen (both oxidised and reduced) depositions at 10 km x 10 km spatial  
140 resolution across the UK. Further details of the model are given elsewhere (Dore et al., 2015;

141 Metcalfe et al., 2005; Whyatt et al., 2007). Here, HARM has been run using 2005 emissions estimates  
142 for SO<sub>2</sub>, NO<sub>x</sub> and NH<sub>3</sub> sources within the UK and the rest of Europe. An illustrative, gap closure type,  
143 scenario was then applied to simulate a possible 2020 emission situation involving a 35% reduction  
144 in SO<sub>2</sub> emissions and a 33% reduction in NO<sub>x</sub> emissions (no reduction was applied to NH<sub>3</sub> emissions).  
145 This 2020 scenario was developed before the RGP was agreed, but is broadly consistent with the  
146 UK's current Gothenburg commitments (DEFRA, 2015). Our SO<sub>2</sub> emissions lie within the likely ranges  
147 for 2020, but our NO<sub>x</sub> emissions are a little high. It is also proposed that UK NH<sub>3</sub> emissions will  
148 decline by 2020, by around 12% from the figure used here. Because our results are likely to be  
149 influenced by the absolute magnitude of the deposition reduction as well as the spatial distribution  
150 of any reduction, our illustrative or hypothetical reduction should be within the bounds of current  
151 projections.

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153 Policymakers require that any model used for environmental policy formulation should reproduce  
154 real world behaviour adequately. In the present context, this means that an acid deposition model  
155 should reproduce the observed acid deposition fields (see for example Dore et al, 2015; Fagerli et al.,  
156 2003; NEG-TAP, 2001; RoTAP, 2012). However, any comparison of model results with observations is  
157 never perfect. Inevitably, there is likely to be good agreement for some sites or species and not with  
158 others. There are inadequacies and simplifications in the model together with site dependent factors  
159 influencing the observations. Here, the view is taken that it is difficult to find a set of model input  
160 parameters that uniquely fit the available observations. There may be a number of sets of  
161 parameters, or combinations of parameters that are 'acceptably' consistent with the available  
162 observations. This is known as equifinality (Beven, 2006) and results from the difficulty of deciding  
163 between competing parameter sets and models, given the limitation of the observations. Equifinality  
164 implies uncertainty and is the basis for our exploration of uncertainty within HARM. We have  
165 approached this by adopting the Generalised Likelihood Uncertainty Estimation (GLUE) framework.

166 In a previous study using HARM, Page et al. (2008) identified a subset of 11,699 HARM model runs  
167 that 'adequately' represented observed acid deposition data, allowing the production of deposition  
168 uncertainty distributions across the UK. This subset of 'acceptable' model parameter sets has been  
169 used in this study to provide distributions of deposition for 2005 and 2020. Details of the parameter  
170 set 'acceptance' criteria and the Monte Carlo parameter set sampling procedure are given in Page et  
171 al. (2008).

## 172 2.2 Critical loads for soils

173 Critical loads for soils were defined and estimated using the steady state mass balance method for  
174 GB (Hornung et al., 1995). CLs were assigned using the dominant soil type at a spatial scale of 1 km x  
175 1 km using the Skokloster categories Class 1 to Class 5 and their distribution across Great Britain (GB)  
176 is shown in Figure 1. Class 1 soils have the lowest buffering capacity (most sensitive) and were  
177 assigned CLs in the range 0 – 0.2 keq ha<sup>-1</sup> yr<sup>-1</sup>. Class 5 soils have the highest buffering capacity and  
178 were assigned CLs greater than 4.0 keq ha<sup>-1</sup> yr<sup>-1</sup>. Soils in Classes 2, 3 and 4 have intermediate levels  
179 of buffering capacity and had their range boundaries set at 0.5, 1.0 and 2.0 keq ha<sup>-1</sup> yr<sup>-1</sup>. Given the  
180 difference in spatial scale between the CL data (1 km x 1 km) and the HARM deposition data (10 km x  
181 10 km), the CL data were aggregated up to the scale of the HARM data, providing the total area for  
182 each Skokloster soil class within each 10 x 10 km grid cell. Aggregating up the CLs in this way does  
183 not change the underlying sensitivity, but masks the spatial distribution and location of the most  
184 sensitive elements within each square. This spatial distribution is only important if there are strong  
185 gradients in deposition within a particular grid square or the assessment of damage is required for a  
186 particular location. At the 10 km x 10 km scale such gradients were not significant and hence the  
187 aggregation process led to no significant loss of accuracy or bias in the CL exceedance.

188 In total, there were 1467 10 km x 10 km grid squares representing England, 258 for Wales and 1047  
189 for Scotland. No corresponding CL data were available for Northern Ireland and so this country was  
190 given no further consideration in this analysis. Here, the effects of incorporating uncertainties



191 associated with the Skokloster CL classifications into the calculation of CL exceedances has been  
192 studied for the 2772 grid squares covering GB, given the uncertain deposition estimates described  
193 above.

194 Uncertainties in the estimation of CLs were first addressed by Zak et al. (1997) who applied the GLUE  
195 approach to the PROFILE model, a steady state geochemical model that is widely used within the CL  
196 community. Heywood et al. (2006b) used coniferous woodland as an example and showed that  
197 uncertainties in GB CLs varied between 14 – 29%. In further work, Heywood et al. (2006c) reviewed  
198 uncertainties in CL assessments across Europe and established the need for a coordinated effort to  
199 characterise uncertainties in CLs. Skeffington et al. (2007) used Monte Carlo methods to obtain the  
200 output distributions of various CL parameters, having quantified the uncertainties in the input  
201 parameters to the CL models. They showed that estimates of the uncertainties in the CLs for acidity  
202 exhibited coefficients of variation which lay between 25 and 61%, across a range of catchments. On  
203 the basis of the uncertainties estimated by Heywood et al. (2006b) and Skeffington et al. (2007), we  
204 take the view that the uncertainties in actual CLs are likely to be smaller, or at most comparable to,  
205 the ranges in the Skokloster classes outlined above.

206 The uncertainty in the CLs within each 10 km x 10 km grid square was addressed by assigning the CL  
207 a probability distribution that was evenly distributed within the particular CL range, that is to say, a  
208 ‘top hat’ function was assumed, as shown in Figure 2. As there was no HARM model estimated CL  
209 exceedance of the least sensitive (Class 5) soils in either 2005 or 2020, they are not discussed in this  
210 paper.

### 211 2.3 Estimating critical loads exceedances and their uncertainties

212 The methodology employed in the estimation of the uncertain CL exceedances for soils is illustrated  
213 in Figure 2. It consisted of a loop over the 2772 GB grid cells. Within this loop, the 11,699 acceptable

214 HARM estimates of total acid deposition for each 10km grid cell were overlaid onto the CL ranges for  
215 each soil class to estimate CL exceedances, as follows:

216  $CL \text{ exceedance (keq ha}^{-1} \text{ yr}^{-1}) = \text{acid deposition load (in keq ha}^{-1} \text{ yr}^{-1}) - CL \text{ (in keq ha}^{-1} \text{ yr}^{-1})$ .

217 The accumulated exceedance (AE) of the CLs in a given grid square was calculated using:

218  $\text{Accumulated Exceedance (keq yr}^{-1}) = CL \text{ exceedance} \times \text{area exceeded}$

219 and summing this over all the soil classes in a given grid square. This calculation was repeated for  
220 each of the soil classes and each of the 10 km x 10 km grid squares.

221 This methodology was then repeated using the 11,699 HARM deposition estimates for the 2020  
222 emission scenario. For each soil class and grid square, the differences in AE (2005 – 2020) were  
223 calculated: these differences were calculated by pairing up the 11,699 HARM estimates for 2005 and  
224 2020 and not drawing them at random from the sets of model runs. The differences in AE were then  
225 ranked in order and the 5<sup>th</sup>-, 25<sup>th</sup>-, 50<sup>th</sup>-, 75<sup>th</sup>- and 95<sup>th</sup>-percentiles were determined for the  
226 distributions of the 11,699 ‘acceptable’ results.

### 227 **3. Estimating 2005 – 2020 differences in critical load exceedance in the Peak District**

228 To illustrate the application of the methodology in Figure 2, attention is turned to a single 10 km x 10  
229 km grid square located in the Peak District National Park, in northern England (see inset Figure 1).

230 Class 1 soils occupied 25% of the surface area of this grid square, Class 2 14%, Class 3 22% and Class  
231 4 25%. Total HARM acid deposition declined from  $1.29^{+0.59}_{-0.40} \text{ keq ha}^{-1} \text{ yr}^{-1}$  (where the quoted  
232 uncertainty range is the 5% - 95% range, equivalent to the  $2 - \sigma$  confidence interval) in 2005 to  $0.93$   
233  $^{+0.39}_{-0.29} \text{ keq ha}^{-1} \text{ yr}^{-1}$  in 2020, giving a reduction in acid deposition of  $0.36^{+0.30}_{-0.11} \text{ keq ha}^{-1} \text{ yr}^{-1}$ .

234 The probability distribution of the HARM model estimates of the difference in AE per class is  
235 illustrated as a box-and-whisker plot in Figure 3. Looking first at the Class 1 (most sensitive) soils, all  
236 11,699 model runs for both 2005 and 2020 gave deposition estimates that exceeded the CL for Class

237 1 soils. The 2005 – 2020 difference in AE for Class 1 soils was found to be  $895^{+493}_{-290}$  keq yr<sup>-1</sup>. On this  
238 basis, the 5% - 95% confidence interval was narrow enough not to encompass zero and it could be  
239 concluded that the difference in AE was statistically significantly different from zero, despite the  
240 uncertainties in the deposition and CLs. However, in Figure 3, it can be seen that the 2 –  $\sigma$   
241 confidence interval was not exactly symmetrical about the 50-percentile value. This lack of  
242 symmetry implies a degree of skewness in the distribution of the differences in the AEs. Statements  
243 about statistical significance based on the assumption of a normal distribution may not be reliable if  
244 there is a high degree of skew. However, on a cautionary basis, if the range between the 50-  
245 percentile and the upper confidence limit was applied at the lower confidence interval, then the 5% -  
246 95% range would still not encompass zero. It was thus concluded that the difference in AE was likely  
247 to be robust, despite the apparent skewness in its probability distribution and the uncertainties in  
248 the deposition and CLs.

249 The deposition loads exceeded the CLs for Class 2 soils in all HARM model runs in both 2005 and  
250 2020. The AE for Class 2 soils was  $1297^{+600}_{-442}$  keq yr<sup>-1</sup> in 2005 and  $795^{+500}_{-300}$  keq yr<sup>-1</sup> in 2020, with a  
251 difference in AE of  $501^{+276}_{-162}$  keq yr<sup>-1</sup>. Since the 2 –  $\sigma$  confidence interval did not encompass zero, it  
252 was concluded that this difference was statistically significant, taking into account the apparent  
253 skewness in its probability distribution. The situation was much the same for Class 3 soils, where the  
254 2005 – 2020 difference in AE was found to be  $763^{+458}_{-394}$  keq yr<sup>-1</sup>, see Figure 3, and again this  
255 difference was considered to be significantly different from zero.

256 Looking at the least sensitive Class 4 soils, all 11,699 model runs gave deposition estimates that  
257 exceeded the CL in 2005, but 75% of the model runs met critical loads in 2020. The 2005 – 2020  
258 difference in AE was found to be  $84^{+511}_{-84}$  keq yr<sup>-1</sup>. The skewness in the distribution for the Class 4  
259 soils is clearly apparent in Figure 3. Uncertainties were so large for the Class 4 soils that they  
260 encompassed zero and so it was unlikely that they could be considered significant because of the  
261 combined uncertainties in the deposition and CLs. We therefore have the situation where in one

262 10km grid square, the most sensitive soils show a large and statistically significant reduction in AE  
263 whereas the least sensitive soils show a small reduction, which is not significant. This contradicts our  
264 conventional notion of environmental protection that if you protect the most sensitive elements in  
265 the environment from damage, then you automatically protect the least sensitive. However,  
266 because CLs were actually met for Class 4 soils in three cases out of four, the small difference in AE  
267 and its lack of statistical significance would not be relevant in policy terms.

#### 268 **4. Estimating 2005 – 2020 differences in critical loads exceedance across GB**

269 The methodology illustrated in Figure 2 was then followed for each of the 2772 10 km x 10 km grid  
270 squares across GB. We found that the differences in AE between 2005 and 2020 for all soil classes (1  
271 – 4) showed that the reductions in emissions in our initial scenario reduced CL exceedances  
272 throughout GB. This implies that non-linearities in the relationship between acid deposition and CL  
273 exceedance were unimportant on the GB scale. This is a reflection of the illustrative emission  
274 reduction scenario chosen, where there was no reduction in the emissions of NH<sub>3</sub> across the UK and  
275 very limited (4%) reduction across the rest of the EMEP area, hence, non-linearities in relation to the  
276 response of S and oxidised N to changes in the emission of NH<sub>x</sub> were minimised.

277 The 2005 – 2020 difference in total AE for Class 1 soils was 354,000 <sup>+145,000</sup><sub>-104,000</sub> keq yr<sup>-1</sup> (see Table 1)

278 The probability distribution of the AE differences is shown as a box-and-whisker plot in Figure 4 and  
279 a 2 – σ confidence range did not encompass zero. Despite the uncertainties in the deposition loads  
280 and CLs, this difference in AE was statistically significant. The spatial distribution in the 50-percentile  
281 reductions in AE for the individual grid squares is shown in Figure 5a. The greatest reductions were  
282 found in southern England, Wales, East Anglia, northern England and in a few scattered locations in  
283 south west Scotland and in the highlands and islands. The 2 – σ ranges in the differences in AE for  
284 the individual grid squares were not evenly distributed about their 50-percentile values. The  
285 dispersion in the AEs about their 50-percentiles showed evidence of skewness, with shorter tails to  
286 low values and longer tails to high values (Figure 4). However, as with the Peak District grid square,

287 this dispersion differed only slightly from that shown by a 'normal' distribution. Consequently, a null  
288 hypothesis that the AE reductions were due to chance could be rejected with a high level of  
289 confidence. On this basis, it was concluded that the reductions in the AEs for Class 1 soils were all  
290 highly significant at the 99.99% level, despite the large uncertainties in the deposition loads and CLs.  
291 Although the changes for this soil class were small (Figure 4) they are likely to be important for these  
292 most acid sensitive environments. There were a small number of grid squares, on the fringes of GB,  
293 where it was difficult to make any robust statement about the policy significance of any reduction in  
294 AE because of severe skewness.

295 The difference in Total AE for Class 2 soils across GB was  $1,275,000^{+460,000}_{-375,000}$  keq yr<sup>-1</sup>, see Table 1  
296 and Figure 4, between 3 – 4 times higher than for Class 1 soils. Again, the 2 –  $\sigma$  confidence range did  
297 not encompass zero and so this difference was highly statistically significant. Although CL  
298 exceedances were generally higher for Class 1 soils, the areas assigned to Class 2 soils were much  
299 larger and so the total AE difference across GB was substantially higher for the latter. Figure 5b  
300 shows the spatial distribution of the 50-percentile AE differences for Class 2 soils for each grid  
301 square. The greatest reductions in AE were found in Wales, Cumbria, south west Scotland and across  
302 the Scottish Highlands. Although the distributions in the AE differences were skewed, the degree of  
303 skewness was considerably less than for Class 1 soils (Figure 4). It was concluded that the reductions  
304 in the AEs for Class 2 soils were all highly significant at the 99.99% level, despite the large  
305 uncertainties in the deposition and CLs. Skewness was a real problem in less than 3% of grid squares,  
306 the bulk of these in the Outer Hebrides. It is difficult to make any robust statement about the  
307 environmental significance of the AE reduction in these locations.

308 The difference in total AE across GB for Class 3 soils was  $1,010,000^{+780,000}_{-565,000}$  keq yr<sup>-1</sup>, see Table 1  
309 and Figure 4. This AE difference was somewhat smaller than for Class 2 soils despite their  
310 substantially larger areal coverage because of their lower CL exceedances. Although the 2 –  $\sigma$   
311 confidence interval did not encompass zero, there was noticeable skewness in the distribution of AE

312 differences. As discussed above, statements about significance may not be reliable if there is a large  
313 amount of skewness. However, as with the Peak District grid square, if the 50-percentile – 95-  
314 percentile range was applied at the lower confidence interval, then the adjusted 5-percentile – 95-  
315 percentile range would still not encompass zero. It was concluded that the difference in total AE was  
316 likely to be robust, despite the uncertainties in the deposition and CLs. Figure 5c shows the spatial  
317 distribution of the 50-percentile differences for the individual grid squares containing Class 3 soils.  
318 The largest reductions were found throughout southern and south west England, south Wales and a  
319 band from the west Midlands and into north west England. In all these regions, the reductions were  
320 likely to be highly significant. However in the regions where the reductions were much smaller and  
321 close to zero, skewness was again a real, issue. In ~ 25% of the grid squares, it was considered likely  
322 that the reductions in AE were not significant. This resulted from the situation where CLs and  
323 deposition loads were comparable in magnitude so the combination of uncertainties has become  
324 overwhelming in the estimation of these small AEs.

325 The difference in total AE across GB for Class 4 soils was found to be  $42,000^{+275,000}_{-41,000}$  keq yr<sup>-1</sup>, see  
326 Table 1 and Figure 4. The spatial distribution of the 50-percentile differences for the individual grid  
327 squares containing Class 4 soils is shown in Figure 5d. The difference in AE is small and highly  
328 uncertain (the 2-σ confidence range encompasses zero) compared with the above same values for  
329 Class 1 – 3 soils. Deposition and CLs were closely comparable in magnitude and so the uncertainties  
330 in these quantities have been magnified in the estimation of AE differences to the extent that AE and  
331 its differences have become unreliable indicators of ecosystem status for Class 4 soils. Given the  
332 relative insensitivity of this class of soils to acidification it is, however, quite feasible that the 2020  
333 scenario would deliver ecosystem protection.

## 334 **5. Discussion and Conclusions**

335 In the Introduction, we posed two policy related questions: The first question was if the current  
336 models and the current CL approaches are too uncertain to identify whether proposed emissions

337 reductions will deliver discernible environmental improvement; the second question concerned the  
338 impact of the change in the optimisation target from CL exceedance to accumulated exceedance.  
339 We have applied the GLUE methodology to address the uncertainties in deposition models and in  
340 the CLs. We have then developed a realistic hypothetical scenario for 2020 and quantified the  
341 uncertainties in the estimates of the differences in AE between 2005 and 2020. The 2- $\sigma$  confidence  
342 limits for the AE difference for Class 1 – 3 soils in the vast majority of GB locations do not encompass  
343 zero (see Figure 4) and so are likely to be statistically significant. In relation to question one, we can  
344 therefore say with some confidence that reductions in emissions of the order of 35% will lead to  
345 reductions in AE which are not ‘lost in the noise’ in the deposition and CL modelling. These findings  
346 are consistent with those of other studies for the UK (Helliwell et al., 2014; Majeko et al., 2009;  
347 Oxley et al., 2013;) using a range of modelling approaches. It is notable, however, that only the  
348 Helliwell et al. study (using the MAGIC model) attempted to include uncertainty in their assessment,  
349 primarily in relation to model inputs (parametric uncertainty). Far from being too uncertain for  
350 policy use, we have been able to make a first attempt at quantifying uncertainties in both  
351 deposition and CL at the GB scale and to demonstrate that the uncertainties are small enough that  
352 they can be employed to develop robust policy assessments. To follow on from Uusitalo et al. (2015,  
353 see Introduction) we can use this approach to give policy makers a more realistic picture of possible  
354 outcomes in tackling this particular ‘wicked problem’.

355 The second question concerned the impact of the change in environmental target from simple CL  
356 exceedance (or not), to an index of success represented by AE. Using the standard CL approach, with  
357 a single value applied to a deposition grid cell, the degree of protection was assessed only on a true  
358 or false basis (see Introduction). If the outcome of running a future emissions scenario was false (ie  
359 CL was still exceeded), policy makers were left with the impression that the proposed emissions  
360 reductions would fail to deliver environmental protection. In contrast, using the AE index gives a  
361 broader measure of better or worse relative to the starting situation, even if CL are not met  
362 completely. In our 2020 scenario, based on our 11,699 model runs, CLs for Class 4 soils would be

363 met 98% of the time. For Class 3 soils this declined to 67%, for Class 2 soils to 27% and for Class 1  
364 soils (most sensitive) to slightly less than 1% (fewer than 116 runs of the 11,699). Only on the most  
365 extreme deposition and CL uncertainty outcomes would Class 1 and 2 soils be protected. This  
366 suggests that emissions reductions in line with current commitments would do little to protect the  
367 most acid sensitive environments across GB (see Table 1). A simple estimate of the magnitude of  
368 emission reduction needed to provide full protection (based on extrapolation from the 2020 results)  
369 indicated that an emission reduction of around 45% would be needed to protect Class 4 soils  
370 completely (compared with 35% in our 2020 scenario) and of around 85% for Class 3 soils. Only very  
371 extreme (and probably impractical) reductions would offer protection to the most sensitive soils  
372 (Class 1). The change of optimisation target from meeting CL to the use of AE has, however, allowed  
373 us to make progress in terms of policy assessment for the most sensitive soils in the face of  
374 uncertainties in deposition models and the CLs themselves.

375 As the science in deposition modelling and CL assessments develops, there should be a narrowing  
376 (bounding) of uncertainties (see Introduction) and this should lead to a narrowing of the  
377 uncertainties in the emission reductions required to meet critical loads for Class 1 soils. There are  
378 reasons to suppose that some deposition estimates for GB have been overestimated (Dore et al.,  
379 2015; see Hall and Smith 2015 for a specific example) and so our conclusions may well have  
380 underestimated the likely improvement in environmental protection afforded by our initial  
381 hypothetical emission scenario. It could be, however, that current emissions reduction targets will  
382 never be able to protect the most acid sensitive environments and that the recovery of both aquatic  
383 and terrestrial ecosystems could take decades, in spite of the marked decrease in exceedance since  
384 the peak in the 1970s and 1980s (De Wit et al., 2015).

385 The importance of both considering and communicating uncertainty has come to the fore recently  
386 because of the debate around this issue in relation to anthropogenic climate change. The idea that a  
387 quantitative approach to uncertainty should be incorporated into environmental policy making has,



388 however, been around for more than 20 years (see Frey, 1992 in relation to the US EPA). As Cooke  
389 (2015) observes ‘There are formidable pitfalls when reasoning under uncertainty, into which both  
390 the scientific community and the general population repeatedly fall’ (p. 8), but there is no doubt that  
391 handling uncertainty in its various forms is now a key part of developing environmental policy in a  
392 variety of domains, as was suggested by Schneider and Kuntz-Duriseti (2002). We have set out one  
393 approach to achieving this, focusing on the implications of taking uncertainty into account in  
394 controlling emissions of acidifying pollutants. It should certainly play a part in developing strategies  
395 for policy initiatives such as the latest iteration of the Convention on Long-range Transboundary Air  
396 Pollution (Gothenburg Protocol, see Introduction) as it attempts to provide the scientific basis and  
397 an effects based approach to addressing a widening range of atmospheric pollutant issues and their  
398 interactions with climate change and biodiversity (UNECE, 2016). The point of this study was to show  
399 how uncertainties could be handled rather than to make a formal assessment of acid deposition  
400 policies, but it is evident that in this case, as in others, uncertainty cannot be used as a reason to  
401 limit action (Drouet et al., 2015).

#### 402 **Acknowledgements**

403 Original development of HARM was supported by UK Department of Environment, Food and Rural  
404 Affairs (DEFRA) and the UK Environment Agency.

405

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515

516 FIGURES

- 517 • Figure 1 – Single Column
- 518 • Figure 2 – Double Column (for legibility)
- 519 • Figure 3 – Single Column
- 520 • Figure 4 – Single Column
- 521 • Figure 5 – Double Column (4 maps)

522 Figure 1. Critical loads in  $\text{keq ha}^{-1} \text{yr}^{-1}$  for the dominant soil type at a spatial scale of 10 km x 10 km  
523 for Great Britain using the Skokloster categories Class 1 (most sensitive: in black) to Class 5 (least  
524 sensitive: in blue) estimated using the steady state mass balance method (Hornung et al., 1995).  
525 Inset shows detail for Peak District grid square.

526 Figure 2. A sketch illustrating the methodology adopted for the estimation of the  
527 frequency distributions of the differences in accumulated critical loads exceedance in a  
528 given 10km grid square between 2005 and 2020. The upper plots show the CL ranges for  
529 individual soil classes as coloured bars, a) Class 1, b) Class 2, c) Class 3, d) Class 4. The  
530 divisions within these bars indicate sampling within these ranges. The upper middle plots  
531 show accumulated exceedance for each individual soil class under the 2005 (in black) and  
532 2020 (in blue) scenarios. The lower middle plots show the difference (reduction) in  
533 accumulated exceedance for each individual soil class between 2005 and 2020. The  
534 bottom plot (e) shows accumulated exceedance for all soil classes under the 2005 (black)  
535 and 2020 (blue) scenarios.

536 Figure 3. Box-and-whisker plots of the dispersion in the estimates of the reductions in  
537 accumulated exceedance between 2005 and 2020 for each soil class in the Peak District  
538 grid cell.

539

540 Figure 4. Box-and-whisker plots of the dispersion in the estimates of the reductions in

541 accumulated exceedance between 2005 and 2020 for each soil class across GB.

542

543 Figure 5. Spatial variations in the 50-percentile points of the distribution of the estimates of the  
544 reduction in accumulated CL exceedance between 2005 and 2020 for a) Class 1 soils, b) Class 2 soils,  
545 c) Class 3 soils and d) Class 4 soils.

546

547 TABLES

548 Table 1. Percentile points in the reduction in AE between 2005 and 2020 for each Skokloster soil  
549 class across GB in  $\text{keq yr}^{-1}$ .

550 Table 1.

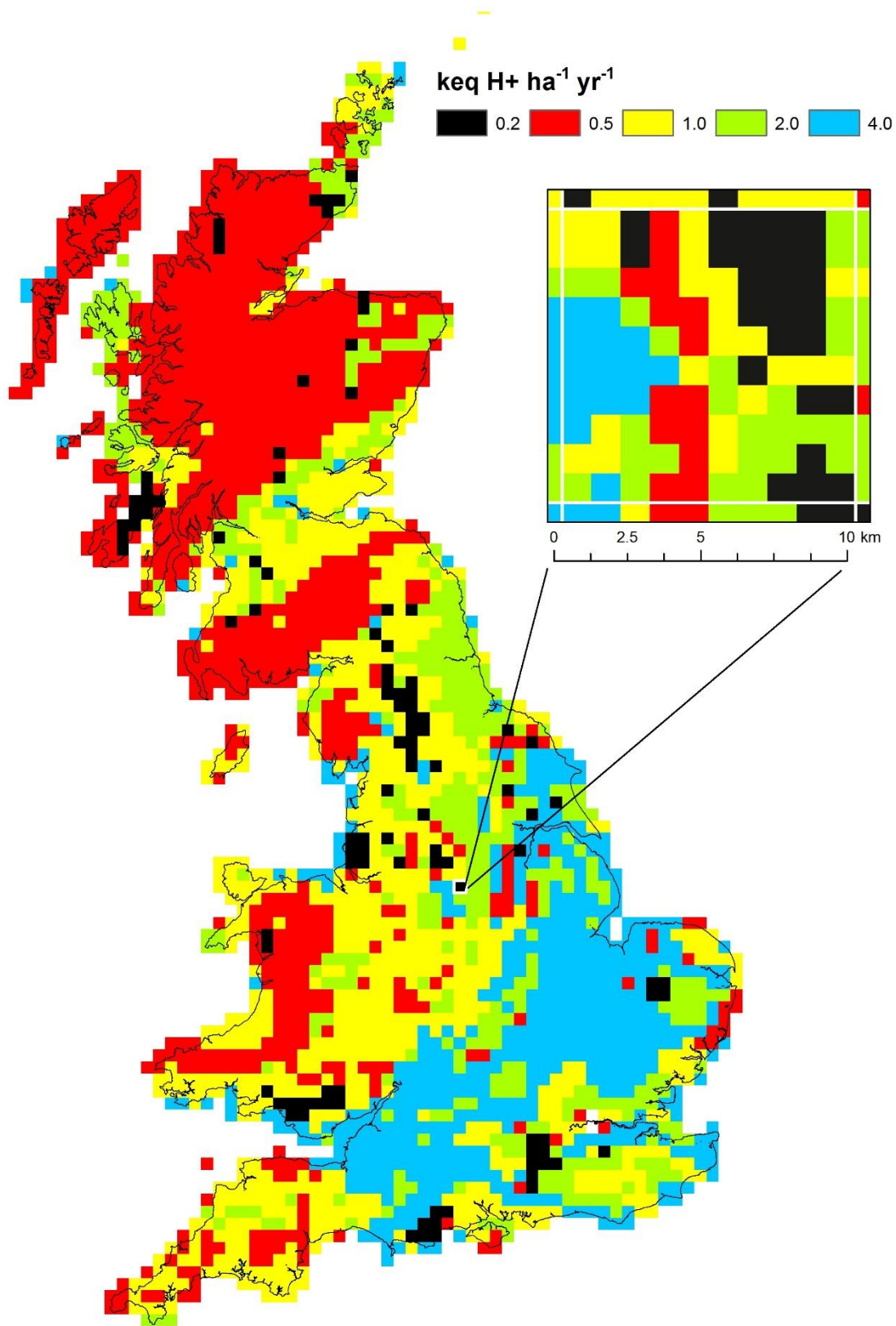
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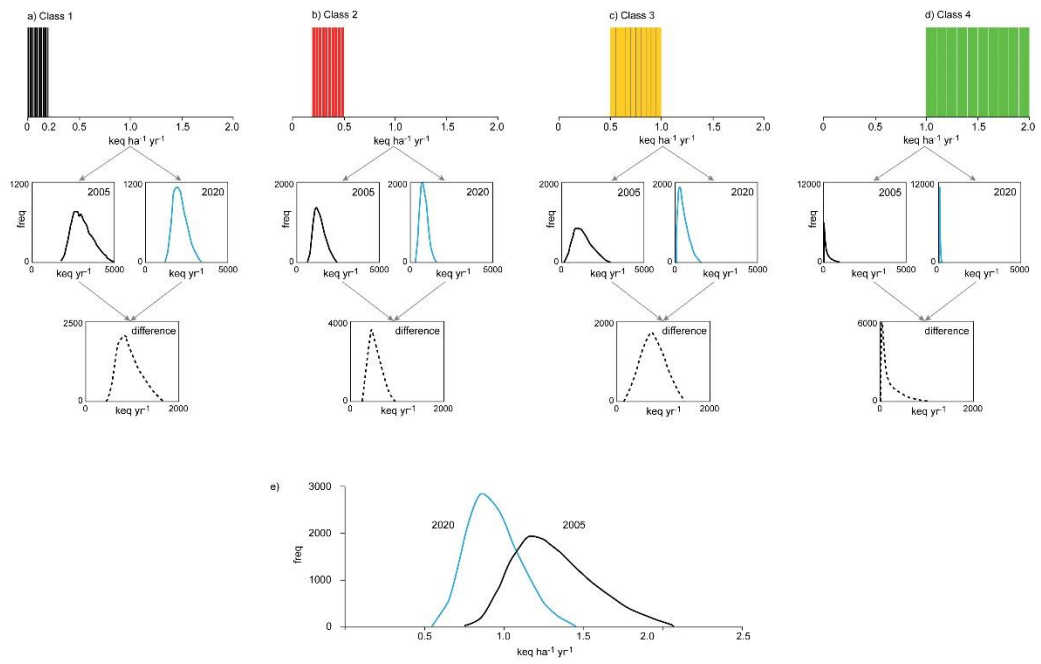
<b>Percentile</b>	<b>Class 1</b>	<b>Class 2</b>	<b>Class 3</b>	<b>Class 4</b>	<b>All classes</b>
<b>5%-ile</b>	250,000	900,000	445,000	1,000	1,596,000
<b>16%-ile</b>	283,000	1,030,000	620,000	6,000	1,939,000
<b>25%-ile</b>	303,000	1,100,000	725,000	12,000	2,140,000
<b>50%-ile</b>	354,000	1,275,000	1,010,000	42,000	2,681,000
<b>75%-ile</b>	415,000	1,465,000	1,345,000	111,000	3,336,000
<b>84%-ile</b>	445,000	1,565,000	1,515,000	167,000	3,692,000
<b>95%-ile</b>	499,000	1,735,000	1,790,000	317,000	4,341,000

552

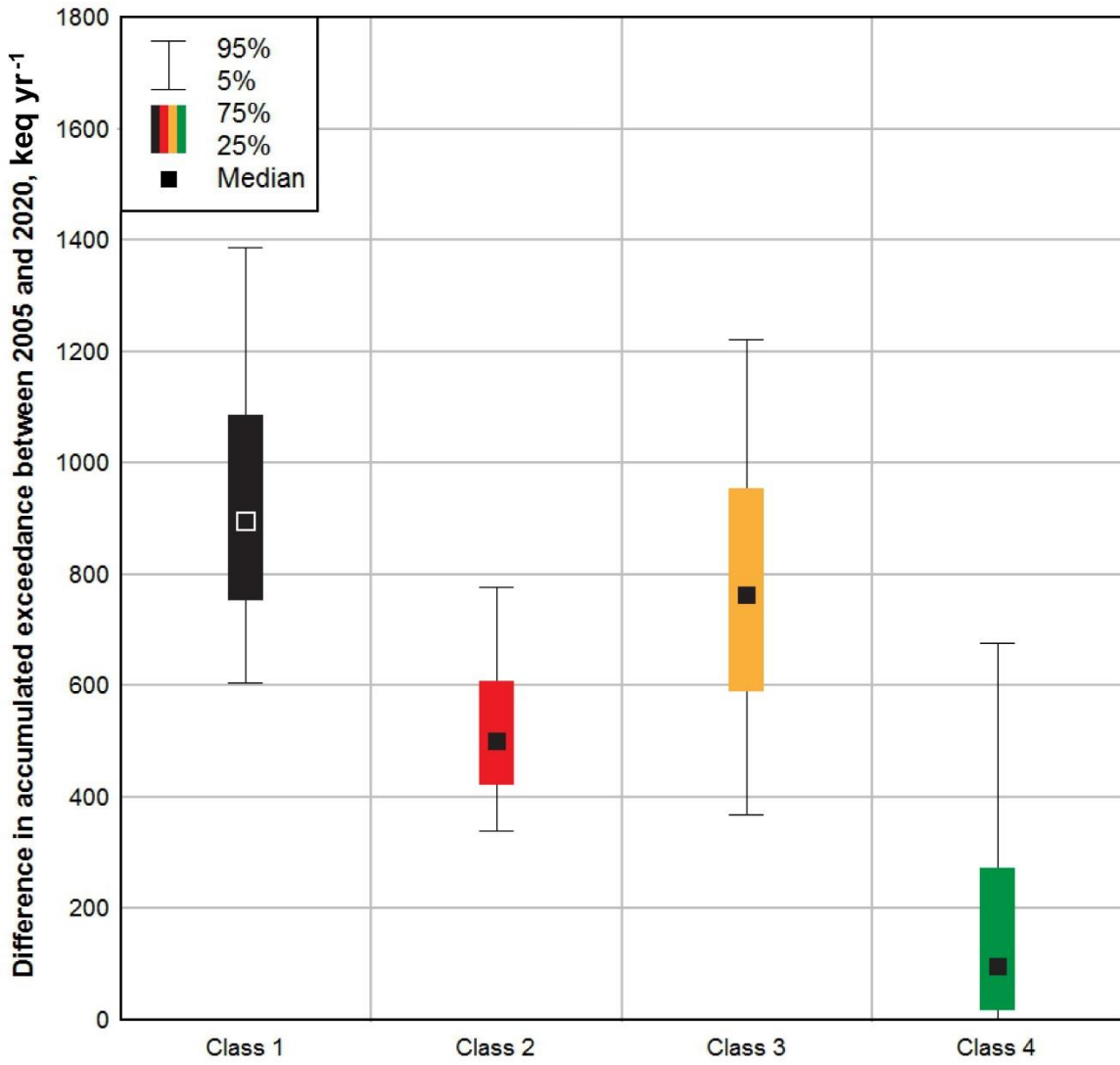
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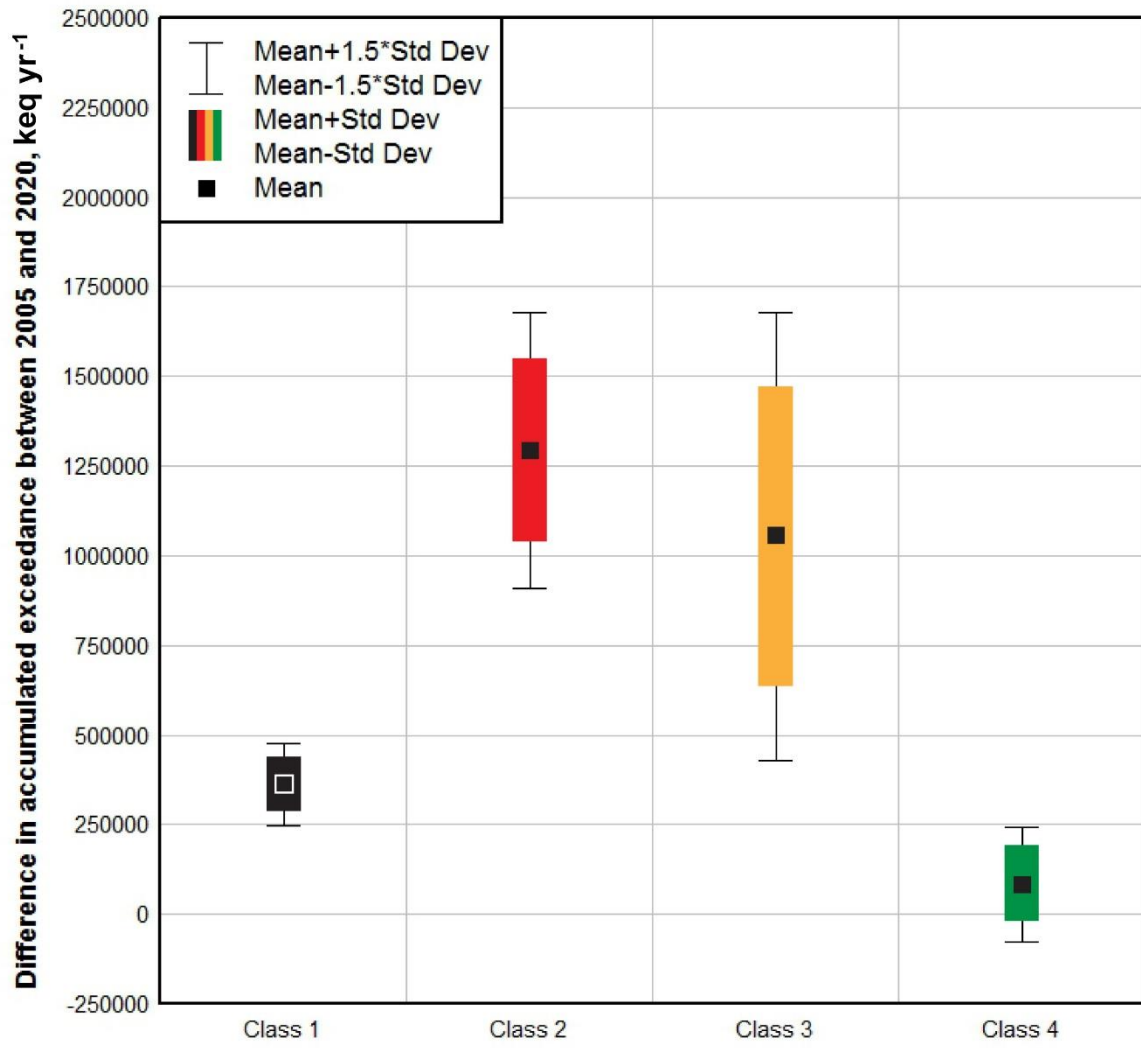


558 Figure 3

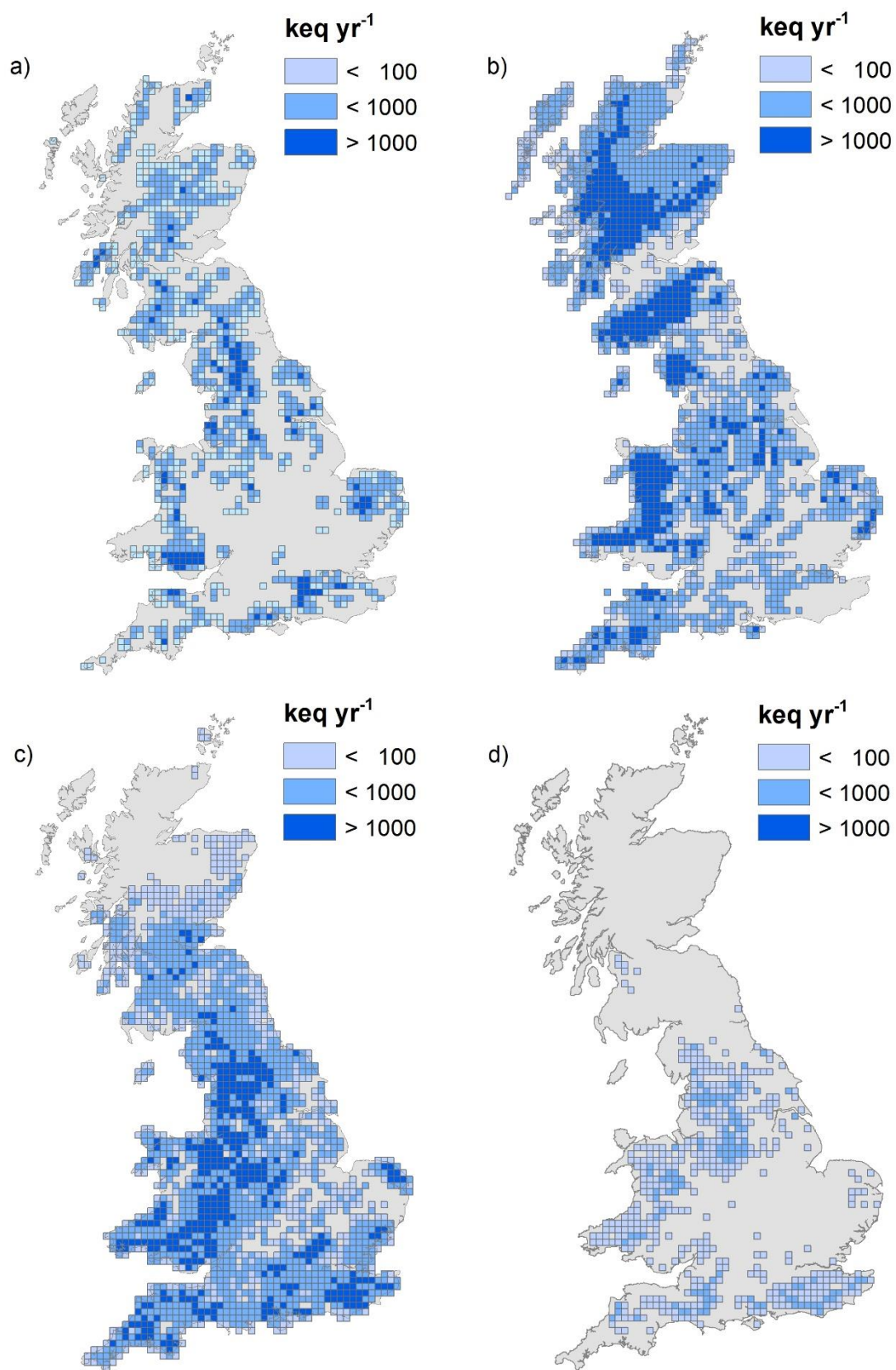


559

560 Figure 4



561



564

565 Vitae

566 Duncan Whyatt is a senior lecturer at Lancaster University. He is a geographer with over 25 years'  
567 experience of applying geospatial techniques in environmental research at local, national and  
568 regional scales. He uses GIS to visualise and analyse spatial data from different sources including  
569 pollution models. He has expertise in running a range of models to address different aspects of air  
570 pollution.

571 Sarah Metcalfe is Professor of Earth and Environmental Dynamics in the School of Geography at the  
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574 Review Group on Acid Rain, the Critical Loads Advisory Group and the National Expert Group on  
575 Transboundary Air Pollution and carried out research for the UK's devolved administrations and the  
576 Environment Agency.

577 Professor Richard (Dick) Derwent took a degree in 1968 and a PhD in 1971 from the  
578 University of Cambridge in physical chemistry. Dick Derwent has spent much of his research  
579 career studying air pollution. Initially, this carried out in the Air Pollution Division, Warren  
580 Spring Laboratory, then at the Harwell Laboratory and finally at the Meteorological Office,  
581 Bracknell. In 2003, he took early retirement and became a self-employed consultant on air  
582 pollution.

583 Trevor Page is a senior research associate at Lancaster University, UK. His interests are primarily in  
584 environmental systems modelling with a focus on hydrological and geochemical fluxes through  
585 catchments. Specifically, his work includes model uncertainty analyses coupled with evaluating the  
586 value of different types of data for improving model process-representation and model predictions.  
587 Much of his work has utilised Generalised Likelihood Uncertainty Estimation as a framework for  
588 these assessments.

589