

## Article (refereed) - postprint

---

Tipping, Edward; Davies, Jessica A.C.; Henrys, Peter A.; Jarvis, Susan G.; Smart, Simon M.. 2021. **Long-term effects of atmospheric deposition on British plant species richness.**

© 2020 Elsevier B.V.

This manuscript version is made available under the CC BY-NC-ND 4.0 license

<https://creativecommons.org/licenses/by-nc-nd/4.0/>



This version is available at <http://nora.nerc.ac.uk/id/eprint/530012>

Copyright and other rights for material on this site are retained by the rights owners. Users should read the terms and conditions of use of this material at <https://nora.nerc.ac.uk/policies.html#access>.

**This is an unedited manuscript accepted for publication, incorporating any revisions agreed during the peer review process. There may be differences between this and the publisher's version. You are advised to consult the publisher's version if you wish to cite from this article.**

The definitive version was published in *Environmental Pollution*, 281, 117017. <https://doi.org/10.1016/j.envpol.2021.117017>

The definitive version is available at <https://www.elsevier.com/>

Contact UKCEH NORA team at  
[noraceh@ceh.ac.uk](mailto:noraceh@ceh.ac.uk)

1 *ENVPOL-D-20-03728*

2 *Revision submitted to Environmental Pollution, March 2021*

3

4 **Long-term effects of atmospheric deposition on British plant species richness**

5 **Edward Tipping<sup>a,\*</sup>, Jessica A.C. Davies<sup>b</sup>, Peter A. Henrys<sup>a</sup>, Susan G. Jarvis<sup>a</sup>, Simon M. Smart<sup>a</sup>**

6

7 <sup>a</sup> UK Centre for Ecology & Hydrology, Lancaster Environment Centre, Lancaster LA1 4AP, UK

8 <sup>b</sup> Lancaster Environment Centre, Pentland Centre for Sustainability in Business, Lancaster University,  
9 Lancaster LA1 4YX, UK

10

11

12 Correspondence to: Dr Edward Tipping

13 [et@ceh.ac.uk](mailto:et@ceh.ac.uk)

14 ++44 (0) 1524 595800

## 15 **Abstract**

16 The effects of atmospheric pollution on plant species richness ( $n_{sp}$ ) are of widespread concern. We  
17 carried out a modelling exercise to estimate how  $n_{sp}$  in British semi-natural ecosystems responded to  
18 atmospheric deposition of nitrogen ( $N_{dep}$ ) and sulphur ( $S_{dep}$ ) between 1800 and 2010. We derived a  
19 simple four-parameter equation relating  $n_{sp}$  to measured soil pH, and to net primary productivity  
20 (NPP), calculated with the N14CP ecosystem model. Parameters were estimated from a large data set  
21 ( $n = 1156$ ) of species richness in four vegetation classes, unimproved grassland, dwarf shrub heath,  
22 peatland, and broadleaved woodland, obtained in 2007. The equation performed reasonably well in  
23 comparisons with independent observations of  $n_{sp}$ . We used the equation, in combination with  
24 modelled estimates of NPP (from N14CP) and soil pH (from the CHUM-AM hydrochemical model), to  
25 calculate changes in average  $n_{sp}$  over time at seven sites across Britain, assuming that variations in  $n_{sp}$   
26 were due only to variations in atmospheric deposition. At two of the sites, two vegetation classes were  
27 present, making a total of nine site/vegetation combinations. In four cases,  $n_{sp}$  was affected about  
28 equally by pH and NPP, while in another four the effect of pH was dominant. The ninth site, a chalk  
29 grassland, was affected only by NPP, since soil pH was assumed constant. Our analysis suggests that  
30 the combination of increased NPP, due to fertilization by  $N_{dep}$ , and decreased soil pH, primarily due to  
31  $S_{dep}$ , caused an average species loss of 39% (range 23 – 100%) between 1800 and the late 20<sup>th</sup> Century.  
32 The modelling suggests that in recent years  $n_{sp}$  has begun to increase, almost entirely due to  
33 reductions in  $S_{dep}$  and consequent increases in soil pH, but there are also indications of recent slight  
34 recovery from the eutrophying effects of  $N_{dep}$ .

35 *Keywords*        *modelling; net primary productivity; nitrogen deposition; plant species richness; soil*  
36 *pH; sulphur deposition*

37

## 38 *Capsule*

39 *Modelling the long-term effects of atmospheric deposition indicates that both nutrient enrichment and*  
40 *soil acidification reduced plant species richness, which is now recovering from acidification.*

## 41 1. Introduction

42 Over the past two centuries, nitrogen (N) and sulphur (S) deposition have impacted terrestrial  
43 ecosystems globally (Vitousek et al, 1997; Larssen et al., 1999; Sutton et al., 2011; Fowler et al., 2012).  
44 Forms of both deposited elements cause soil acidification (van Breemen et al., 1984), while N also acts  
45 as a nutrient (Matson et al., 2002). Among the ecological effects caused by these pollutants, a major  
46 concern is plant species loss in natural and semi-natural (i.e. managed but not fertilized) ecosystems  
47 (Bobbink et al., 2010; Azevedo et al., 2013).

48 Empirical field evidence connecting deposition with declines in plant species richness ( $n_{sp}$ ; number of  
49 species per unit area) comes largely from spatial surveys, covering locations with different deposition  
50 loads ( $N_{dep}$ ,  $S_{dep}$ ). For example, Stevens et al. (2004) and Maskell et al. (2010) related spatial variations  
51 in  $n_{sp}$  to  $N_{dep}$  in Britain. Similar findings were made for sites across NW Europe by Dupré et al. (2010).  
52 For the continental USA, Simkin et al. (2016) reported a positive dependence of  $n_{sp}$  on  $N_{dep}$  at low  $N_{dep}$   
53 (up to about  $1 \text{ gN m}^{-2} \text{ a}^{-1}$ ) and a negative dependence over higher ranges of  $N_{dep}$ . With regard to pH,  
54 a number of studies have been published showing a positive dependence of  $n_{sp}$  on pH in temperate  
55 and boreal ecosystems (e.g. Grime 1973; Gough et al., 2000; Peet et al., 2003; Stevens et al., 2004;  
56 Dupré et al., 2010), strongly suggesting that soil acidification reduces  $n_{sp}$ .

57 Field evidence for changes in  $n_{sp}$  over time, as a result of changes in deposition rates, is comparatively  
58 rare. Two exceptions are recent studies by Rose et al. (2016) and Mitchell et al. (2018), which have  
59 demonstrated increases in plant diversity over recent decades at several sites in Britain. Rose et al.  
60 (2016) attributed the increases principally to the reversal of soil acidification, emphasising reductions  
61 in  $S_{dep}$ , and they identified “a pressing need to distinguish more clearly between the spatial impacts of  
62 acidification and eutrophication on species richness.”

63 Here we address the issue of long-term (1800-2010) temporal change in  $n_{sp}$  in Britain using process-  
64 based models, driven by measured and modelled deposition rates. We account for soil acidification  
65 with CHUM-AM, a model that simulates soil and surface water chemistry, and which has been  
66 calibrated and used for long-term simulations at a range of locations to describe responses to both  
67 acid and metal deposition (Ashmore et al., 2004; Tipping et al., 2006, 2007, 2010; Tipping & Chaplow,  
68 2012). The nutrient effects of  $N_{dep}$  are simulated with the N14CP model (Davies et al., 2016, 2017),  
69 which deals with the interlinked biogeochemical cycles of carbon, nitrogen and phosphorus, and has  
70 been used to calculate national scale changes in NPP in British semi-natural ecosystems. Because these  
71 ecosystems are assumed mostly to be N-limited, like many temperate natural and semi-natural  
72 ecosystems (Elser *et al.* 2007; LeBauer & Treseder 2008; Lee *et al.* 2010; Field *et al.* 2017), N14CP-  
73 calculated NPP responded strongly to the increases in  $N_{dep}$  over the simulation period (Tipping et al.,

74 2019). The exception was ombrotrophic peatland, which is likely phosphorus-limited (Toberman et al.,  
75 2015; Schillereff et al., 2016). The significance of NPP is in providing the mechanism for changes in  $n_{sp}$ ,  
76 primarily via competition effects (Grime, 1973; Huston, 2014, Fraser et al., 2015; Grace et al., 2016),  
77 thereby enabling  $n_{sp}$  to be linked with the nutrient effects of  $N_{dep}$ .

78 We analysed plant richness data obtained in the British Countryside Survey of 2007  
79 (<http://www.countrysidesurvey.org.uk/>) for four vegetation classes, namely acid and neutral  
80 grassland, dwarf shrub heath, peatland and broadleaved woodland. We parameterized a simple model  
81 relating the values of  $n_{sp}$  to (a) measured soil pH at the sampling sites and (b) NPP calculated with the  
82 N14CP model. Then, again employing N14CP-calculated NPP values, we used the model to predict  
83 changes of  $n_{sp}$  over time at six field sites to which the CHUM-AM model had been calibrated in previous  
84 work. We also performed predictive calculations for a chalkland site at which soil acidification was  
85 assumed not to have occurred.

86 We report simulations of  $n_{sp}$  over the period 1800-2010 for the combined effects of changes in soil pH  
87 and NPP, and for each factor separately. The results provide a retrospective account of the paths to  
88 present conditions, partition the  $n_{sp}$  changes according to the two drivers (pH, NPP), and offer an  
89 approach to forecasting future changes in  $n_{sp}$  for different scenarios of pollutant deposition. In its long-  
90 term approach, our study complements that of Clark et al. (2018), who calculated exceedances of  
91 Critical Loads of air pollutants for the period 1800-2025 in the USA. Whereas Clark et al. considered  
92 changes over time in the vulnerability of various indicators, including plant community composition,  
93 to atmospheric pollution, our focus is on the mechanistic biogeochemical effects of pollution and the  
94 quantitative responses of plant species richness.

## 95 2. Methods

### 96 2.1. Plant species richness and soil pH data

97 Plant species richness data,  $n_{sp}$ , expressed as the number of different species per quadrat for each  
98 vegetation class, and measured soil pH, were obtained from Countryside Survey (CS) results from  
99 1978, 1998 and 2007 (<http://www.countrysidesurvey.org.uk/>); see Table 1. Bryophytes were excluded  
100 because these have not been exhaustively recorded in the CS quadrats. Data from four vegetation  
101 classes were analysed, namely unimproved grassland, dwarf shrub heath, peatland, and broadleaved  
102 woodland (Jackson, 2000). Peatland includes bog and peat-dominated heathland. Unimproved  
103 grassland comprises neutral and acid grassland not subject to agricultural improvement. For brevity,  
104 we refer to the vegetation classes, as grassland, heathland, peatland and woodland.

105 Values of  $n_{sp}$  were available for 4 m<sup>2</sup> quadrats in 1998 and 2007, and for 200 m<sup>2</sup> quadrats in all three  
106 of the survey years. These quadrat sizes are appropriate for sampling species richness in the target  
107 habitats; 200m<sup>2</sup> has been consistently used for British broadleaved woodlands (Smart et al., 2014)  
108 while 4m<sup>2</sup> is recommended for grassland and dwarf shrub heath (Rodwell, 2006). Sampling was carried  
109 out as close as possible to the same date in every survey to avoid seasonal effects.

110 We used data from the most extensive survey, that of 2007, for model fitting, and data from 1978 and  
111 1998 for model testing. Some  $n_{sp}$  values in the 4 m<sup>2</sup> quadrat data were zero, because at this scale, no  
112 live material was present at the time of sampling, at sites with bare rock, areas of bare peat, and dense  
113 stands of the rhizomatous fern *Pteridium aquilinum*. For surveys with results for both 4 m<sup>2</sup> and 200  
114 m<sup>2</sup> quadrats, the  $n_{sp}$  values for the two quadrat sizes were strongly correlated, as demonstrated with  
115 the 2007 data in Fig. S1.

116 We also used relevant data from the literature, for comparison. Data on species richness in 1 m<sup>2</sup>  
117 quadrats as a function of pH in two Derbyshire (UK) valleys, Cressbrookdale and Lathkilddale, came  
118 from Grime (1973), samples collected in 1965-1971. Lathkilddale was ungrazed and sporadically  
119 burned, and Cressbrookdale was grazed by sheep and cattle. At both locations, sites with a range of  
120 pH were sampled. Stevens et al. (2004), reported species richness in 4 m<sup>2</sup> quadrats samples collected  
121 in 2002 and 2003 from acid grassland sites at 68 locations on a transect of  $N_{dep}$  (with varying pH) across  
122 Britain.

### 123 2.2. The CHUM-AM model

124 The hydrochemistry model CHUM-AM was described in detail by Tipping et al. (2006, 2007). It deals  
125 with hydrology, C, N and S cycling, cation sorption to soil organic matter, mineral precipitation, and

126 weathering, in a soil-rock system that includes a topsoil compartment. Water and solutes are  
127 transferred to surface waters. The model was driven by measured and modelled  $N_{\text{dep}}$ ,  $S_{\text{dep}}$ , and base  
128 cation deposition data from 1975, and for earlier years by deposition estimated from known or  
129 assumed emissions, as described in detail by Tipping et al. (2006, 2007, 2010) and Tipping and Chaplow  
130 (2012); see Fig. S2. Weathering rate constants are calibrated to match observed soil and water  
131 chemistries. The key output for the present work was the annual average pH of topsoil, typically to a  
132 depth of 15 cm.

133 The model outputs used here were from the previous studies referenced above with simulations from  
134 1400 CE to 2010 for a number of small catchments (Fig. 1, Table 2). The CHUM modelling is for the  
135 lumped catchment, and therefore we have to use the same pH values for each class of vegetation  
136 considered at the site. For example, the River Etherow catchment has both grassland and peatland,  
137 and the same pH values were assumed to apply to both.

### 138 *2.3. The N14CP model*

139 The model is described in detail in Davies et al. (2016), also in Tipping et al. (2017, 2019). It describes  
140 plant-soil pools and fluxes of C, N and P, driven by climate,  $N_{\text{dep}}$ ,  $S_{\text{dep}}$ , base cation deposition,  
141 weathering of base cations and P, and vegetation history. The model simulates, on four quarterly time  
142 steps within each year, the growth and decay of plant biomass, N fixation, the production of litter and  
143 its incorporation into soil organic matter (SOM), the immobilisation of nutrients (N, P) by SOM, SOM  
144 turnover, leaching losses, and denitrification. The fixation of N from the atmosphere is positively  
145 related to temperature and P availability, but does not depend on vegetation class. It is down-  
146 regulated by  $N_{\text{dep}}$  such that no additional N from  $N_{\text{dep}}$  becomes available until the  $N_{\text{dep}}$  rate outstrips  
147 the potential fixation rate. The peatland module is described by Janes-Bassett et al. (2020); peatland  
148 is assumed to be P-limited, and therefore no effect of  $N_{\text{dep}}$  on NPP is predicted.

149 Within the N14CP formulation, soil pH, calculated from an annual balance of cations and anions, acts  
150 as a modifier to organic matter decomposition, and therefore affects N cycling and thereby NPP.  
151 However, the resultant pH dependence of decomposition following the model's parameterization was  
152 found to be small, and so in practice variations in soil pH have little influence on the simulated NPP.  
153 Because the soil pH estimates with N14CP are highly approximate, the results from the more  
154 chemically-detailed CHUM-AM model were preferred.

155 The N14CP model, driven by vegetation class, climatic variables and modelled atmospheric pollutant  
156 deposition, is used to simulate ecosystem C-N-P cycling, including NPP over the Holocene. We  
157 assumed that the vegetation classes at the present sites had been constant since the period of tree

158 clearance in Britain (see Tipping et al. 2017). Annual values of  $N_{\text{dep}}$  and  $S_{\text{dep}}$  resolved spatially (5 km x  
159 5 km grid squares) over the period 1800–2010, and taking vegetation class into account, were  
160 estimated as in previous modelling reported in Tipping et al. (2017); see Fig. S2. The key outputs for  
161 the present work were the average NPP ( $\text{gC m}^{-2} \text{a}^{-1}$ ) by year for each vegetation class in 5 km x 5 km  
162 grid squares across Britain. These are referred to as  $\text{NPP}_{\text{calc}}$ .

#### 163 *2.4. Data analysis*

164 Following Grime (1973) and Peet et al. (2003), in order to establish general trends in the data, we  
165 averaged values of  $n_{\text{sp}}$  according to pH and  $\text{NPP}_{\text{calc}}$ . This was done first with ordered pH values and  
166 second with ordered  $\text{NPP}_{\text{calc}}$ . The number of data for each variable in each averaged bin was set to the  
167 integer nearest to the square root of the total number of data.

168 We used Microsoft Excel to perform linear regression analyses. Results were checked for normality  
169 using Q-Q plots, and where necessary outliers were identified as points deviating by more than three  
170 standard deviations from the expected value.

171 Data fitting was performed using Microsoft Excel Solver. Models were compared using the Akaike  
172 information criterion ( $\Delta\text{AIC}$ ); Akaike weights ( $w_{\text{AIC}}$ ) were interpreted as conditional probabilities  
173 (Wagenmakers and Farrell, 2004). We estimated model parameter uncertainty using a Monte Carlo  
174 method implemented in Microsoft Excel (Hu et al., 2015). This involved repeated resampling of  $n_{\text{sp}}$   
175 from the values predicted with the best-fit model, adding perturbations from the assumed normal  
176 distribution of root mean squared deviations. Each derived set of  $n_{\text{sp}}$  was then fitted with the model  
177 to yield a parameter set. This was done 1000 times, and the parameter standard errors were  
178 calculated from the results. To estimate ranges of predicted values, the model was run repeatedly  
179 (1000 times) with parameter sets drawn from the normal distribution of values described by the  
180 parameter means and standard errors.



### 181 3. Results

#### 182 3.1. Dependence of $n_{sp}$ on pH and $NPP_{calc}$

183 Trends in  $n_{sp}$  with measured soil pH and  $NPP_{calc}$  are shown in Fig. 2, by averaged data for the four  
 184 vegetation classes in 4 m<sup>2</sup> quadrat samples collected in 2007. The N14CP model assumes peatland to  
 185 be P-limited, consequently there is little variation spatial variation in  $NPP_{calc}$  for this vegetation class,  
 186 and so the results are not plotted. Values of  $n_{sp}$  clearly increase with pH, and in two cases (heathland,  
 187 woodland) they clearly decline with  $NPP_{calc}$ . For grassland, the decline with NPP is less definite. Very  
 188 similar trends were evident in the 200 m<sup>2</sup> quadrat data for 2007 (Fig. S3).

189 Increasing  $n_{sp}$  with pH, and declining  $n_{sp}$  with NPP, are expected from the published literature (see  
 190 Introduction). However, there is no evidence of a positive relationship at low  $NPP_{calc}$ , as would be  
 191 expected from a unimodal dependence on NPP (Grime, 1973; Fraser et al., 2015). In case this might  
 192 reflect the high degree of N enrichment in the ecosystems that had developed by 2007, we plotted  
 193 the data (200 m<sup>2</sup> quadrats) for 1978, when N enrichment would have been less, but again found no  
 194 evidence for positive relationships in plots of  $n_{sp}$  against  $NPP_{calc}$  at low  $NPP_{calc}$ .

195 The pH dependence of  $n_{sp}$  (Fig. 2) is similar in shape to published values for temperate and boreal field  
 196 sites, with a relatively steep rise in the low pH region, and a flattening off at higher pH (Grime, 1973;  
 197 Gough et al., 2000; Peet et al., 2003; Schuster and Diekmann, 2003; Vonlanthen et al., 2006; Chytrý  
 198 et al., 2007; Peppler-Lisbach and Kleyer, 2009; Riesch et al., 2018). Therefore to model the  
 199 dependence of  $n_{sp}$  on pH at constant NPP we adopted an equation of the form

$$200 \quad n_{sp} = \alpha' (\text{pH} - \text{pH}_0)^{\beta'} \quad (1)$$

201 Here  $\alpha'$  and  $\beta'$  are constants, and  $\beta'$  has a value between 0 and 1. The value of  $\text{pH}_0$  is the pH at which  
 202 the soil is too acid for significant plant survival. In the above-cited literature, plots of  $n_{sp}$  against pH  
 203 suggest that  $\text{pH}_0$  has values near to 3, and the plots in Fig. 2 are in line with this. For simplicity, the  
 204 decline in  $n_{sp}$  with  $NPP_{calc}$  was assumed to be a linear relationship, requiring one additional parameter  
 205  $\gamma$ . This led to a full model equation as follows

$$206 \quad n_{sp} = \alpha (\text{pH} - \text{pH}_0)^\beta (1 - \gamma NPP_{calc}) \quad (2)$$

207 We explored how many parameters could be justifiably fitted to describe the  $n_{sp}$  values of the four  
 208 vegetation classes.

209 The first step was to fit the model to the grassland, heathland and woodland data sets separately for  
 210 the 4 m<sup>2</sup> quadrats sampled in 2007, using individual data points (not the averaged values of Fig. 2).  
 211 Four versions of the model were tested (Table 3). Version 1 included all four parameters, in version 2  
 212 pH dependence was eliminated, in version 3 NPP dependence was eliminated, and in version 4 a linear

213 pH dependence was forced. Table 3 shows that for each of the three vegetation classes, the values of  
 214  $w_{AIC}$ , interpreted as the probability of the model being correct, are very low for model versions 2 and  
 215 3, which means that both pH dependence and NPP dependence need to be included in the best model.  
 216 Overall, the  $w_{AIC}$  values for versions 1 and 4 are similar, making it difficult to choose between them.  
 217 However, the  $pH_0$  values fitted with version 1 are similar for the three vegetation classes (grassland  
 218 2.89, heathland 2.90, woodland 3.30), and consistent with observations reported in the literature and  
 219 shown in Fig. 2. On the other hand, with version 4 ( $\beta$  set to 1.0), the  $pH_0$  values (0.91, 2.04, -2.02  
 220 respectively) vary more, and are less realistic. Therefore we favour version 1 of the model. Table 4  
 221 shows the fitted parameters.

222 In the case of peatland, because there is essentially no dependence of  $n_{sp}$  on  $NPP_{calc}$ , only two model  
 223 versions were tested, the first with  $\beta$  optimised, and the second with  $\beta$  set to 1.0. These gave  $w_{AIC}$   
 224 values of 0.35 and 0.65 respectively. Again these are quite similar, and again the  $pH_0$  value of 3.04,  
 225 obtained with the first version, is close to the values for the other vegetation classes, and more  
 226 realistic than the value of 1.99 obtained with the second version. The fitted parameters for peatland  
 227 with the first model version are presented in Table 4.

228 The second step was to consider whether the total number of fitted parameters could be reduced  
 229 from the total of 17 required to parameterize the model separately for the four vegetation classes  
 230 (Table 4). We fitted the entire data set ( $n = 1156$ ) with three models, A, B and C (Table 5). Model A  
 231 retained the 17 parameters already fitted, while model B had common values of  $\alpha$ ,  $\beta$ ,  $\gamma$  and  $pH_0$  for all  
 232 four vegetation classes. For model C, the vegetation classes were distinguished by fitting a specific  
 233 value of  $\alpha$  for each class, with common values of  $\beta$ ,  $\gamma$  and  $pH_0$ . The values of  $w_{AIC}$  (Table 5) show that  
 234 models B and C are both clearly superior to model A, and that model C is preferable to model B.  
 235 Therefore, we chose model C for the predictive calculations.

236 Model C accounts for 23.9 % of the total variance in  $n_{sp}$  ( $n = 1156$ ). Its parameter values and their  
 237 standard errors are shown in Table 6. The parameter uncertainties are fairly modest, suggesting that  
 238 the parameter values are reasonably well defined, bearing in mind the large scatter in the data. A plot  
 239 of observed against calculated  $n_{sp}$  values is shown in Fig. 3. Plots to show what the fitted model  
 240 predicts for heathland and woodland are shown in Fig. S4; as would be expected from the similar  
 241 values of  $\alpha$  for the non-woodland classes (Table 6), results for grassland and peatland are similar to  
 242 those for heathland. Parameter values derived from the 200 m<sup>2</sup> quadrat data are shown in Table S1.

243 *3.2. Testing the model against independent data*

244 We compared model predictions with values of  $n_{sp}$  for different vegetation classes extracted from CS  
245 survey results for 1978 and 1998, at sites that had not been surveyed in 2007. Because the 1978 survey  
246 did not include 4 m<sup>2</sup> quadrats, we restricted the comparisons to 200 m<sup>2</sup> quadrat values, using the  
247 parameters for Model C shown in Table S1. The observed and predicted values of  $n_{sp}$  (Fig. S5) were in  
248 reasonable agreement ( $r^2_{adj} = 0.161$ ,  $p < 0.0001$ ,  $n = 336$ ). The slope of 0.92 in a plot of observed vs  
249 predicted  $n_{sp}$  did not differ from unity.

250 We also applied the grassland model to two published data sets for  $n_{sp}$  in grassland (Fig. 4). For the  
251 Derbyshire dales data of Grime (1973) the measured pH values were used to drive the model, with a  
252 single modelled NPP value. For the Stevens et al. (2004) data, we used measured pH and modelled  
253 NPP values to drive the model. We found highly significant correlations between observed and  
254 calculated values (see caption to Fig. 4), but the observed values were appreciably greater than the  
255 observed and predicted values for the CS plots; see Discussion.

256 Another approach was to compare changes in  $n_{sp}$  occurring at CS plots between either 1978 and 1998  
257 or 1978 and 2007. We extracted results for 551 sites (237 grassland, 96 heathland, 156 peatland, 62  
258 woodland), and compared observed and calculated changes in  $n_{sp}$  (Fig. S6). Both losses and gains of  
259 species were expected and found. They were weakly ( $r^2_{adj} = 0.049$ ) but significantly ( $p < 0.0001$ )  
260 correlated.

261 We also tested the ability of the CHUM-AM model to simulate changes in soil pH, by comparing  
262 observed results for the CS sites with the modelled changes for the CHUM-AM sites. We restricted the  
263 CS sites analysed to those with an average pH of between 3 and 5, to match the range of CHUM-AM  
264 values. The CS and CHUM-AM locations are not the same, and so the comparison is only of general  
265 trends across Britain. The average increase in pH between 1978 and 1998 for 174 CS sites was 0.31  
266 (standard error, SE = 0.04), while that for 144 sites from 1978 to 2007 was 0.33 (SE = 0.05). The  
267 corresponding average modelled changes for the six CHUM-AM sites were, for 1978-1998, 0.22 (SE =  
268 0.03) and, for 1978-2007, 0.32 (SE = 0.05). The differences between the observed and modelled values  
269 are not significant ( $p > 0.05$ ). Another comparison that can be made is with measured changes  
270 reported by the National Soil Inventory (NSI) of England and Wales (Kirk et al., 2010). Between 1978  
271 and 2003, average annual increases in soil pH, attributed principally to reductions in acid deposition,  
272 were 0.011 for semi-natural grassland (235 sites) and 0.013 for deciduous woodland (104 sites). These  
273 correspond to total pH changes of 0.275 and 0.325 respectively, comparable to the modelled average  
274 pH change for the six CHUM-AM sites of 0.288 over the same 25-year period.

275 *3.3. Predicted temporal changes 1800-2010*

276 Calculated changes in soil pH (CHUM-AM) and NPP (N14CP) at the study sites (Fig. 1) are shown in  
277 Figs. 5 and S7. Except for the South Downs site, soil pH is calculated to have been at its lowest in the  
278 1970s, then to have increased by 0.2 – 0.5 units over the period to 2010. For the South Downs chalk  
279 grassland we assumed that there had been no significant change in soil pH, owing to the high buffering  
280 capacity of the soil conferred by the calcium carbonate; we assumed a constant pH of 7.5 (Goulding,  
281 2016). We applied Model C, driven by the modelled pH and NPP values, to calculate  $n_{sp}$  for 4 m<sup>2</sup>  
282 quadrats from 1800 to 2010 at the sites for which CHUM-AM had been calibrated. Results are plotted  
283 in Fig. 5 and Fig. S7, and summarized in Table 7. Fig. S8 shows estimates of uncertainty in the calculated  
284 values of  $n_{sp}$ .

285 In two of the nine site/vegetation combinations of Table 7, namely Lochnagar/heathland and Old  
286 Lodge/woodland, the  $NPP_{calc}$  values for the early years of the simulations were less than the lowest  
287  $NPP_{calc}$  values for the period (1978-2007) with observed  $n_{sp}$  values, i.e. values used for data fitting.  
288 Therefore in these two cases the predictions of  $n_{sp}$  are outside the range of  $NPP_{calc}$  for which Model C  
289 is calibrated. However, the 1800 values are only slightly less than the 1978-2007 minima, and so it is  
290 unlikely that the effects on predicted  $n_{sp}$  are significant.

291 The overall pattern of change over the period 1800 to 2010 is of an initial decline in  $n_{sp}$  until around  
292 1980, followed by an increase. The declines were caused both by decreases in soil pH and increases in  
293 NPP, whereas the subsequent recoveries are due almost entirely to pH reversal. The River Etherow  
294 site is calculated to have responded most in terms of  $n_{sp}$  change, owing to the high levels of  
295 atmospheric pollution it has received over a prolonged period. The least sensitive sites are Lochnagar  
296 and the South Downs, the former because of its remoteness from pollution sources, the latter because  
297 of the lack of change in soil pH.

298 The comparative effects of variations in soil pH and NPP on the predicted variations of  $n_{sp}$  over time  
299 (Table 7, Fig. 5, Fig. S7) vary among the sites, depending upon vegetation class, soil response to  
300 acidifying agents, and atmospheric deposition (Table 2). In four cases, Cote Gill, Mosedale Beck, and  
301 the two Old Lodge sites, the effects of pH and NPP variations are similar. For Lochnagar and River  
302 Etherow/grassland pH effects dominate, but there are contributions from NPP. For the two peatland  
303 sites, River Etherow and Great Dun Fell, the effects of NPP variation are near to zero, because of the  
304 assumed P limitation, as discussed in Section 3.1, and so soil pH variation is the sole factor. The chalk  
305 grassland of the South Downs is calculated to respond only to changes in NPP, since atmospheric  
306 deposition of acidifying agents is assumed not to affect soil pH.

307 The summary data in Table 7 show that all nine site/vegetation combinations are calculated to have  
308 suffered declines in  $n_{sp}$  since 1800, the greatest, 100 %, being for both grassland and peatland at the

309 River Etherow, while the smallest, 23 %, was for heathland at Lochnagar. On average, the decline was  
310 39 %. Species richness recoveries are calculated for most of the sites over recent years (from about  
311 1990 onwards), the annual rates ranging from zero to 0.095 species a<sup>-1</sup> with an average of 0.046  
312 species a<sup>-1</sup>. The highest rates are estimated for peatland vegetation, and for sites with high S  
313 deposition (cf. Table 2).

## 314 4. Discussion

### 315 4.1. Modelling species richness

316 The model combines the two distinct drivers, resulting in a model (equation 2) that gives stronger  
317 relationships than either driver separately, which is necessary to take into account both acidification  
318 and eutrophication effects on  $n_{sp}$ . The best version (Model C, Table 5), distinguishes vegetation classes  
319 only in terms of the parameter  $\alpha$ , which provides a general measure of species richness in a vegetation  
320 class; the  $\alpha$  values suggest that woodland tends to be the most species-rich of the four classes, while  
321 grassland, heathland and peatland are quite similar.

322 Ideally, the construction of a model relating  $n_{sp}$  to soil pH and NPP would be based on empirical  
323 measurements of both driving variables at each site where the richness is determined. We have such  
324 data for pH, but NPP estimates have not been part of the CS studies, and therefore we used values  
325 modelled as averages over 5 km x 5 km grid cells. Another limitation in the modelling is that the  
326 available data for parameterization and testing refer only to relatively recent times, i.e. from 1978  
327 onwards. This means that there are relatively few values of  $n_{sp}$ , from remote relatively unpolluted  
328 sites, that might approach near-pristine conditions. Therefore, application of the model to unpolluted  
329 conditions involves some extrapolation, as noted in Section 3.3.

330 The results of tests on independent data provide support for the parameterizations from 2007 data.  
331 Earlier CS data for 1978 and 1998 are predicted quantitatively and without bias, with parameters  
332 obtained by fitting the 2007 data (Fig. S5), and changes in  $n_{sp}$  at the CS plots are also captured (Fig.  
333 S6). However, although the predicted values of  $n_{sp}$  are strongly correlated with data reported in two  
334 other studies (Fig. 4), the absolute reported values are in the upper range of CS values and so are  
335 underpredicted. This is probably because these other studies (Grime, 1973; Stevens et al., 2004) used  
336 internally more homogenous examples of each grassland type associated with stable, well-established  
337 grazing regimes. The random sampling approach used in CS results in a noisier sample, more  
338 representative of the range of variation in the wider countryside where assignment to each grassland  
339 habitat type is *post hoc* rather than predetermined during study design (Maskell et al., 2010).  
340 Additional support for the model, in terms of the structure of equation (2), is that it predicts that  
341 slopes of  $n_{sp}$  against NPP should become more negative with increasing pH (Fig. S4), behaviour that  
342 has been reported for the Park Grass experiments at Rothamsted (Crawley et al., 2005).

343 We did not find evidence for the operation of the unimodal (hump back) model of Grime (1973), in  
344 which lower  $n_{sp}$  at low NPP would be expected. This applied to both the 2007 data and the data from  
345 1978, when the ecosystems would have been less enriched in N, and therefore perhaps more likely to

346 reveal a positive relationship of  $n_{sp}$  to  $NPP_{calc}$  at low  $NPP_{calc}$ . The lack of evidence of unimodal  
347 behaviour is perhaps not surprising given the crude and highly-averaged nature of our NPP estimates,  
348 which do not reflect the considerable small-scale spatial variations in measured values (Tipping et al.,  
349 2019). Consequently, our ranges of  $NPP_{calc}$  are more restricted than the ranges of productivity in  
350 studies that do provide evidence for the unimodal model (Huston, 2014; Fraser et al., 2015; Simkin et  
351 al., 2016).

352 Although the model accounts for a significant amount of the variance in  $n_{sp}$ , a considerable amount  
353 (76.1 %) remains unaccounted for, and consequently uncertainty (95 % CL) in the predicted values is  
354 about  $\pm 30$  %, depending upon the vegetation class (Fig. S8). It must result from controls on  $n_{sp}$  other  
355 than soil pH and NPP, including land management, grazing, small-scale soil variation, climate and  
356 species pool effects, micronutrient levels, predation, competition, pathogens, seed dispersal, and  
357 stochastic events or short-range phenomena such as weather-induced population dynamics (Grace et  
358 al., 2016). These other controlling factors may cause  $n_{sp}$  to be greater or smaller than average. If these  
359 other factors are constant over time, then relative changes due to variations in soil pH and NPP might  
360 still hold, but if they vary temporally then this will not be true. We therefore cannot claim to be  
361 simulating actual  $n_{sp}$  over time at any particular place, nor even relative values. The outputs of the  
362 calculations are best thought of as estimations of idealised relative change in  $n_{sp}$  over time, due to  
363 variations in soil pH and NPP, as affected only by  $N_{dep}$  and  $S_{dep}$ .

364 In our analysis climate plays only a minor role in determining  $NPP_{calc}$ . This is principally because the  
365 N14CP model assumes that NPP is controlled by a single limiting factor, which for semi-natural sites  
366 in Britain is N availability. Temperature has only a minor effect via its influence on the turnover of soil  
367 organic matter, and the associated modification of N cycling rates (Tipping et al., 2017). With regard  
368 to soil pH, the soil chemical reactions assumed in the CHUM-AM model are only slightly affected by  
369 temperature. For neither N14CP nor CHUM-AM do trends in rainfall exert much influence. Therefore  
370 our estimates of  $n_{sp}$  are hardly dependent upon climatic variation.

#### 371 *4.2. Different approaches to understanding atmospheric deposition effects*

372 Previous studies of plant species richness across Britain and in Europe have mainly used modelled  
373 estimates of  $N_{dep}$ , with some studies including  $S_{dep}$ , as explanatory variables introduced into statistical  
374 regression models (Stevens et al., 2004; Maskell et al., 2010; Dupré et al., 2010; Mitchell et al., 2018).  
375 There are difficulties with such an approach because the N and S linger in the ecosystem, so that  
376 observed  $n_{sp}$  at a given point in time may not be directly related to deposition in the same year. In the  
377 case of acidification, there is not a general, direct relationship between pollutant deposition and soil  
378 pH, since variations in soil chemistry, especially weathering rates, also control pH. Thus whilst

379 measured pH is a good predictor of  $n_{sp}$ ,  $N_{dep}$  and  $S_{dep}$  are less effective. In the case of  $N_{dep}$ , although  
380 correlations can be established between species loss and contemporary deposition rates, on a spatial  
381 basis, it is less easy to extend these to take temporal change into account. Again, the ecosystem retains  
382 deposited N which changes relatively slowly in response to changes in deposition rates. As pointed  
383 out by Stevens (2016) the use of cumulative N deposition, while taking into account the long-term  
384 enrichment of ecosystems with N, does not allow for N loss and associated ecosystem recovery to  
385 occur. This has led to the recent recommendation of a moving 30-year cumulative average (Rowe et  
386 al., 2017; Payne et al., 2019).

387 The modelling reported here is a more fundamental approach, based on process understanding using  
388 previously published studies to parameterize a particular function form. Relating richness to  
389 biogeochemical cycling permits a starting  $n_{sp}$  to be calculated, based on the acquisition of N by fixation  
390 in earlier years, which was superseded by  $N_{dep}$  during the period of the simulation (1800-2010). By  
391 distinguishing the separate dependences of  $n_{sp}$  on soil pH and  $NPP_{calc}$ , equation (2) permits the  
392 separate effects of  $N_{dep}$  and  $S_{dep}$  to be estimated. However, whereas modelling NPP at a large scale is  
393 feasible, at least in terms of averages, pH is difficult to predict, since it can be highly spatially variable.  
394 Thus the CHUM-AM modelling has depended upon calibration to individual catchments. For both  
395 variables, and biogeochemistry as a whole, large scale modelling gives only average values, which will  
396 therefore always restrict the accuracy of prediction, until such time as measured data at high  
397 resolution become available to drive biogeochemical models. With regard to how plants respond to  
398 changes in NPP and pH, we have assumed here that, although changes in NPP and pH result from  
399 gradual changes in soil properties,  $n_{sp}$  responds to them instantaneously in a given year. It seems likely  
400 that there would actually be some lag in the response (Posch et al., 2004; Clark et al., 2013; Rowe et  
401 al., 2017), but it is difficult to test this, given the high degree of scatter in the available data.

#### 402 4.3. Long-term changes in $n_{sp}$

403 The long term simulations of  $n_{sp}$  using Model C rely on the estimations of pH and NPP at the different  
404 sites over time. The pH values refer to averages over each of the catchments, while the NPP values  
405 are averages for each vegetation class over 5 km x 5 km grid cells. Changes of surface water pH  
406 calculated with CHUM-AM over 100-200 years are realistic by comparison with estimates of pH change  
407 estimated from diatom sediment records (Tipping & Chaplow, 2012). Furthermore, CHUM-AM was  
408 calibrated using data covering periods during which surface water pH changed appreciably in most  
409 cases, which strengthens belief in the calculations. That soil pH is reasonably estimated is shown from  
410 comparisons of changes in CHUM-AM-predicted values for 1978-1998 and 1978-2007 with CS  
411 measurements for similarly-acid soils, and with results from the NSI surveys (Section 3.2). Although it



412 seems logical for continued depositional inputs of N over time to have increased productivity in N-  
413 limited ecosystems, such as the grassland, heathland and woodland considered here, empirical  
414 evidence of long term change in NPP is lacking. We have previously presented evidence that modelled  
415 NPP is compatible with available measured values for sites in Britain, albeit in a correlation with high  
416 scatter, and that NPP is related to  $N_{dep}$  (Tipping et al., 2019). If space-for-time substitution holds, then  
417 it can be assumed that NPP has increased over time. Moreover, increases in soil C over periods of  
418 several decades were consistent, through the N14CP model, with a long-term N fertilization effect of  
419  $N_{dep}$  on NPP (Tipping et al., 2017).

420 The long term modelling results (Table 7, Figs. 5 and S7) show declines in  $n_{sp}$  of between 23 and 100  
421 % from an assumed pristine condition in 1800 to the latter part of the 20<sup>th</sup> Century, after which  
422 recovery took place. In 2010, the simulated  $n_{sp}$  values averaged 76 % of the 1800 ones, compared to  
423 54 % at the times of maximum species loss. Nearly all of the calculated recoveries result from the  
424 reversal of soil acidification, although in recent years slight decreases in NPP are simulated, with  
425 consequent small upturns in  $n_{sp}$ . Since peatland NPP is supposed not to depend upon  $N_{dep}$ , the  
426 recovery of  $n_{sp}$  at the peatland sites, owing to acidification reversal, is calculated to be nearly  
427 complete. Again, we emphasise that these are changes for idealised conditions, driven only by  $N_{dep}$   
428 and  $S_{dep}$ .

429 These findings are consistent with the results of plant trait analyses in British semi-natural terrestrial  
430 ecosystems by Maskell et al. (2010) and Stevens et al. (2010), who concluded that acidification had  
431 had a greater effect than eutrophication on species decline in acid grasslands and heathlands. Of the  
432 five site/vegetation examples that fall into these vegetation categories, soil pH does indeed dominate  
433 in two cases (River Etherow, Lochnagar), whereas the pH and NPP effects are about equal for the other  
434 three (Cote Gill, Mosedale Beck, Old Lodge). Furthermore, Maskell et al. concluded that  
435 eutrophication was the dominant effect in calcareous grassland, and again this is borne out by our  
436 results for the South Downs site. Our modelling results are also reasonably consistent with measured  
437 changes in  $n_{sp}$  over recent decades at sites in the UK Environmental Change Network; Rose et al. (2016)  
438 found average increases of 0.14, 0.066 and 0.059 species per year in 4 m<sup>2</sup> quadrats as the averages  
439 for lowland, upland and woodland respectively over the period 1993 to 2012 (slightly later than in our  
440 modelling). The average rate of change from the data of Table 7 is 0.046 species per year, comparable  
441 to the values of Rose et al.

442 The analysis reported here follows applications of the N14CP model to calculate long-term increases  
443 in soil C (Tipping et al., 2017) and NPP (Tipping et al., 2019), both driven by  $N_{dep}$  fertilization. Despite  
444 the coherence of the modelling and results, the evidence for these effects of  $N_{dep}$  is not yet conclusive;  
445 in particular the prediction of an increase in NPP due to fertilization by  $N_{dep}$ , although statistically

446 significant, is tenuous, owing to the extremely noisy data (Tipping et al., 2019). If the trends are  
447 correct, however, they mean that  $N_{\text{dep}}$  has promoted C storage while diminishing plant diversity, and  
448 so there is a conflict between two desirable environmental outcomes.

449 Since  $S_{\text{dep}}$  in Britain has declined so markedly in recent decades, variations in  $N_{\text{dep}}$  are likely to have  
450 the most effect on  $n_{\text{sp}}$  in the future. Our calculations suggest that a slight recovery from eutrophication  
451 has taken place already, reflecting decreases in  $N_{\text{dep}}$  in recent years. Continued  $N_{\text{dep}}$  at present levels,  
452 or further declines, would be expected to lead to further increases in  $n_{\text{sp}}$  at the N-limited non-peatland  
453 sites. This prediction differs from that of Stevens et al. (2016) who forecast continued declines in  
454 richness, based on cumulative  $N_{\text{dep}}$ . However, their conceptual model did not take account of  
455 terrestrial N turnover, and the loss of N from ecosystems under lowered  $N_{\text{dep}}$ , which are features of  
456 the N14CP model, and would also be taken into account by the use of moving average cumulative N  
457 deposition (Rowe et al., 2017; Payne et al., 2019).

#### 458 *4.4 Developments and wider applications*

459 In the work described here, we combined measured soil pH and modelled NPP, both of which are  
460 strongly affected by atmospheric pollution, as drivers of the  $n_{\text{sp}}$  model, and used spatial data for  
461 calibration. To improve this analysis, in future work, measurements of NPP at CS sites where  $n_{\text{sp}}$  is  
462 determined would provide valuable additional data for model calibration. Another possibility is to  
463 make use of NPP estimated from remote sensing, drawing on high spatial resolution mapping studies  
464 (Tebbs et al., 2017). With regard to further model testing and improved calibration in the British  
465 context, it could be informative to analyse results from the unfertilised experimental plots of the  
466 Rothamsted Park Grass experiment (Crawley et al., 2005; Silvertown et al., 2006), for which time series  
467 data covering more than 150 years on plant species diversity and soil pH are available, together with  
468 measurements of herbage mass, obtained in the management of the site by annual mowing.

469 The parameterizations of equation (2) reported here cannot be regarded as universal, but specific to  
470 the particular conditions in these British locations. To explore the wider applicability of the model,  
471 analyses might be conducted at larger scales. For example, the large set of species richness and soil  
472 pH data for the USA reported by Simkin et al. (2018) could be analysed, with modelled NPP or NPP  
473 estimated by remote sensing (Zhao et al., 2005). Another possibility is to make use of the data  
474 assembled by Fraser et al. (2015) for grasslands worldwide. These two sets cover wider ranges of  $N_{\text{dep}}$   
475 and NPP than experienced by locations in Britain, and so might include low-NPP sites where  $n_{\text{sp}}$   
476 increases with NPP. If so, a more complex form of equation (2) would be required, to take account of  
477 the unimodal relationship.

478 In the present study, to conduct temporal modelling, we took advantage of previous acidification  
479 modelling with CHUM-AM at a small number of well-calibrated catchments. To provide a more  
480 comprehensive analysis for Britain, it would be necessary to calibrate the topsoil part of CHUM-AM or  
481 another dynamic soil acidification model such as VSD (Posch and Reinds, 2009) to individual CS plots,  
482 using measured pH and deposition history. This would permit  $n_{sp}$  to be simulated at both large spatial  
483 and long temporal scales. Similar approaches could be applied more widely, at regional and perhaps  
484 global scales; the key is to relate soil pH and NPP to atmospheric deposition, which is rare in current  
485 large-scale biogeochemical models or dynamic global vegetation models. Such modelling could lead  
486 to useful forecasts of species richness responses to pollution, given appropriate future scenarios of  
487  $N_{dep}$  and  $S_{dep}$ .

488

## 489 5. Conclusions

- 490 • A model of plant species richness ( $n_{sp}$ ) based on simulated NPP and measured soil pH accounts for  
491 24 % of the variation in  $n_{sp}$  at British semi-natural grassland, heathland, peatland and woodland  
492 sites.
- 493 • The model is validated through comparison with (a) independent Countryside Survey data, both  
494 absolute  $n_{sp}$  values, and  $n_{sp}$  changes between years, (b) independent published data sets, and (c)  
495 observed recent increases in  $n_{sp}$  at sites within the UK Environmental Change Network.
- 496 • Results for nine site/vegetation cases across Britain suggest that past losses of richness were in  
497 the range 23 to 100 % (average 39 %), depending upon the extent of atmospherically deposited  
498 pollution by nitrogen and sulphur, and on soil and vegetation properties.
- 499 • In four of the nine cases,  $n_{sp}$  was affected about equally by pH and NPP, while in another four the  
500 effect of pH was dominant. The ninth site, a chalk grassland, was affected only by NPP, since soil  
501 pH was assumed constant.
- 502 • Recovery is calculated to have taken place over recent decades, largely due to acidification  
503 reversal, with an average increase of 0.046 species per year over the period 1993 to 2010.
- 504 • There are indications of recovery from the eutrophying effects of N deposition, with slight  
505 increases in predicted  $n_{sp}$  in recent years.

**506 Acknowledgements**

507 We thank UKCEH staff and CS surveyors who have helped with collection, organization and analysis of  
508 Countryside Survey data. The Countryside Survey of 2007 was funded by a partnership of nine  
509 government funded bodies led by the Natural Environment Research Council (NERC) and the  
510 Department for Environment, Food and Rural Affairs (Defra). We are grateful to U. Dragosits, E.J.  
511 Carnell, A.J. Dore, S.J. Tomlinson and M.A. Sutton (UK Centre for Ecology & Hydrology, Edinburgh) for  
512 providing modelled atmospheric deposition data, and to E.C.Rowe (UK Centre for Ecology &  
513 Hydrology, Bangor) for helpful comments on the draft manuscript. We thank the five reviewers for  
514 their helpful comments. This study was partially funded by the NERC Macronutrient Cycles Programme  
515 (Grant nos. NE/J011533/1, NE/J011703/1, NE/J011991/1). and by the NERC UK-SCaPE program  
516 delivering National Capability (Grant no. NE/R016429/1).

**517 Appendix A. Supplementary data**

518 Supplementary data to this article can be found online at  
519 <https://doi.org/10.1016/j.envpol.2021.117017>.

**520 Author contributions**

521 Edward Tipping; Conceptualization, Writing - original draft, Project administration, Investigation,  
522 Software, Methodology, Funding acquisition, Jessica A.C. Davies; Investigation, Software, Writing -  
523 review & editing, Peter A. Henrys; Investigation, Software, Writing - review & editing, Susan G. Jarvis;  
524 Investigation, Software, Writing - review & editing, Simon M. Smart; Conceptualization, Investigation,  
525 Software, Writing - review & editing.

526 **References**

- 527 Ashmore, M., Shotbolt, L., Hill, M., et al. 2004. Further Development of an Effects (Critical Loads) Based  
528 Approach for Cadmium, Copper, Lead and Zinc. Final Report for Defra EPG 1/3/188.
- 529 Azevedo, L.B., van Zelm, R., Hendriks, A.J. et al. 2013. Global assessment of the effects of terrestrial  
530 acidification on plant species richness. *Environ. Pollut.* 174, 10-15.
- 531 Bobbink, R., Hicks, K J. Galloway, J. et al. 2010. Global assessment of nitrogen deposition effects on  
532 terrestrial plant diversity: a synthesis. *Ecol. Appl.* 20, 30–59.
- 533 Chytrý, M., Danihelka, J., Ermakov, N. et al. 2007. Plant species richness in continental southern  
534 Siberia: effects of pH and climate in the context of the species pool hypothesis. *Global Ecol.*  
535 *Biogeogr.* 16 , 668–678.
- 536 Clark, C.M., Morefield, P.E., Gilliam, F.S., Pardo, L.H. 2013. Estimated losses of plant biodiversity in the  
537 United States from historical N deposition (1985–2010). *Ecology* 94, 1441–1448.
- 538 Clark, C.M., Phelan, J., Doraiswamy, P. et al. 2018. Atmospheric deposition and exceedances of critical  
539 loads from 1800-2025 for the conterminous United States. *Ecol. Appl.* 28, 978–1002.
- 540 Crawley, M.J., Johnston, A.E., Silvertown, J. et al. 2005. Determinants of species richness in the Park  
541 Grass experiment. *Amer. Naturalist* 165, 179–192.
- 542 Davies, J.A.C., Tipping, E., Rowe, E.C. et al., (2016a) Long-term P weathering and recent N deposition  
543 control contemporary plant-soil C, N, and P. *Glob. Biogeochem. Cycles* 30, 231–249.
- 544 Dupré, C., Stevens, C.J., Ranke, T. et al., 2010. Changes in species richness and composition in  
545 European acidic grasslands over the past 70 years: the contribution of cumulative atmospheric  
546 nitrogen deposition. *Global Change Biol.* 16, 344–357.
- 547 Elser, J.J., Bracken, M.E.S., Cleland, E.E. et al., 2007. Global analysis of nitrogen and phosphorus  
548 limitation of primary producers in freshwater, marine and terrestrial ecosystems. *Ecol. Lett.*  
549 10, 1135–1142.
- 550 Field, C.D., Evans, C.D., Dise, N.B. et al., 2017. Long-term nitrogen deposition increases heathland  
551 carbon sequestration. *Sci. Tot. Environ.* 592, 426–435.
- 552 Fowler, D., Ashmore, M., Cape, J.N. et al. 2012. Review of Transboundary Pollution (RoTAP)  
553 Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Centre for  
554 Ecology and Hydrology report to the Department for Environment, Food and Rural Affairs.
- 555 Fraser, L.H., Pither, J., Jentsch, A. et al., 2015. Worldwide evidence of a unimodal relationship between  
556 productivity and plant species richness. *Science* 349, 302–305.
- 557 Gough, L., Shaver, G.R., Carroll, J., Royer, D.L., Laundre, J.A. 2000. Vascular plant species richness in  
558 Alaskan arctic tundra: the importance of soil pH. *J. Ecol.* 88, 54-66.

- 559 Goulding, K.W.T. 2016. Soil acidification and the importance of liming agricultural soils with particular  
560 reference to the United Kingdom. *Soil Use Managemnt.* 32, 390–399.
- 561 Grace, J.B., Anderson T.M., Seabloom, E.W. et al. 2016. Integrative modelling reveals mechanisms  
562 linking productivity and plant species richness. *Nature* 529, 390-393.
- 563 Grime, J.P. 1973. Control of species density in herbaceous vegetation. *J. Environ. Managemnt.* 1, 151–  
564 167.
- 565 Hu, W., Xie, J., Chau, H.W., Si, B.G. 2015. Evaluation of parameter uncertainties in nonlinear regression  
566 using Microsoft Excel Spreadsheet. *Environ. Syst. Res.* 4, 4.
- 567 Huston, M.A. 2014. Disturbance, productivity, and species diversity: empiricism vs. logic in ecological  
568 theory. *Ecology* 95, 2382–2396.
- 569 Jackson, D.L. 2000. Guidance on the interpretation of the Biodiversity Broad Habitat Classification  
570 (terrestrial and freshwater types): Definitions and the relationship with other classifications,  
571 JNCC Report No. 307, JNCC, Peterborough, ISSN 0963-8091.
- 572 Janes-Bassett, V., Bassett, R., Rowe, E. et al., 2020. Changes in carbon storage over the Anthropocene:  
573 a national scale analysis of stocks in the terrestrial biosphere. *Anthropocene*, in review.
- 574 Kirk, G.J.D., Bellamy, P.H., Lark R.M. 2010. Changes in soil pH across England and Wales in response to  
575 decreased acid deposition. *Glob. Change Biol.* 16, 3111–3119.
- 576 Larssen T., Seip H.M., Semb, A. 1999. Acid deposition and its effects in China: an overview *Environ. Sci.*  
577 *Policy* 2, 9-24.
- 578 LeBauer, D.S., Treseder, K.K., 2008. Nitrogen limitation of net primary productivity in terrestrial  
579 ecosystems is globally distributed. *Ecology* 89, 371–379.
- 580 Lee, M., Manning, P., Rist, J. et al., 2010. A global comparison of grassland biomass responses to CO<sub>2</sub>  
581 and nitrogen enrichment. *Phil. Trans. R. Soc. Lond. B* 365, 2047–2056.
- 582 Maskell, L.C., Smart, S.M., Bullock, J.M. et al., 2010. Nitrogen deposition causes widespread loss of  
583 species richness in British habitats. *Glob. Change. Biol.* 16, 671–679.
- 584 Matson, P., Lohse, K.A., Hall, S.J. 2002. The globalization of nitrogen deposition: consequences for  
585 terrestrial ecosystems. *Ambio* 31, 113-119.
- 586 Mitchell, R.J., Hewison, R.L., Fielding, D.A. et al., 2018. Decline in atmospheric sulphur deposition and  
587 changes in climate are the major drivers of long-term change in grassland plant communities  
588 in Scotland. *Environ. Poll.* 235, 956-964.
- 589 Payne, R.J., Campbell, C., Britton, A.J. et al., 2019. What is the most ecologically-meaningful metric of  
590 nitrogen deposition? *Environ. Pollut.* 247, 319-331.
- 591 Peet, R.K., Fridley, J.D., Gramling, J.M. 2003. Variation in species richness and species pool size across  
592 a pH gradient in forests of the southern Blue Ridge Mountains. *Folia Geobot.* 38, 391-401.

- 593 Pepler-Lisbach, C., Kleyer, M. 2009. Patterns of species richness and turnover along the pH gradient  
594 in deciduous forests: testing the continuum hypothesis. *J. Veg. Sci.* 20, 984–995.
- 595 Posch, M., Hettelingh, M.J.-P., Slootweg, J., 2004. Dynamic modelling. In: Spranger, T., Lorenz, U.,  
596 Gregor, H. (Eds.), *Manual on Methodologies and Criteria for Modelling and Mapping Critical*  
597 *Loads & Levels and Air Pollution Effects, Risks and Trends.* Umwelt Bundes Amt (Federal  
598 Environment Agency), Berlin, pp. VI\_1–VI\_33.
- 599 Posch, M., Reinds, G.J., 2009. A very simple dynamic soil acidification model for scenario analyses and  
600 target load calculations. *Environ. Mod. Software* 24, 329-340.
- 601 Riesch, F., Stroh, H.G., Tonn, B., Isselstein J. 2018. Soil pH and phosphorus drive species composition  
602 and richness in semi-natural heathlands and grasslands unaffected by twentieth-century  
603 agricultural intensification, *Plant Ecol. Divers.* 11, 239-253.
- 604 Rodwell, J.S. 2006. *National Vegetation Classification: Users' handbook.* Joint Nature Conservation  
605 Committee, Peterborough, UK. ISBN – 13 978 1 86107 574 1. 20 pp.
- 606 Rose, R., Monteith, D.T., Henrys, P. et al., 2016. Evidence for increases in vegetation species richness  
607 across UK Environmental Change Network sites linked to changes in air pollution and weather  
608 patterns. *Ecol. Indic.* 68, 52-62.
- 609 Rowe, E.C., Jones, L., Dise, N.B. et al. 2017. Methods for evaluating the ecological benefits of decreased  
610 nitrogen deposition. *Biol. Conserv.* 212, 454–463.
- 611 Schillereff, D.N., Boyle, J.F., Toberman H. et al. 2016. Long-term macronutrient stoichiometry of UK  
612 ombrotrophic peatlands. *Sci. Tot. Environ.* 572, 1561-1572.
- 613 Schuster, B., Diekmann, M. 2003. Changes in species density along the soil pH gradient: evidence from  
614 German plant communities. *Folia Geobot.* 38, 367-379.
- 615 Simkin, S.M., Allen, E.B., Bowman, W.D. et al. 2016. Conditional vulnerability of plant diversity to  
616 atmospheric nitrogen deposition across the United States. *Proc. Natl. Acad. Sci. USA* 113,  
617 4086-4091.
- 618 Silvertown, J., Poulton, P., Johnston, E. et al. 2006. The Park Grass Experiment 1856–2006: its  
619 contribution to ecology. *J. Ecol.* 94, 801–814.
- 620 Smart, S.M., Ellison, A.M., Bunce, R.G.H. et al. 2014. Quantifying the impact of an extreme climate  
621 event on species diversity in fragmented temperate forests: the effect of the October 1987  
622 storms on British broadleaved woodlands. *J. Ecol.* 102, 1273-1287.
- 623 Stevens, C.J. 2016. How long do ecosystems take to recover from atmospheric nitrogen deposition?  
624 *Biol. Conserv.* 200, 160–167.
- 625 Stevens, C.J., Dise, N.B., Mountford, J.O., Gowing, D.J. 2004. Impact of nitrogen deposition on the  
626 species richness of grasslands. *Science* 303, 1876–1879.



- 627 Stevens, C.J., Payne, R.J., Kimberley A., Smart, S.M. 2016. How will the semi-natural vegetation of the  
628 UK have changed by 2030 given likely changes in nitrogen deposition? *Environ. Pollut.* 208,  
629 879-889.
- 630 Stevens, C.J., Thompson, K., Grime, J.P. et al. 2010. Contribution of acidification and eutrophication to  
631 declines in species richness of calcifuge grasslands along a gradient of atmospheric nitrogen  
632 deposition. *Funct. Ecol.* 24, 478-484.
- 633 Sutton, M.A., Howard, C.M., Erisman, J.W. et al. (Eds.), 2011. *The European Nitrogen Assessment:  
634 Sources, Effects and Policy Perspectives.* Cambridge University Press, Cambridge.
- 635 Tebbs, E.J., Rowland, C.S., Smart, S.M. 2017. Regional-scale high spatial resolution mapping of  
636 aboveground net primary productivity (ANPP) from field survey and landsat data: a case study  
637 for the Country of Wales. *Remote Sens.* 9, 801.
- 638 Tipping, E., Chaplow, J.S. 2012. Atmospheric pollution histories of three Cumbrian surface waters.  
639 *Freshwat. Biol.* 57, 244–259.
- 640 Tipping, E., Davies, J.A.C., Henrys, P.A. et al. 2017. Long-term increases in soil carbon due to ecosystem  
641 fertilization by atmospheric nitrogen deposition demonstrated by regional scale modelling  
642 and observations. *Sci Rep* 7:1890
- 643 Tipping, E., Davies, J.A.C., Henrys, P.A. et al. 2019. Measured estimates of semi-natural terrestrial NPP  
644 in Great Britain: comparison with modelled values, and dependence on atmospheric nitrogen  
645 deposition. *Biogeochem.* 144, 215–227.
- 646 Tipping, E., Lawlor, A.J., Lofts, S. 2006. Simulating the long-term chemistry of an upland UK catchment:  
647 Major solutes and acidification. *Environ. Pollut.* 141, 151–166.
- 648 Tipping, E., Rothwell, J.J., Shotbolt, L., Lawlor, A.J. 2010. Dynamic modelling of atmospherically-  
649 deposited Ni, Cu, Zn, Cd and Pb in Pennine catchments (northern England). *Environ. Pollut.*  
650 158, 1521–1529.
- 651 Tipping, E., Yang, H., Lawlor, A.J. et al. 2007. Trace metals in the catchment, loch and sediments of  
652 Lochnagar: measurements and modelling. In: *Lochnagar: The Natural History of a Mountain  
653 Lake* (Ed. N.L. Rose ), pp. 345–373. Springer, Dordrecht.
- 654 Toberman, H., Tipping, E., Boyle, J.F. et al. 2015. Dependence of ombrotrophic peat nitrogen on  
655 phosphorus and climate. *Biogeochem.* 125, 11–20.
- 656 Van Breemen, N., Driscoll, C.T., Mulder, J. 1984. Acid deposition and internal proton sources in  
657 acidification of soils and waters. *Nature* 307, 599–604.
- 658 Vitousek, P. M., Aber, J.D., Howarth, R.W. 1997. Human alteration of the global nitrogen cycle: sources  
659 and consequences. *Ecol. Appl.* 7, 737–750.

- 660 Vonlanthen, C.M., Kammer, P.M., Eugster, W. et al. 2006. Alpine vascular plant species richness: the  
661 importance of daily maximum temperature and pH. *Plant Ecol.* 184,13 –25.
- 662 Wagenmakers, E.-J., Farrell,S. 2004. AIC model selection using Akaike weights *Psych. Bull. Rev.* 11, 192-  
663 196.
- 664 Zhao, M. T, Heinsch, F.A., Nemani, R.R., Running, S.W. 2005. Improvements of the MODIS terrestrial  
665 gross and net primary production global data set. *Rem. Sens. Environ.* 95, 164–176.

666 Table 1. Summary of Countryside Survey field data; the values are the numbers of plots sampled for  
 667 species richness and soil pH in each vegetation class, by year of sampling and quadrat size.

668

Vegetation class	Year	1978	1998	2007	2007
	Quadrat m <sup>2</sup>	200	200	4	200
grassland		90	81	573	573
heathland		218	226	189	189
peatland		96	216	223	223
woodland		43	58	171	171

669

670 Table 2. Catchments for which CHUM-AM has been calibrated, and used for simulation of long-term variation in  $n_{sp}$ . The total deposition values ( $\text{g m}^{-2} \text{a}^{-1}$ )  
 671 (non-marine for  $S_{dep}$ ) refer to non-tree vegetation, and are given for years when deposition rates were approximately maximal. The references are to studies  
 672 in which topsoil pH was modelled; soil chemistry was not modelled for the chalk soil of the South Downs.

673

Catchment	Location	Area km <sup>2</sup>	Vegetation	N <sub>dep</sub>		S <sub>dep</sub>	Reference
				1990	1970		
Cote Gill	Central Pennines	5.0	grassland	2.9	6.0	Tipping et al., 2010	
River Etherow	South Pennines	13.0	peatland & grassland	3.4	6.0	Tipping et al., 2010	
Great Dun Fell	North Pennines	0.2	peatland	2.3	2.3	Tipping et al., 2010	
Lochnagar	Cairngorms	0.9	heathland	0.8	2.1	Tipping et al., 2007	
Mosedale Beck	Lake District	3.7	grassland	3.2	5.2	Tipping and Chaplow, 2012	
Old Lodge	Ashdown Forest	2.4	heathland & woodland	2.3	2.1	Ashmore et al., 2004	
-	South Downs	-	grassland	2.4	-		

674

675

676 Table 3. Comparison of different versions of the equation(2) model, fitted to data for separate vegetation classes. Key:  $n_{\text{param}}$  = number of parameters, SS =  
 677 sum of squared deviations, AIC = Akaike Information Criterion,  $\Delta\text{AIC}$  = difference in AIC between a model and the best model,  $w_{\text{AIC}}$  = AIC weight.

Version	Parameters fitted	$n_{\text{param}}$	Grassland (n = 573)			Heathland (n = 189)			Woodland (n = 171)		
			SS	$\Delta\text{AIC}$	$w_{\text{AIC}}$	SS	$\Delta\text{AIC}$	$w_{\text{AIC}}$	SS	$\Delta\text{AIC}$	$w_{\text{AIC}}$
1	$\alpha, \beta, \gamma, \text{pH}_0$	4	18935	0.27	0.466	2341	1.20	0.340	3981	0.00	0.797
2	$\alpha, \gamma (\beta = 0, \text{pH}_0 = 0)$	2	23395	117.5	0.000	3079	49.00	0.000	4540	18.50	0.000
3	$\alpha, \beta, \text{pH}_0 (\gamma = 0)$	3	19514	15.54	0.000	2420	5.48	0.040	4326	12.23	0.002
4	$\alpha, \gamma, \text{pH}_0 (\beta = 1)$	3	18992	0.00	0.534	2351	0.00	0.620	4093	2.75	0.201

678

679

680 Table 4. Parameters with model version 1 from Table 3, fitted to vegetation classes separately. The  
 681 value of  $\gamma$  for peatland was assumed to be the average of the values for the other three vegetation  
 682 classes.

Parameter	Grassland	Heathland	Woodland	Peatland
$\alpha$	10.4	8.1	15.2	10.3
$\beta$	0.557	0.677	0.297	0.573
$10^3 \gamma$	-1.23	-0.92	-1.04	-1.06
$\text{pH}_0$	2.89	2.90	3.30	3.04

683

684

685

686 Table 5. Results of fitting equation (2) to the combined data set ( $n = 1156$ ). See Table 3 for key to  
 687 abbreviations.

Model	Parameters fitted	$n_{\text{param}}$	SS	$\Delta\text{AIC}$	$W_{\text{AIC}}$
A	$\alpha, \beta, \gamma, \text{pH}_0$ for each class	15	28100	8.95	0.08
B	common $\alpha, \beta, \gamma, \text{pH}_0$	4	28472	2.15	0.252
C	$\alpha$ for each class; common $\beta, \gamma, \text{pH}_0$	7	28272	0.00	0.739

688

689

690

691 Table 6. Parameters adopted for predictive modelling with model C. The standard errors were  
 692 obtained from Monte Carlo analysis (Section 2.4). SE = standard error.

693

parameter	value	SE
$\alpha$ grassland	11.7	1.3
$\alpha$ heathland	11.2	1.3
$\alpha$ peatland	11.6	1.3
$\alpha$ woodland	13.9	2.0
$\beta$	0.458	0.070
$10^3 \gamma$	-1.12	0.097
$\text{pH}_0$	3.20	0.25

694

695 Table 7. Summary of long-term simulations of average  $n_{sp}$ . Recent increases in  $n_{sp}$  refer to the period  
 696 1993-2010.

697

Site	Vegetation	$n_{sp}$ 1800	minimum $n_{sp}$	$n_{sp}$ 2010	maximum fractional decrease	recent increase $n_{sp}$ yr <sup>-1</sup>
Cote Gill	grassland	13.2	7.8	9.3	0.41	0.047
River Etherow	grassland	8.0	0.0	5.3	1.00	0.083
River Etherow	peatland	6.8	0.0	6.1	1.00	0.095
GDF	peatland	8.7	6.1	8.5	0.30	0.067
Lochnagar	heath	9.5	7.3	8.1	0.23	0.017
Mosedale Beck	grassland	13.0	7.5	8.5	0.42	0.038
Old Lodge	heathland	6.4	3.5	4.7	0.45	0.034
Old Lodge	woodland	7.8	3.6	4.8	0.54	0.032
South Downs	grassland	18.7	14.1	14.5	0.25	0.000

698

699 **Figure captions**

700 Fig. 1. Location map, showing the sites for which CHUM-AM model has been calibrated.

701 Fig. 2. Averaged data for samples from 4 m<sup>2</sup> quadrats in 2007. The numbers of values averaged were:  
702 grassland 24, heathland 14, peatland 15, woodland 13. The error bars are standard deviations; when  
703 they are not visible in the x-axis data, they fall within the points.

704 Fig. 3. Observed  $n_{sp}$  vs  $n_{sp}$  calculated with Model C, for 4 m<sup>2</sup> quadrats, sampled in the 2007 Countryside  
705 Survey. Results for all four vegetation classes are shown. The 1:1 line is shown.

706 Fig. 4. Observed  $n_{sp}$  for grassland published by Grime (1973) for two dales in Derbyshire, and for the  
707 British survey of Stevens et al. (2004), vs values predicted with Model C, compared with the CS data  
708 for 2007 (from Fig. 3). Linear regression results: Lathkilldale, slope = 1.76,  $r^2_{adj} = 0.42$ ,  $p < 0.0001$ ,  $n =$   
709 39; Cressbrookdale, slope = 3.07,  $r^2_{adj} = 0.71$ ,  $p < 0.0001$ ,  $n = 34$ ; Stevens' survey, slope = 1.71,  $r^2_{adj} =$   
710 0.64,  $p < 0.0001$ ,  $n = 61$ .

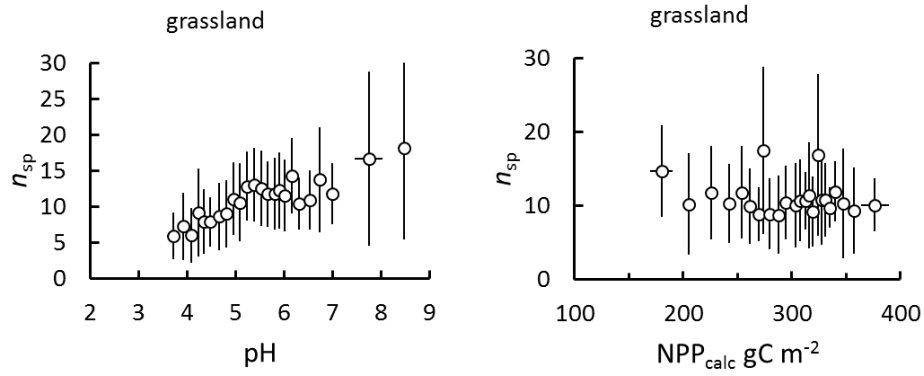
711 Fig. 5. Long-term variations in soil pH,  $NPP_{calc}$  and calculated  $n_{sp}$  (4 m<sup>2</sup> quadrats) at four sites. In the  
712 right-hand  $n_{sp}$  panels, the thick solid line refers to variations driven by both pH and  $NPP_{calc}$ , the thin  
713 solid line is for constant pH (1800 value) and the dotted line is for constant  $NPP_{calc}$  (1800 value).  
714 Constant pH was assumed for the South Downs site, because the chalk soil has high buffering capacity  
715 (Section 3.3). Plots for the other five site/vegetation combinations are shown in Fig. S7.



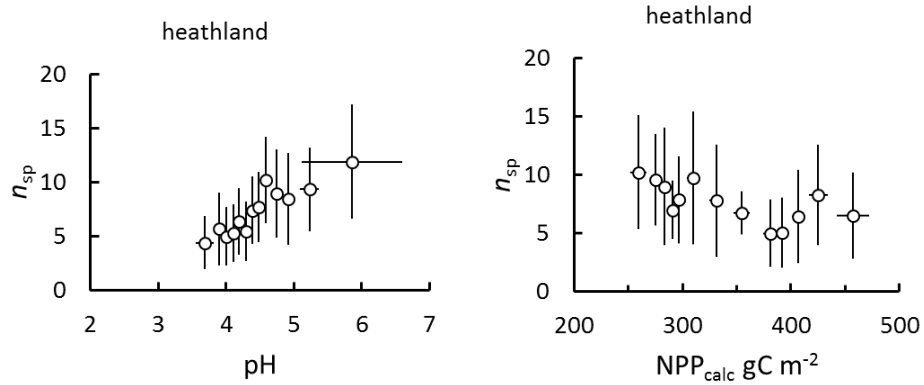


716

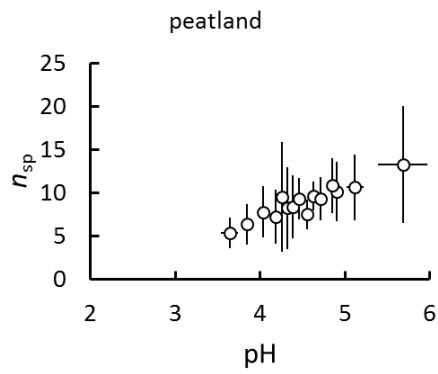
717 Fig. 1



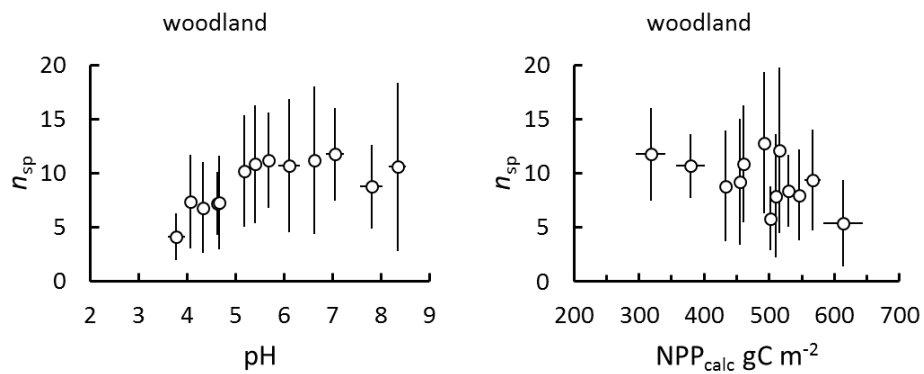
718



719

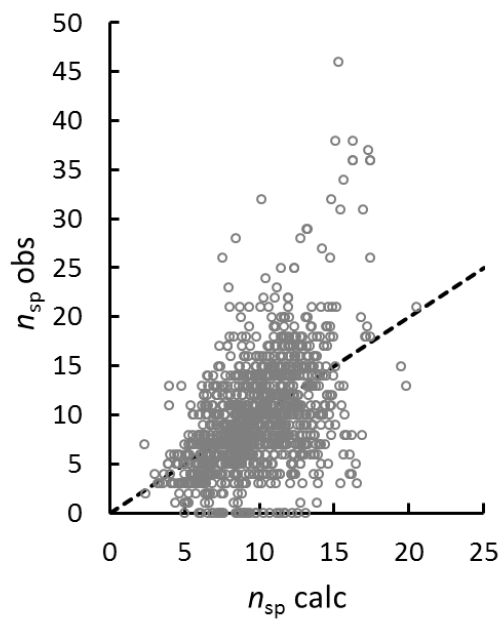


720



721

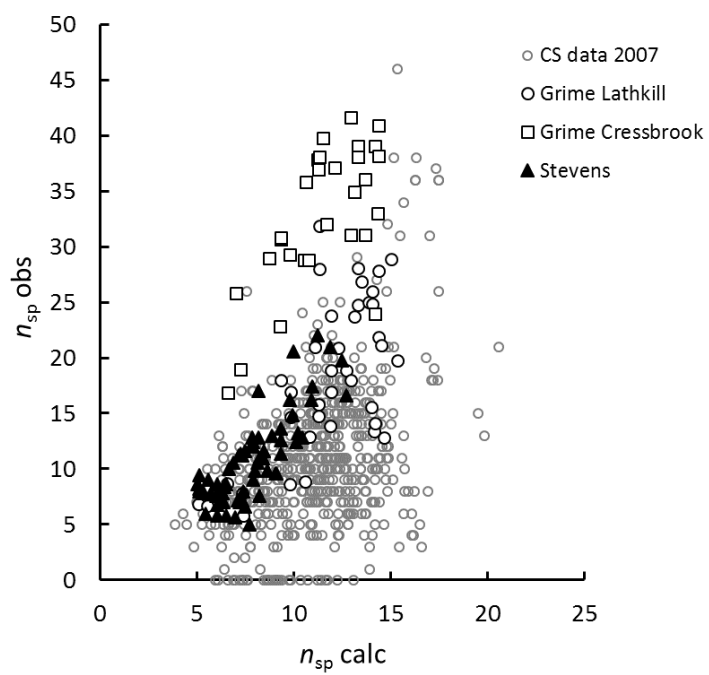
722 Fig. 2.



723

724 Fig. 3.

725

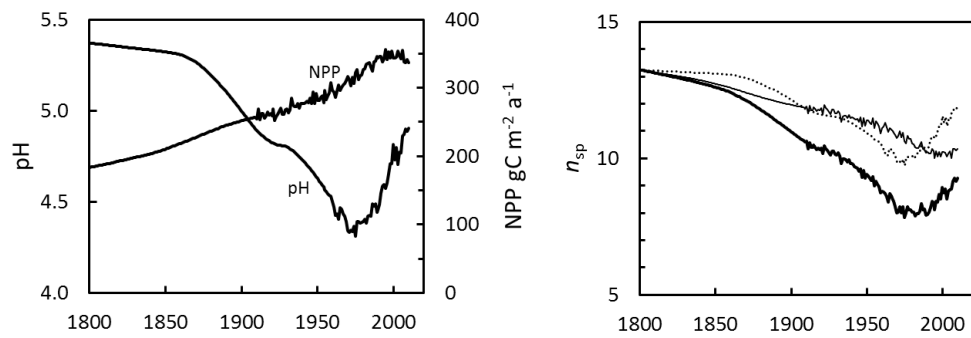


726

727 Fig. 4.

728

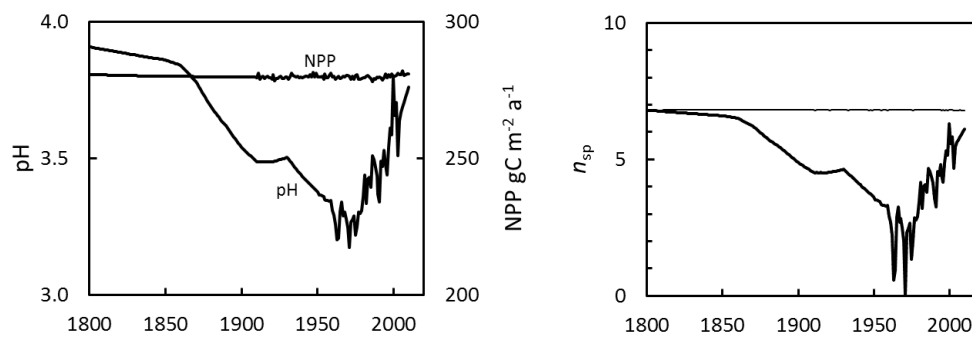
## Cote Gill (grassland)



729

730

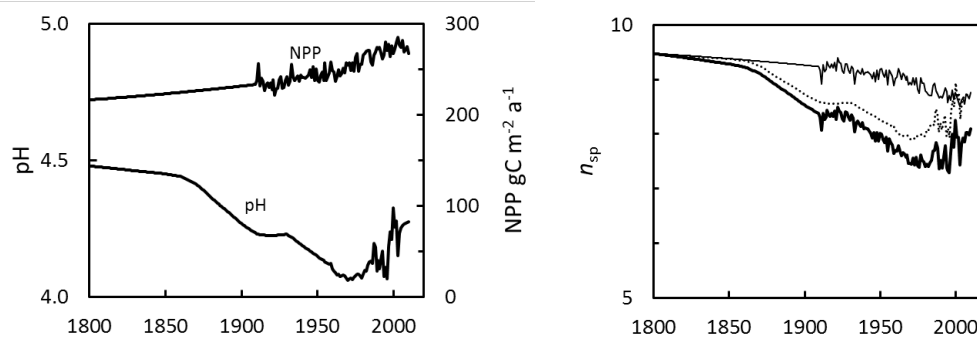
## River Etherow (peatland)



731

732

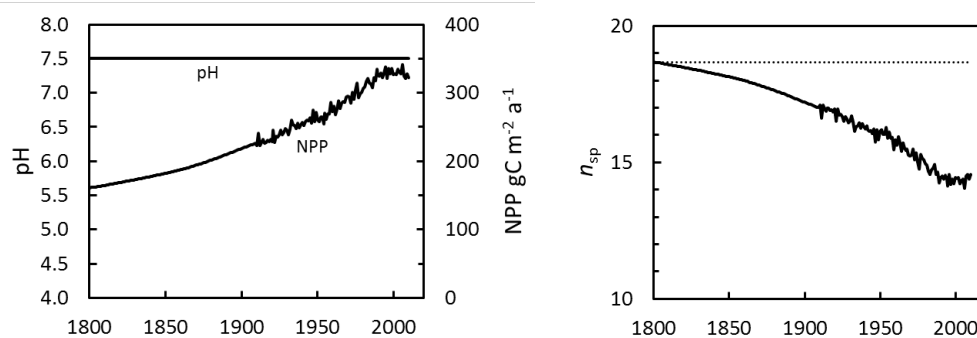
## Lochnagar (heathland)



733

734

## South Downs (grassland)



735

736 Fig. 5