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1 **What is the most ecologically-meaningful metric of nitrogen deposition?**

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18

19 **ABSTRACT**

20 Nitrogen (N) deposition poses a severe risk to global terrestrial ecosystems, and managing  
21 this threat is an important focus for air pollution science and policy. To understand and  
22 manage the impacts of N deposition, we need metrics which accurately reflect N deposition  
23 pressure on the environment, and are responsive to changes in both N deposition and its  
24 impacts over time. In the UK, the metric typically used is a measure of total N deposition  
25 over 1-3 years, despite evidence that N accumulates in many ecosystems and impacts from  
26 low-level exposure can take considerable time to develop. Improvements in N deposition  
27 modelling now allow the development of metrics which incorporate the long-term history of  
28 pollution, as well as current exposure. Here we test the potential of alternative N deposition  
29 metrics to explain vegetation compositional variability in British semi-natural habitats. We  
30 assembled 36 individual datasets representing 48,332 occurrence records in 5,479 quadrats  
31 from 1,683 sites, and used redundancy analyses to test the explanatory power of 33  
32 alternative N metrics based on national pollutant deposition models. We find convincing  
33 evidence for N deposition impacts across datasets and habitats, even when accounting for  
34 other large-scale drivers of vegetation change. Metrics that incorporate long-term N  
35 deposition trajectories consistently explain greater compositional variance than 1-3 year N  
36 deposition. There is considerable variability in results across habitats and between similar  
37 metrics, but overall we propose that a thirty-year moving window of cumulative deposition is  
38 optimal to represent impacts on plant communities for application in science, policy and  
39 management.

40 KEYWORDS: air pollution; biodiversity; cumulative deposition; vegetation; community  
41 ecology; environmental change; nitrogen deposition.

42 CAPSULE: Measures of nitrogen deposition which incorporate long-term pollution history  
43 explain more spatial variance in plant communities than those which do not.

44 HIGHLIGHTS:

- 45 • We present a large study of N deposition impacts on British vegetation.
- 46 • N deposition consistently explains spatial variability in vegetation composition.
- 47 • Metrics based on long-term pollution histories are superior to current deposition.
- 48 • We propose thirty-year cumulative deposition as an optimum metric.

49

## 50 INTRODUCTION

51 Nitrogen (N) deposition is recognised as one of the most severe threats to ecosystems,  
52 arguably exceeded only by climate and land-use change as a hazard to global terrestrial  
53 biodiversity (Bobbink et al., 2010; Dise et al., 2011; Sala et al., 2000). The global budget of  
54 reactive compounds of N is now dominated by anthropogenic production and, while  
55 emissions and deposition are beginning to plateau and decline in some developed countries,  
56 N deposition is rapidly increasing in the developing world (Fowler et al., 2013; Fowler et al.,  
57 2015; Galloway et al., 2004; Galloway et al., 2008).

58 Nitrogen deposition impacts terrestrial ecosystems through multiple mechanistic pathways.  
59 At high concentrations nitrogen, particularly as gaseous ammonia and aerosols, can cause  
60 direct toxic effects on plants and other organisms (Cape et al., 2009; Pearson and Stewart,  
61 1993). N deposition to soils may lead to acidification, base cation depletion and mobilisation  
62 of potentially toxic metals (Bowman et al., 2008; Horswill et al., 2008). Nitrogen deposition  
63 can also increase the susceptibility of organisms to secondary stressors such as climatic  
64 extremes, pathogens and predators (Carroll et al., 1999; Mitchell et al., 2003; Throop and  
65 Lerdau, 2004). However, the impact-pathway which has attracted greatest attention is  
66 eutrophication. Increased nutrient supply may shift the competitive balance between  
67 species, ultimately leading to the exclusion of taxa that are poor competitors for resources  
68 (Bobbink et al., 2010; Wedin and Tilman, 1993). The consequences of these combined  
69 impacts include loss of biodiversity, changed taxonomic and trait assemblages and erosion  
70 of important ecosystem services, ultimately imposing significant societal costs (Jones et al.,  
71 2014; Stevens et al., 2006; Stevens et al., 2010).

72 Managing the environmental impacts of N deposition is an important concern for  
73 environmental policy-makers, managers and regulators. Common roles include the  
74 permitting of new industrial and agricultural emission sources, legislating on appropriate  
75 technologies and monitoring and reporting of impacts to national and international bodies.  
76 These roles require *metrics* of N deposition which reflect its pressure on the environment.  
77 Currently the metric used in most applications is 'current deposition', usually defined over a  
78 period of 1-3 years. In the United Kingdom current N deposition is typically estimated using  
79 the empirical Concentration Based Estimated Deposition model (CBED) (Smith et al., 2000).  
80 CBED output is produced annually based on measured pollutant concentrations, wet  
81 deposition and meteorological data, and is available as both single-year and three-year  
82 means (the latter intended to smooth-out meteorological variation). These 'current  
83 deposition' data are used as part of the UK's national-scale reporting and to assess  
84 'background' deposition when considering the impact of additional pollution in permit  
85 applications (Hall et al., 2017). Current deposition data produced in similar ways are also  
86 used internationally by environmental managers and policy-makers.

87 There are a number of reasons why current deposition data may not optimally represent N  
88 deposition as it affects the environment. Experimental studies show that N deposition  
89 impacts take considerable time to develop (Phoenix et al., 2012). Many long-term studies  
90 have shown hysteresis over 1-3 years but have ultimately shown large change over time-  
91 periods of a decade or more (Clark and Tilman, 2008). Similarly, some studies of recovery  
92 have shown limited recovery when N additions are ceased (Isbell et al., 2013). Nitrogen  
93 deposition increases N stocks and concentrations in soil and plant tissue, and increases  
94 primary production, leading to greater N in above- and below-ground pools (Meter et al.,

95 2016; Pornon et al., 2018; Rowe et al., 2014). Nitrogen deposition therefore tends to have  
96 *cumulative* impacts as these pools build up over time. Time-scales of species response will  
97 depend on the autecology of the species concerned and may vary dependent on their  
98 nitrogen sources and those of their competitors. Short-term modelled N deposition estimates  
99 will also be affected by atmospheric conditions during that period, particularly in terms of  
100 precipitation, and may not always correlate well with longer time-periods. The ecological  
101 impacts of N deposition are primarily long-term processes which are likely to be imperfectly  
102 characterised over a period of less than three years.

103 An alternative framework is to consider N deposition over a much longer period. Studies  
104 synthesising experimental results through time have found that a strong basis for doing this  
105 is by calculating the *total accumulated dose* of nitrogen, including both experimental  
106 treatments and background deposition (Phoenix et al., 2012). Similarly, studies investigating  
107 the impacts of N deposition in the landscape have included cumulative atmospheric  
108 deposition as an explanatory variable (Duprè et al., 2010; Payne et al., 2011). However, a  
109 limitation to previous cumulative N deposition calculations has been that they are typically  
110 based on re-scaling current deposition values using national scaling factors (Duprè et al.,  
111 2010; Fowler et al., 2005). In theory this produces a metric which is fully correlated with  
112 current deposition and therefore adds no independent predictive power (Rowe et al., 2014).  
113 In practice cumulative deposition is often calculated from a different baseline (typically 1996-  
114 98 in the United Kingdom) and may include measured data for the recent past (Payne, 2014;  
115 Payne et al., 2017), meaning that correlations are weaker (Payne et al., 2011).  
116 Nevertheless, it is clear that cumulative deposition calculations have been unable to fully  
117 account for the changing spatial distribution of N deposition over time. In the UK this  
118 situation has now been changed by the development of better modelling of long-term N  
119 deposition. Recent work has estimated spatially distributed historic N emissions back to  
120 1800 and used FRAME (Fine Resolution Atmospheric Multipollutant Exchange: (Dore et al.,  
121 2012; Dore et al., 2007; Matejko et al., 2009)), an atmospheric chemistry and transport  
122 model, to produce estimates of N deposition (Dragosits et al., 2016; Tipping et al., 2017).  
123 There is the potential to build on this to produce a range of indices of N deposition that more  
124 realistically represent long-term N deposition as it affects the environment. However, it is  
125 unclear which of many possible options would be most appropriate. Rowe et al. (2014) and  
126 Rowe et al. (2017) have proposed thirty years of cumulative deposition above the critical  
127 load as a useful measure of N deposition pressure for 'soil based ecosystems'. However this  
128 is not currently based on any empirical analysis.

129 The aim of this study is to test the explanatory power of alternative metrics of N deposition  
130 with large vegetation datasets in order to propose an optimal metric.

## 131 **METHODS**

132 In order to quantify the power of alternative potential metrics of N deposition we compiled  
133 multiple large-scale vegetation datasets, calculated alternative N metrics based on long-term  
134 deposition trajectories, and used ordination to test the explanatory power of these metrics in  
135 explaining vegetation assemblage variability while accounting for other potential controls on  
136 vegetation. We addressed the impacts of N deposition on semi-natural vegetation, focussing  
137 on Great Britain (GB) due to the recent development of long-term N deposition modelling,  
138 strong gradients in N deposition and availability of large-scale vegetation datasets.

139 **Vegetation data**

140 We first assembled large-scale vegetation datasets. In order to be included, datasets  
141 required large-scale spatial coverage, species-level plant identification, and precise  
142 locational information (the latter excludes some ecological surveillance datasets). We  
143 ultimately identified 11 studies and 36 individual datasets which met these criteria and were  
144 available for this project (Figure 1; Table 1). These datasets have been produced for a range  
145 of purposes including classifying vegetation types, quantifying temporal change and  
146 identifying N deposition impacts and indicators. Partly due to these varying motivations the  
147 datasets also differed in terms of when the survey was conducted, quadrat size, the  
148 grouping of species and the specificity of the habitats targeted (Table 1). Given these  
149 differences, the combination of individual datasets into larger datasets is fraught with  
150 complexity and we considered it more practical to analyse them separately.

151 We made a number of adjustments to the original datasets prior to analysis. We first aimed  
152 to focus our analysis on meaningful habitat datasets. Studies conducted in the context of  
153 understanding air pollution impacts have often been targeted at specific vegetation  
154 communities, often a single UK National Vegetation Classification (NVC) category. However,  
155 other datasets are much broader in their coverage, including studies which have deliberately  
156 aimed to maximise the range of habitats sampled. In these latter datasets the degree of  
157 replication within a specific NVC category is often limited. For each dataset we made a  
158 decision regarding the maximum degree of habitat specificity which would still allow  
159 adequate sample size. We ultimately focussed our analysis on datasets with differing  
160 taxonomic resolution for the differing surveys, ranging from the specific (e.g. 'U4 acid  
161 grasslands' for the Stevens et al. (2004) dataset) to the general (e.g. 'all grasslands' for the  
162 Ross et al. (2012) dataset). Some of the datasets comprised re-surveys of older datasets  
163 and for these we focused solely on the re-survey component.

164 We next aimed to focus our analyses at a spatial scale which was meaningful for the  
165 identification of N deposition impacts. Although N sources can sometimes have very  
166 localised impacts, most impacts are diffuse and widely distributed. UK national pollutant  
167 deposition models typically have an output resolution of 5 km x 5 km, making it impossible to  
168 attribute finer-scale plant community variability to N deposition. Most of the datasets we  
169 considered are based on designs with a number of quadrats (typically 4-5) positioned in a  
170 relatively small 'site' (often <1 ha) which will typically fall within a single model cell. For these  
171 datasets we analysed mean vegetation cover data for each such site. However, other  
172 datasets –particularly those originally designed for vegetation classification– are based on  
173 quadrats which may be widely scattered across the landscape. For these datasets we  
174 aggregated data by calculating the mean species coverage of all quadrats within the 5 km x  
175 5 km cells of the .deposition datasets.

176 The total pool of analysed data represents 48,332 occurrence records in 5,479 quadrats  
177 from 1,683 sites (Table 1). For discussion we categorised the individual datasets into five  
178 groups: heathlands, grasslands, wetlands, montane (encompassing alpine heaths and  
179 grasslands) and sand dune habitats (Table 2). The majority, but not all, datasets included  
180 species composition of all plants including bryophytes, lichens and vascular species.

181 **Nitrogen deposition modelling and data**

182 We developed a range of potential N deposition metrics for each location using recently-  
183 developed hind-casted deposition modelling for the UK based on spatially distributed historic  
184 N emissions data and the FRAME model (Dragosits et al., 2016; Tipping et al., 2017) The  
185 FRAME model is an atmospheric chemistry transport model which simulates the emissions  
186 of nitrogen compounds, their vertical diffusion and horizontal transport, atmospheric  
187 chemical transformation and deposition to the surface by wet and dry processes. N  
188 deposition modelling for this study was based on ground coverage of low-growing semi-  
189 natural species, as suited to the habitats considered (N deposition estimates for woodland  
190 are generally higher, due to a higher deposition velocity, notably for NH<sub>3</sub>). The underlying  
191 emissions data is currently available for six time-steps: 1800, 1900, 1950, 1970, 1990 and  
192 2010. These years were selected based on data availability and likely changes in air  
193 pollution, including initial industrial development (19<sup>th</sup> century), the advent and widespread  
194 implementation of the Haber-Bosch process (first half of 20<sup>th</sup> century), the peak in emissions  
195 (late 20<sup>th</sup> century) and subsequent decline. Based on this modelling, we produced grid-cell  
196 specific deposition chronologies for all 5 km x 5 km cells containing vegetation data with  
197 changes between the six tie-points calculated using linear interpolation. We compared these  
198 results to current deposition, as estimated using the standard CBED model used in UK  
199 policy and management. Given the broad spatial and temporal scope of the study we  
200 focused on total N deposition, accepting that somewhat different effects may be produced by  
201 reduced and oxidised forms of N, and by dry and wet deposition (Sheppard et al., 2011;  
202 Stevens et al., 2011; Van den Berg et al., 2008; van den Berg et al., 2016).

### 203 **Nitrogen deposition metrics**

204 We calculated a number of N deposition metrics based on alternative approaches to  
205 summarising the grid-cell deposition chronologies across the available time-steps. We first  
206 considered cumulative N deposition from a static starting-point, an approach used in a  
207 number of previous studies (Payne et al., 2011; Stevens et al., 2016). We considered five  
208 variants based on each of the available time-steps, i.e. cumulative deposition from 1800,  
209 1900, 1950, 1970 and 1990 up to the time of vegetation survey. These metrics –in which  
210 values can only increase over time– reflect the possibility that deposited N gradually  
211 accumulates in ecosystems producing progressively intensifying impacts, while regime-shifts  
212 mean that rapid recovery in vegetation composition is unlikely in at least the medium term  
213 (Isbell et al., 2013; Payne et al., 2017). Cumulative deposition was calculated from the  
214 deposition chronologies using the trapezoidal area method based on all available time-steps  
215 between the start year and the latest year of survey.

216 We next considered a moving window of cumulative N deposition, with deposition calculated  
217 for the years preceding vegetation survey. We assessed metrics based on cumulative  
218 deposition over windows of 5, 10, 20, 30, 50, 100, 150 and 200 years. These metrics reflect  
219 the accumulation of N in ecosystems over time but also the expectation that recovery will  
220 occur if deposition is reduced. N is likely to be gradually lost from ecosystems over time (due  
221 to denitrification, fire, grazing, leaching etc.) but there is uncertainty in the speed of  
222 ecological recovery due to factors such as the loss of seed-banks, leading to hysteresis  
223 (Basto et al., 2015). Such a moving window of deposition has been suggested as a useful  
224 indicator of N deposition pressure in policy (Rowe et al., 2017; Rowe et al., 2014). Linear  
225 interpolation was used to calculate deposition at the beginning and end of moving window  
226 periods and cumulative deposition calculated based on the trapezoidal area method.

227 Our third group of metrics was related, but included the critical load as a threshold; metrics  
228 were calculated based on cumulative deposition above the critical load. These alternatives  
229 embed the assumption that the critical load achieves its stated purpose of being a 'floor'  
230 below which there are no impacts. In this formulation it is only cumulative deposition *above*  
231 *the critical load* that is likely to have ecological impacts. One example of this class of metrics  
232 is the '30-year cumulative deposition above critical load' metric recently proposed by Rowe  
233 et al. (2014). Critical load values used in these calculations were based on current UNECE  
234 values (Bobbink and Hettelingh, 2011) valid for the UK, using the lowest point of range as  
235 generally implemented in UK policy. Where the vegetation communities sampled spanned  
236 habitats with different critical loads, we selected the lowest value. Linear interpolation was  
237 used to calculate the year at which critical load was first exceeded and, where deposition fell  
238 sufficiently, last exceeded, and cumulative deposition calculated as above.

239 A related alternative metric is to simply consider the number of years that the critical load is  
240 exceeded. The assumption here is that it is the *duration* of damaging quantities of N  
241 deposition which is the key attribute associated with ecological impacts, rather than the  
242 loading *per se*. Linear interpolation was used to identify the timing of first and last (where  
243 applicable) critical load exceedance, and the time-period between these points was  
244 calculated. We finally considered the maximum and minimum N deposition that a grid-cell  
245 has received in the modelled period. These metrics reflect the possibility that plant  
246 community variability may be best explained by the greatest or least N deposition pressure  
247 that the ecosystem has received over an extended time period.

248 Within these general classes there is an almost limitless diversity of metrics that could be  
249 calculated, but since most are strongly conceptually linked and highly correlated, we  
250 focussed on the 33 metrics listed in Table 3. We compared the explanatory power of these  
251 metrics for UK vegetation to those of current N deposition based on the CBED model (Smith  
252 et al., 2000), as currently used in most UK science and management. We considered both  
253 single- and three-year mean deposition values.

## 254 **Ordination**

255 We tested the link between vegetation community composition and N deposition metrics  
256 using (partial) redundancy analysis (RDA)(van den Wollenberg, 1977). RDA is an extension  
257 of principal components analysis (PCA) which attempts to summarise the variation in a set of  
258 multivariate response variables attributable to one or more explanatory variables. Partial  
259 RDA extends classical RDA by attempting to remove the effect of ('partial out') one or more  
260 co-variates (Borcard et al., 1992). We implemented RDA in R using the function *rda* in the  
261 *vegan* package (Oksanen et al., 2007; R Development Core Team, 2014). Vegetation data  
262 were Hellinger-transformed prior to analysis (Legendre and Gallagher, 2001; Rao, 1995) to  
263 allow the use of RDA in situations where species may be expected to show unimodal  
264 responses to their environment (Legendre and Gallagher, 2001). The significance of results  
265 was assessed by permutation tests (999 permutations) and summarised in terms of  
266 explained variance and P-value. Our analyses focused on overall vegetation composition,  
267 accepting that different metrics may be appropriate for different species and plant functional  
268 types.

269 We took three complementary strategies to account for other environmental factors which  
270 might affect vegetation composition in these habitats. We first tested the explanatory power



271 of each N deposition metric as sole predictor of plant community composition. This test  
272 quantifies the maximum proportion of variance which may be explained by each metric,  
273 ignoring the fact that some of this apparent relationship may actually be driven by other,  
274 correlated, variables. In our second test we made decisions on what are likely to be other  
275 important variables for which we have data. We included climate variables (mean annual  
276 precipitation: MAP, and mean annual temperature: MAT, both from the Hijmans et al. (2005)  
277 dataset), altitude (from the Shuttle Radar Tomography Mission dataset of Farr et al. (2007))  
278 and 'historic peak' S deposition (86-88 data from the CBED model of Smith et al. (2000)) as  
279 covariates in all of these analyses. These analyses with covariates partialled out provide a  
280 more realistic quantification of explained variance but results are partially determined by a  
281 *priori* judgements of likely importance. In our final set of tests we also included covariates but  
282 with these selected on statistical grounds, rather than prior expectations. In these tests we  
283 used a larger pool of potential covariates including the environmental data used above  
284 (MAT, MAP, Altitude, peak S deposition) but also other variables where available. Some  
285 datasets included considerable contextual environmental data, but these were not available  
286 for all datasets. We included all available environmental variables with a conceivable link to  
287 large-scale vegetation variability in a pool of variables available for selection for each  
288 dataset. We used the automated model-building approach of the *ordistep* function in *vegan*  
289 (Oksanen et al., 2007) to construct an optimum model by stepwise selection of variables,  
290 with variables alternately removed and added until the model remained unchanged.  
291 Inclusion decisions were made on the basis of permutation-based significance tests (999  
292 permutations). Stepwise selection was conducted using all environmental variables -other  
293 than those related to nitrogen deposition- to identify an optimum suite of co-variates. This  
294 suite of co-variates was then used in a final RDA with each nitrogen deposition metric as an  
295 explanatory variable. The process was repeated afresh for every analysis, so each includes  
296 a degree of randomness. These analyses provide a more objective alternative to a *priori*  
297 selection of covariates but the use of permutation tests mean results may vary between  
298 runs, there is a risk that covariates identified may not be the most ecologically plausible, and  
299 selected covariates might differ between different N deposition metrics.

300 Each of the above approaches has been applied in previous studies relating plant  
301 communities to nitrogen deposition, and collectively they provide a robust range of  
302 complementary information on the explanatory power of N deposition metrics. We ultimately  
303 conducted 3,564 individual ordinations for each of the 36 vegetation datasets, 33 N  
304 deposition metrics and three approaches to co-variates. This inevitably produces very  
305 complex results. We propose that a useful metric should be consistently significant in these  
306 analyses ( $P < 0.05$ ) and explain a maximal proportion of compositional variance. Therefore  
307 we suggest that a useful measure to assess the relative performance of alternative N  
308 deposition metrics across analyses is the mean significant variance explained, with non-  
309 significant analyses assigned a zero-score. Collectively these analyses ultimately enable us  
310 to answer the question: what is the most ecologically-informative metric of nitrogen  
311 deposition?

## 312 **RESULTS AND DISCUSSION**

### 313 **Properties of the datasets**

314 The pool of vegetation data assembled spans most major UK semi-natural habitats, with the  
315 exception of woodlands. Sampling locations are widely distributed (Fig. 1; Supplementary

316 Fig. 1) but, due to the inclusion of three large Scotland-specific datasets (Table 1) the overall  
317 data has a bias towards the north of Britain. As the northern Highlands and Western Isles  
318 are the least-polluted regions of the UK, the overall dataset also has a high representation of  
319 sites with low N deposition, but with high variability within and between individual habitat  
320 datasets. Most datasets also capture considerable variability in other environmental controls  
321 on vegetation (Table 2).

322 All studied sites have experienced an increase in N deposition over the time period  
323 considered (Fig. 2). A typical trajectory would be similar to Fig 2A, with a gradual increase  
324 through the 19<sup>th</sup> and early 20<sup>th</sup> centuries, increasing rapidly in the late 20<sup>th</sup> century and then  
325 declining to 2010. However, there is considerable variability across sites. In some sites the  
326 decline between 1990 and 2010 is more (Fig. 2B), or less (Fig. 2C) steep, and in a minority  
327 of sites there is no decrease at all (e.g. Fig. 2D). In some sites the initial increase is earlier  
328 (Fig. 2C) or later (Fig. 2D). In most sites the critical load value is exceeded by the late 20<sup>th</sup>  
329 century and remains exceeded (Fig. 2A), while in some sites the critical load is never  
330 exceeded (Fig. 2E) or is exceeded and then subsequently no longer exceeded (Fig. 2F).  
331 Given the general similarity in many trajectories, there are correlations between many of the  
332 metrics derived from these data (Supplementary Table 1). Correlations are particularly  
333 strong within 'families' of metrics, particularly over similar time periods. Correlations are  
334 notably weaker between current N deposition and longer-term metrics.

### 335 **Nitrogen deposition and British vegetation**

336 The first clear finding of our ordination analyses is that N deposition consistently explains  
337 significant variance in the composition of British plant communities (Fig. 3). Across all  
338 vegetation datasets and co-variate approaches it is rare that at least one N metric does not  
339 explain significant variance (Supplementary Figure 2). The proportion of variance explained  
340 is typically small, but this is unsurprising given the many and varied controls on vegetation.  
341 Variance explained by N deposition metrics was greatest in analyses without co-variates and  
342 least in analyses with stepwise selection of co-variates, suggesting that some co-variates  
343 available for the stepwise model-building but not selected *a priori* may have been important  
344 for some habitats. N deposition is clearly an important control on UK vegetation which can  
345 be robustly identified in field data; however its impact is likely to often be subordinate to  
346 factors such as land-use and climate.

347 The majority of published spatial gradient studies addressing N deposition impacts on  
348 vegetation have deliberately targeted sites with a range of N deposition and have aimed to  
349 minimise the impact of co-variates. These designs will have increased the probability of  
350 identifying N deposition impacts. By contrast, many of the datasets addressed here did not  
351 consider N in their sampling design. That N is still shown to be significant in most analyses  
352 provides convincing evidence for the impact of N deposition. Our dataset also includes a  
353 number of habitats with comparatively restricted distributions which have not been  
354 considered in previous studies, including coastal cliffs and tall grass mires (Supplementary  
355 Figure 2). Our results provide the first evidence for N deposition impacts occurring widely in  
356 these habitats in the UK landscape.

357 Individual species correlations with N are not the primary focus of this study but we note that  
358 consistent significant correlations (Supplementary Table 2) mostly match other evidence. For  
359 instance, negative correlations between N and *Racomitrium lanuginosum* in heath and

360 montane habitats (Jones et al., 2002; Van Der Wal et al., 2003), *Plantago lanceolata*  
361 (Mountford et al., 1993) and *Lotus corniculatus* in dunes and grasslands (Stevens et al.,  
362 2016) and positive correlations between N and *Festuca ovina* (Hartley and Mitchell, 2005) in  
363 grassland and montane habitats and *Deschampsia flexuosa* in heathland habitats (Barker et  
364 al., 2004) are all well-established from independent studies.

### 365 **Optimum metrics**

366 Given the number of individual vegetation datasets and metrics, combined with the three  
367 approaches to considering co-variates, there is considerable complexity in results  
368 (Supplementary Figure 2). Straightforward results should not be expected when dealing with  
369 large and diverse datasets from 'real world' landscapes, but it is possible to draw some  
370 general conclusions.

371 The first clear result is that current deposition generally performed poorly compared to  
372 metrics which consider long-term N deposition trajectories. Whether based on a single year  
373 or a three-year mean, current N deposition typically explained lower variance and was less  
374 frequently significant at  $P < 0.05$  than most other N deposition metrics (Supplementary Figure  
375 2). For instance, considering the aggregated significant results with step-wise model-building  
376 (Fig. 3), 3-year current deposition was the worst-performing metric overall, explaining 56%  
377 lower mean significant variance than the best-performing metric. This result supports  
378 considerable previous research suggesting that the long-term history of N deposition is an  
379 important determinant of current status (Phoenix et al., 2012).

380 The conclusion that long-term metrics tend to out-perform current deposition holds for most -  
381 but not all- of the component datasets (Supplementary Figure 2). The most notable  
382 exception is for sand dune habitats where current N deposition more frequently explained  
383 significant variance than long-term metrics (Supplementary Figure 2M-O). In some analyses,  
384 for some dune habitats, current N deposition also explained a larger proportion of variance  
385 and across all dune analyses it was rare for greater variance to be explained by long-term  
386 than current metrics. This distinctive response of sand dune habitats is interesting as, in a  
387 recent field study, Aggenbach et al. (2017) found that high N deposition does not necessarily  
388 lead to increases in N pools, with model simulations suggesting a mechanism whereby N  
389 deposition suppresses symbiotic fixation of atmospheric  $N_2$ . While these results are solely  
390 for calcareous dunes they imply a plausible mechanism whereby N deposition may lead to  
391 vegetation change but without sustained increases in N stock. It is also likely that less N is  
392 retained in dunes than other systems due to limited soil organic matter. The absence of a  
393 cumulative impact of N on soil stocks might thereby explain the apparently superior  
394 correlations with current than long-term N in dune habitats.

395 In the United Kingdom, current and longer-term N deposition values are the products of  
396 different pollutant deposition models: the empirically-based CBED for current deposition  
397 (Smith et al., 2000) and the chemical transport model FRAME (Dore et al., 2007) for longer-  
398 term deposition. Results from the two models are strongly correlated and are frequently used  
399 in tandem. However, it is possible that an unquantified proportion of the difference in metric  
400 performance detected here is due to differences in model performance. This possibility has  
401 implications for policy given that permitting decisions and much national reporting are based  
402 exclusively on CBED.

403 The second clear overall result is that metrics which do not embed the habitat-specific critical  
404 load value have consistently superior performance over those which do. For instance,  
405 considering the aggregated significant results with step-wise model-building (Fig. 3),  
406 cumulative N deposition metrics which do not embed the critical load explain 22% greater  
407 mean significant variance than those which do. This difference is even more marked in the  
408 analyses without co-variates (+26%) or with *a priori* selected co-variates (+31%). Previous  
409 work has advocated a metric based on cumulative deposition above the critical load  
410 (CUM.CL.30Y) for application in UK policy (Rowe et al., 2014). This metric typically performs  
411 better than current deposition (DEP.CUR.3) but is considerably weaker than an equivalent  
412 metric which does not embed the critical load (CUM.30Y)(Fig. 3).

413 In some datasets from low-deposition regions there were few if any sites with N deposition  
414 above the critical load and metrics which embedded the critical load consequently included  
415 many zeroes. These metrics unsurprisingly explained little or no variance. More surprisingly  
416 however, in many of these datasets, many metrics which did not embed the critical load *did*  
417 explain significant variance. For instance, all of the tall grass mire locations were below the  
418 critical load: metrics which embedded the critical load explained no variance but all metrics  
419 which did not embed the critical load did explain variance in analyses without co-variates.  
420 There are two possible explanations for this result: either the apparent correlations are  
421 spurious or, N deposition is having impacts at deposition levels below the critical load. We  
422 consider the former possibility unlikely given that the result is robust to the inclusion of co-  
423 variates for many large-scale controls on vegetation and is replicated across several  
424 datasets. These results therefore provide evidence for sub-critical load impacts.

425 Generally, the best performing metrics are those based on cumulative N deposition without  
426 embedding the critical load. Choosing between cumulative deposition from a fixed starting  
427 point and cumulative deposition over a moving window is difficult on statistical grounds as  
428 metrics are highly correlated and results consequently similar (Fig. 3). Moving window  
429 metrics typically explain fractionally more variance when considering stepwise selection of  
430 co-variates. We propose that moving windows are also likely to be more useful in practice as  
431 they allow for gradual decreases over time, whereas cumulative deposition from a fixed start  
432 point can only increase (Rowe et al., 2014). There is similar difficulty in selecting amongst  
433 different cumulative periods as these metrics are also typically highly correlated. Based on  
434 the stepwise selection of covariates approach (Fig. 3), which is arguably the most robust, the  
435 greatest mean proportion of significant variance was explained by CUM.30Y: a thirty year  
436 moving window of N deposition. This metric also performed competitively without covariates  
437 and with *a priori* selected covariates. Thirty years is the period of cumulative deposition  
438 previously identified on the basis of expert opinion by Rowe et al. (2017) and Rowe et al.  
439 (2014) and used in modelling N deposition impacts by Payne et al. (2017). This period of  
440 deposition therefore has some prior existence in science and policy. The 30 year cumulative  
441 deposition metric offers superior explanatory power to current deposition alone. For  
442 instance, considering analyses without co-variates, across all 36 vegetation datasets thirty  
443 year cumulative deposition explained 23% more variance than single-year current deposition  
444 and explained significant variance ( $P < 0.05$ ) in six datasets (17%) in which current deposition  
445 did not (Fig. 4). On this basis we suggest that cumulative deposition over a thirty year  
446 moving window is a good candidate for the most ecologically-meaningful metric. We focus  
447 on overall plant communities across habitats but we acknowledge it is possible that different  
448 metrics may be most useful when the conservation interest is in particular groups of plants.

449 For instance, there is some experimental evidence that shorter time-scales might be more  
450 relevant to bryophytes and lichens than to vascular plants (Jones, 2005; Rowe et al., 2014).  
451 This might imply that shorter periods of cumulative deposition could be appropriate where  
452 bryophytes are the central focus. Similarly, our results imply that shorter deposition periods  
453 might be more optimal for sand dunes than for other habitats. However we believe that there  
454 is value in selecting a single metric and propose thirty year cumulative deposition as a strong  
455 candidate for this role.

## 456 **CONCLUSIONS**

457 This is the largest study thus-far to assess the role of N deposition as a cause of variability in  
458 UK vegetation, in terms of both sample size and the range of habitats considered. Nitrogen  
459 deposition is significant in most analyses. The size of the effect is often smaller than that of  
460 other drivers of change, but is nevertheless consistent and widespread. These results add to  
461 the increasing body of evidence that N deposition is having far-reaching impacts in UK  
462 habitats. A related conclusion is that there is evidence for N deposition impacts even in  
463 datasets where most or all of the sites are below the critical load, strongly implying that  
464 current critical loads may be set too high for at least some habitats. Finally, our study  
465 provides convincing evidence that current N deposition –as widely used in science and  
466 policy- is not the most meaningful metric to represent N deposition as it affects vegetation. It  
467 is highly probable that many impacts of N pollution develop incrementally over time and that  
468 metrics which incorporate this history better explain spatial patterns of pollution impacts in  
469 the UK landscape. One implication of this finding is that as N deposition falls, recovery is  
470 unlikely to be rapid. We propose thirty years of cumulative deposition as a more ecologically-  
471 informative metric of N deposition for further development and application.

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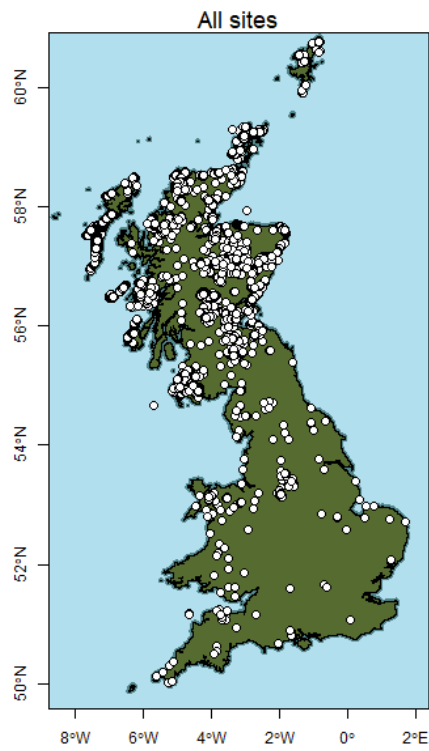
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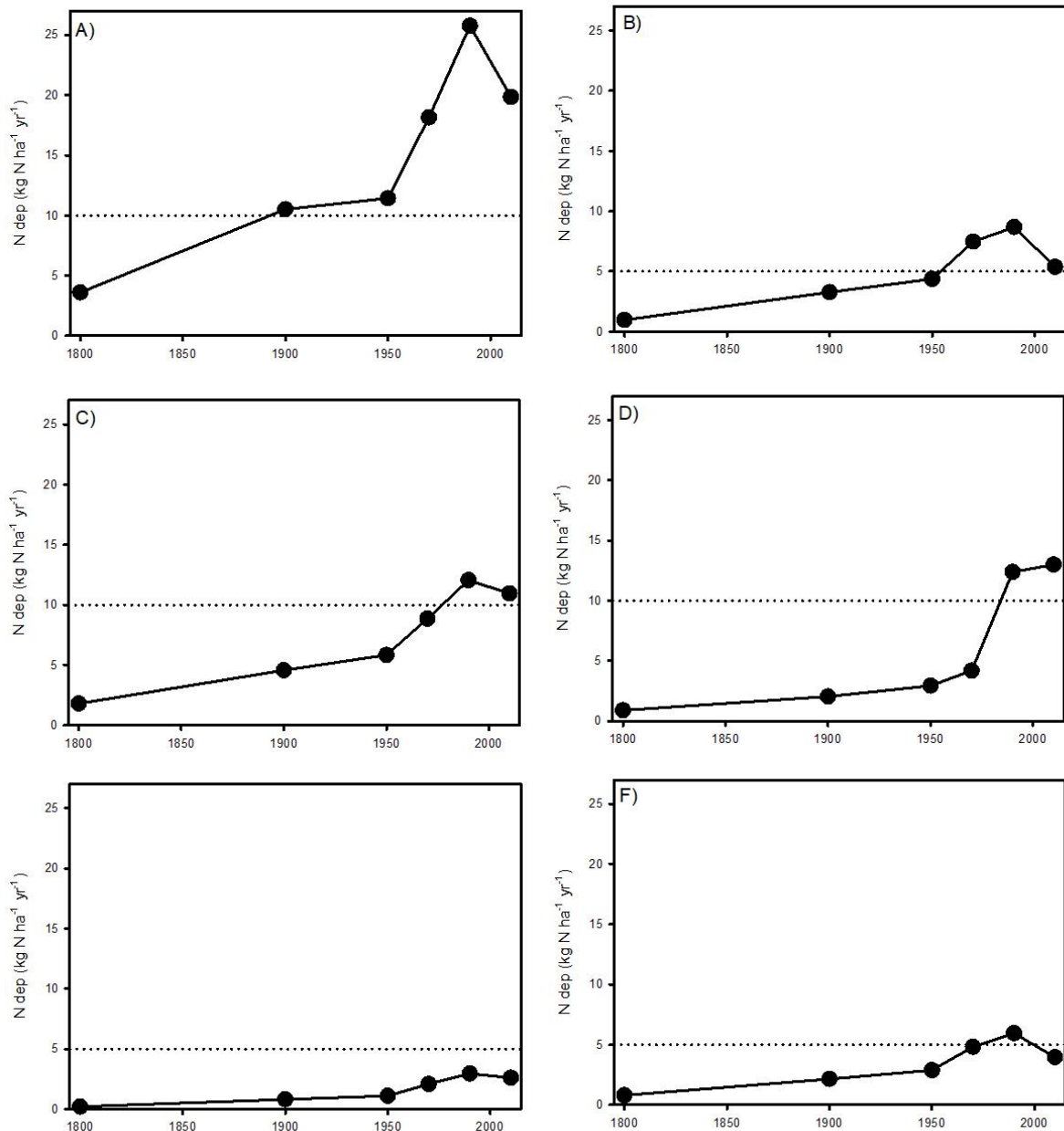
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741 Figure 1. Distribution of sampling sites across all surveys. See Supplementary Figure 1 for  
742 mapping of individual studies and Tables 1 and 2 for details of surveys.

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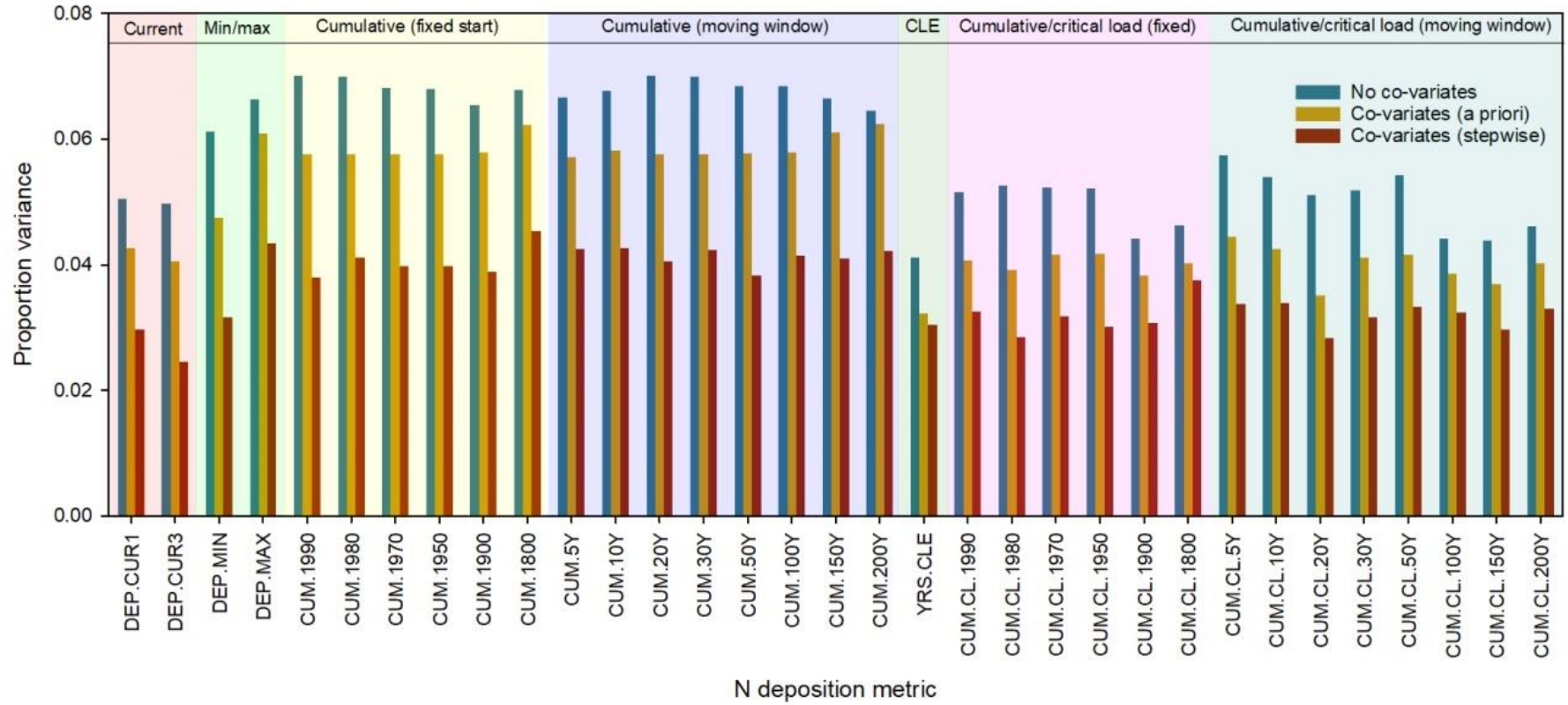


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746 Figure 2. N deposition chronologies for selected sites included in the vegetation surveys. A)  
 747 Coed Poeth (EDM); B) Allt Cragach (PAYN); C) Gladhouse (B.AGRASS); D) Hilldavale  
 748 (B.CHEATH); E) Bennadrove (PAYN) and F) Cawdow (TU.BOG). N deposition values are  
 749 vegetation-specific estimates for low-growing semi-natural habitats (e.g. heaths, bogs,  
 750 grasslands, montane). Plots show modelled tie-points (circles) and interpolated trends  
 751 (lines). Dotted horizontal lines show critical loads for the habitats concerned. For dataset  
 752 codes refer to Table 2.

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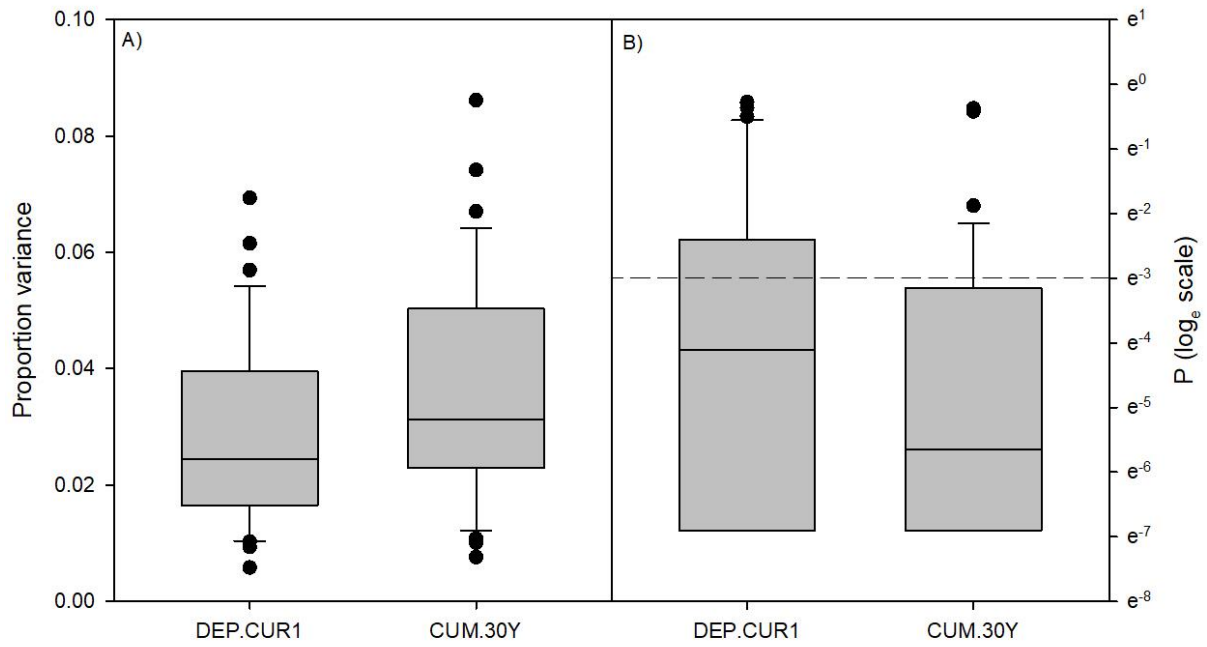
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755 Figure 3. Compositional variance explained by alternative N deposition metrics for all habitats. Background shading denotes different 'families'  
 756 of metrics. See Table 3 for metric codes.

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761 Figure 4. Comparison of single-year current deposition (DEP.CUR1) and 30 year cumulative  
762 deposition (CUM.30Y) for all vegetation datasets (without co-variates) in terms of explained  
763 variance (A) and P-value (B). Dashed horizontal line shows P=0.05.

764

Table 1. Key details of the component vegetation datasets utilised in this study.

Name and references	Key details	Individual datasets
Terrestrial Umbrella nitrogen gradient surveys (Field et al., 2014).	Vegetation survey was conducted in four broad habitats across Great Britain in 2009: bog (Eunis class D1), upland heaths, lowland heaths (both Eunis F4.2) and sand dunes (Eunis B1.4). 22-29 sites were surveyed for each habitat with locations selected to span the N deposition gradient. In each site five, 2 m x 2 m quadrats were positioned in a homogeneous area using random numbers. Cover of vascular plants and mosses was estimated, liverworts were not included in the survey. The acid grassland dataset also included in the published paper is a subset of the Stevens et al. (2004) dataset listed below and was not considered separately.	Terrestrial Umbrella- bogs; Terrestrial Umbrella- lowland heaths; Terrestrial Umbrella- sand dunes; Terrestrial Umbrella- upland heaths
Edmondson regional heathland survey (Edmondson et al., 2013).	Fourteen heathland sites were sampled in England and Wales in 2005. Sites were selected on the basis of consistent vegetation type (NVC H12). Five 50 cm x 50 cm quadrats were positioned randomly in each site and moss and liverwort cover recorded as presence-absence (higher plants were not surveyed).	Edmondson- heather moorlands
Moorland regional survey (Caporn et al., 2014).	Twenty two heathland sites were surveyed in northern England, north Wales and eastern Scotland in 2006. Sites were late building phase NVC H12 upland heathlands, selected to span the N deposition gradient. Presence-absence of all plant species (including liverworts) was recorded in each of five, 50 cm x 50 cm quadrats in each site.	Moorland Regional Survey- heaths
Stevens acid grassland survey (Stevens et al., 2006; Stevens et al., 2004).	Sixty four acid grassland sites (NVC U4) were surveyed across Britain in 2002 and 2003. Sites were randomly selected based on mapped habitat distribution to span the N deposition gradient with additional criteria around site size and accessibility. Five sampling points were randomly selected within a 100 m x 100 m area. At each point a 2 m x 2 m quadrat was surveyed and species cover estimated.	Stevens- acid grasslands
McVean and Ratcliffe survey and resurvey (McVean and Ratcliffe, 1962; Ross et al., 2012)	Surveys of plant communities in the northwest Scottish Highlands were undertaken between 1952 and 1959 with the aim of producing a phytosociological classification of the vegetation. Plant surveys were conducted on the Domin scale in quadrats which varied in size from 1-4 m <sup>2</sup> (the latter most frequent), recording all species including bryophytes and lichens. A resurvey project was undertaken in 2007-2008 with original survey plots relocated with as much accuracy as feasible (Ross et al., 2010). Re-survey vegetation surveys followed the original methodology in as much detail as possible, including using quadrats of the same size. Re-surveys were conducted based on percentage cover-estimates which for comparability were subsequently converted to Domin scores. Only the re-survey dataset, consisting of 254 individual records, was used in this study. Analyses were based on quadrats grouped into Wetland, Moorland, Grassland and Alpine Heathland classes following the original authors.	McVean- alpine; McVean- grassland; McVean- moorlands; McVean- wetlands
Armitage <i>Racomitrium</i> heath survey (Armitage et al., 2014)	Thirty six <i>Racomitrium</i> heath sites were surveyed across Europe, of which here we focus on 27 UK sites in Wales, Cumbria, the Southern Uplands and the Highlands of Scotland. Sites were selected to span the geographic range of the habitat while covering a range of environmental drivers. In each site between 8 and 16, 1 m x 1 m quadrats were equally-spaced in an area of between 1ha and 1km <sup>2</sup> . The cover of all species (including bryophytes and lichens) was estimated.	Armitage- <i>Racomitrium</i> heaths
Birse and Robertson surveys (Birse, 1980, 1984; Birse and	This dataset is the product of a large survey project over two time periods. Original surveys were conducted between 1958 and 1987 with the aim of producing a phytosociological classification and re-surveys were conducted between 2004 and 2014. Re-surveys followed the original protocols as closely as possible and only this re-survey dataset was used here. Quadrat sizes ranged from 1m <sup>2</sup> to more than 9m <sup>2</sup> but were typically 4 m <sup>2</sup> . Re-surveys were conducted based	Birse- acid grasslands; Birse- calcareous; grasslands; Birse- Calluna heaths; Birse- Lolium grasslands; Birse-



Robertson 1976) and re-surveys (Britton et al., 2009; Britton et al., 2017a; Britton et al., 2017b; Mitchell et al., 2017)	on percentage cover estimates which, for comparability with the original study, have been converted to Domin scores and reconverted to percentages. Cover of rock and bare ground were not considered in the analysed data. We considered habitats as grouped by the survey authors, focussing on those which were more abundant: <i>Calluna</i> heath (NVC: H10,H11,H12,H13,H15,H17), <i>Vaccinium</i> heath (H18,H19,H20), <i>Racomitrium</i> heath (U10), acid grassland (U1d,U1e,U4a,U4c,U4d,U4e,U13,U20), calcareous grassland (CG2,CG10,CG11), <i>Lolium</i> grassland, (MG6,MG7), mesotrophic grassland (U4b,SD8,MG1,MG3,MG5,MC9), wet grassland, (M6,M10,M22,M23,M24,M25,M26,M27,MG9, MG10,SD17) swamps (S9,S19,S11,S19,S27,S28), and springs (M32,M37). Where quadrats were intermediate between NVC classes they were included in both options. Data were aggregated to the 5 km x 5 km resolution of the N deposition model.	mesotrophic grasslands; Birse- <i>Racomitrium</i> heaths; Birse- springs; Birse- swamps; Birse- <i>Vaccinium</i> heaths; Birse- wet grasslands
Scottish coastal (re)survey (Lewis et al., 2016; Pakeman et al., 2015; Pakeman et al., 2016; Pakeman et al., 2017; Shaw et al., 1983)	Original surveys were conducted between 1975 and 1977 (most frequently 1976) as part of the Scottish Coastal Survey project (Shaw et al., 1983). Repeat surveys were conducted between 2009 and 2013 (most frequently 2010) with original locations located based on available information from the original survey (Pakeman et al., 2017). Only the re-survey dataset was used in the analyses presented here. A minimum of five, 5 m x 5 m quadrats were recorded for each site. Vascular plant cover was estimated by species and lichen and bryophyte cover was estimated collectively. The data are from 91 individual coastal locations but some of the sites are large so rather than simply aggregating quadrat results by these sites we aggregate on the basis of grid cells used by the N deposition models. The data were divided into 15 broad habitats, as defined by the original authors (Pakeman et al., 2015), of which 10 had sufficient data to warrant detailed analysis. 18 unidentified species, some taxa only identified to genus and some sites without full details were removed prior to analysis.	Scottish Coastal- acid grasslands; Scottish Coastal- cliffs; Scottish Coastal- dune slacks; Scottish Coastal- fixed dunes; Scottish Coastal- heathlands; Scottish Coastal- mobile dunes; Scottish Coastal tall grass mire; Scottish Coastal- unimproved grasslands; Scottish Coastal- wet grasslands; Scottish Coastal- wet heathlands
CEH sand dunes surveys (Aggenbach et al., 2017; Beaumont et al., 2014; Field et al., 2014; Jones et al., 2008; Jones et al., 2004)	This dataset focuses on selected sand dune systems in a limited number of locations around the UK coast. Cover was estimated as a percentage for each species in a 2x2m quadrat. Here the quadrats were grouped to the level of a 5 km x 5 km cell in the N deposition model. Previous studies have considered the dataset in four broad habitat types: dune slacks, semifixed dunes, acid dune grassland and fixed dune grassland. However the spatial distribution of the sites is limited giving small dataset sizes once grouped by model cells so we group the semifixed dunes, acid dune grassland and fixed dune grasslands as a single 'dune grasslands' category. The full dataset as used in some previous analyses incorporates data also included in the Terrestrial Umbrella dataset listed above and sites outside the UK; these quadrats were excluded here.	CEH dune grasslands; CEH dune slacks
Payne peatlands survey (Payne, unpublished)	Peatland sampling sites were selected based on random points positioned on the British Geological Survey UK peat map. Data considered here is for 33 sites which were field-classified as upland bog in a semi-natural condition (excluding e.g. afforested sites). In each site all plants with the exception of liverworts were surveyed in four, 50x50cm quadrats randomly located immediately adjacent to the randomly-selected coordinates or nearest locatable peat. Plant cover was recorded on the Domin scale and is here converted to relative abundance using the Domin2.6 conversion (Currall, 1987).	Payne- bogs
Britton <i>Racomitrium</i> heath survey (Britton et al., 2018)	This survey targeted <i>Racomitrium</i> heath in the UK uplands. Sites were selected to maximise the N deposition gradient and within each site a homogeneous 1ha study area was selected. 8-10 1m <sup>2</sup> quadrats were surveyed per site with species cover estimated to the nearest 1%. All species were recorded, with liverworts grouped into a single category. Species cover recorded as "<1%" was here given a value of 0.5% and non-plant categories (bare ground, litter etc) were excluded. Quadrats were aggregated by sites.	Britton- <i>Racomitrium</i> heaths

Table 2. Full details of vegetation and environmental data for analysed vegetation datasets. Showing key details of datasets, summary codes used elsewhere in this paper, environmental details, habitat groupings, number of sampling sites used in final analysis (n) and additional variables included in stepwise model-building. Critical loads are based on the lowest point of the range in the most recent compilation (Bobbink and Hettelingh, 2011), using established EUNIS habitat conversions. For comparison, the total current N deposition gradient of Great Britain is 2.6-44.6 kg N ha<sup>-1</sup> yr<sup>-1</sup> (CBED 2014 data) but all habitats will not be found across this full gradient.

Dataset	Code	n	Quadrats	Species	Current N dep range (kg ha <sup>-1</sup> yr <sup>-1</sup> )	Critical load value (kg ha <sup>-1</sup> yr <sup>-1</sup> )	Mean annual temperature (°C)	Mean annual precipitation (mm)	Altitude (m)	Additional environmental variables included in pool available for selection.
<b>Heathlands</b>										
Birse- Calluna heaths	B.CHEATH	67	142	233	4.5-26.3	10	3.6-8.5	772-1894	22-938	Aspect; slope.
Birse- Vaccinium heaths	B.VHEATH	33	56	152	7.9-26.3	10	3.5-8.3	725-2062	176-1041	Aspect; slope.
Edmondson- heather moorlands	EDM	14	70	19	20.2-28.7	10	6.8-8.8	998-1347	330-510	Mean annual temperature; mean annual precipitation; growing degree days; ozone.
McVean- moorlands	MCV.MOOR	79	79	200	3.9-19.6	10	3.3-8.4	887-1735	39-925	Aspect; slope.
Moorland Regional Survey- heaths	MRS	22	110	50	6.9-33.7	10	4.5-9.0	952-1318	280-530	Mean annual temperature; mean annual precipitation; litter % Nitrogen.
Scottish Coastal- heathlands	SC.HEATH	36	138	173	2.7-11.8	10	6.7-8.9	641-1484	0-76	-
Scottish Coastal- wet heathlands	SC.WHEATH	38	107	174	2.9-10.7	10	7.4-8.9	639-1563	0-93	-
Terrestrial Umbrella- lowland heaths	TU.LH	27	135	87	4.8-18.1	10	6.2-10.3	598-1113	0-280	Growing degree days; mean annual precipitation; slope; soil loss on ignition; soil pH; ozone.
Terrestrial Umbrella- upland heaths	TU.UH	24	120	78	5.6-29.5	10	5.3-9.2	815-1842	255-706	Growing degree days; mean annual precipitation; slope; soil

										loss on ignition; soil pH; ozone.
<b>Grasslands</b>										
Birse- acid grasslands	B.AGRASS	42	61	192	4.6-21.8	10	3.1-8.2	725-1903	25-927	Aspect; slope.
Birse- calcareous grasslands	B.CGRASS	41	71	209	5.8-21.6	15	3.6-8.6	798-1939	4-859	Aspect; slope.
Birse- Lolium grasslands	B.LGRASS	46	58	96	4.6-19.0	10	6.3-8.7	708-1789	7-347	Aspect; slope.
Birse- mesotrophic grasslands	B.MGRASS	73	96	178	4.0-23.3	10	5.3-8.8	672-1886	5-416	Aspect; slope.
Birse- wet grasslands	B.WGRASS	56	80	248	3.3-31.1	10	4.8-8.8	672-1892	0-750	Aspect; slope.
McVean- grassland	MCV.GRASS	56	56	218	5.1-18.8	10	4.3-8.1	979-2067	117-1008	Aspect; slope.
Scottish Coastal- acid grasslands	SC.AGRASS	53	186	230	2.7-11.2	10	7.1-8.9	641-1487	0-76	-
Scottish Coastal- cliffs	SC.CLIFF	38	60	175	2.8-10.7	5	6.6-8.9	653-1480	0-46	-
Scottish Coastal- unimproved grasslands	SC.UGRASS	76	270	296	2.7-9.0	10	7.1-8.9	641-1563	0-80	-
Scottish Coastal- wet grasslands	SC.WGRASS	57	156	224	2.9-9.0	10	7.1-8.9	663-1498	0-118	-
Stevens- acid Grasslands	CS.AGRASS	64	320	181	7.7-40.9	10	6.0-10.3	568-1989	15-500	Radiation index; cutting; management index; mean maximum temperature; mean minimum temperature; mean annual precipitation; topsoil pH; Olsen P; total C.
<b>Wetlands</b>										
Birse- springs	B.SPRI	25	44	191	5.3-20.4	15	3.6-7.1	853-1677	315-1084	Aspect; slope.
Birse- swamps	B.SWAM	33	48	160	3.6-20.9	15	5.7-8.3	655-1528	4-524	Aspect; slope.
McVean- wetlands		28	28	170	5.1-15.8	5	3.8-8.1	1002-1822	144-945	Aspect; slope.
Payne- bogs	PAYN	33	132	81	3.4-29.2	5	4.5-8.6	815-1790	9-693	-
Scottish Coastal tall grass mire	SC.TGM	51	114	233	2.7-10.7	15	6.7-8.9	648-1563	0-109	-
Terrestrial Umbrella-bogs	TU.BOG	29	145	97	4.8-26.7	5	4.4-9.7	755-1778	9-564	Growing degree days; mean annual precipitation; slope; soil

										pH; ozone; hydrological index.
<b>Montane habitats</b>										
Armitage-Racomitrium heaths	ARM.RHE	26	298	58	8.9-47.9	5	2.9-7.7	1064-2118	690-1103	-
Birse- Racomitrium heaths	B.RHE	77	134	214	5.8-31.2	5	3.4-8.0	745-1956	14-1114	Aspect; slope.
Britton- Racomitrium heaths	BRI.RHE	15	148	66	6.0-34.7	5	2.9-7.8	1183-1754	712-1026	-
McVean- alpine	MCV.ALP	91	91	191	4.9-19.4	5	2.9-7.5	1039-1822	295-1145	Aspect; slope.
<b>Sand dune habitats</b>										
CEH dune grasslands	CEH.DUG R	34	235	345	3.4-13.1	10	8.1-11.1	603-1105	0-15	-
CEH dune slacks	CEH.SLAC	29	285	362	2.8-11.4	10	8.1-11.1	603-1156	0-29	-
Scottish Coastal-dune slacks	SC.SLAC	65	198	246	2.7-11.8	10	6.9-8.9	648-1480	0-73	-
Scottish Coastal-fixed dunes	SC.FDU	121	960	310	2.7-11.8	10	6.6-8.9	646-1656	0-118	-
Scottish Coastal-mobile dunes	SC.MDU	60	128	136	2.7-11.8	10	6.5-8.9	642-1653	0-109	-
Terrestrial Umbrella-sand dunes	TU.SD	24	120	190	3.9-12.5	8	8.0-10.4	603-1108	0-119	Growing degree days; mean annual precipitation; slope; soil loss on ignition; soil pH; altitude; ozone.

Table 3. Metrics of N deposition considered in this study.

<b>Metric family</b>	<b>Metric</b>	<b>Code</b>
Current deposition	Current deposition over year of survey.	DEP.CUR1
	Three-year mean prior to year of survey.	DEP.CUR3
Minimum/Maximum deposition	Minimum deposition 1800 onwards.	DEP.MIN
	Maximum deposition 1800 onwards.	DEP.MAX
Cumulative deposition based on a fixed start date.	Cumulative deposition since 1990.	CUM.1990
	Cumulative deposition since 1980.	CUM.1980
	Cumulative deposition since 1970.	CUM.1970
	Cumulative deposition since 1950.	CUM.1950
	Cumulative deposition since 1900.	CUM.1900
	Cumulative deposition since 1800.	CUM.1800
Cumulative deposition over a moving window of years.	Cumulative deposition over 5 years prior to survey.	CUM.5Y
	Cumulative deposition over 10 years prior to survey.	CUM.10Y
	Cumulative deposition over 20 years prior to survey.	CUM.20Y
	Cumulative deposition over 30 years prior to survey.	CUM.30Y
	Cumulative deposition over 50 years prior to survey.	CUM.50Y
	Cumulative deposition over 100 years prior to survey.	CUM.100Y
	Cumulative deposition over 150 years prior to survey.	CUM.150Y
	Cumulative deposition over 200 years prior to survey.	CUM.200Y
Critical load exceedance (CLE)	Years of deposition above critical load.	YRS.CLE
Cumulative deposition over the critical load, based on a fixed start date.	Cumulative deposition above critical load since 1990.	CUM.CL.1990
	Cumulative deposition above critical load since 1980.	CUM.CL.1980
	Cumulative deposition above critical load since 1970.	CUM.CL.1970
	Cumulative deposition above critical load since 1950.	CUM.CL.1950
	Cumulative deposition above critical load since 1900.	CUM.CL.1900
	Cumulative deposition above critical load since 1800.	CUM.CL.1800
Cumulative deposition over the critical load, based on a moving window of years.	Cumulative deposition above critical load over 5 years prior to survey.	CUM.CL.5Y
	Cumulative deposition above critical load over 10 years prior to survey.	CUM.CL.10Y
	Cumulative deposition above critical load over 20 years prior to survey.	CUM.CL.20Y
	Cumulative deposition above critical load over 30 years prior to survey.	CUM.CL.30Y
	Cumulative deposition above critical load over 50 years prior to survey.	CUM.CL.50Y
	Cumulative deposition above critical load over 100 years prior to survey.	CUM.CL.100Y
	Cumulative deposition above critical load over 150 years prior to survey.	CUM.CL.150Y
	Cumulative deposition above critical load over 200 years prior to survey.	CUM.CL.200Y