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

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ABSTRACT

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

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

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

HIGHLIGHTS:

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

INTRODUCTION

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

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

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

There are a number of reasons why current deposition data may not optimally represent N deposition as it affects the environment. Experimental studies show that N deposition impacts take considerable time to develop (Phoenix et al., 2012). Many long-term studies have shown hysteresis over 1-3 years but have ultimately shown large change over time-periods of a decade or more (Clark and Tilman, 2008). Similarly, some studies of recovery have shown limited recovery when N additions are ceased (Isbell et al., 2013). Nitrogen deposition increases N stocks and concentrations in soil and plant tissue, and increases primary production, leading to greater N in above- and below-ground pools (Meter et al., 2016; Pornon et al., 2018; Rowe et al., 2014). Nitrogen deposition therefore tends to have *cumulative* impacts as these pools build up over time. Time-scales of species response will depend on the autecology of the species concerned and may vary dependent on their nitrogen sources and those of their competitors. Short-term modelled N deposition estimates will also be affected by atmospheric conditions during that period, particularly in terms of precipitation, and may not always correlate well with longer time-periods. The ecological impacts of N deposition are primarily long-term processes which are likely to be imperfectly characterised over a period of less than three years.

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

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

METHODS

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

Vegetation data

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

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

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

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

Nitrogen deposition modelling and data

We developed a range of potential N deposition metrics for each location using recently-developed hind-casted deposition modelling for the UK based on spatially distributed historic N emissions data and the FRAME model (Dragosits et al., 2016; Tipping et al., 2017) The FRAME model is an atmospheric chemistry transport model which simulates the emissions of nitrogen compounds, their vertical diffusion and horizontal transport, atmospheric chemical transformation and deposition to the surface by wet and dry processes. N deposition modelling for this study was based on ground coverage of low-growing semi-natural species, as suited to the habitats considered (N deposition estimates for woodland are generally higher, due to a higher deposition velocity, notably for NH_3). The underlying emissions data is currently available for six time-steps: 1800, 1900, 1950, 1970, 1990 and 2010. These years were selected based on data availability and likely changes in air pollution, including initial industrial development (19^{th} century), the advent and widespread implementation of the Haber-Bosch process (first half of 20^{th} century), the peak in emissions (late 20^{th} century) and subsequent decline. Based on this modelling, we produced grid-cell specific deposition chronologies for all 5 km x 5 km cells containing vegetation data with changes between the six tie-points calculated using linear interpolation. We compared these results to current deposition, as estimated using the standard CBED model used in UK policy and management. Given the broad spatial and temporal scope of the study we focused on total N deposition, accepting that somewhat different effects may be produced by reduced and oxidised forms of N, and by dry and wet deposition (Sheppard et al., 2011; Stevens et al., 2011; Van den Berg et al., 2008; van den Berg et al., 2016).

Nitrogen deposition metrics

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

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

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

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

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

Ordination

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

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

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

RESULTS AND DISCUSSION

Properties of the datasets

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

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

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

Nitrogen deposition and British vegetation

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

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

Individual species correlations with N are not the primary focus of this study but we note that consistent significant correlations (Supplementary Table 2) mostly match other evidence. For instance, negative correlations between N and *Racomitrium lanuginosum* in heath and montane habitats (Jones et al., 2002; Van Der Wal et al., 2003), *Plantago lanceolata* (Mountford et al., 1993) and *Lotus corniculatus* in dunes and grasslands (Stevens et al., 2016) and positive correlations between N and *Festuca ovina* (Hartley and Mitchell, 2005) in grassland and montane habitats and *Deschampsia flexuosa* in heathland habitats (Barker et al., 2004) are all well-established from independent studies.

Optimum metrics

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

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

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

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

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

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

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

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

CONCLUSIONS

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

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Author contributions: RiJP conceived the study, conducted data analysis and wrote the first draft of the manuscript. UD, ST, EJC and AJD conducted N deposition modelling. AJB, RJM, RoJP, LJ, LCR, LR, CJS, CF, SJMC, JC, JLE and RiJP designed and conducted vegetation surveys. All authors contributed design suggestions and interpretation and commented on the manuscript

REFERENCES

Aggenbach, C.J.S., Kooijman, A.M., Fujita, Y., van der Hagen, H., van Til, M., Cooper, D., Jones, L., 2017. Does atmospheric nitrogen deposition lead to greater nitrogen and carbon accumulation in coastal sand dunes? Biological Conservation 212, 416-422.
Armitage, H.F., Britton, A.J., van der Wal, R., Woodin, S.J., 2014. The relative importance of nitrogen deposition as a driver of Racomitrium heath species composition and richness across Europe. Biological Conservation 171, 224-231.
Barker, C.G., Power, S.A., Bell, J.N.B., Orme, C.D.L., 2004. Effects of habitat management on heathland response to atmospheric nitrogen deposition. Biological Conservation 120, 41-52.
Basto, S., Thompson, K., Phoenix, G., Sloan, V., Leake, J., Rees, M., 2015. Long-term nitrogen deposition depletes grassland seed banks. Nature communications 6, 6185.
Beaumont, N.J., Jones, L., Garbutt, A., Hansom, J.D., Toberman, M., 2014. The value of carbon sequestration and storage in coastal habitats. Estuarine, Coastal and Shelf Science 137, 32-40.
Birse, E., 1980. Plant communities of Scotland - a preliminary phytocoenonia. Macaulay Institute for Soil Research, Aberdeen.
Birse, E., 1984. The phytocoenonia of Scotland - additions and revision. Macaulay Institute for Soil Research, Aberdeen.
Birse, E., Robertson , J., 1976. Plant communities and soils of the lowland and southern upland regions of Scotland. Macaulay Institute for Soil Research, Aberdeen.
Bobbink, R., Hettelingh, J.P., 2011. Review and revision of empirical critical loads and dose-response relationships : Proceedings of an expert workshop, Noordwijkerhout, 23-25 June 2010. RIVM, The Netherlands.
Bobbink, R., Hicks, K., Galloway, J., Spranger, T., Alkemade, R., Ashmore, M., Bustamante, M., Cinderby, S., Davidson, E., Dentener, F., 2010. Global assessment of nitrogen deposition effects on terrestrial plant diversity: a synthesis. Ecological applications 20, 30-59.
Borcard, D., Legendre, P., Drapeau, P., 1992. Partialling out the Spatial Component of Ecological Variation. Ecology 73, 1045-1055.
Bowman, W.D., Cleveland, C.C., Halada, Ĺ., Hreško, J., Baron, J.S., 2008. Negative impact of nitrogen deposition on soil buffering capacity. Nature Geoscience 1, 767.
Britton, A.J., Beale, C.M., Towers, W., Hewison, R.L., 2009. Biodiversity gains and losses: Evidence for homogenisation of Scottish alpine vegetation. Biological Conservation 142, 1728-1739.
Britton, A.J., Hester, A.J., Hewison, R.L., Potts, J.M., Ross, L.C., 2017a. Climate, pollution and grazing drive long-term change in moorland habitats. Applied Vegetation Science 20, 194-203.
Britton, A.J., Hewison, R.L., Mitchell, R.J., Riach, D., 2017b. Pollution and climate change drive long-term change in Scottish wetland vegetation composition. Biological Conservation 210, 72-79.
Britton, A.J., Mitchell, R.J., Fisher, J.M., Riach, D.J., Taylor, A.F.S., 2018. Nitrogen deposition drives loss of moss cover in alpine moss–sedge heath via lowered C : N ratio and accelerated decomposition. New Phytologist 218, 470-478.
Cape, J.N., van der Eerden, L.J., Sheppard, L.J., Leith, I.D., Sutton, M.A., 2009. Evidence for changing the critical level for ammonia. Environmental Pollution 157, 1033-1037.
Caporn, S.J., Carroll, J.A., Dise, N.B., Payne, R.J., 2014. Impacts and indicators of nitrogen deposition in moorlands: Results from a national pollution gradient study. Ecological Indicators 45, 227-234.
Carroll, J.A., Caporn, S.J.M., Cawley, L., Read, D.J., Lee, J.A., 1999. The effect of increased deposition of atmospheric nitrogen on Calluna vulgaris in upland Britain. New Phytologist 141, 423-431.
Clark, C.M., Tilman, D., 2008. Loss of plant species after chronic low-level nitrogen deposition to prairie grasslands. Nature 451, 712-715.
Currall, J.E.P., 1987. A transformation of the Domin scale. Vegetatio 72, 81-87.

Dise, N.B., Ashmore, M.R., Belyazid, S., Bobbink, R., De Vries, W., Erisman, J.W., Spranger, T., Stevens, C., van den Berg, L., 2011. Nitrogen as a threat to European terrestrial biodiversity, in: Sutton, M. (Ed.), The European nitrogen assessment: sources, effects and policy perspectives. Cambridge University Press.

Dore, A., Kryza, M., Hall, J., Hallsworth, S., Keller, V., Vieno, M., Sutton, M., 2012. The influence of model grid resolution on estimation of national scale nitrogen deposition and exceedance of critical loads. Biogeosciences 9, 1597-1609.

Dore, A.J., Vieno, M., Tang, Y.S., Dragosits, U., Dosio, A., Weston, K.J., Sutton, M.A., 2007. Modelling the atmospheric transport and deposition of Sulphur and Nitrogen over the United Kingdom and assessment of the influence of SO_2 emissions from international shipping. Atmospheric Environment 41, 2355-2367.

Dragosits, U., Tomlinson, S.J., Carnell, E.J., Dore, A.J., Misselbrook, T., Tipping, E., 2016. Historic trends in N and S deposition in the UK- 1800 to present, Committee on Air Pollution Effects Research. Centre for Ecology and Hydrology.

Duprè, C., Stevens, C.J., Ranke, T., Bleeker, A., Peppler-Lisbach, C., Gowing, D.J.G., Dise, N.B., Dorland, E.D.U., Bobbink, R., Diekmann, M., 2010. Changes in species richness and composition in European acidic grasslands over the past 70 years: the contribution of cumulative atmospheric nitrogen deposition. Global Change Biology 16, 344-357.

Edmondson, J., Terribile, E., Carroll, J.A., Price, E.A.C., Caporn, S.J.M., 2013. The legacy of nitrogen pollution in heather moorlands: Ecosystem response to simulated decline in nitrogen deposition over seven years. Science of The Total Environment 444, 138-144.

Farr, T.G., Rosen, P.A., Caro, E., Crippen, R., Duren, R., Hensley, S., Kobrick, M., Paller, M., Rodriguez, E., Roth, L., Seal, D., Shaffer, S., Shimada, J., Umland, J., Werner, M., Oskin, M., Burbank, D., Alsdorf, D., 2007. The Shuttle Radar Topography Mission. Reviews of Geophysics 45, n/a-n/a.

Field, C.D., Dise, N.B., Payne, R.J., Britton, A.J., Emmett, B.A., Helliwell, R.C., Hughes, S., Jones, L., Lees, S., Leake, J.R., 2014. The role of nitrogen deposition in widespread plant community change across semi-natural habitats. Ecosystems 17, 864-877.

Fowler, D., Coyle, M., Skiba, U., Sutton, M.A., Cape, J.N., Reis, S., Sheppard, L.J., Jenkins, A., Grizzetti, B., Galloway, J.N., 2013. The global nitrogen cycle in the twenty-first century. Phil. Trans. R. Soc. B 368, 20130164.

Fowler, D., O'Donoghue, M., Muller, J.B.A., Smith, R.I., Dragosits, U., Skiba, U., Sutton, M.A., Brimblecombe, P., 2005. A chronology of nitrogen deposition in the UK between 1900 and 2000. Water, Air, & Soil Pollution: Focus 4, 9-23.

Fowler, D., Steadman, C.E., Stevenson, D., Coyle, M., Rees, R.M., Skiba, U.M., Sutton, M.A., Cape, J.N., Dore, A.J., Vieno, M., Simpson, D., Zaehle, S., Stocker, B.D., Rinaldi, M., Facchini, M.C., Flechard, C.R., Nemitz, E., Twigg, M., Erisman, J.W., Butterbach-Bahl, K., Galloway, J.N., 2015. Effects of global change during the 21st century on the nitrogen cycle. Atmos. Chem. Phys. 15, 13849-13893.

Galloway, J.N., Dentener, F.J., Capone, D.G., Boyer, E.W., Howarth, R.W., Seitzinger, S.P., Asner, G.P., Cleveland, C.C., Green, P.A., Holland, E.A., 2004. Nitrogen cycles: past, present, and future. Biogeochemistry 70, 153-226.

Galloway, J.N., Townsend, A.R., Erisman, J.W., Bekunda, M., Cai, Z., Freney, J.R., Martinelli, L.A., Seitzinger, S.P., Sutton, M.A., 2008. Transformation of the nitrogen cycle: recent trends, questions, and potential solutions. Science 320, 889-892.

Hall, J., Smith, R., Dore, A.J., 2017. Trends Report 2017: Trends in critical load and critical level exceedances in the UK. DEFRA, London.

Hartley, S.E., Mitchell, R.J., 2005. Manipulation of nutrients and grazing levels on heather moorland: changes in Calluna dominance and consequences for community composition. Journal of Ecology 93, 990-1004.

Hijmans, R.J., Cameron, S.E., Parra, J.L., Jones, P.G., Jarvis, A., 2005. Very high resolution interpolated climate surfaces for global land areas. International Journal of Climatology 25, 1965-1978.

Horswill, P., O'Sullivan, O., Phoenix, G.K., Lee, J.A., Leake, J.R., 2008. Base cation depletion, eutrophication and acidification of species-rich grasslands in response to long-term simulated nitrogen deposition. Environmental Pollution 155, 336-349.
Isbell, F., Tilman, D., Polasky, S., Binder, S., Hawthorne, P., 2013. Low biodiversity state persists two decades after cessation of nutrient enrichment. Ecology letters 16, 454-460.
Jones, L., Provins, A., Holland, M., Mills, G., Hayes, F., Emmett, B., Hall, J., Sheppard, L., Smith, R., Sutton, M., Hicks, K., Ashmore, M., Haines-Young, R., Harper-Simmonds, L., 2014. A review and application of the evidence for nitrogen impacts on ecosystem services. Ecosystem Services 7, 76-88.
Jones, M.L.M., 2005. Effects of nitrogen and simulated grazing on two upland grasslands. University of Sheffield, Sheffield.
Jones, M.L.M., Oxley, E.R.B., Ashenden, T.W., 2002. The influence of nitrogen deposition, competition and desiccation on growth and regeneration of Racomitrium lanuginosum (Hedw.) Brid. Environmental Pollution 120, 371-378.
Jones, M.L.M., Sowerby, A., Williams, D.L., Jones, R.E., 2008. Factors controlling soil development in sand dunes: evidence from a coastal dune soil chronosequence. Plant and Soil 307, 219-234.
Jones, M.L.M., Wallace, H.L., Norris, D., Brittain, S.A., Haria, S., Jones, R.E., Rhind, P.M., Reynolds, B.R., Emmett, B.A., 2004. Changes in Vegetation and Soil Characteristics in Coastal Sand Dunes along a Gradient of Atmospheric Nitrogen Deposition. Plant Biology 6, 598-605.
Legendre, P., Gallagher, E.D., 2001. Ecologically meaningful transformations for ordination of species data. Oecologia 129, 271-280.
Lewis, R.J., Marrs, R.H., Pakeman, R.J., Milligan, G., Lennon, J.J., 2016. Climate drives temporal replacement and nested-resultant richness patterns of Scottish coastal vegetation. Ecography 39, 754-762.
Matejko, M., Dore, A.J., Hall, J., Dore, C.J., Błaś, M., Kryza, M., Smith, R., Fowler, D., 2009. The influence of long term trends in pollutant emissions on deposition of sulphur and nitrogen and exceedance of critical loads in the United Kingdom. Environmental Science & Policy 12, 882-896.
McVean, D.N., Ratcliffe, D.A., 1962. Plant communities of the Scottish Highlands. A study of Scottish mountain, moorland and forest vegetation. HMSO, London.
Meter, K.J.V., Basu, N.B., Veenstra, J.J., Burras, C.L., 2016. The nitrogen legacy: emerging evidence of nitrogen accumulation in anthropogenic landscapes. Environmental Research Letters 11, 035014.
Mitchell, C.E., Reich, P.B., Tilman, D., Groth, J.V., 2003. Effects of elevated CO2, nitrogen deposition, and decreased species diversity on foliar fungal plant disease. Global Change Biology 9, 438-451.
Mitchell, R.J., Hewison, R.L., Britton, A.J., Brooker, R.W., Cummins, R.P., Fielding, D.A., Fisher, J.M., Gilbert, D.J., Hester, A.J., Hurskainen, S., Pakeman, R.J., Potts, J.M., Riach, D., 2017. Forty years of change in Scottish grassland vegetation: Increased richness, decreased diversity and increased dominance. Biological Conservation 212, 327-336.
Mountford, J.O., Lakhani, K.H., Kirkham, F.W., 1993. Experimental Assessment of the Effects of Nitrogen Addition Under Hay- Cutting and Aftermath Grazing on the Vegetation of Meadows on a Somerset Peat Moor. Journal of Applied Ecology 30, 321-332.
Oksanen, J., Kindt, R., Legendre, P., O'Hara, B., Stevens, M.H.H., Oksanen, M.J., Suggests, M., 2007. The vegan package. Community ecology package 10.
Pakeman, R.J., Alexander, J., Beaton, J., Brooker, R., Cummins, R., Eastwood, A., Fielding, D., Fisher, J., Gore, S., Hewison, R., Hooper, R., Lennon, J., Mitchell, R., Moore, E., Nolan, A., Orford, K., Pemberton, C., Riach, D., Sim, D., Stockan, J., Trinder, C., Lewis, R., 2015. Species composition of coastal dune vegetation in Scotland has proved resistant to climate change over a third of a century. Global Change Biology 21, 3738-3747.
Pakeman, R.J., Alexander, J., Brooker, R., Cummins, R., Fielding, D., Gore, S., Hewison, R., Mitchell, R., Moore, E., Orford, K., Pemberton, C., Trinder, C., Lewis, R., 2016. Long-term

impacts of nitrogen deposition on coastal plant communities. Environmental Pollution 212, 337-347.
Pakeman, R.J., Hewison, R.L., Lewis, R.J., 2017. Drivers of species richness and compositional change in Scottish coastal vegetation. Applied Vegetation Science 20, 183-193.
Payne, R., 2014. The exposure of British peatlands to nitrogen deposition, 1900–2030. Mires and Peat 14, 1-9.
Payne, R.J., Dise, N.B., Field, C.D., Dore, A.J., Caporn, S.J., Stevens, C.J., 2017. Nitrogen deposition and plant biodiversity: past, present, and future. Frontiers in Ecology and the Environment 15, 431-436.
Payne, R.J., Stevens, C.J., Dise, N.B., Gowing, D.J., Pilkington, M.G., Phoenix, G.K., Emmett, B.A., Ashmore, M.R., 2011. Impacts of atmospheric pollution on the plant communities of British acid grasslands. Environmental Pollution 159, 2602-2608.
Pearson, J., Stewart, G.R., 1993. The deposition of atmospheric ammonia and its effects on plants. New Phytologist 125, 283-305.
Phoenix, G.K., Emmett, B.A., Britton, A.J., Caporn, S.J.M., Dise, N.B., Helliwell, R., Jones, L., Leake, J.R., Leith, I.D., Sheppard, L.J., 2012. Impacts of atmospheric nitrogen deposition: responses of multiple plant and soil parameters across contrasting ecosystems in long-term field experiments. Global Change Biology 18, 1197-1215.
Pornon, A., Boutin, M., Lamaze, T., 2018. Contribution of plant species to the high N retention capacity of a subalpine meadow undergoing elevated N deposition and warming. Environmental Pollution.
R Development Core Team, 2014. R: A language and environment for statistical computing. R foundation for Statistical Computing.
Rao, C.R., 1995. A review of canonical coordinates and an alternative to correspondence analysis using Hellinger distance. Qüestiió 19, 23-63.
Ross, L.C., Woodin, S.J., Hester, A., Thompson, D.B., Birks, H.J.B., 2010. How important is plot relocation accuracy when interpreting re-visitation studies of vegetation change? Plant Ecology & Diversity 3, 1-8.
Ross, L.C., Woodin, S.J., Hester, A.J., Thompson, D.B.A., Birks, H.J.B., 2012. Biotic homogenization of upland vegetation: patterns and drivers at multiple spatial scales over five decades. Journal of Vegetation Science 23, 755-770.
Rowe, E., Jones, L., Dise, N., Evans, C., Mills, G., Hall, J., Stevens, C.J., Mitchell, R., Field, C., Caporn, S., 2017. Metrics for evaluating the ecological benefits of decreased nitrogen deposition. Biological Conservation 212, 454-463.
Rowe, E., Jones, L., Stevens, C., Vieno, M., Dore, A., Hall, J., Sutton, M.A., Mills, G., Evans, C., Helliwell, R., Britton, A., Mitchell, R., Caporn, S., Dise, N., Field, C., Emmett, B., 2014. Measures to evaluate benefits to UK semi-natural habitats of reductions in nitrogen deposition. Final report on REBEND project (Defra AQ0823; CEH NEC04307). Centre for Ecology and Hydrology, Bangor.
Sala, O.E., Chapin, F.S., Armesto, J.J., Berlow, E., Bloomfield, J., Dirzo, R., Huber-Sanwald, E., Huenneke, L.F., Jackson, R.B., Kinzig, A., 2000. Global biodiversity scenarios for the year 2100. Science 287, 1770-1774.
Shaw, M., Hewett, D., Pizzey, J., 1983. Scottish coastal survey. Institute of Terrestrial Ecology, Bangor, Gwynedd, UK.
Sheppard, L.J., Leith, I.D., Mizunuma, T., Neil Cape, J., Crossley, A., Leeson, S., Sutton, M.A., Dijk, N., Fowler, D., 2011. Dry deposition of ammonia gas drives species change faster than wet deposition of ammonium ions: evidence from a long-term field manipulation. Global Change Biology 17, 3589-3607.
Smith, R., Fowler, D., Sutton, M., Flechard, C., Coyle, M., 2000. Regional estimation of pollutant gas dry deposition in the UK: model description, sensitivity analyses and outputs. Atmospheric Environment 34, 3757-3777.
Stevens, C.J., Dise, N.B., Gowing, D.J., Mountford, J.O., 2006. Loss of forb diversity in relation to nitrogen deposition in the UK: regional trends and potential controls. Global Change Biology 12, 1823-1833.

Stevens, C.J., Dise, N.B., Mountford, J.O., Gowing, D.J., 2004. Impact of nitrogen deposition on the species richness of grasslands. Science 303, 1876-1879.
Stevens, C.J., Manning, P., Van den Berg, L.J.L., De Graaf, M.C.C., Wamelink, G.W., Boxman, A.W., Bleeker, A., Vergeer, P., Arroniz-Crespo, M., Limpens, J., 2011. Ecosystem responses to reduced and oxidised nitrogen inputs in European terrestrial habitats. Environmental Pollution 159, 665-676.
Stevens, C.J., Payne, R.J., Kimberley, A., Smart, S.M., 2016. How will the semi-natural vegetation of the UK have changed by 2030 given likely changes in nitrogen deposition? Environmental Pollution 208, 879–889.
Stevens, C.J., Thompson, K., Grime, J.P., Long, C.J., Gowing, D.J.G., 2010. Contribution of acidification and eutrophication to declines in species richness of calcifuge grasslands along a gradient of atmospheric nitrogen deposition. Functional ecology 24, 478-484.
Throop, H.L., Lerdau, M.T., 2004. Effects of nitrogen deposition on insect herbivory: implications for community and ecosystem processes. Ecosystems 7, 109-133.
Tipping, E., Davies, J.A.C., Henrys, P.A., Kirk, G.J.D., Lilly, A., Dragosits, U., Carnell, E.J., Dore, A.J., Sutton, M.A., Tomlinson, S.J., 2017. Long-term increases in soil carbon due to ecosystem fertilization by atmospheric nitrogen deposition demonstrated by regional-scale modelling and observations. Scientific Reports 7, 1890.
Van den Berg, L., Peters, C., Ashmore, M., Roelofs, J., 2008. Reduced nitrogen has a greater effect than oxidised nitrogen on dry heathland vegetation. Environmental Pollution 154, 359-369.
van den Berg, L.J., Jones, L., Sheppard, L.J., Smart, S.M., Bobbink, R., Dise, N.B., Ashmore, M.R., 2016. Evidence for differential effects of reduced and oxidised nitrogen deposition on vegetation independent of nitrogen load. Environmental Pollution 208, 890-897.
van den Wollenberg, A.L., 1977. Redundancy analysis an alternative for canonical correlation analysis. Psychometrika 42, 207-219.
Van Der Wal, R., Pearce, I., Brooker, R., Scott, D., Welch, D., Woodin, S., 2003. Interplay between nitrogen deposition and grazing causes habitat degradation. Ecology letters 6, 141-146.
Wedin, D., Tilman, D., 1993. Competition Among Grasses Along a Nitrogen Gradient: Initial Conditions and Mechanisms of Competition. Ecological Monographs 63, 199-229.

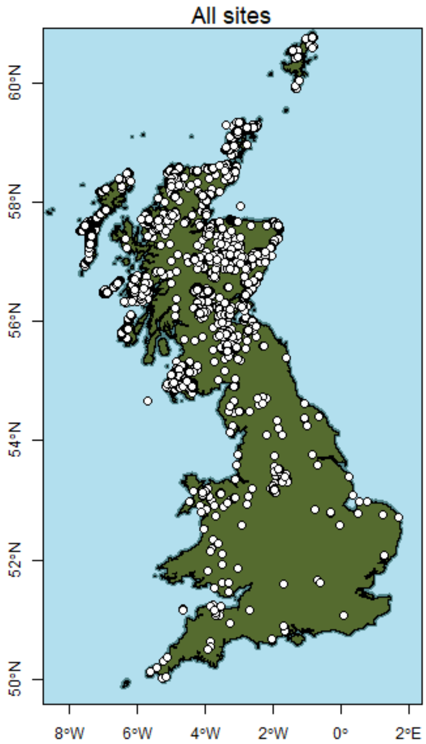

Figure 1. Distribution of sampling sites across all surveys. See Supplementary Figure 1 for mapping of individual studies and Tables 1 and 2 for details of surveys.

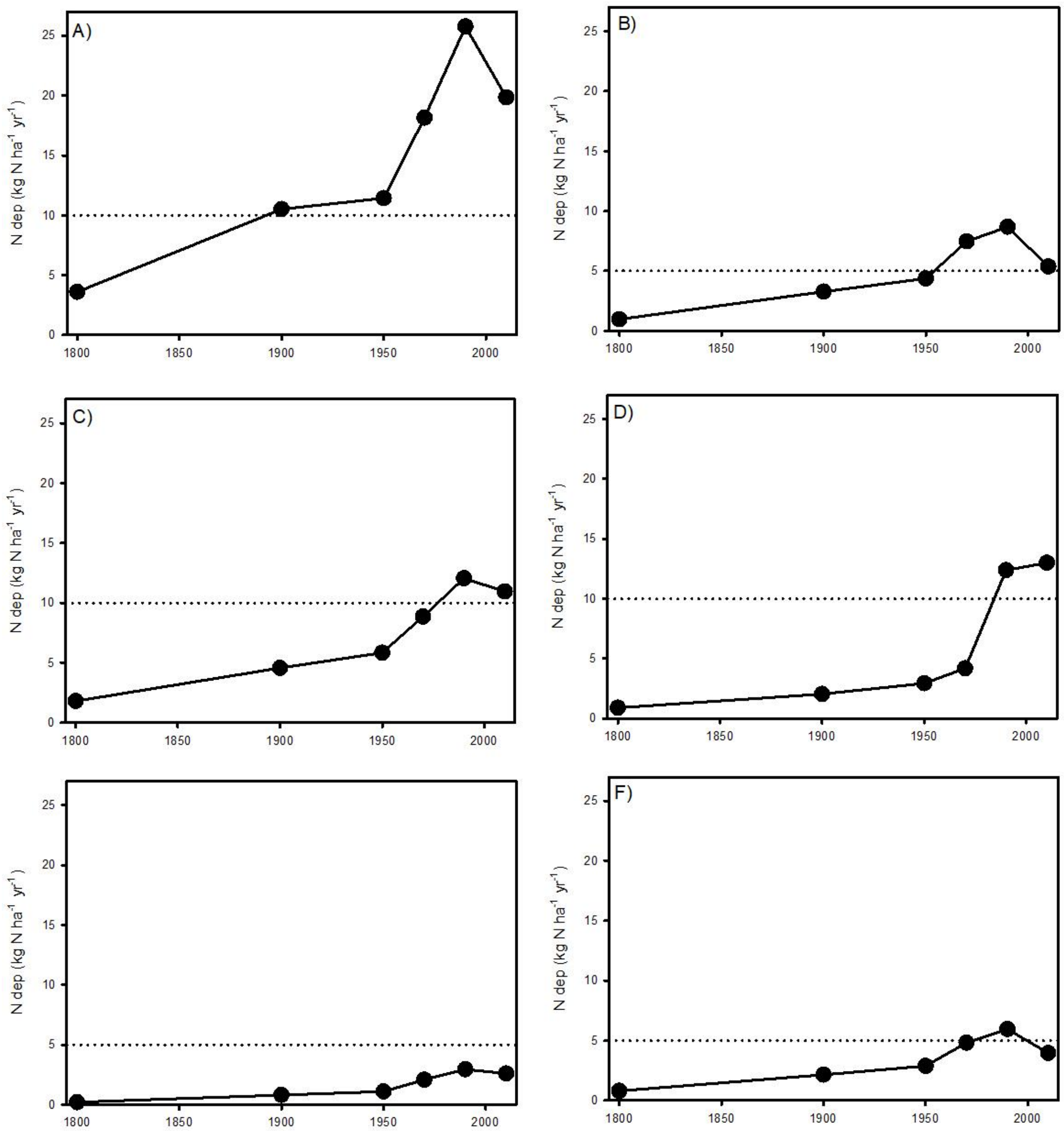

Figure 2. N deposition chronologies for selected sites included in the vegetation surveys. A) Coed Poeth (EDM); B) Allt Cragach (PAYN); C) Gladhouse (B.AGRASS); D) Hilldavale (B.CHEATH); E) Bennadrove (PAYN) and F) Cawdow (TU.BOG). N deposition values are vegetation-specific estimates for low-growing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

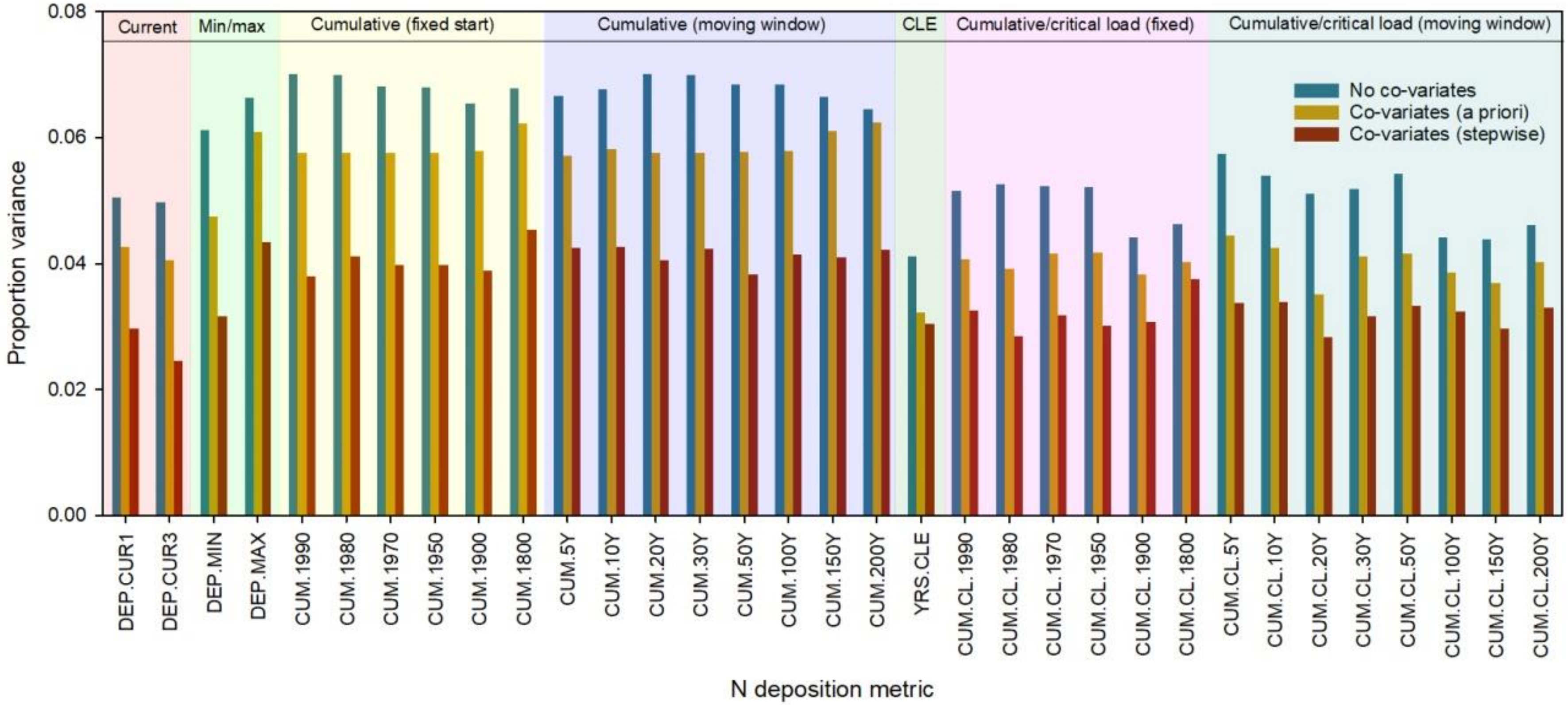

Figure 3. Compositional variance explained by alternative N deposition metrics for all habitats. Background shading denotes different ‘families’ of metrics. See Table 3 for metric codes.

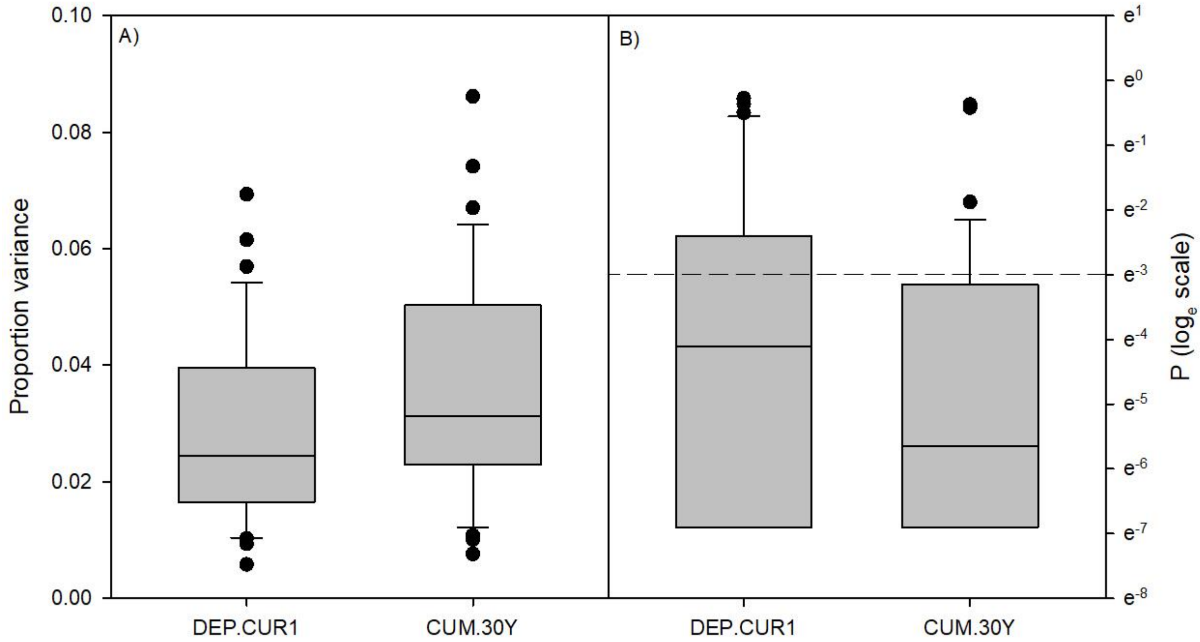

Figure 4. Comparison of single-year current deposition (DEP.CUR1) and 30 year cumulative deposition (CUM.30Y) for all vegetation datasets (without co-variates) in terms of explained variance (A) and P-value (B). Dashed horizontal line shows P=0.05.

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 and1959 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^2 (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 *Racomitrium* heath survey (Armitage et al., 2014)	Thirty six *Racomitrium* 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^2. The cover of all species (including bryophytes and lichens) was estimated.	Armitage- *Racomitrium* 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^2 to more than 9m^2 but were typically 4 m^2. 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: *Calluna* heath (NVC: H10,H11,H12,H13,H15,H17), *Vaccinium* heath (H18,H19,H20), *Racomitrium* heath (U10), acid grassland (U1d,U1e,U4a,U4c,U4d,U4e,U13,U20), calcareous grassland (CG2,CG10,CG11), *Lolium* 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- Racomitrium heaths; Birse- springs; Birse- swamps; Birse- Vaccinium 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 *Racomitrium* heath survey (Britton et al., 2018)	This survey targeted *Racomitrium* 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^2 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- *Racomitrium* 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^{-1} yr^{-1} (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^{-1} yr^{-1})	Critical load value (kg ha^{-1} yr^{-1})	Mean annual temperature (°C)	Mean annual precipitation (mm)	Altitude (m)	Additional environmental variables included in pool available for selection.
Heathlands										
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.
Grasslands										
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.
Wetlands										
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.
Montane habitats										
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.
Sand dune habitats										
CEH dune grasslands	CEH.DUGR	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.

Metric family	Metric	Code
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