

# An exceedance score for the assessment of the impact of nitrogen deposition on habitats in the UK

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## ARTICLE INFO

### Keywords:

Nitrogen deposition  
Uncertainty  
Critical loads  
Eutrophication  
Integrated assessment modelling

## ABSTRACT

Large areas of nitrogen-sensitive habitats are currently estimated to be in exceedance of their critical loads (CLs) as indicators for protection from nitrogen deposition. In the UK, deposition estimates from the semi-empirical Concentration Based Estimated Deposition (CBED) model are used for official reporting of current exceedances. The UK Integrated Assessment Model (UKIAM) framework is designed to provide future projections of concentrations and deposition due to projected changes in emissions. UKIAM has been extended to provide alternative deposition estimates aligned with those of CBED, and the results combined with the range in habitat CL values to create an exceedance score, leading to a probabilistic evaluation of CL exceedances. The utility of the method is demonstrated by analysing a series of hypothetical scenarios. It is shown that NH<sub>3</sub> mitigation is likely to be four times more effective in reducing CL exceedances in the UK than the mitigation of NO<sub>x</sub> emissions.

## 1. Introduction

By 2030 the UK government plans to reduce ammonia (NH<sub>3</sub>) and nitrogen oxide (NO<sub>x</sub>) emissions by 16% and 73%, respectively, relative to emissions in 2005 as outlined in the 2019 Clean Air Strategy (DEFRA, 2019). In addition to the well-documented human health impacts of these pollutants, they have a significant impact on the health of ecosystems through direct effects from atmospheric concentrations and when deposited in wet (NO<sub>3</sub><sup>-</sup>, NH<sub>4</sub><sup>+</sup>) and dry (NH<sub>3</sub>, HNO<sub>3</sub>, NO<sub>x</sub>) forms. This impact includes the eutrophication of soils and freshwater, leading to the loss of species that become less competitive when nitrogen (N) availability is increased; and acidification, leading to effects such as reduced fertility and nutrient deficiencies and the loss of acid-sensitive species. Sulphur emissions also contribute significantly to acidification: however the deposition of sulphur oxides (SO<sub>x</sub>) is no longer the main cause of acidification in the UK following a 94% decrease in the UK SO<sub>2</sub> emissions between 1970 and 2010 (RoTAP, 2012). As of 2017, 39% of UK ecosystems were considered to be in exceedance of their critical loads (CLs) for acidity, while 58% were considered to be in excess of their eutrophication CL (Rowe et al., 2020).

The exceedance of CLs has long been used as a method to evaluate the harm caused to specific habitats by the deposition of reactive

nitrogen, and has proven particularly useful for target setting and policy development (e.g. the UNECE's Gothenburg Protocol). CLs are estimates of the deposition rate below which a habitat is not considered to be significantly harmed according to current knowledge (Nilsson and Grennfelt, 1988). For N deposition, CLs are evaluated empirically, based on experiment and field observations, and agreed at expert workshops at the UNECE level under the Convention on Long-range Transboundary Air Pollution. Given the empirical nature of their derivation, there is a degree of subjectivity in their evaluation, in addition to uncertainties underlying the observations on which these judgements are made. Further, some studies (e.g. Armitage et al., 2014; Payne et al., 2013) have suggested that vegetation changes incrementally with N deposition, with no threshold below which no effects are seen. Given these factors, there is a question as to how meaningful the exceedance of a single limit value (i.e. the CL value) is in assessing the harm caused to habitats through eutrophication. Despite this, CL values do provide a useful measure of the varying degrees of resilience of different habitats to excess nitrogen. Exceedances of CL values also continue to be commonly used for policy development in many European countries, including the UK (e.g. Trends Report (Rowe et al., 2020)).

In the UK the Concentration Based Estimated Deposition (CBED) model is used for official reporting of CL exceedances of deposition for

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<https://doi.org/10.1016/j.envsoft.2022.105355>

Received 3 August 2021; Received in revised form 1 February 2022; Accepted 5 February 2022

Available online 8 February 2022

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sensitive habitats (Hall et al., 2015). The CBED model estimates dry deposition rates using a “big leaf” model (Smith et al., 2000); combining gas and particulate concentration maps, constrained to measurements, with maps of vegetation cover. The model accounts for vegetation-specific deposition velocities and includes a simple model of the complex bi-directional exchange of ammonia due to stomatal emission. Wet deposition is estimated by combining spatially distributed measurements of concentrations in precipitation with annual precipitation maps and a two-dimensional seeder-feeder rainfall (“seeder” rain from high level cloud falls through lower hill “feeder” clouds) model (Dore and Choularton, 1992) to estimate wet deposition rates (Smith and Fowler, 2000). Considerable uncertainty is associated with these deposition estimates, particularly in areas of higher altitude and precipitation, where deposition rates are greatest. This is due to a number of reasons, including a shortage of measurements at high altitude and the added complexity of orographic enhancement. The direct use of concentrations in precipitation measurements as input to the model also means that there is a shortage of measurements for model validation. Smith and Fowler (2000) showed that uncertainty in the seeder-rain enhancement factor, specified as a parameter of the seeder-feeder model, has a significant effect on the model output. This factor, which is used to scale the estimated deposition due to feeder rain, is assumed to be equal to 2 based on a single set of experiments conducted on the Great Dun Fell over the course of a few days (Choularton et al., 1988). While reasonable agreement has been shown with subsequent measurements (Dore et al., 2001; Beswick et al., 2003), such validation datasets are scarce. Further, measurements of wet deposition from bulk collectors are known to overestimate wet deposition as they can also capture a degree of dry deposition (e.g. Fowler and Cape 1984; González Benítez et al., 2009; Cape et al., 2011).

In contrast, Atmospheric Chemical Transport Models (ACTMs) are generally found to underestimate deposition of reactive N when compared to measurements in the UK (Dore et al., 2015) and across Europe (Fagerli et al., 2021). To what extent this discrepancy is due to bias in the models or the measurements is currently not well understood. One cause could be an underestimation of NH<sub>3</sub> emissions; the UK National Atmospheric Emission Inventory estimates were recently shown to give lower UK NH<sub>3</sub> emissions when compared to estimates derived from satellite observations (Marais et al., 2021). Further research is required before these differences are fully understood, and a substantial increase in measurement sites is likely required.

The UK Integrated Assessment Model (UKIAM) is a model framework, consisting of a family of physically-based models, used to investigate the impact of future emissions scenarios on UK air quality and ecosystem health (Oxley et al., 2013; ApSimon et al., 2021). A scenario can be run within an hour and possible outputs include national concentration and deposition maps, estimates of exceedances of CLs and a full breakdown of source apportionment; allowing the identification of the most harmful sources by habitat and by region. UKIAM also underestimates deposition relative to measurements and the CBED model. The degree of underestimation by UKIAM relative to CBED is of the same order as the mean underestimation of the ACTMs considered by Dore et al. (2015) relative to measurements. The areas of greatest disagreement between UKIAM and CBED correspond to the areas of greatest uncertainty in deposition, which tend to be areas of higher altitude and precipitation where complex wet deposition processes occur, such as occult and seeder-feeder deposition.

In this paper we outline a new methodology implemented within the UKIAM framework for the assessment of the impact of N deposition on sensitive habitats in the UK on a national and regional level. The approach is based on the Joint Nature Conservation Committee’s (JNCC) Nitrogen Decision Framework’s method designed for national level evaluation while accounting for the uncertainty in both the N deposition and CL estimates (Jones et al., 2016). We make use of CLs as a measure of the varying sensitivities of habitats to eutrophication, but remove the dependence on the exceedance of a single limit value by introducing an

exceedance score based on two CL values; a minimum and a maximum. These two CL values are combined with two deposition estimates based on two fundamentally different models, UKIAM and CBED, providing an upper and lower value in each grid square and used to create an exceedance scale. As the two deposition models used are based on inherently different methodologies, they are unlikely to share the same biases, therefore combining the two provides a more robust analysis than using a single model prediction. Further, the exceedance scale removes the dependency of the analysis on a single, imprecise limit value and avoids step changes in exceedance which can occur when a single limit value is used.

The method is used to assess the state of habitat exceedance of nitrogen deposition for the base year, 2016, in addition to two sets of hypothetical scenarios designed to explore an effective strategy for reducing exceedances in the UK and the degree of abatement required to reach UK Government targets.

## 2. Method

### 2.1. The UKIAM framework

A brief overview of the components of the UKIAM framework relevant to ecosystem assessment is provided here. For a detailed description of a recent version of the framework see ApSimon et al. (2021).

The framework brings together inputs from several independent models. These include ASAM (ApSimon et al., 1994), used to calculate the imported pollutant contribution from other countries; BRUTAL (Oxley et al., 2009), a bottom-up traffic model which estimates the emissions from traffic; and UKIAM5, a 5 km resolution sub-model used to estimate concentrations and deposition due to UK sources in addition to international shipping. NO<sub>x</sub> emissions from shipping are substantial and have a significant impact both on ecosystem health and on air quality in the UK (ApSimon et al., 2019). UKIAM5 estimates deposition for future scenarios by scaling Source-Receptor (S-R) footprints of deposition, generated by an ACTM, to reflect the change in emissions relative to a base case.

The S-R footprints are generated by reducing the emissions from each source individually relative to a base case, before calculating the deposition rate using the ACTM. The difference between the calculated deposition and the base case is then used to calculate a map of the change in deposition per unit change in emission, i.e. the S-R footprint. This is done separately for each pollutant.

For the work presented in this study the FRAME (Fine Resolution Atmospheric Multi-pollutant Exchange) model (Singles et al., 1998) is used to generate the UK S-R footprints; however, any atmospheric dispersion model could be used to generate the S-R footprints and implemented within the UKIAM framework. This scaling of S-R footprints depends on the assumption that concentration and deposition estimates vary linearly with emissions. This linear assumption has been shown to be acceptable for variations in emissions of  $\pm 40\%$  (Aleksankina et al., 2018), i.e. within this range the effect of non-linearity is acceptable relative to other uncertainties. The framework also includes the UKIAM1 sub-model, used to generate 1 km resolution maps of concentrations for primary pollutants (PM and NO<sub>x</sub>/NO<sub>2</sub>), for example used to develop policy aimed at meeting the UK’s targets for PM<sub>2.5</sub> concentrations set out in the UK’s Clean Air Strategy (DEFRA, 2019). However, in this paper we focus on the utility of the UKIAM framework in evaluating N deposition and corresponding CL exceedances at a national level.

In total UKIAM considers 94 UK sources. These are divided between ten SNAP (Selected Nomenclature for Air Pollution) sectors as defined in the UK’s National Atmospheric Emissions Inventory (NAEI). Table 1 shows the sectors and the associated emissions of NO<sub>x</sub> and NH<sub>3</sub>. International shipping in the sea areas surrounding the UK is not included in the NAEI but is also considered. The NH<sub>3</sub> emissions are dominated by SNAP 10; agriculture. These NH<sub>3</sub> emissions are mainly due to livestock,

**Table 1**  
SNAP sectors and major NH<sub>3</sub> and NO<sub>x</sub> sources for 2016 (base year).

SNAP sector	NH <sub>3</sub> emissions (ktonnes)	NO <sub>x</sub> emissions (ktonnes)
1. Combustion in energy and transformation industries	0.2	139.4
2. Non-Industrial combustion plants	1.4	41.1
3. Combustion in manufacturing industry	0.3	135.6
4. Production processes	4.2	0.6
5. Extraction and distribution of fossil fuels and geothermal energy	0.0	1.9
6. Solvent and other product use	1.2	0.0
7. Road transport	3.4	295.1
8. Other mobile sources and machinery	0.3	171.6
9. Waste treatment and disposal	13.0	1.3
10. Agriculture	239.9	0.0
Beef	55.5	0.0
Dairy	63.5	0.0
Pigs	18.5	0.0
Layers	9.3	0.0
Other poultry	25.9	0.0
Sheep	3.9	0.0
Other livestock	6.9	0.0
Fertiliser	56.2	0.0
11. Natural	16.8	0.4
International shipping in sea areas around UK	0.0	665.2
<b>Total</b>	<b>280.6</b>	<b>1502.1</b>

but fertiliser also contributes a significant amount (56 ktonnes). NO<sub>x</sub> emissions are distributed between several SNAP sectors, mainly SNAP 1, 3, 7 and 8, in addition to international shipping. The importance of including international shipping is evident from the very high emission of NO<sub>x</sub> surrounding the UK from this sector (ApSimon et al., 2019).

The imported contribution from other countries is also significant and the magnitude is estimated using ASAM, which uses S-R footprints derived from the European-scale model EMEP (Simpson et al., 2012). These EMEP S-R footprints are used to calculate the total deposited N from European emissions. This contribution is then spatially distributed across the UK using FRAME S-R deposition footprints. EMEP is used to provide the magnitude of the contribution from other countries reaching the UK because FRAME is known to underestimate long-range transport of ions such as NO<sub>3</sub><sup>-</sup> and NH<sub>4</sub><sup>+</sup> due to an assumption of constant drizzle which overestimates washout of these pollutants. However FRAME then gives a more detailed spatial mapping of the deposition reflecting important orographic enhancement effects on wet removal over land at higher altitudes. The use of both EMEP and FRAME therefore allows both a reasonable estimate of the contribution from other countries and the enhanced washout in areas of higher precipitation. The maps for the imported contribution from other countries and for shipping is provided in the supplementary material (Figs. S4 and S5).

Different deposition velocities are assumed for short habitats, such as grasses and dwarf shrub heath, than for taller habitats, such as woodlands. Two separate maps are used; the first for short habitats which is referred to as the “moorland” deposition map, and a second for woodland, referred to as the “woodland” deposition map. These deposition values are calculated for all grid squares regardless of whether a given habitat exists within the grid square and therefore do not represent the actual deposition, which depends on the area of moorland and woodland habitats within a grid square. The moorland and woodland maps for deposition derived from beef production in 2016 are provided in the supplementary material as an example (Fig. S2).

The sources given in Table 1 are broken down further into 94 individual sources; the breakdown for agriculture is shown. The latest version of UKIAM (version 6R) allows the emissions from different regions of the UK (England, Wales, Scotland, Northern Ireland and London) to be varied independently, although the distribution within these regions remain fixed.

## 2.2. Nitrogen-sensitive habitats

In total 13 nitrogen-sensitive habitats are considered for analysis. These are given in Table 2 along with the range of CL values. These habitats match those used for official reporting in the UK (Rowe et al., 2020). The UKIAM includes a library of 1 km × 1 km maps indicating the area of land covered by each habitat in each grid square, along with the appropriate recommended CL value for those habitats for which this value is not constant across the country. Therefore, while depositions are estimated at 5 km resolution, all exceedance statistics and maps are generated at 1 km resolution due to the higher resolution of the eco-systems data.

Along with minimum and maximum CLs, a recommended value (CLrec) within this range is also given for each habitat, although increasingly use of the minimum CL value is recommended for this. Here the recommended CL value is used to calculate the exceedance unless otherwise stated, while the minimum and maximum values are used to derive the exceedance score (Section 2.4).

## 2.3. Accumulated exceedance

The Accumulated Exceedance (AE) is used as a metric for the level of exceedance of the recommended CL for a particular habitat at the national or regional level. The AE for a habitat is calculated as follows:

$$AE \text{ (kg/year)} = \text{exceedance (kg/ha/year)} \times \text{exceeded area (ha)}$$

The Average Accumulated Exceedance (AAE) is used as a metric of the exceedance across all N-sensitive habitats. The AAE is calculated by dividing the total AE for all habitats by the total area of all habitats (Hall et al., 2015).

**Table 2**  
Nitrogen sensitive habitats considered for analysis.

Habitat	Area (km <sup>2</sup> )	EUNIS habitat class	Habitat type	CLmin-CLmax range (kg N/ha/year)	CLrec (kg N/ha/year)
Acid grassland dry/wet	15213	E1.7 & E3.52	Short	10-15/10-20	10/15
Calcareous grassland	3565	E1.26	Short	15-25	15
Dwarf shrub heath (wet & dry)	24776	F4.11 & F4.2	Short	10-20	10
Montane	5487	E4.2	Short	5-10	7
Bog	3128	D1	Short	5-10	8, 9, 10 <sup>a</sup>
Managed coniferous woodland	8370	G3	Tall	5-15	12
Managed broadleaved woodland	7473	G1	Tall	10-20	12
Beech woodland (unmanaged)	718	G1.6	Tall	10-20	15
Acidophilous oak woodland (unmanaged)	1407	G1.8	Tall	10-15	10
Scots Pine woodland (unmanaged)	201	G3.4	Tall	5-15	12
Other unmanaged woodland	1747	G4	Tall	5-15	12
Dune grassland	257	B1.4	Short	8-15	9 or 12 <sup>b</sup>
Saltmarsh	276	A2.53/54/55	Short	20-30	25

<sup>a</sup> Spatially varied and dependent on local rainfall.

<sup>b</sup> 9 used for acid dunes and 12 for non-acid.

### 2.4. Exceedance score

The exceedance score is derived in order to provide a more stable and reliable indicator of ecosystem protection than provided by the exceedance of a single CL value estimated using a single deposition value. The method is based on that designed for national-scale evaluation (“Factor 1” score) outlined in the JNCC’s Nitrogen Decision Framework (Jones et al., 2016).

We use the minimum and maximum deposition values for a given habitat in each grid square to provide an indicator of the uncertainty (rather than the 95% confidence interval used in the Nitrogen Decision Framework), as illustrated in Fig. 1. The two deposition estimates consist of the UKIAM estimate, and the UKIAM-Scaled estimate. The UKIAM-Scaled estimate is generated by multiplying the UKIAM deposition in each grid square by the ratio of the CBED and UKIAM values for the 2016 base year:

$$N_{UKIAM-Scaled}^i = N_{UKIAM}^i \times \left( \frac{N_{CBED}^{2016}}{N_{UKIAM}^{2016}} \right)^i \text{ for each grid square } i.$$

The resulting UKIAM-Scaled map of deposition combines important spatial information based on the empirical CBED model, for example areas where measured deposition is greater than the modelled UKIAM deposition, with estimates of the relative reduction in deposition estimated by UKIAM. This allows us to incorporate the range in deposition estimates into our analysis of future scenarios, which is otherwise not possible for CBED due to its dependence on measurements. We then define our minimum and maximum deposition values for each grid square as:

$$\begin{aligned} N_{min}^i &= \min(N_{UKIAM}^i, N_{UKIAM-Scaled}^i), \\ N_{max}^i &= \max(N_{UKIAM}^i, N_{UKIAM-Scaled}^i). \end{aligned} \quad (1)$$

This is done separately for the woodland and moorland deposition estimates. The maps of  $N_{min}$  and  $N_{max}$  for moorland and woodland are shown in Fig. S6. The difference between the deposition estimates provided by the two models varies in magnitude across the UK (Fig. S7). Maps of the ratio of deposition given by CBED and UKIAM are shown in Fig. S8. A statistical comparison of the UKIAM deposition estimates with those of CBED is provided in the supplementary material. UKIAM generally predicts lower values of deposition, particularly in areas of higher precipitation and higher altitude such as much of Scotland, Wales and the lake district, where the uncertainty in deposition estimates and measurements are greatest.

For the base year the UKIAM-Scaled estimate is equal to the CBED estimate since it is the base year values which are used to derive the ratio maps. However, the method can also be used to provide a second estimate of deposition for future scenarios, which are not available directly from CBED.

CL values provide a useful measure of the varying degrees of resilience of different habitats to excess nitrogen. For example, montane is a particularly sensitive habitat and therefore has a lower recommended CL value (7 kg N/ha/yaer) than a more resilient (yet still sensitive) habitat, calcareous grassland, which has a higher recommended CL

value (15 kg N/ha/yaer). The damage per unit N deposited is likely to be higher for montane than calcareous grassland and CLs provide a measure of this difference. Each habitat is assigned a recommended critical load (CLrec) in addition to a minimum (CLmin) and a maximum (CLmax), reflecting the imprecise derivation of CLs. We make use of this range as an indicator of the habitat’s resilience to N deposition, and therefore the likelihood with which a habitat will survive estimated rates of deposited N.

The higher and lower deposition values within each grid square are identified (equation (1)). Six scores, P0, P1, P4 and P5, are defined ranging from highly unlikely to be in exceedance to highly likely to be in exceedance, and P2 and P3 which are defined as marginal due to CL estimates and deposition estimates, respectively (Fig. 1). The score is assigned to each habitat grid square,  $i$ , as follows:

$$P_i = \begin{cases} P_0 & \text{if } N_{max}^i < CL_{min}, \\ P_1 & \text{if } N_{min}^i < CL_{min} \text{ and } N_{max}^i > CL_{min} \text{ and } N_{max}^i < CL_{max}, \\ P_2 & \text{if } N_{min}^i > CL_{min} \text{ and } N_{max}^i < CL_{max}, \\ P_3 & \text{if } N_{min}^i < CL_{min} \text{ and } N_{max}^i > CL_{max}, \\ P_4 & \text{if } N_{min}^i > CL_{min} \text{ and } N_{min}^i < CL_{max} \text{ and } N_{max}^i > CL_{max}, \\ P_5 & \text{if } N_{min}^i > CL_{max}. \end{cases} \quad (2)$$

## 3. Results

### 3.1. Base year assessment

#### 3.1.1. UKIAM deposition estimates and source apportionment

Maps of the total deposition of N, NO<sub>x</sub> and NH<sub>x</sub> estimated by UKIAM for 2016 are given in Fig. 2(a–c). Note the different scale used for the NO<sub>x</sub> and NH<sub>x</sub> components as compared with the total N map. The deposition of NH<sub>x</sub> greatly exceeds that of NO<sub>x</sub>. The greatest deposition occurs in areas of higher altitude and precipitation that are downwind of many emissions sources, such as Wales and north west England. Here orographic enhancement increases the rate of deposition through processes such as occult deposition (Crossley et al., 1992) and seeder-feeder effect (Fowler et al., 1988). The deposition given by UKIAM in Scotland is much lower than the other regions of the UK due to the distance of much of Scotland from major pollution sources. The distribution of NH<sub>x</sub> deposition correlates closely with the distribution of agriculture emissions (Fig. S3) since a large proportion of NH<sub>x</sub> is often deposited locally. Some NO<sub>x</sub> deposition occurs in urban areas which are large sources of NO<sub>x</sub> emissions, but most of the NO<sub>x</sub> deposition occurs after conversion to NO<sub>3</sub><sup>-</sup> during long-range transport, with removal in precipitation enhanced over areas of higher altitude.

Table 3 provides the contribution of UK sources, international shipping and the imported contribution from Europe for each UK region as calculated by UKIAM. This source apportionment is not available for CBED since only the total NH<sub>x</sub> and NO<sub>x</sub> depositions are available - see Fig. 2d–f. Maps of the NH<sub>x</sub> and NO<sub>x</sub> deposition due to these sources are provided in the supplementary material (Figs. S4 and S5).

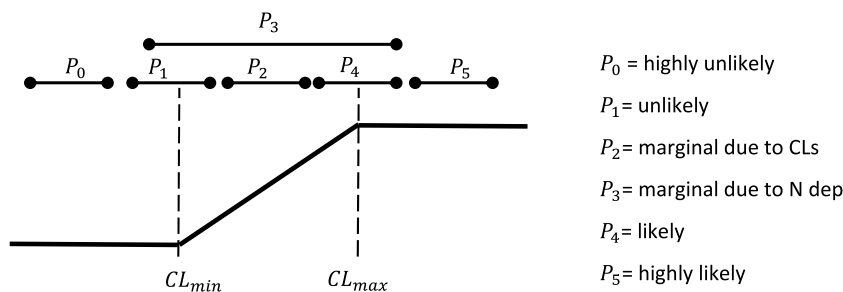


Fig. 1. Illustration of exceedance scores.

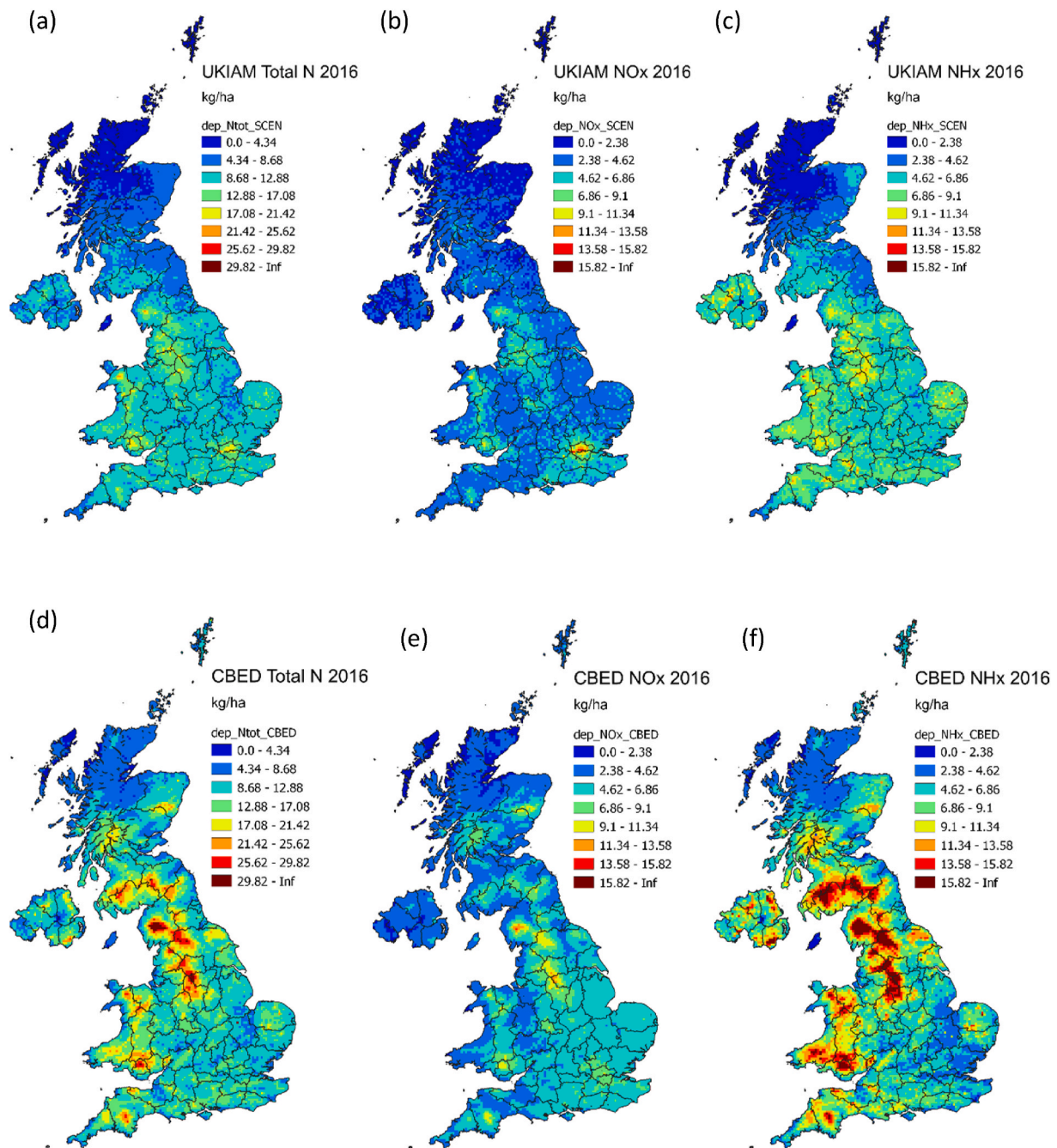


Fig. 2. UKIAM 2016 deposition estimates for (a) N, (b) NO<sub>x</sub> and (c) NH<sub>x</sub> and CBED Average 2015–2017 deposition estimates for (d) N, (e) NO<sub>x</sub> and (f) NH<sub>x</sub>. Units given for all maps in kg-N/ha.

Table 3  
2016 UKIAM deposition source apportionment. Units = ktonnes-N.

		National	England	Scotland	Wales	Northern Ireland
UK Sources	Total NH <sub>x</sub>	97.8	63.6	17.6	10.6	6.0
	Agriculture NH <sub>x</sub>	81.0	51.2	15.3	8.9	5.6
	Total NO <sub>x</sub>	49.7	33.1	9.8	5.0	1.8
	Road transp. NO <sub>x</sub>	21.0	14.8	3.4	1.8	1.0
International Shipping	Total N	13.4	8.8	2.3	1.9	0.4
Imported from Europe	Total N	58.4	35.2	11.3	7.5	4.4
All Sources	NH <sub>x</sub>	132.0	84.5	22.8	15.4	9.3
	NO <sub>x</sub>	87.3	56.1	18.2	9.6	3.4

UK emissions of  $\text{NH}_x$  make by far the greatest contribution to the deposition budget in the UK (97.8 ktonnes), twice that of UK  $\text{NO}_x$  sources (49.7 ktonnes). A large proportion of the UK  $\text{NH}_x$  contribution is due to agriculture emissions (81.0 ktonnes), while the largest contributor to  $\text{NO}_x$  deposition within the UK is road transport (21.0 ktonnes). The relative contribution of agriculture is particularly high in Northern Ireland where it constitutes 93% of the deposited  $\text{NH}_x$ .

The imported contribution of total nitrogen is also provided in Table 3 for international shipping and other European countries. The international shipping contribution is entirely  $\text{NO}_x$  since  $\text{NH}_x$  emissions from shipping are negligible. This contribution is mainly concentrated in the south of England and Wales, near the major shipping lanes (see Fig. S4). The contribution from other countries is highest in Wales, Northern Ireland and the south east of England due to their proximity to other countries. The  $\text{NH}_x$  contribution from across the border in Ireland is particularly high (Fig. S5).

### 3.1.2. CBED deposition estimates and model comparison

We refer to the base case as 2016 because UKIAM uses NAEI emission estimates for this year. However, it should be noted that the CBED deposition values are in fact derived from a three-year average spanning 2015–2017.

Fig. 2(d–f) shows this three-year average N,  $\text{NO}_x$  and  $\text{NH}_x$  deposition in the UK. It is immediately evident that CBED gives significantly higher deposition than UKIAM (Fig. 2(a–c)). This is particularly true in areas of higher altitude such as much of Scotland, Wales and the north west of England. Conversely, UKIAM gives higher deposition values in urban areas, where London in particular stands out. Despite these differences, the areas of highest deposition rates are generally in agreement between the two models. Maps of the difference between the two models and the ratio of deposition values are shown in Fig. 3 and Fig. 4 respectively.

Table 4 shows the deposition budget in each region of the UK as predicted by the two models. With the exception of Scotland, UKIAM estimates the total deposited  $\text{NH}_x$  and  $\text{NO}_x$  to be 83% and 80%, respectively, of that estimated by CBED. However, in Scotland the deposition is 46% and 60% of that given by CBED for  $\text{NO}_x$  and  $\text{NH}_x$ , respectively.

A statistical comparison of the two models is given in the

supplementary material.

### 3.1.3. Critical load exceedances

Fig. 5 shows the AAE for all ecosystems calculated using the recommended critical loads (Table 2) given by (a) UKIAM and (b) CBED. Due to the higher deposition predicted by CBED, the exceedance tends to be higher than for UKIAM across much of the UK. This is particularly true in Wales and the north of England. In Scotland, the exceedance predicted by CBED is also higher than that by UKIAM, however it is considerably lower than that for the other regions of the UK (Table 5). Over half of the N-sensitive habitat area considered is in Scotland, therefore the UK-wide statistics are heavily dependent on the exceedances in this region. As the situation in Scotland is not reflective of that in the rest of the UK, it is worth considering the average exceedance across England, Wales and Northern Ireland separately. These are also given in Table 5 and can be seen to be significantly higher than the UK-wide average. The significance of agriculture is evident from both models with both predicting high exceedance in areas near high agricultural  $\text{NH}_3$  emissions, shown in Fig. S3 (a).

### 3.2. Exceedance score

Table 6 shows the percentage area of habitat attributed to each exceedance score for 2016. For the UK, 46.5% of the N-sensitive habitat area is either highly unlikely or unlikely in exceedance. This lies between the % area not in exceedance of the recommended CL estimated by CBED and UKIAM independently in Section 3.1.3 of 42% and 57%, respectively. Similarly, for Scotland 71.3% of the N-sensitive habitat area is either highly unlikely or unlikely in exceedance, which lies between the independently estimated range of 65–87%. For Wales and N. Ireland, the percentage area in these lowest two exceedance scores is lower than the range derived from the two models. However, for these two regions the percentage area assigned to P2 and P3 is much greater, indicating a greater proportion of habitat area near the exceedance limit. Therefore, the estimated proportion of protected habitat area in these two regions is less certain than that for England and Scotland which have a lower proportion of habitat area assigned to the P2 and P3 scores.

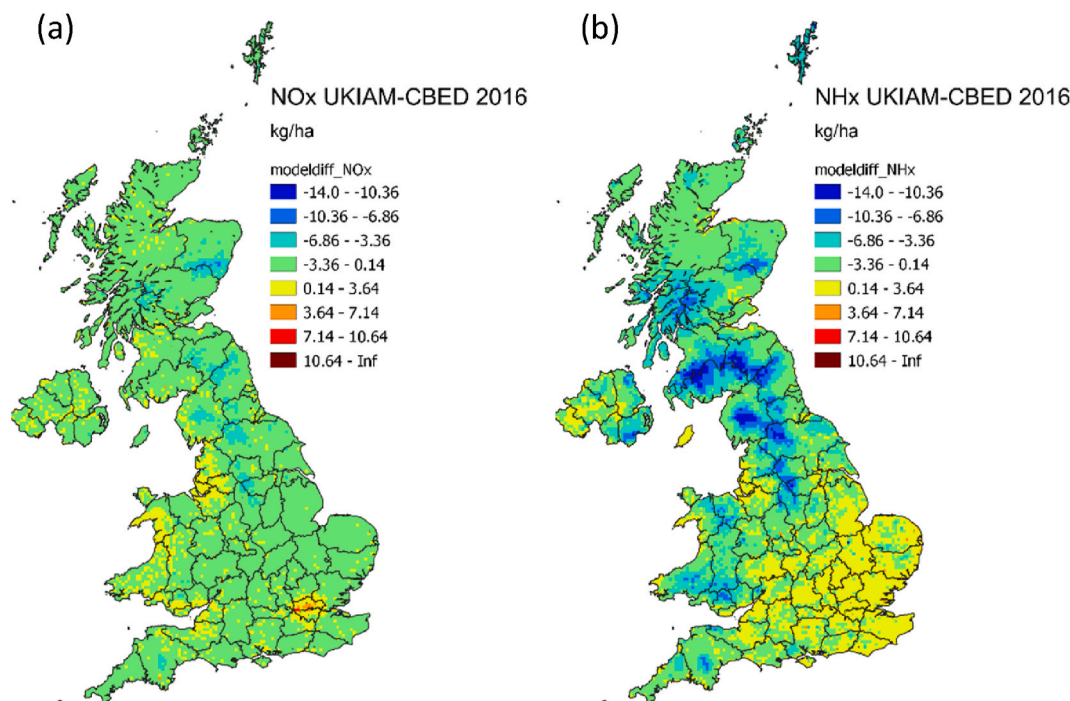


Fig. 3. 2016 model difference (UKIAM - CBED) for (a)  $\text{NO}_x$  and (b)  $\text{NH}_x$  deposition.

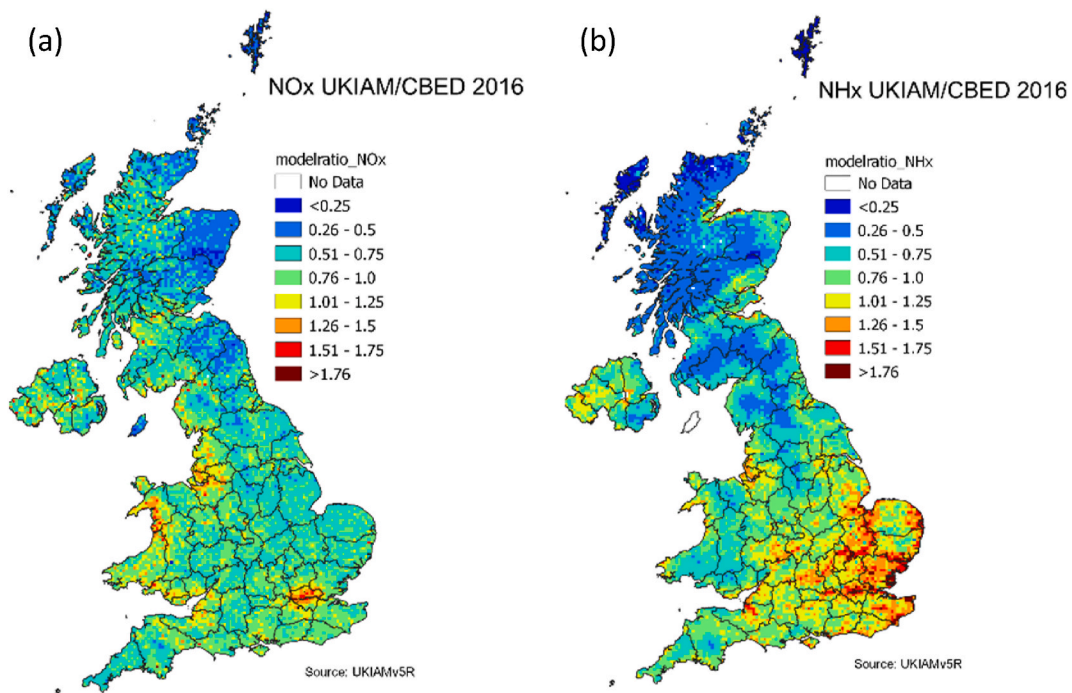


Fig. 4. 2016 UKIAM/CBED ratio for (a) NO<sub>x</sub> deposition and (b) NH<sub>x</sub> deposition.

Table 4  
2016 total N deposition by region for UKIAM and CBED.

		National	England	Scotland	Wales	Northern Ireland	England, Wales & N. Ireland
NH <sub>x</sub> (kt-N)	UKIAM	132.0	84.1	23.0	15.3	9.3	109
	CBED	180.8	97.8	50.0	21.2	11.8	130.8
	UKIAM/CBED %	73	86	46	72	79	83
NO <sub>x</sub> (kt-N)	UKIAM	87.3	55.8	18.0	9.6	3.4	69.3
	CBED	116.5	71.6	30.1	10.4	4.4	86.4
	UKIAM/CBED %	75	78	60	92	78	80

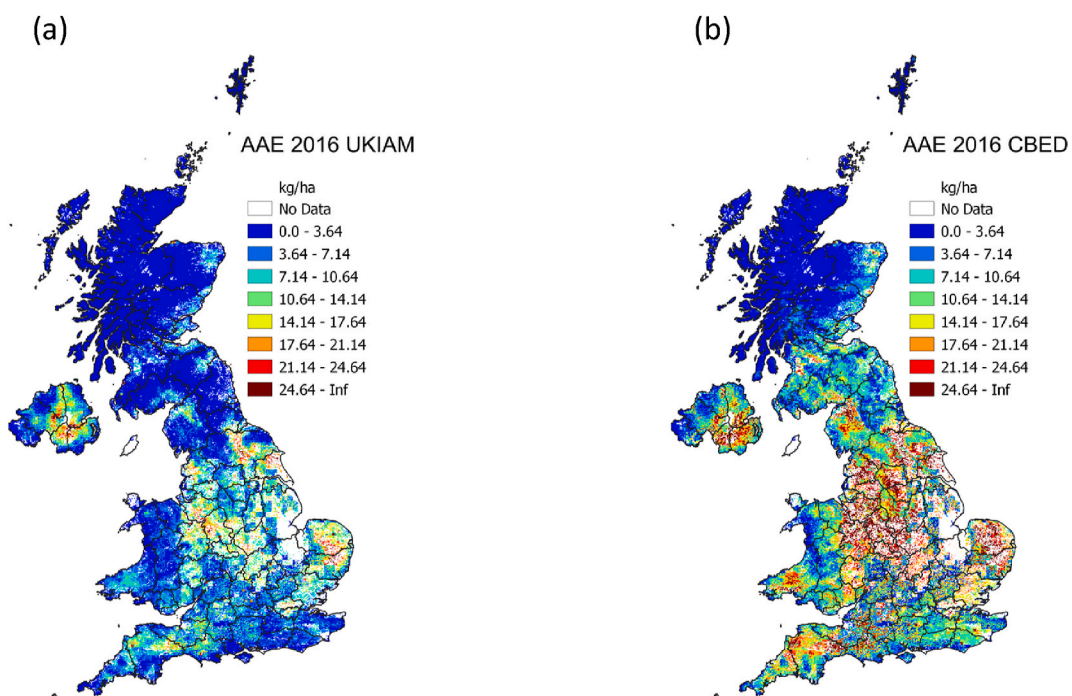


Fig. 5. 2016 Average Accumulated Exceedance for all ecosystems for (a) UKIAM and (b) CBED.

**Table 5**  
2016 Average Accumulated Exceedance for each region of the UK.

	Habitat area (km <sup>2</sup> )	% area exceeded		Average Accumulated Exceedance (kg/ha)	
		UKIAM	CBED	UKIAM	CBED
England	19324	86	95	6.2	12.3
Wales	6822	88	88	5.1	8.6
N. Ireland	3427	83	84	6.0	7.9
Scotland	43053	13	35	0.3	1.9
UK	72625	43	58	2.6	5.6
England, Wales & N. Ireland	29572	86	93	5.9	10.9

**Table 6**  
Exceedance index for each region of the UK 2016. P0 = highly unlikely, P1 = unlikely, P2 = marginal due to CLs, P3 = marginal due to N dep. Estimates, P4 = likely, P5 = highly likely.

	P0	P1	P2	P3	P4	P5
England	2.4	5.4	17.6	0.8	28.4	45.4
Wales	2.2	2.3	34.5	0.2	24.1	36.7
N. Ireland	3.4	3.9	35.3	0.2	15.5	41.7
Scotland	52.2	21.3	11.8	1.2	11.0	2.5
UK	32.0	14.5	16.6	0.9	17.1	19.0
England, Wales & N. Ireland	2.5	4.5	23.5	0.6	25.9	42.9

The regional differences in habitat protection are evident with a much more positive outlook in Scotland than the remainder of the UK.

For each region the percentage area of habitats which lie within the P2 score is much greater than that for P3. The percentage area of habitats given the P3 score is very low, including in Scotland despite the large differences in deposition here between the two models. While the difference between the deposition estimates of the two models often exceeds the range in habitat CLs, this tends to occur for grid squares for which both deposition estimates are greater than the minimum CL value, leading to a P4 or P5 score. Whether a habitat area is deemed to be in exceedance or not is therefore considerably more sensitive to the range in critical load values than the range in deposition estimates.

Fig. 6 (and Table S4 in supplementary material) shows the exceedance scores for each habitat across the UK in 2016. It is immediately

evident that woodland habitats are at greatest risk, other than scots pine which is entirely within Scotland, with “other unmanaged woodlands” in particular danger.

A large variation in exceedance is estimated for bog, with 35.8% in category P0 and 34.5% in category P5. This indicates a large variation in the habitat’s exceedance depending on location. This variation is not captured by the % area in exceedance and AAE statistics presented in Table S2 (supplementary material). While the % area in exceedance given by UKIAM and CBED are fairly consistent with the highly likely in exceedance percentage, at 36% and 41%, respectively, the variation in exceedance of the bog areas is not clear from the AAE values, which are relatively low (1.7 kg/ha and 3.2 kg/ha). UKIAM can also provide a regional breakdown of these statistics (Tables S5–S8), which reveals that this variation is due to low exceedances in Scotland and high exceedances elsewhere.

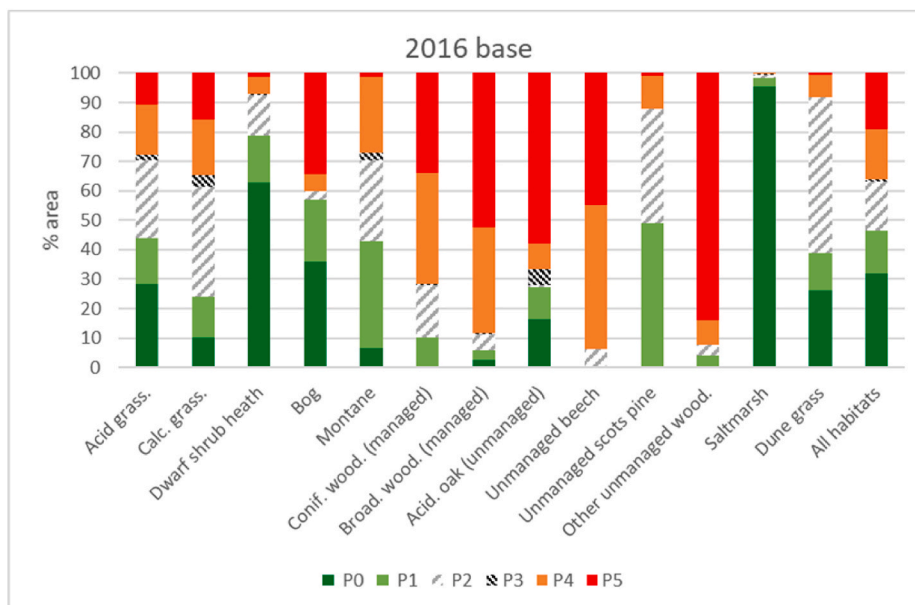
The scale provided by this approach also highlights the uncertain picture for calcareous grassland, scots pine and dune grass, where large proportions of the habitats lie within the P2 category (37.3%, 38.7% and 53.1%, respectively), indicating that for a large proportion of grid squares the deposition estimates lie within the CL range.

### 3.3. Scenario analysis

We first consider three scenarios as an initial investigation into the most effective strategy to reduce CL exceedance in the UK. These “selective” scenarios include a 40% reduction in all imported emissions (imported from other countries and sea areas, including international shipping), a 40% reduction in all UK NO<sub>x</sub> emissions and a 40% reduction in all UK NH<sub>3</sub> emissions. The absolute contribution of each of these sources to the total N deposition, in addition to the spatial distribution of the deposition due to each of these sources, varies significantly, as seen in Table 3 and Figs. S4 and S5. Due to these differences their relative impacts on CL exceedances are likely to vary significantly. By individually reducing each of these components by an equal proportion we are able to explore their relative impacts on exceedances.

We then consider four “blanket” scenarios abating all NH<sub>3</sub> and NO<sub>x</sub> emissions, both domestic and imported, by 20%, 30%, 40% and 50%, in order to investigate the degree of improvement that can be expected from varying degrees of abatement.

Fig. 7 shows a map of the reduction in AAE for each scenario as given by UKIAM. It is clear that the 40% reduction in UK NH<sub>3</sub> emissions is the



**Fig. 6.** % Area of habitat in each exceedance category for 2016 base case.



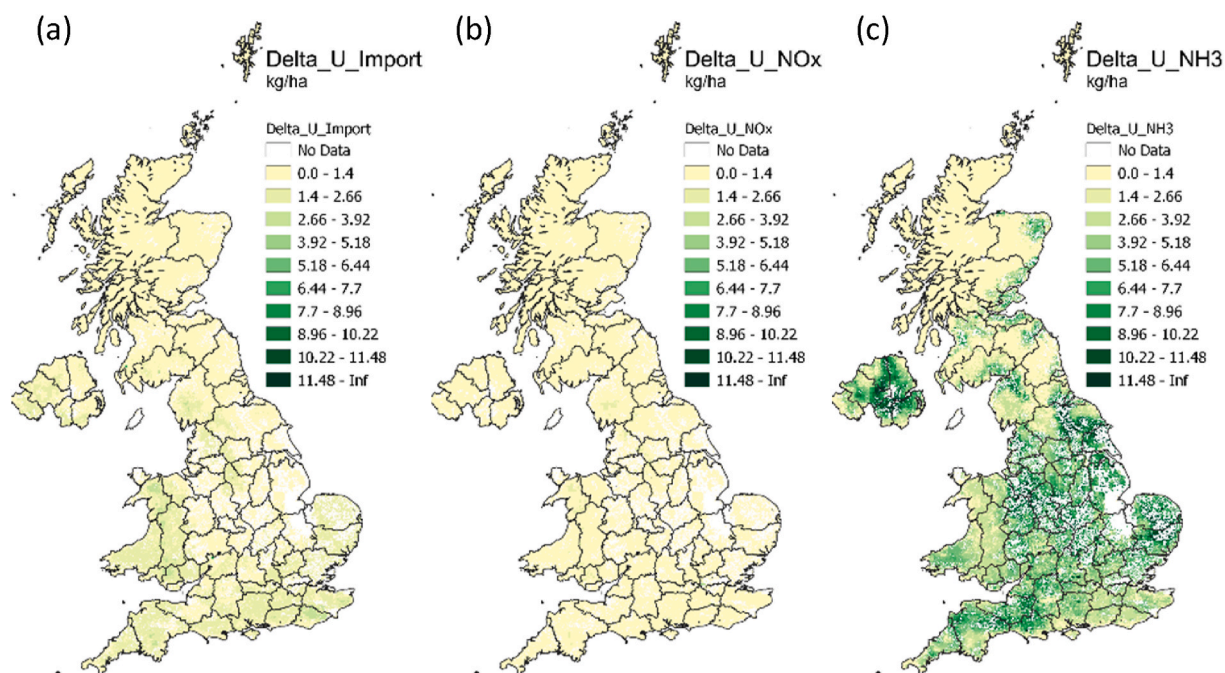


Fig. 7. Map of reduction in AAE (kg/ha) for the (a) abated imported contribution, (b) abated UK NO<sub>x</sub> and (c) abated UK NH<sub>3</sub> scenarios relative to the 2016 base case as given by UKIAM.

scenario which most effectively reduces the AAE. For this scenario both UKIAM and UKIAM-Scaled (see Fig. S9) predict significant reductions in AAE across much of England, Wales, Northern Ireland and southern Scotland. The reductions predicted for the abated imported contribution and abated UK NO<sub>x</sub> emissions are much lower. For the abated imported contribution scenario, the reductions are highest in Wales, the north west of England and the south east of England, which correspond to the areas of higher deposition of imported emissions. The reduction in AAE due to UK NO<sub>x</sub> abatement is low across the entire UK, with slightly higher values in the north west of England.

Table 7 provides the reduction in N deposition for each scenario relative to the 2016 base case, in addition to the change in % area of habitat in the lower two and higher two exceedance categories; representing “highly unlikely or unlikely in exceedance” (P0+P1) and “likely or highly likely in exceedance” (P4+P5). The upper and lower values shown for the deposition represent the changes predicted by UKIAM and UKIAM-Scaled. The UK NH<sub>3</sub> abatement scenario leads to a greater reduction in deposition, by roughly a factor of 2, than the other two selective scenarios. From the selective scenarios, only the UK NH<sub>3</sub> abatement scenario achieves the 2030 target of 17% reduction in deposited N outlined in UK’s Clean Air Strategy (DEFRA, 2019).

The greater reduction in deposition by a factor of 2 from the abatement of NH<sub>3</sub> as compared to the other two selective abatement scenarios is unsurprising given that NH<sub>3</sub> emissions contribute twice the amount to the total N deposition budget (Table 3). However, the increase in the %

area of habitats in categories P0+P1 for the UK NH<sub>3</sub> abatement scenario is a factor four greater than that for the NO<sub>x</sub> abatement scenario, and three times greater than the imported abated scenario. A similar comparison is seen for the reduction in the % area of habitats in categories P4+P5. This increased factor is due to the different spatial distributions of the deposition from these sources, with a greater proportion of the deposited N from NH<sub>3</sub> affecting sensitive habitats. Therefore, the abatement of UK NH<sub>3</sub> emissions is likely to be a much more effective strategy for reducing CL exceedances than the abatement of UK NO<sub>x</sub> emissions, and is likely to provide much greater benefits for sensitive habitats than those gained from emission reductions outside the UK.

The UK NO<sub>x</sub> abatement scenario leads to the lowest improvement in habitat area in exceedance, lower than the reduced imported contribution scenario. This is perhaps unsurprising since much of the deposited NO<sub>x</sub> is from long-range transport and the total reduction in European NO<sub>x</sub> emissions for the imported scenario is considerably greater than that for the UK NO<sub>x</sub> scenario; for the imported scenario all European NO<sub>x</sub> emissions outside the UK is reduced by 40%, including international shipping. However, the reduction in deposited N for the two scenarios is of a similar order. This suggests that N deposited due to imported emissions has a proportionally greater impact on CL exceedances than that deposited due to UK NO<sub>x</sub> emissions as it is more likely to deposit in areas containing sensitive habitats.

For the blanket reduction scenarios, the rate of decrease in deposited N with each 10% increment reduction of reactive N emitted is nearly

Table 7

Reduction in UK emissions, deposited N and % area of habitat in the upper and lower exceedance categories for each scenario relative to the 2016 base case. The ranges provided for deposition are derived from the values given by UKIAM and UKIAM-Scaled.

Abatement scenario	Δ UK NO <sub>x</sub> emissions (kt)	Δ UK NH <sub>3</sub> emissions (kt)	Δ N dep (kt-N)	% change P0+P1	% change P4+P5
40% Import	0.0	0.0	23.9–32.9 (11–15%)	4.8	–5.1
40% UK NO <sub>x</sub>	334.9	0.0	20.2–26.3 (9–12%)	3.3	–3.1
40% UK NH <sub>3</sub>	0.0	112.3	38.2–52.9 (17–24%)	12.7	–14.1
20% NO <sub>x</sub> & NH <sub>3</sub>	167.4	56.1	41.2–56.0 (19–26%)	10.6	–10.6
30% NO <sub>x</sub> & NH <sub>3</sub>	251.2	84.2	61.9–84.1 (28–38%)	16.5	–17.1
40% NO <sub>x</sub> & NH <sub>3</sub>	334.9	112.3	82.5–112.1 (38–51%)	23.5	–25.1
50% NO <sub>x</sub> & NH <sub>3</sub>	418.6	140.3	103.1–140.1 (47–64%)	31.8	–31.3

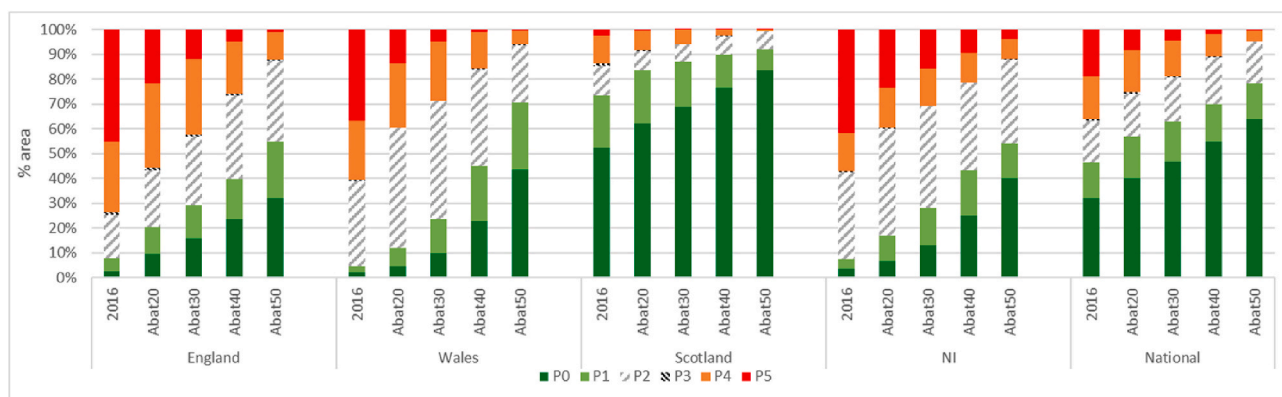


Fig. 8. % Area of all habitats in each exceedance category for each blanket scenario by region.

linear; the small non-linearity is due to cross-pollutant effects ( $\text{SO}_2$  emissions are kept constant for all scenarios). The % change in habitat area in the P0+P1 categories also decreases fairly linearly at the national level, however this is not the case by region, as seen in Fig. 8. In Wales, there is an increasing rate of improvement as abatement is increased, suggesting that in Wales high abatement policies are required in order to see the most significant improvements.

It is also clear from Fig. 8 that there remains a significant variation in habitat exceedance between each region, even for the 50% abated scenario. In this case, the national % area of habitats in the P0+P1 categories is up to 78%. Such an improvement would contribute significantly to the achievement of the target set by the UK government in the 25 Year Environment Plan (DEFRA, 2018) of restoring 75% of protected sites to a favourable condition. However, given that each devolved administration in the UK sets their own environmental targets, in reality this target applies only to England, for which the % area is significantly lower at 55%. This is still a very large improvement on the base year value of 8%, and there is a large proportion of habitat area near the exceedance limit with 32% of habitat area assigned to the P2 or P3 score.

It should also be noted that there remains a high degree of variation in exceedance between habitats for all considered scenarios (see Fig. S10 in the supplementary material). The proportion of woodland habitats unlikely to be in exceedance remains low across the scenarios, with the % area of unmanaged woodland given the P0+P1 scores as low as 8% for the 50% abated scenario. However, there is again a large proportion of habitat area near the exceedance limit, reflected by the prominence of the P2 score at 50%.

#### 4. Discussion

The analysis of the 2016 base year given here highlights the urgent need for action within the UK to protect nitrogen-sensitive habitats from eutrophication. Outside Scotland, the vast majority of woodland habitats are highly likely to be in exceedance of their eutrophication CLs, with high levels of exceedance predicted by both UKIAM and CBED models. Large areas of short habitats such as grasslands and bogs are also likely to be in exceedance, with saltmarsh being the only habitat which is currently almost entirely below its CL.

The 50% abatement scenario gives an indication of what could be achieved were European nations to meet the highly ambitious target proposed by the UNEP's Colombo Declaration (United Nations, 2019) of a 50% reduction in all N waste by 2030. It is clear from the rather modest gains that are achieved by the lower abatement scenarios that reductions of the order targeted by the Colombo directive are required if these N-sensitive habitats in the UK are to be protected. The significantly greater reductions in exceedances achieved through the abatement of  $\text{NH}_3$  as compared to  $\text{NO}_x$  also highlights that significant abatement of  $\text{NH}_3$  emissions is key to achieve the UK government's target of restoring 75% of

protected sites to a favourable condition (25 Year Environment Plan (DEFRA, 2018)). It is unlikely that the UKNECD targets of a 16% reduction in  $\text{NH}_3$  and a 73% reduction in  $\text{NO}_x$  emissions will be sufficient.

It is important to note that here we consider all sensitive habitat areas, not only those assigned for protection, for example SSSI (Sites of Special Scientific Interest) sites. While the modelling suggests that a considerable decrease in national emissions is required, this alone will not be enough to protect these sites and therefore additional local measures will also likely be necessary, as suggested in the Nitrogen Futures report (Dragosits et al., 2020).

The outlook in Scotland is more positive than the other regions of the UK, despite considerable uncertainty in the deposition estimates there. This uncertainty is reflected in the large discrepancy between the two model predictions in Scotland. The causes of the disagreement are not yet fully understood, however both models result in lower exceedances in Scotland than the remainder of the UK (Table 5). The exceedance score shows that most of the habitat area in Scotland lies within the lower P0 and P1 categories, providing confidence in this regional outlook despite the uncertainty in deposition estimates.

For each scenario, the P2 category is much more prominent than the P3 category. This does not mean that the ranges in the CL estimates are necessarily greater than the ranges seen in deposition estimates. However, the areas where the absolute difference between the deposition estimates is large correspond to the areas where deposition is high, and therefore both deposition estimates tend to be in exceedance of the minimum CL value, leading to a P4 or P5 category. In areas where the deposition estimates are of a similar order to the CL range, the absolute difference between the two deposition estimates tend to be smaller than the CL range, leading to a P2 category. This remains true for the abatement scenarios, for which the % area in the P3 category remain low despite areas of very high deposition being much reduced. Marginal evaluations of CL exceedance are therefore in the most part due to the range in CL values rather than the range in deposition estimates.

Finally, it should be noted that restoring a habitat area to N deposition values below their CLs does not guarantee protection or restoration of the habitat to a past state. First, there is evidence that there is no threshold of N deposition below which no effects are seen (e.g. Armitage et al., 2014; Payne et al., 2013). Secondly, a particular habitat could disappear from the area before N deposition levels are reduced below the CL. Despite this, the CL values do provide useful indicators of habitat resilience to N deposition.

#### 5. Conclusions

The UKIAM framework has been extended to provide a second set of nitrogen deposition estimates based on those given by the CBED model. This second set of deposition values is used to provide higher and lower CL exceedance estimates at a national and regional level for nitrogen sensitive habitats. The difference between the two models is used as an

indicator for the uncertainty, which is in turn projected to future scenarios by scaling the UKIAM projections by the 2016 UKIAM/CBED ratio. It is unlikely that both models share the same biases and as a result a more robust analysis is provided. Further, an exceedance score has been developed, using the two deposition values and the minimum and maximum CL estimates, removing the dependency of the analysis on a single, imprecise limit value and avoiding step changes in exceedance which can occur when a single limit value is used.

Applying the method to the base year, 2016, it is shown that the vast majority of woodland habitats in the UK are likely to be in considerable exceedance of their CLs. Outside of Scotland, where deposition is lower in the most part, large areas of short habitats are also likely in exceedance.

Three hypothetical scenarios are considered in order to explore the varying impacts of N emissions from different sources on CL exceedances in the UK. It is shown that UK NH<sub>3</sub> emissions contribute a disproportionate amount to exceedances as compared to UK NO<sub>x</sub> and imported emissions, with a 40% reduction in NH<sub>3</sub> emissions leading to a factor four greater decrease in habitat exceedance, relative to the 2016 base year, than the other two scenarios. Four further scenarios are used to explore what level of protection is feasible given increasing abatement levels. It is shown that a reduction in the order of 50% of all NO<sub>x</sub> and NH<sub>3</sub> sources, a level consistent with the Colombo declaration target, is likely required to reach the UK government's target of restoring 75% of protected sites to a favourable condition (DEFRA, 2018).

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Acknowledgement

This work reflects the personal views of the authors. Although the modelling work has been supported by the UK Department of Environment, Food and Rural Affairs, the findings and recommendations discussed here are those of the authors and do not necessarily represent the views of Defra.

## Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.envsoft.2022.105355>.

## References

- Aleksankina, K., Heal, M.R., Dore, A.J., Van Oijen, M., Reis, S., 2018. Global sensitivity and uncertainty analysis of an atmospheric chemistry transport model: the FRAME model (version 9.15.0) as a case study. *Geosci. Model Dev. (GMD)* 11, 1653–1664. <https://doi.org/10.5194/gmd-11-1653-2018>.
- ApSimon, H., Warren, R.F., Wilson, J.J.N., 1994. The abatement strategies assessment model – ASAM: applications to reductions of sulphur dioxide emissions across Europe. *Atmos. Environ.* 28 (4), 649–663.
- ApSimon, H., Oxley, T., Woodward, H., July, 2019. The Contribution of Shipping Emissions to Pollutant Concentrations and Nitrogen Deposition across the UK. DEFRA Contract ECM\_53210: Support for National Air Pollution Control Strategies.
- ApSimon, H., Oxley, T., Woodward, H., Mehlig, D., Dore, A., Holland, M., 2021. The UKIAM model for source apportionment and air pollution policy applications to PM<sub>2.5</sub>. *Environ. Int.* 153, 106515. <https://doi.org/10.1016/j.envint.2021.106515>.
- 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. *Biol. Conserv.* 171, 224–231.
- Beswick, K.M., Choularton, T.W., Inglis, D.W.F., Dore, A., Fowler, D., 2003. Influences on long-term trends in ion concentration and deposition at Holme Moss. *Atmos. Environ.* 37, 1927–1940.
- Cape, J.N., Cornell, S.E., Jickells, T.D., Nemitz, E., 2011. Organic nitrogen in the atmosphere — where does it come from? A review of sources and methods. *Atmos. Res.* 102, 30–48.

- Choularton, T.W., Gay, M.J., Jones, A., 1988. The influence of altitude on wet deposition. Comparison between field measurements at Great Dun Fell and the predictions of a seeder-feeder model. *Atmos. Environ.* 22 (7), 1363–1371.
- Crossley, A., Wilson, D.B., Milne, R., 1992. Pollution in the upland environment. *Environ. Pollut.* 75, 81–87.
- DEFRA, 2018. A Green Future: Our 25 Year Plan to Improve the Environment. <https://www.gov.uk/government/publications/25-year-environment-plan>.
- DEFRA, 2019. Clean Air Strategy. <https://www.gov.uk/government/publications/clean-air-strategy-2019>.
- Dore, A.J., Choularton, T.W., 1992. Orographic rainfall enhancement in the mountains of the lake district and snowdonia. *Atmos. Environ.* 26A (3), 357–371.
- Dore, A.J., Choularton, T.W., Inglis, D.W.F., 2001. Monitoring Studies of Precipitation and Cap Cloud Chemistry at Holme Moss in the Southern Pennines. *Water Air Soil Pollut. Focus* 1, 381–390. <https://doi.org/10.1023/A:1013116919017>.
- Dore, A.J., Carslaw, D.C., Braban, C., Cain, M., Chemel, C., Conolly, C., Derwent, R.G., Griffiths, S.J., Hall, J., Hayman, G., Lawrence, S., Metcalfe, S.E., Redington, A., Simpson, D., Sutton, M.A., Sutton, P., Tang, Y.S., Vieno, M., Werner, M., Whyatt, J. D., 2015. Evaluation of the performance of different atmospheric chemical transport models and inter-comparison of nitrogen and sulphur deposition estimates for the UK. *Atmos. Environ.* 119, 131–143. <https://doi.org/10.1016/j.atmosenv.2015.08.008>.
- Dragosits, U., Carnell, E.J., Tomlinson, S.J., Misselbrook, T.H., Rowe, E.C., Mitchell, Z., Thomas, I.N., Dore, A.J., Levy, P., Zwagerman, T., Jones, L., Dore, C., Hampshire, K., Raoult, J., German, R., Pridmore, A., Williamson, T., Marner, B., Hodgins, L., Laxen, D., Wilkins, K., Stevens, C., Zappala, S., Field, C., Caporn, S.J.M., 2020. Nitrogen Futures. JNCC Report No. 665. JNCC, Peterborough. ISSN 0963-8091.
- Fagerli, H., Basart, S., 2021. Evaluation of PM and its chemical components modelled by regional models in CAMS [conference presentation]. In: International Technical Meeting on Air Pollution Modelling 2021. Barcelona, Spain. [https://atmosphere.copernicus.eu/sites/default/files/custom-uploads/CAMS-5thGA/day2/Fagerli%20H\\_Met%20Norway\\_Chemistry%20aerosol%20modelling.pdf](https://atmosphere.copernicus.eu/sites/default/files/custom-uploads/CAMS-5thGA/day2/Fagerli%20H_Met%20Norway_Chemistry%20aerosol%20modelling.pdf).
- Fowler, D., Cape, J.N., 1984. On the episodic nature of wet deposited sulphate and acidity. *Atmos. Environ.* 18 (9), 1859–1866.
- Fowler, D., Cape, J.N., Leith, I.D., Choularton, T.W., Gay, M.J., Jones, A., 1988. The influence of altitude on rainfall composition. *Atmos. Environ.* 22, 1355–1362.
- González Benítez, J.M., Cape, J.N., Heal, M.R., van Dijk, N., Vidal Díez, A., 2009. Atmospheric nitrogen deposition in south-east Scotland: quantification of the organic nitrogen fraction in wet, dry and bulk deposition. *Atmos. Environ.* 43 (26), 4087–4094.
- Hall, J., Curtis, C., Dore, T., Smith, R., 2015. Methods for the Calculation of Critical Loads and Their Exceedances in the UK. Centre for Ecology and Hydrology. Report to Defra under contract AQ0826.
- Jones, L., Hall, J., Strachan, I., Field, C., Rowe, E., Stevens, C.J., Caporn, S.J.M., Mitchell, R., Britton, A., Smith, R., Bealey, B., Masante, D., Hewison, R., Hicks, K., Whitfield, C., Mountford, E., 2016. A decision framework to attribute atmospheric nitrogen deposition as a threat to or cause of unfavourable habitat condition on protected sites. JNCC. JNCC, Peterborough.
- Marais, E.A., Pandey, A.K., Van Damme, M., Clarisse, L., Coheur, P.F., Shephard, M.W., Cady-Pereira, K.E., Misselbrook, T., Zhu, L., Luo, G., Yu, F., 2021. UK ammonia emissions estimated with satellite observations and GEOS-Chem. *J. Geophys. Res. Atmos.* 126, e2021JD035237.
- Nilsson, J., Grennfelt, P., 1988. 15. Critical Loads for Sulphur and Nitrogen. Miljørapport, Denmark.
- Oxley, T., Valiantis, M., Elshkaki, A., ApSimon, H., 2009. Background, road and urban transport modelling of air quality limit values (the BRUTAL model). *Environ. Model. Software* 24, 1036–1050.
- Oxley, T., Dore, A.J., ApSimon, H., Hall, J., Kryza, M., 2013. Modelling future impacts of air pollution using the multi-scale UK Integrated Assessment Model (UKIAM). *Environ. Int.* 61, 17–35.
- Payne, R.J., Dise, N.B., Stevens, C.J., Gowing, D.J., Partners, B.E.G.I.N., 2013. Impact of nitrogen deposition at the species level. *Proc. Natl. Acad. Sci. U.S.A.* 110 (3), 984–987.
- RoTAP, 2012. Review of Transboundary Air Pollution: Acidification, Eutrophication, Ground Level Ozone and Heavy Metals in the UK. Contract Report to the Department for Environment, Food and Rural Affairs. Centre for Ecology & Hydrology.
- Rowe, E.C., Mitchell, Z., Tomlinson, S., Levy, P., Banin, L.F., Sawicka, K., Martín Hernandez, C., Dore, A., 2020. Trends Report 2020: Trends in Critical Load and Critical Level Exceedances in the UK. Report to Defra under Contract AQ0843, CEH Project NEC05708. [https://uk-air.defra.gov.uk/library/reports/report\\_id=1001](https://uk-air.defra.gov.uk/library/reports/report_id=1001).
- Simpson, D., Benedictow, A., Berge, H., Bergstrom, R., Emberson, L.D., Fagerli, H., Flechard, C.R., Hayman, G.D., Gauss, M., Jonson, J.E., Jenkin, M.E., Nyiri, A., Richter, C., Semeena, V.S., Tsyro, S., Tuovinen, J.P., Valdebenito, A., Wind, P., 2012. The EMEP MSC-W chemical transport model – technical description. *Atmos. Chem. Phys.* 12, 7825–7865.
- Singles, R., Sutton, M.A., Weston, K.J., 1998. A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain. *Atmos. Environ.* 32 (3), 393–399. [https://doi.org/10.1016/S1352-2310\(97\)83467-X](https://doi.org/10.1016/S1352-2310(97)83467-X).
- Smith, R.I., Fowler, D., Sutton, M.A., Flechard, C., Coyle, M., 2000. Regional estimation of pollutant gas deposition in the UK: model description, sensitivity analyses and outputs. *Atmos. Environ.* 34, 3757–3777.
- Smith, R.I., Fowler, D., 2000. Uncertainty in estimation of wet deposition of sulphur. *Water Air Soil Pollut. Focus* 1, 341–354.
- United Nations, 2019. Colombo Declaration on Sustainable Nitrogen Management. United Nations Global Campaign on Sustainable Nitrogen Management.