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The role of indicator choice in quantifying the threat of atmospheric ammonia to the ‘Natura 2000’ network

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ARTICLE INFO

Keywords:

Ammonia
Critical levels
FRAME
Natura 2000
Nitrogen
Habitats

ABSTRACT

New ‘critical levels’ (CLE) for assessing the effects of atmospheric ammonia on sensitive ecosystems have recently been adopted by the United Nations Economic Commission for Europe (UNECE) of 1 and 3 [2–4] $\mu\text{g NH}_3 \text{ m}^{-3}$ of ambient air (including water vapour), for different species sensitivities and their associated habitats. Based on these values, we examined how indicator choice affects estimates of stock-at-risk in the European ‘Natura 2000’ network.

We applied an atmospheric model, FRAME, to estimate surface air concentrations of ammonia at 5 km and 1 km resolution for the UK network of Natura sites, optionally including calibration with the National Ammonia Monitoring Network. As a base indicator, we estimated the overall percentage area of the UK Natura network that exceeded critical level thresholds (‘Area Weighted Indicator’, AWI). We compared this with an alternative approach, estimating the percentage number of Natura sites where the critical level was exceeded (‘Designation Weighted Indicator’, DWI), which we consider more relevant under the terms of the Habitats Directive.

Using the AWI (with 1 km calibrated ammonia), we estimate that 11.2%, 1.3% and 0.2% area of the UK Natura network exceeds the critical level values of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$, respectively. By contrast, using the DWI, the equivalent exceedances are 59.1%, 23.6% and 9.8%. The highest regional exceedance (DWI, critical level 1 $\mu\text{g NH}_3 \text{ m}^{-3}$) was calculated for England (91.9% exceeded), and the lowest for Scotland (24.0% exceeded). High resolution maps show that the larger threat estimated by the DWI approach is explained by (i) an anti-correlation between NH_3 concentration and Natura site area and (ii) the fact that exceedance over part of a Natura site is considered to represent a threat to the integrity of the whole site.

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1. Introduction

High levels of atmospheric ammonia (NH_3) may cause adverse effects on the environment through a range of processes, including eutrophication effects on biodiversity, acidification

of soils and particulate matter effects on human health (Erisman and Sutton, 2008). The magnitude of the ecological effects is generally assessed in relation to either atmospheric N deposition (including the contribution of other forms of reactive nitrogen) or to atmospheric NH_3 concentrations. The

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1462-9011/\$ – see front matter © 2010 Elsevier Ltd. All rights reserved.
doi:10.1016/j.envsci.2010.09.010

environmental effects thresholds for each are referred to as critical loads and critical levels, respectively (Achermann and Bobbink, 2003; Sutton et al., 2009c).

The critical level (CLE) of an air pollutant, such as NH_3 , is defined as the concentration in the atmosphere above which direct adverse effects on receptors, such as plants, ecosystems or materials, may occur according to present knowledge (Posthumus, 1988). Use of the critical level approach has some advantages over the critical loads approach, in that it is much easier to measure NH_3 air concentrations than total nitrogen deposition, and the indicator considers the specific effects of NH_3 rather than all reactive nitrogen compounds. As such, critical levels for NH_3 are in principle well suited as a tool for practical environmental management (Sutton et al., 2009a). By contrast, it should be noted that critical levels for ammonia, represent a less-comprehensive approach than the application of the critical loads approach, which integrates the effects of all forms of reactive nitrogen and can take account of ecosystem dependent deposition rates. As such, the critical loads approach has been widely used in guiding emissions reduction strategies, e.g. in the Gothenburg Protocol of the United Nations Economic Commission for Europe, Convention on Long Range Transboundary Air Pollution (Hettelingh et al., 2008).

Until recently, the NH_3 critical levels for vegetation used by the UNECE, were those agreed in Egham (United Kingdom) in 1992 (van der Eerden et al., 1994), which were based on measurements and observations from the 1980s, mostly from the Netherlands. Several values were set, ranging from short-term (hourly) exposure to annual exposure, with the annual critical level set at $8 \mu\text{g NH}_3 \text{ m}^{-3}$. While this value was derived from the best information available at the time, there were several limitations. Not the least of these was the fact that this critical level was much less precautionary than the equivalent NH_3 concentration that would (with appropriate deposition velocities) lead to exceedance of nitrogen critical loads (Burkhardt et al., 1998; Cape et al., 2009a,b; Sutton et al., 2009a). For example, even in the absence of other nitrogen deposition, typically around $2\text{--}3 \mu\text{g NH}_3 \text{ m}^{-3}$ would be sufficient to exceed critical loads. The result was that this former value of the critical level for NH_3 was largely redundant and little used as a tool for environmental management.

At the UNECE Edinburgh Ammonia Workshop, more recent scientific evidence for setting critical level values for NH_3 was evaluated, and new long-term critical level values set for different vegetation types (Cape et al., 2009a,b), which were adopted for use in the LRTAP Convention (UNECE, 2007; Mills et al., 2008; Sutton et al., 2009b). New values of the critical level were set for two main vegetation/habitat groups, based on observed changes to species composition in the field. Most of the evidence came from studies in the United Kingdom, but there was corroborative evidence from Italy, Portugal and Switzerland (see papers in Sutton et al., 2009c). The critical level values were set for long-term exposure, reflecting a protection period over decades, although, recognizing the uncertainties, it could not be concluded that the levels set would provide protection for longer than 20–30 years. The most sensitive vegetation types were identified as lichens and bryophytes. For these plant groups, and habitats where they are important to ecosystem integrity, the long term critical

level was set at $1 \mu\text{g NH}_3 \text{ m}^{-3}$. Although different species of mosses and lichens have different sensitivities to ammonia, this level protects the most sensitive species within these groups.

There was less evidence available to quantify the concentrations at which long-term effects of NH_3 caused species changes in communities of higher plants. Recognizing the uncertainties, a long-term critical level for higher plants was set at $3 \mu\text{g NH}_3 \text{ m}^{-3}$, together with an uncertainty range of $2\text{--}4 \mu\text{g NH}_3 \text{ m}^{-3}$ (Cape et al., 2009a,b). While the mid-point thus represents the best estimate of the critical level for higher plants, depending on the degree of precaution appropriate to different ecological and regulatory contexts, the lower or the higher value might be applied (UNECE, 2007; Sutton et al., 2009c).

The availability of the new critical level estimates is particularly relevant for assessing ecological condition under the terms of the Habitats Directive (Council Directive 92/43/EEC on the Conservation of natural habitats and of wild fauna and flora), and the associated Birds Directive (Council Directive 79/409/EEC on the conservation of wild birds). These seek to protect Europe's natural resources, especially the most seriously threatened habitats and species across Europe.

Article 3 of the Habitats Directive requires the establishment of a European network of sites that will make a significant contribution to conserving the 189 habitat types and 788 species identified in Annexes I and II of the Directive (as amended). The listed habitat types and species are those meeting the Directive's criteria and thus considered to be most in need of conservation at a European level (<http://www.jncc.gov.uk/page-1464>).

The Birds Directive requires the establishment of Special Protection Areas (SPAs), while the Habitats Directive requires Special Areas of Conservation (SAC) to be designated for non-bird species, and particularly for habitats.

The Birds Directive provides no formal criteria for selecting SPAs, so the JNCC, on behalf of the statutory country conservation agencies and government, published SPA Selection Guidelines for use in the UK. Each SPA has been selected according to the principles laid out in the selection guidelines. In the context of ammonia, if critical levels are exceeded then the vegetation could be damaged such that the habitat of the designated site's species could be compromised.

Together, SPAs and SAC make up the 'Natura 2000' network, to be implemented by all EU Member States, and representing a flagship for biodiversity protection in the European Union.

In this study, we have sought to investigate how the recently established critical levels could be used to develop indicators to assess the ammonia threat to the Natura 2000 network in the UK. Specifically, we aimed to investigate different approaches to quantifying the Natura 2000 stock-at-risk. For this purpose, the new critical level values provide simple thresholds which can be compared with estimated NH_3 concentrations, as derived from a combination of models and measurements (Sutton et al., 2001; Fournier et al., 2005a,b).

For this study we applied an atmospheric transport model, Fine Resolution Atmospheric Multi-pollutant Exchange

(FRAME) (Singles et al., 1998; Fournier et al., 2005a,b; Dore et al., 2007), which has been widely used to generate spatial patterns of measured ammonia concentrations and deposition of nitrogen across the UK (Sutton et al., 2001), as well as temporal trends (Sutton et al., 2006; Tang et al., 2009; Matejko et al., 2009). The FRAME model has in most instances been applied for the whole of the UK at 5 km resolution. Until recently, computational requirements prevented practical application at a higher resolution, while the input NH_3 emissions data are also normally available at 5 km resolution (Dragosits et al., 1998; Hellsten et al., 2008). We have advanced this for the present study by developing a 1 km resolution application of FRAME, run on a Linux cluster. In addition to normal methodological uncertainties, the main constraint is the need to protect confidentiality of input data at this scale (see Section 2). With these restrictions, the comparison of the 5 km and 1 km FRAME applications highlights the importance of model scale for the present assessment.

In addition to wanting to know whether the critical level is exceeded at certain locations, we were interested to investigate how to measure the overall ammonia threat to the Natura 2000 network. In England, there is a Public Service Agreement target for nationally designated sites known as Sites of Special Scientific Interest (many of which are also designated as part of SAC and SPAs). The public service agreement aims to ensure that 95% of the total area of Sites of Special Scientific Interest in England is brought into favourable or recovering condition by 2010 (HM Treasury 2004, Chapter 13, DEFRA).

An option then would be to follow this method of assessment by investigating how much total area of the Natura 2000 reserves are exceeded by the new critical levels of ammonia. In the context of the Habitats Directive, however, there is concern that this Public Service Agreement target is not sufficiently precautionary. Some SAC are very large (e.g. in remote mountain areas), while others are very small. The most extensive localities (e.g. The Wash and North Norfolk Coast, Caithness and Sutherland Peatlands) exceed 100,000 ha, while the smallest are less than one hectare. Such an averaging approach based on 95% of the total area would take little account of the status of such small sites, being weighted by status of the larger remote sites. Depending on the relationship to ammonia exposure, this could affect the overall assessment of threat. Secondly, there is concern that an approach based on total area is inappropriate given the precautionary principles of the Habitats Directive.

SAC designation requires Member States to establish conservation measures which correspond to the ecological requirements of Annex I habitats and Annex II species present on the site (Article 6.1), and to take appropriate steps to avoid deterioration of the natural habitats and habitats of species, as well as significant disturbance of species, for which the site is designated (Article 6.2). An assessment of site condition should aim to show if the critical level is exceeded over any part of an SAC, since a threat to the integrity of part of a site is considered as representing a threat to the integrity of the whole (Frost, 2004). For example, if atmospheric NH_3 exceeds the critical level even for a small percentage of the area of an SAC, this would still be considered as representing a threat to the integrity of the whole SAC.

As a general principle, SAC boundaries have been drawn closely around the qualifying habitat types or the habitats of species for which the sites have been selected, taking into account the need to ensure that the site operates as a functional whole for the conservation of the habitat type(s) or species and to maintain sensible management units. Buffer zones have generally not been included as part of SACs (<http://www.jncc.gov.uk/page-1475>). Under the Habitats Directive, Assessment: “Coherence of a site’s ecological structure and function, across its whole area or the habitats, complex of habitats and/or populations of species for which the site is or will be classified” Habitats Regulations Guidance, 1997 Article 6(3) of the Habitats Directive (92/43/EEC).

For the present study, we therefore applied FRAME to estimate the distribution of NH_3 across the UK at both 5 km and 1 km resolution, also considering the consequences of calibration with atmospheric monitoring data. We then defined and applied two indicators to assess the extent of ammonia exceedance in the UK Natura 2000 network: an ‘Area Weighted Indicator’ (AWI) and a ‘designated weighted indicator’ (DWI). We report the resulting estimates of critical level exceedance for each indicator and consider the implications for assessing habitat stock-at-risk due to effects of ammonia.

2. Methods

2.1. Atmospheric modelling of ammonia

The Fine Resolution Atmospheric Multi-pollutant Exchange (FRAME) model is a statistical Lagrangian trajectory model that can be used to estimate the spatial distribution of reactive nitrogen surface concentrations and deposition to the UK. Here we provide a summary of the main model features. FRAME was initially developed to focus specifically on ammonia. Singles et al. (1998) provide a detailed description and Fournier et al. (2003) describe the parallelisation of the model. Further development of the model to a robust multi-chemical species tool for estimating deposition of sulphur and nitrogen is described in Fournier et al. (2004) and Dore et al. (2007). The model was generally found to be able to represent the spatial distribution of measured values of annual wet deposition of sulphur and nitrogen, as well as average annual concentrations of gases and aerosols. However, obtaining a good correlation with the gases which make a large contribution to nitrogen deposition (nitric acid and ammonia) presented a greater challenge. In the case of ammonia, this was attributed to the highly variable spatial pattern of emissions, which varied on a scale smaller than the 5 km resolution of the model. For this research, the model was adapted to enable a horizontal resolution on any given domain down to 1 km \times 1 km.

FRAME has a finer scale vertical resolution than most other regional scale dispersion models. A column of air with 33 layers of varying thickness from 1 m at the surface, increasing to 100 m in thickness at heights above 100 m is modelled with the diffusion between the layers calculated using a finite volume method. As ammonia is emitted

almost entirely from low level sources, a fine vertical resolution in the model is important in order to accurately represent surface concentrations. Separate horizontal trajectories of the air column are run at 1° resolution for all grid edge points.

Wet deposition is calculated with scavenging coefficients and a constant drizzle approach, using precipitation rates calculated from a climatological map of annual average precipitation of the British Isles combined with a directional orographic rainfall model (Fournier et al., 2005a,b). For the 5 km model, rainfall was averaged from 1 km estimates, to explore the importance of the resolution of emissions and model with all else being equal.

Dry deposition is calculated individually to five land categories (arable, forest, moorland, grassland and urban). For ammonia, dry deposition is calculated at each grid square using a canopy resistance model (Singles et al., 1998). Wind frequency and wind speed roses (from a 10-year period of radiosonde data, 1991–2000; Dore et al., 2006) are used to give the appropriate weighting to directional deposition and concentration for the calculation of total deposition and concentration. The model chemistry includes gas phase and aqueous phase reactions of oxidised sulphur and oxidised nitrogen and conversion of NH_3 to ammonium sulphate and ammonium nitrate aerosol.

A maximum time step can be set prior to each simulation with the model, but FRAME also has an adaptive time step to ensure that for every grid square that a trajectory passes over, the model goes through at least one iteration of the diffusion equation. However, a single iteration may not be sufficient for the proper mixing and deposition to be calculated. It was therefore necessary to set a maximum time step of 20 s for the 1 km model. In contrast, the 5 km model has a maximum time step of 120 s. These settings force sufficient iterations of vertical diffusion to be carried out to ensure proper mixing and deposition over each grid square. Tests showed no significant difference in distribution or total concentration of NH_3 with further reductions in maximum time step for the respective model resolutions.

2.2. Ammonia monitoring data and calibration of model estimates

Surface air concentrations of measured NH_3 were taken from the UK National Ammonia Monitoring Network (Sutton et al., 2001; Tang et al., 2009). This network comprises measurements at around 100 sites, using a combination of active denuder sampling (DELTA: DENuder for Long-Term Atmospheric sampling, Sutton et al., 2001) and passive sampling (ALPHA: Adapted Low-cost, Passive High Absorption samplers, Tang et al., 2001). The verification data used here were for 2003 (Tang et al., 2008). The detection limit of the denuder is $0.02 \mu\text{g NH}_3 \text{ m}^{-3}$. Paired sampling at the cleanest UK site showed an average Root Mean Square Difference of 9% for individual duplicate samples and 11% for annual means. These figures provide an upper limit, values at sites with more typical concentrations will be generally <5% (Sutton et al., 2001).

The National Atmospheric Monitoring Network covers a wide distribution of monitoring sites including measurements

in both agricultural and semi-natural areas, while avoiding the immediate vicinity of agricultural areas. Vieno (2005) has shown that there is a significant difference in the relationship between FRAME and the National Atmospheric Monitoring Network sites for (a) monitoring sites over mixed agricultural surfaces, and (b) monitoring sites over semi-natural and forest vegetation, with the model overestimating concentrations at the latter. For the present study which focuses on estimation of NH_3 concentrations over SAC and SPAs, we therefore focus exclusively on the comparison with National Atmospheric Monitoring Network measurements over semi-natural and forest vegetation, as identified by Vieno (2005).

2.3. Geographical analysis of ammonia indicators for Natura 2000

The Geographic Information System software ArcGIS was used to analyse the extent that the estimated NH_3 concentrations encroach on the UK Natura 2000 sites. ArcGIS was used to perform areal analyses on modelled NH_3 air concentrations. These analyses quantified the total combined area of SAC and SPAs exceeding the critical levels of NH_3 as input to calculate the Area Weighted Indicator (AWI). In addition, the total number of SAC and SPAs with any fraction of their area exceeded by a critical level of NH_3 was determined as input to calculate the Designation Weighted Indicator (DWI).

The frequency of modelled air concentrations of NH_3 over the UK was summed and then grouped into levels. The levels are defined as, less than $1 \mu\text{g NH}_3 \text{ m}^{-3}$, $1\text{--}2 \mu\text{g NH}_3 \text{ m}^{-3}$, $2\text{--}3 \mu\text{g NH}_3 \text{ m}^{-3}$, and above $3 \mu\text{g NH}_3 \text{ m}^{-3}$.

The overall threat to the Natura 2000 network in relation to ammonia concentrations was assessed according to two approaches.

In the first method, we estimate the overall percentage area of the UK Natura network within which each 1, 2 and $3 \mu\text{g NH}_3 \text{ m}^{-3}$ critical level threshold of NH_3 air concentration is exceeded. The overall percentage area (AWI) within the UK, and within each of the four UK countries, was found by summing the tabulated areas and dividing by the total areal coverage of reserve within the appropriate region. For the second method (DWI), the total number of sites with some part of the area exceeded (i.e. any fraction of the area of a site) by the three critical level values in each devolved region was calculated.

We define the two indicators in equation form as follows.

Area Weighted Indicator (AWI) is defined as:

$$\text{AWI}_{\text{CLE}} = \sum_j \text{area exceeded site}(j)_{\text{CLE}} \quad (1)$$

$$\text{where area exceeded site}(j)_{\text{CLE}} = \sum_i A(i) \text{ for all } A(i) \text{ in site}(j)$$

and where $A(i)$ is any area in site j with mean concentration \geq critical level (CLE).

Designation Weighted Indicator (DWI) is defined as:

$$\text{DWI}_{\text{CLE}} = \sum_j \text{site}(j) \text{ for all } j \quad (2)$$

where $\text{site}(j)$ contains any area with mean concentration \geq critical level (CLE).

Where the subscript ‘critical level’ on AWI and DWI in the above equations refers to the indicator for a given critical level, i.e. 1, 2 or 3 $\mu\text{g NH}_3 \text{ m}^{-3}$.

2.3.1. Emissions

Agricultural emission sources (livestock manures, cultures with fertilisers and field burning) were distributed using the AENEID model (Atmospheric Emissions for National Environmental Impacts Determination) (Dragosits et al., 1998; Hellsten et al., 2008), which incorporates agricultural census data, land cover data, agricultural practice information (e.g. fertiliser application rates, stocking densities). Detailed emission source strength estimates were derived for the main livestock emission components (livestock housing, manure storage, land spreading of manures, livestock grazing) and fertiliser application rates to different crops. Fertiliser N application rates, are not provided by a census, but are provided from the British Survey of Fertiliser Practice (BSFP, e.g. BSFP 2009). Overall, emissions from fertiliser application to crops and conserved grassland in the UK only contribute approx. 14% of the total agricultural emissions, compared with, e.g. 57% from cattle.

The census data (as described above) are an essential requirement for producing maps of ammonia emissions. This method has been published in peer reviewed papers and the annually produced spatial emissions (at 5 km scale) are published annually in the UK National Atmospheric Emissions Inventory and are the best available for the UK.

The non-agricultural sources were spatially distributed using a combination of population census data, landcover data, data from the devolved authorities on landfill sites and sewage works and mammal distribution data (Dragosits et al., 2009).

For the purpose of this assessment, emission estimates for the year 2003 were applied, given the availability of matching datasets for 1 km and 5 km resolution NH_3 emissions.

2.4. Emissions uncertainties

Uncertainties in the emissions mapping may relate to the magnitude of emissions and/or the spatial location of emissions. These can also be affected by seasonal variability in environmental conditions and farming practice.

The most important input data to the AENEID model are the annual agricultural census statistics. Census data are collected at the holding (farm) level but aggregated to a coarser spatial resolution, e.g. aggregated to the level of ‘parishes’, which represent variably sized districts for handling information associated with each village. Since a farm may overlap several aggregation zones, NH_3 emissions in each zone have an associated uncertainty, especially in small aggregation zones. The emissions maps therefore change depending on the type of aggregation zones applied.

The AENEID approach distributes agricultural census data as NH_3 sources in the landscape within each aggregation zone, onto landcover types where the emissions are most likely to occur. Therefore, uncertainties in the landcover dataset and the assumptions applied to allocate emissions to landcover types influence the spatial location of emissions. The locations of emissions from land types with intensive farming (e.g. pigs

and poultry) carry more uncertainty since they tend to be point sources but emissions are only known at parish level.

In our usual procedures, the AENEID model is applied at a 1 km resolution, but aggregated into a coarser resolution of $5 \times 5 \text{ km}^2$, to reduce some of the uncertainties in the modelling process. For the present study 1 km resolution emission estimates of AENEID were carried through to the atmospheric transport modelling in FRAME. This has the potential to improve the result, as the variability within each 5 km grid cell may be considerable.

2.5. Handling confidentiality of agricultural datasets

A major practical challenge in modelling ammonia over the UK is that the input agricultural datasets are confidential. These data are collected as part of annual farm census for each of the devolved regions of the UK (England, Scotland, Wales and Northern Ireland). When using these data for years prior to 2004 (as in the present study), the data agreements require that results should not be reported in such a way that the statistics relating to individual farm holdings could be derived (e.g. for individual statistics an aggregation to at least 5 holdings is required).

To ensure that no confidential information is disclosed in our usual modelling of NH_3 emissions at 5 km grid resolution, the following steps are applied:

- (a) redistribution of parish or parish group data for England, Scotland and Wales (or 5 km gridded data for northern Ireland) to a 1 km grid, based on weighted landcover re-allocation of the livestock and crop sources according to the methodology of Dragosits et al. (1998) and Hellsten et al. (2008);
- (b) averaging of the land-cover weighted, redistributed emissions to 5 km resolution (which also averages out uncertainties);
- (c) reporting of NH_3 emissions at 5 km resolution only for two aggregate classes of (i) total livestock NH_3 emissions and (ii) total fertilizer-related crop and grass NH_3 emissions.

Thus with this approach, the landcover-weighted redistribution of emissions and the combination of the data to aggregated NH_3 emissions classes prevent the identification of original statistics for individual holdings.

The modelling of NH_3 concentrations at 1 km resolution is more challenging to ensure that no individual statistics at holding level are disclosed. For the purpose of the present study, the following approach was therefore adopted:

- (a) spatial redistribution of parish/parish group etc livestock and crop statistics, weighted by landcover to provide 1 km resolution estimates (as above);
- (b) combination of all NH_3 emissions (including non-agricultural sources) into a single emissions file at 1 km, which is not disseminated, but is used as confidential input to the 1 km FRAME model;
- (c) further spatial redistribution of the information in the FRAME model, as NH_3 emissions are dispersed to provide estimated NH_3 concentrations (and potentially deposition estimates, not reported here);

- (d) reporting of the resulting NH_3 concentrations only as colour banded maps or as a regression plots for comparison with the National Ammonia Monitoring Network;
- (e) reporting of SAC and SPA related statistics from the analysis grouped for the main devolved regions (England, Scotland, Wales, Northern Ireland).

By this approach the possibility to disclose confidential farm holdings statistics is avoided.

3. Results

3.1. National area exceedance of ammonia critical levels

3.1.1. Spatial pattern of ammonia across the UK

The overall modelled field of NH_3 concentration at 1 km resolution is shown in Fig. 1. This map shows the classifications of NH_3 concentration, corresponding to exceedance of the critical levels described in the introduction. Also shown

Surface Concentration of NH_3 exceedance (μg^{-3})

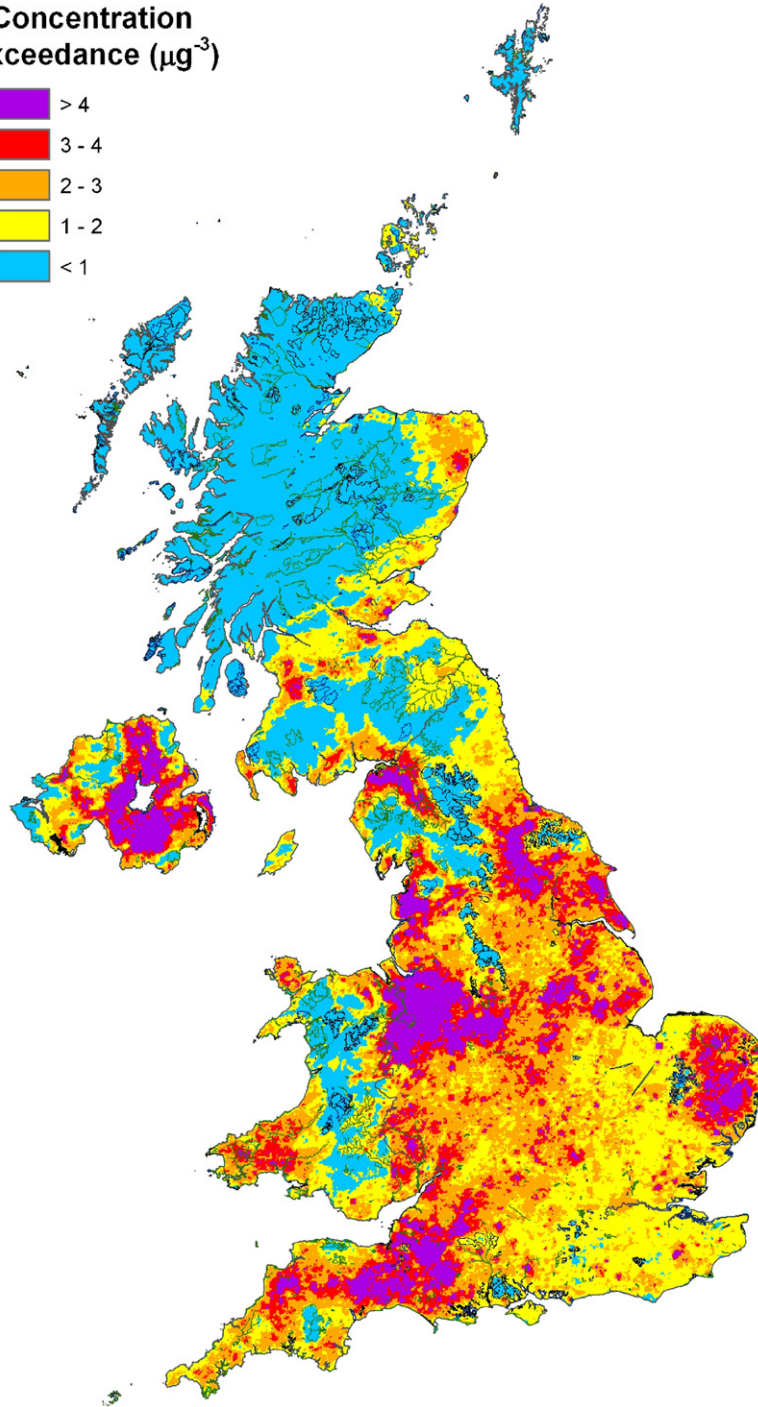


Fig. 1 – National map of ammonia concentrations modelled by FRAME at 1 km resolution (uncalibrated) with SAC and SPA boundaries indicated by green and dark blue borders, respectively (some SAC are also SPAs, indicated by black borders). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

Table 1 – Summary statistics for the percentage area of the UK where critical levels of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ are exceeded. Data are shown for England, Wales, Scotland, Northern Ireland and the UK as a whole (uncalibrated 5 km and 1 km models).

FRAME application	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
5 km model	1 $\mu\text{g m}^{-3}$	97%	82%	29%	94%	73%
5 km model	2 $\mu\text{g m}^{-3}$	75%	40%	10%	73%	51%
5 km model	3 $\mu\text{g m}^{-3}$	37%	14%	2%	53%	24%
1 km model	1 $\mu\text{g m}^{-3}$	93%	68%	26%	85%	69%
1 km model	2 $\mu\text{g m}^{-3}$	61%	38%	9%	65%	42%
1 km model	3 $\mu\text{g m}^{-3}$	27%	14%	2%	43%	19%

are the Natura 2000 network Special Protection Areas (SPAs) and Special Areas of Conservation (SAC).

The most obvious regional pattern is for higher NH_3 concentrations in the South East with lower concentrations in the North West. Upland areas of the UK are characterised by low concentrations of NH_3 , while high concentrations occur in the Welsh border area, East Anglia and Northern Ireland.

Ammonia levels in the devolved regions of the UK where critical levels of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ are exceeded are given in Table 1. The values support the observations from the map with England having the highest regional exceedance for the 1 $\mu\text{g NH}_3 \text{ m}^{-3}$ critical level of 93% and Scotland the lowest of 26%, based on the 1 km model.

3.2. Area-weighted exceedance of the critical level for Natura 2000 UK sites

3.2.1. Natura 2000 site location and NH_3 concentration

The percentage of total areas of SAC and SPAs exceeded by each critical level represents the Area Weighted Indicator (AWI). The values in Table 2 show that for SAC, the percentage of area exceeded by critical levels of ammonia is less than for broad country-wide areas, e.g. the devolved regions and the UK (Table 1). A similar observation can be made for SPAs (see Table 6). The AWI is highest for Natura reserves in England and Northern Ireland. Both Wales and Scotland score significantly lower AWI values with Scottish SAC clearly showing the least percentage of area exceeded, see Table 2.

3.2.2. Model resolution (5 km versus 1 km FRAME)

The 5 km model estimates almost twice the total area of SAC and SPAs exceeded by 1 $\mu\text{g NH}_3 \text{ m}^{-3}$ than the 1 km model for the UK and almost 3 times the area exceeded by

2 $\mu\text{g NH}_3 \text{ m}^{-3}$. Within individual countries (England, Scotland, Wales and Northern Ireland), the differences can be even higher. SPAs in Wales are estimated by the 1 km model to have 8.5% of their area exceeded by 1 $\mu\text{g NH}_3 \text{ m}^{-3}$ whereas the 5 km model predicts 27.6% exceeded, more than a factor of 3.

Fig. 2 shows that there are major differences for the Dartmoor SAC and New Forest SAC. The 1 km model simulates that these SAC are almost entirely free of any critical level exceedance whereas the 5 km model predicts most of their area exceeded by 1 $\mu\text{g NH}_3 \text{ m}^{-3}$ and a small area exceeded by 2 $\mu\text{g NH}_3 \text{ m}^{-3}$.

3.3. Designation Weighted Indicator

The values in Table 3 show that Scotland has the least number of SAC and SPAs exceeded by each critical level for the calibrated concentrations of NH_3 when the Designation Weighted Indicator (DWI) is applied. England has the greatest number exceeding the critical level of 1 $\mu\text{g NH}_3 \text{ m}^{-3}$ for SAC and SPAs but NI has the largest number exceeding critical levels of 2 $\mu\text{g NH}_3 \text{ m}^{-3}$ for SAC and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ for SPAs. The DWI statistics show that more reserves are exceeded by critical levels of NH_3 than would be apparent from the AWI approach.

3.4. Comparison of the model with measured NH_3 concentrations

For the subset of National Atmospheric Monitoring Network measurement sites (see Fig. 3) that include only semi-natural vegetation and forest sites, regression analyses show that 1 km FRAME gives an improvement in slope, intercept and correlation coefficient compared to 5 km FRAME, see Fig. 4.

Table 2 – Summary statistics for the Area Weighted Indicator (AWI): the percentage area of the UK SAC network where NH_3 air concentrations of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ are exceeded in England, Wales, Scotland, Northern Ireland and the UK as a whole (uncalibrated 5 km and 1 km models).

FRAME application	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
5 km model	1 $\mu\text{g m}^{-3}$	79.0%	39.5%	3.3%	77.0%	39.6%
5 km model	2 $\mu\text{g m}^{-3}$	19.8%	9.0%	0.6%	23.3%	9.9%
5 km model	3 $\mu\text{g m}^{-3}$	4.0%	2.1%	0.0%	10.0%	2.2%
1 km model	1 $\mu\text{g m}^{-3}$	40.9%	21.3%	2.4%	31.7%	20.8%
1 km model	2 $\mu\text{g m}^{-3}$	6.3%	6.1%	0.4%	11.1%	3.7%
1 km model	3 $\mu\text{g m}^{-3}$	1.6%	1.6%	0.0%	2.9%	0.9%

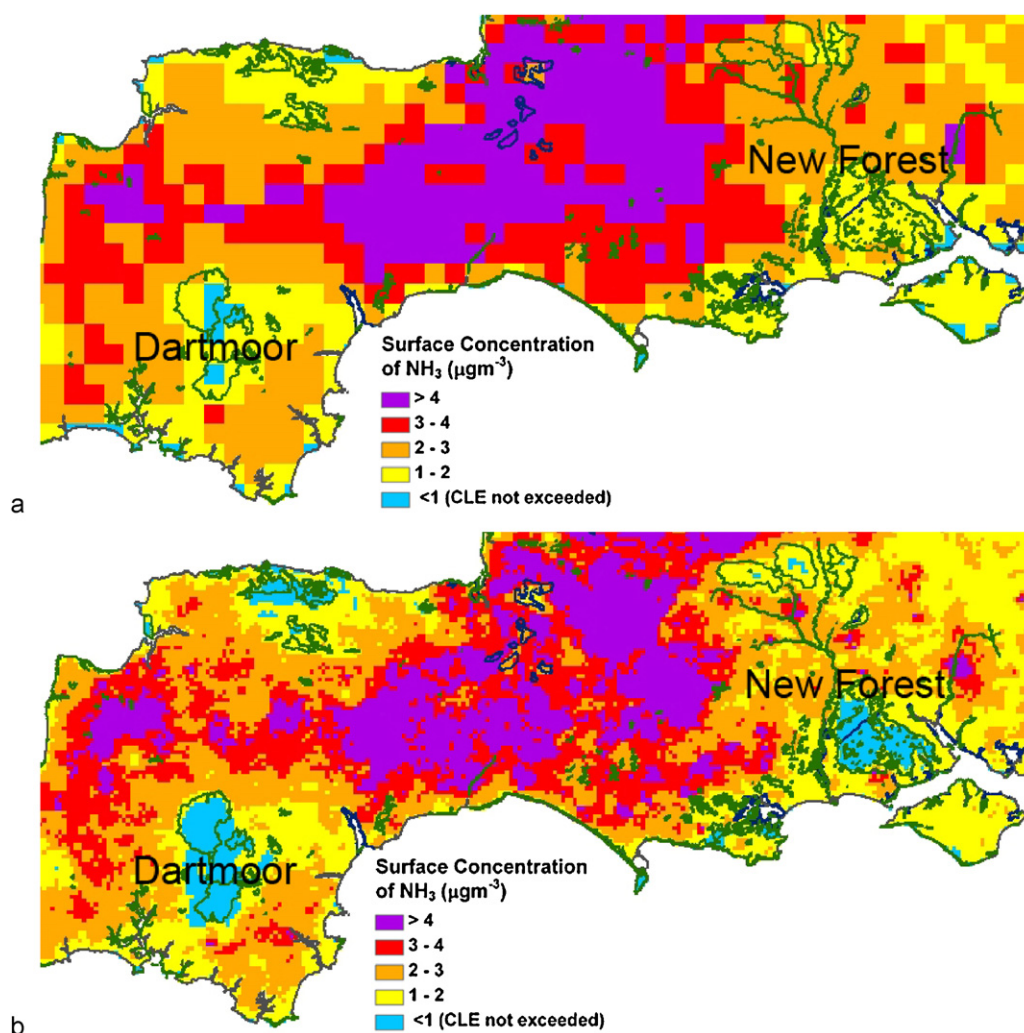


Fig. 2 – The difference between exceedance of NH_3 critical levels for two applications of the FRAME model: (a) from the 5 km resolution FRAME application and (b) from the 1 km FRAME application. The model outputs shown here are uncalibrated. Green boundaries show boundaries of Special Areas of Conservation (SAC). The model comparison reveals major differences for the Dartmoor and New Forest Special SAC (largely shown in blue for the 1 km model). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

Table 3 – Summary statistics for the Designation Weighted Indicator (DWI): the percentage number of UK SAC and SPAs where there is some exceedance of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ in England, Wales, Scotland, Northern Ireland and the UK as a whole (uncalibrated 1 km statistics only).

Natura 2000 component	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
SAC	1 $\mu\text{g m}^{-3}$	97.1%	91.5%	30.0%	84.9%	68.9%
SAC	2 $\mu\text{g m}^{-3}$	67.8%	59.8%	11.4%	64.2%	44.1%
SAC	3 $\mu\text{g m}^{-3}$	29.7%	28.0%	4.6%	32.1%	19.1%
SPAs	1 $\mu\text{g m}^{-3}$	98.8%	94.7%	41.3%	83.3%	65.7%
SPAs	2 $\mu\text{g m}^{-3}$	70.7%	42.1%	15.2%	58.3%	36.7%
SPAs	3 $\mu\text{g m}^{-3}$	40.2%	15.8%	8.0%	41.7%	19.8%
All	1 $\mu\text{g m}^{-3}$	97.5%	92.1%	34.1%	84.6%	68.0%
All	2 $\mu\text{g m}^{-3}$	68.5%	56.4%	12.8%	63.1%	41.9%
All	3 $\mu\text{g m}^{-3}$	32.4%	25.7%	5.9%	33.8%	19.3%

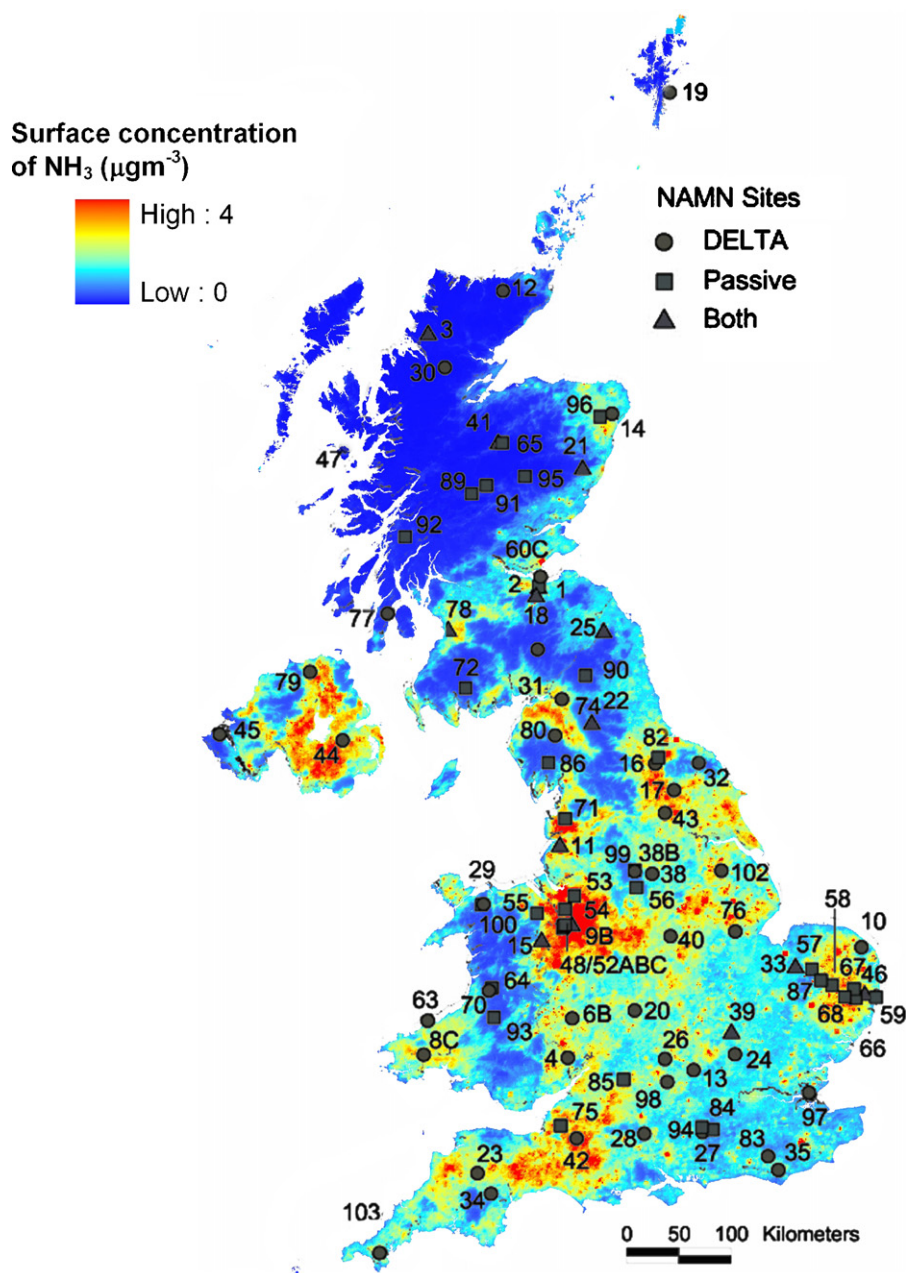


Fig. 3 – Map of National Ammonia Monitoring Network sites (with numbered points) overlain on the UK ammonia concentrations map at 1 km resolution, calibrated to measured concentrations at semi-natural and woodland sites.

However, the regression in Fig. 5 suggests that there is only a modest performance benefit of the higher resolution FRAME (1 km) when comparing with National Atmospheric Monitoring Network sites of all land-cover types.

To illustrate the 1 km FRAME model performance more clearly for the semi-natural and forest National Atmospheric Monitoring Network sites, Fig. 6 plots the results on a logarithmic scale, marking each monitoring point. An analysis of the regression shows that the intercept ($-0.03 \mu\text{g NH}_3 \text{ m}^{-3}$) is not statistically significant and allows for easy calibration of the model, by simply correcting for the slope in Fig. 6.

3.5. Comparison of the calibrated and uncalibrated models

In Table 4 we present the 1 km model AWI statistics, calibrated for NH_3 concentrations by using the regression with the measurements from seminatural/forest sites. The calibrated NH_3 concentrations exceeding $1 \mu\text{g NH}_3 \text{ m}^{-3}$ cover about 11% of the total area of SAC and SPAs exceeded which is half the area of the uncalibrated concentrations. For the higher critical levels, the difference in the AWI statistics between the calibrated and uncalibrated concentrations significantly increases. A similar observation can be made for the DWI

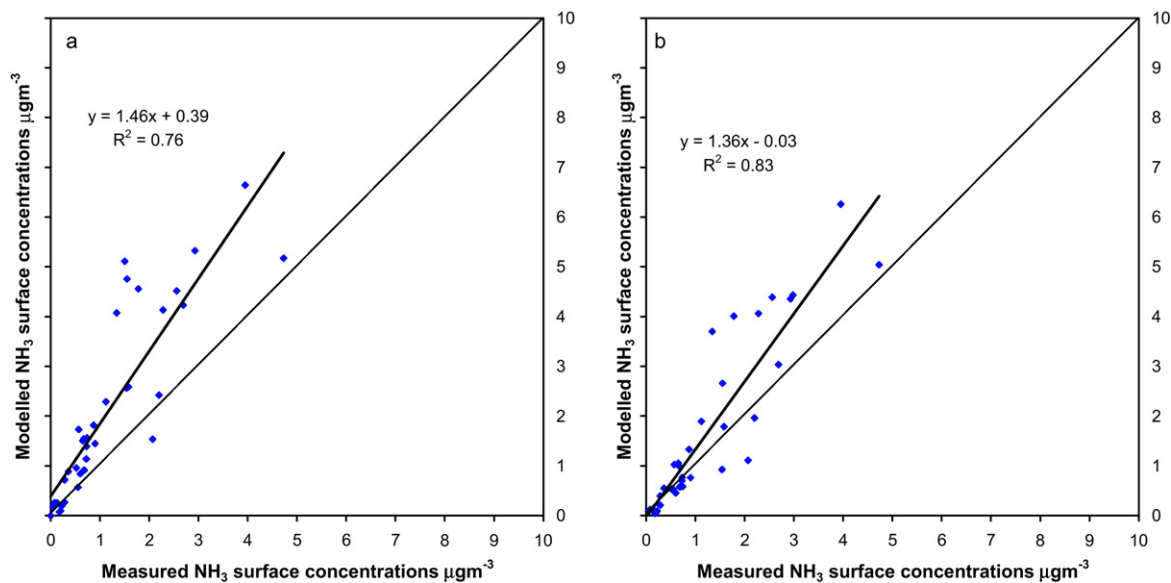


Fig. 4 – Comparison of FRAME modelled surface concentrations of ammonia with measurements from the UK National Ammonia Monitoring Network (Sutton et al., 2001) for sites classed as semi-natural vegetation or forest for the period 2002–2004. (a) 5 km FRAME and (b) 1 km FRAME.

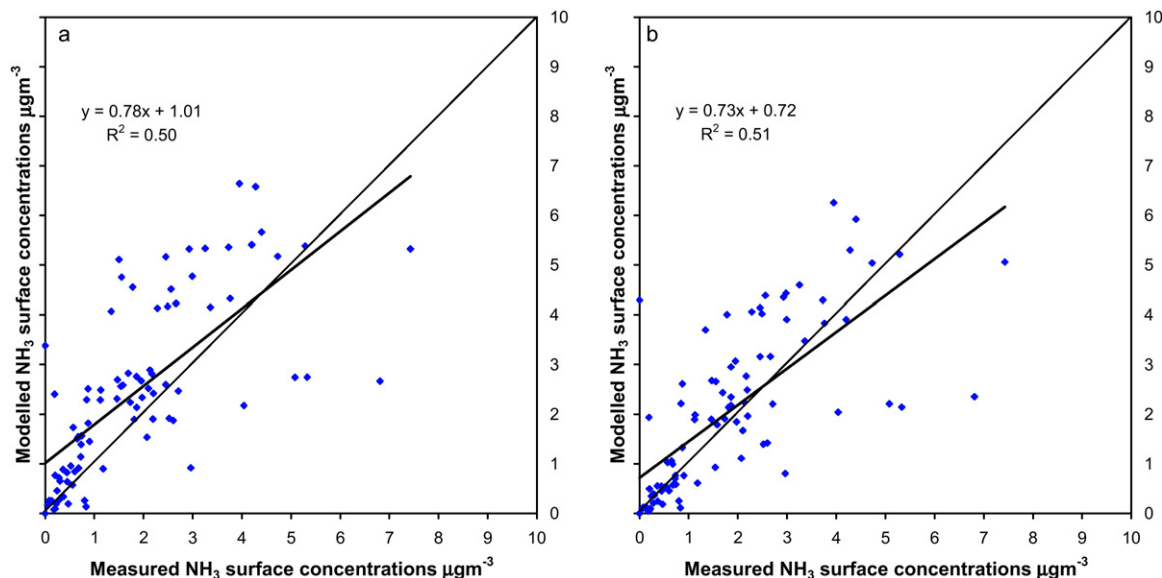


Fig. 5 – Comparison of FRAME modelled surface concentrations of ammonia with measurements from the UK National Ammonia Monitoring Network (Sutton et al., 2001) for all sites for the period 2002–2004. (a) 5 km FRAME and (b) 1 km FRAME.

Table 4 – Summary statistics for the Area Weighted Indicator (AWI): the percentage area of UK SAC and SPAs separately where NH₃ air concentrations of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ are exceeded in England, Wales, Scotland, Northern Ireland and the UK as a whole (calibrated 1 km statistics only).

Natura 2000 component	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
SAC	1 $\mu\text{g m}^{-3}$	20.6%	13.2%	1.2%	22.3%	10.9%
SAC	2 $\mu\text{g m}^{-3}$	2.3%	2.3%	0.1%	4.7%	1.4%
SAC	3 $\mu\text{g m}^{-3}$	0.4%	0.4%	0.0%	0.3%	0.2%
SPAs	1 $\mu\text{g m}^{-3}$	23.9%	4.6%	1.4%	59.3%	11.6%
SPAs	2 $\mu\text{g m}^{-3}$	2.4%	0.3%	0.0%	16.5%	1.2%
SPAs	3 $\mu\text{g m}^{-3}$	0.5%	0.3%	0.0%	1.0%	0.2%
All	1 $\mu\text{g m}^{-3}$	22.2%	9.7%	1.3%	29.5%	11.2%
All	2 $\mu\text{g m}^{-3}$	2.3%	1.5%	0.1%	7.0%	1.3%
All	3 $\mu\text{g m}^{-3}$	0.5%	0.4%	0.0%	0.5%	0.2%

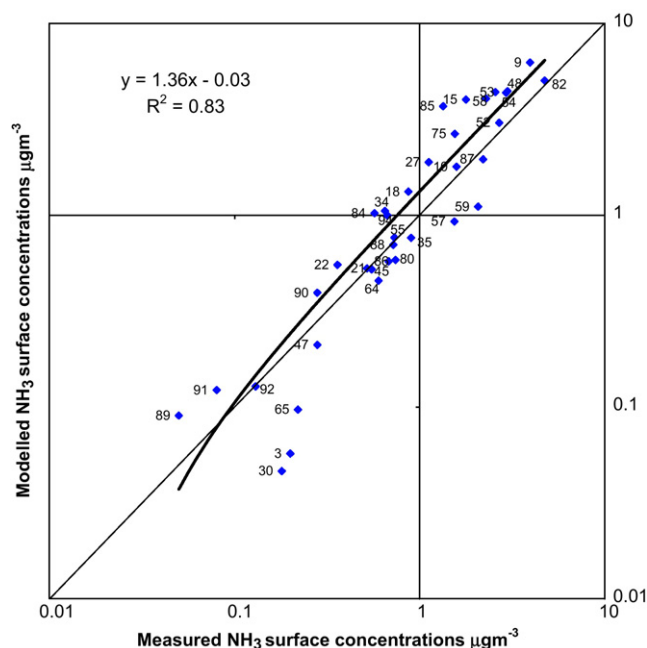


Fig. 6 – Comparison of FRAME modelled surface concentrations of ammonia with measurements from the UK National Ammonia Monitoring Network (Sutton et al., 2001) for the period 2002–2004, showing sites where monitoring is conducted at locations with semi-natural vegetation or woodland. Sites are numbered as shown in Fig. 4. Although the figure is plotted with log axes for better visibility, the fitted line represents a simple linear regression.

statistic, with fewer numbers of reserves exceeded by each critical level than with the uncalibrated model, compare Tables 3 and 5.

3.6. Case studies illustrating the spatial interactions for ammonia and SAC

Three case studies within the UK serve to reveal that low NH_3 concentrations are often found on SAC and SPAs since they are not farmed intensively. However there is a distinction

between small SAC and large ones and between those in remote and source areas.

Fig. 7 highlights the location of the Peak District, Fenn's Moss and Brown's Moss SAC, each of which are estimated to exceed the DWI for at least critical level $1 \mu\text{g NH}_3 \text{ m}^{-3}$. However, it can be seen that while Brown's Moss and Fenn's Moss show a very high level of exceedance, because of their small areas, they would contribute very little to the AWI statistic.

A similar example is illustrated by the northern edge of Dartmoor, for which ammonia concentrations exceed the $1 \mu\text{g NH}_3 \text{ m}^{-3}$ critical level, while other parts of Dartmoor are not exceeded (see Fig. 8), so that the AWI would tend to give a low result. By contrast, the small reserves of the Culm Grasslands exceed the critical level, with concentrations of $2\text{--}3 \mu\text{g NH}_3 \text{ m}^{-3}$. The whole area of these sites is compromised by exceedance of critical level which would be represented by the DWI statistic but contribute insignificantly to the AWI statistic.

Small area SAC are also affected by high ammonia concentrations in Northern Ireland, while the large SAC tend to be located in cleaner coastal regions (see Fig. 9). Thus overall, while 22% of the SAC area in Northern Ireland (the AWI) exceeds the $1 \mu\text{g m}^{-3}$ critical level, 74% of the SAC sites (the DWI) show exceedance of the $1 \mu\text{g m}^{-3}$ critical level (Tables 3 and 4).

4. Discussion and conclusions

4.1. Atmospheric modelling performance and ammonia critical levels

4.1.1. Importance of model resolution at the regional scale

The values in Table 1 are calculated from the uncalibrated modelled NH_3 air concentrations at 1 km and 5 km resolution. This shows that the differences in critical level exceedances between the 5 km resolution model and the 1 km model are small at country wide scales. The 5 km model indicates a slightly higher percentage of area exceeded by each of the three critical levels. At the country-wide scale, the small difference in modelled NH_3 exceedance of critical levels between the 5 km resolution model and the 1 km model (see Table 1) is explained by localised high peaks of emissions that are better resolved in the 1 km model but smoothed-out by the 5 km model.

Table 5 – Summary statistics for the Designation Weighted Indicator (DWI): the percentage number of UK SAC and SPAs where there is some exceedance of 1, 2 and $3 \mu\text{g NH}_3 \text{ m}^{-3}$ in England, Wales, Scotland, Northern Ireland and the UK as a whole (calibrated 1 km statistics only).

Natura 2000 component	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
SAC	$1 \mu\text{g m}^{-3}$	91.6%	78.0%	22.4%	73.6%	61.2%
SAC	$2 \mu\text{g m}^{-3}$	36.4%	37.8%	5.9%	41.5%	24.3%
SAC	$3 \mu\text{g m}^{-3}$	17.6%	14.6%	1.3%	7.5%	9.7%
SPAs	$1 \mu\text{g m}^{-3}$	92.7%	78.9%	26.8%	75.0%	54.0%
SPAs	$2 \mu\text{g m}^{-3}$	43.9%	31.6%	9.4%	41.7%	21.8%
SPAs	$3 \mu\text{g m}^{-3}$	20.7%	15.8%	2.9%	33.3%	10.1%
All	$1 \mu\text{g m}^{-3}$	91.9%	78.2%	24.0%	73.8%	59.1%
All	$2 \mu\text{g m}^{-3}$	38.3%	36.6%	7.2%	41.5%	23.6%
All	$3 \mu\text{g m}^{-3}$	18.4%	14.9%	1.9%	12.3%	9.8%

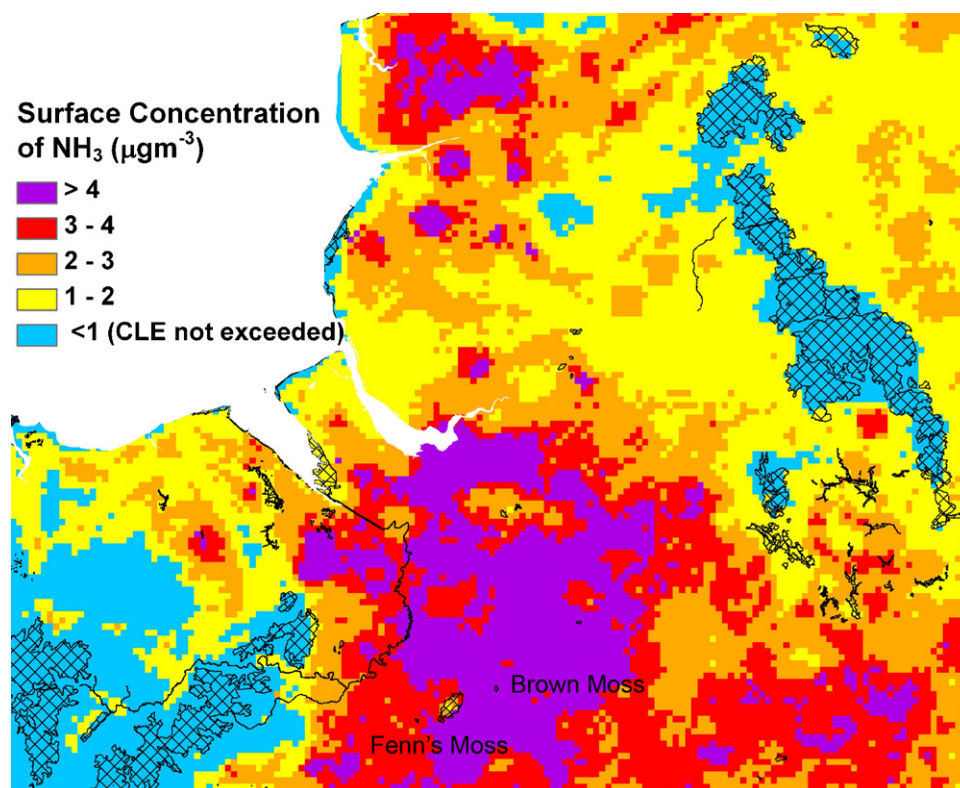


Fig. 7 – Spatial pattern of estimated NH_3 concentration (1 km resolution, calibrated FRAME) and the location of Special Areas of Conservation (SAC) for part of north-west England and north Wales. Large SAC areas are associated with $\text{NH}_3 < \text{critical level}$ ($1 \mu\text{g m}^{-3}$), while small SAC, such as Fenn's Moss and Brown Moss occurring in agricultural regions exceed the critical levels.

4.1.2. Importance of model resolution at the reserve scale

Our development of the 1 km resolution model was motivated by concern over the sub-grid scale variability at 5 km resolution, the effect of which is well illustrated at reserve scales (see Section 3.2 and Fig. 2). The 5 km resolution model evens out variations, so that the majority of Dartmoor and all of the New Forest are exceeded by the $1 \mu\text{g NH}_3 \text{ m}^{-3}$ critical level, whereas the 1 km model estimates these areas to be mainly less than $1 \mu\text{g NH}_3 \text{ m}^{-3}$, with the critical level only being exceeded near the SAC edges.

At large scales, this grid smoothing makes little difference to critical level exceedance. By contrast, at the SAC and SPA scale, the grid smoothing effect can change the classification (critical level exceedance) of entire reserves up to the scale of tens of kilometres. If a coarse spatial resolution is applied, information about emission peaks or low emissions occurring at a more local level will be lost. Applying the 5 km resolution emission values in an atmospheric transport model may also underestimate exceedances in semi-natural areas in close proximity to intensive agricultural areas.

4.1.3. The FRAME model compared with National Atmospheric Monitoring Network measurements

Fig. 4(a) shows that the 5 km resolution FRAME model slightly overestimates ammonia concentrations in semi-natural sites, mainly because of sub-grid scale variability in ammonia emissions. This is still apparent to a lesser extent with the new 1 km resolution FRAME application (see Fig. 4(b)). It is notable

that the correlation between modelled concentrations versus measurements is higher for the semi-natural sites than the overall network (which also includes sites in agricultural areas). This can be explained by the sites typically being further from sources, so that uncertainties in local emissions estimates are to some extent averaged out.

4.1.4. Calibration of the model

In this study we have used monitoring sites in semi-natural and woodland land types to calibrate the modelled ammonia surface concentrations. Although these sites are not necessarily representative of all of the nature reserves, the calibrated NH_3 concentrations can reduce some of the uncertainties in the model that lead to overestimation (compared to the measured concentrations in semi-natural and forested sites). On this basis, the calibrated estimates might be considered to be more realistic. Using the uncalibrated concentrations could therefore be considered to be a suitably precautionary approach to assessing protection of the Natura 2000 reserves.

4.1.5. Regression analyses

It should be noted that regression analysis used here minimises the square of residuals on the modelled concentrations, since the uncertainties the model estimates (e.g. related to census data, emission factors, atmospheric dispersion, 5 km and 1 km resolution estimates of site based concentrations) are much larger than the uncertainties in

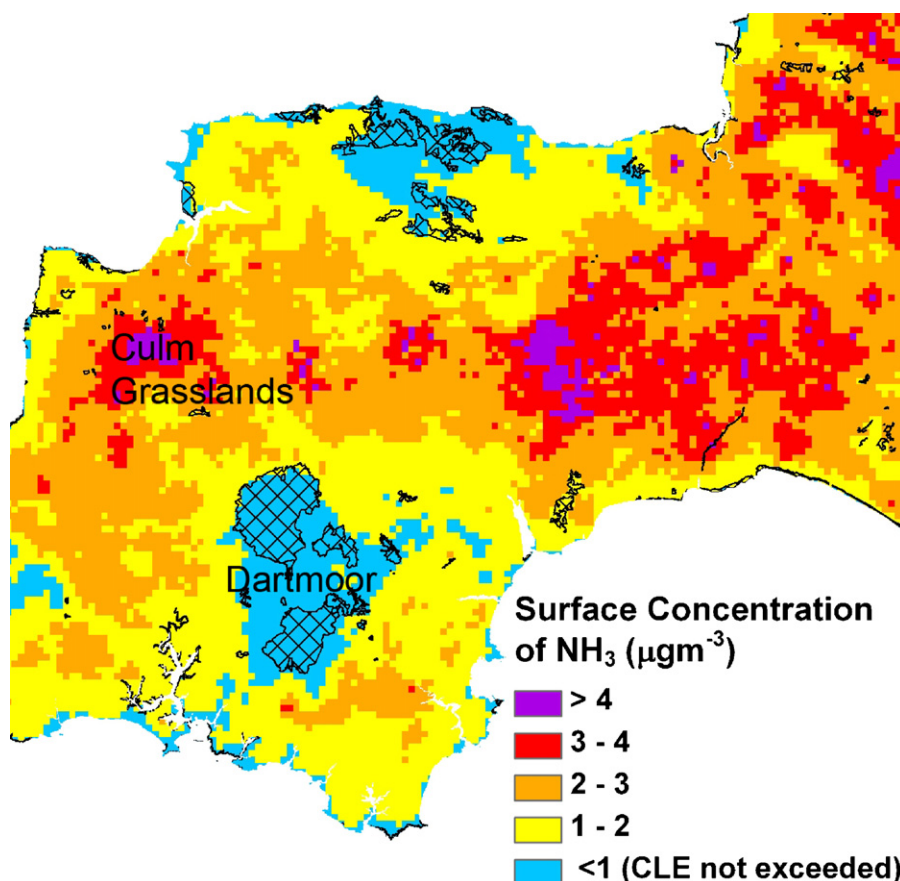


Fig. 8 – Spatial pattern of estimated NH_3 concentration (1 km resolution, calibrated FRAME) and the location of Special Areas of Conservation (SAC) in South West England. With the exception of small areas at the boundaries, most of Dartmoor is $<1 \mu\text{g m}^{-3}$ critical level. By contrast, the Culm Grasslands mostly exceed a critical level of $2 \mu\text{g m}^{-3}$.

the annual average measured concentrations Sutton et al. (2006). Nevertheless, as there is some error in the measured annual mean concentrations (overall $<5\%$), it is relevant to assess the influence of measurement uncertainty on our estimates where the calibrated model is applied.

We therefore carried out a mixed regression analysis for example estimates. Fig. 5b shows that approximately 83% of variance in model 'explained' by the regression ($R^2 = 0.83$). If we consider an upper estimate of 5% in the measured annual mean concentrations, this leaves, 12% of the variance that is not explained by the model, and may be attributed to overall model uncertainty. We therefore applied a mixed regression (x on y and y on x , weighted according to the reciprocal of the uncertainties: measurements weighted 20, model weighed 7.7). This provided the following regression: model (1 km) = $1.44 \times \text{measurement} - 0.13$. Applying this relationship to the UK values for "all sites" in Table 5 gives the following values of exceedance according of the DWI: 57.9%, 21.3% and 9.0%. These values are almost the same as those given in Table 5 (59.1%, 23.6% and 9.8%, respectively), with the differences ($<2\%$) being much smaller than the differences between the AWI and DWI approaches (Tables 4 and 5). The regression approach used here can thus be considered as well suited the model calibration for the purpose of the indicator comparison.

4.2. Comparison of the Area weighted and Designation Weighted Indicators for Natura 2000 sites

4.2.1. Area Weighted Indicator and the effect of modelled spatial resolution

Tables 2 and 6 show the percentage area of SAC and SPAs exceeding the critical levels (the Area Weighted Indicator) is less than percentage exceedance for the UK as a whole, a feature also seen for the devolved regions. The main reason for this is that many SAC and SPAs tend to be located in the less intensively farmed and mountainous regions of the UK with lower-than-average NH_3 concentrations. The SAC and SPAs in these remote areas also tend to be the largest, and therefore have the greatest influence on the statistics of the Area Weighted Indicator. In addition, the largest SAC and SPAs may have high ammonia concentrations near their edges surrounding farming activity, which are greatly diluted towards the centre of the SAC and SPAs.

4.2.2. Comparison of indicators at the reserve scale

Using the Area Weighted Indicator (AWI) (with 1 km calibrated ammonia), we estimate that 11% and 1% area of the UK Natura network exceeds the critical level values of 1 and $2 \mu\text{g NH}_3 \text{ m}^{-3}$, respectively. By contrast, the equivalent exceedances using the Designation Weighted Indicator (DWI) are

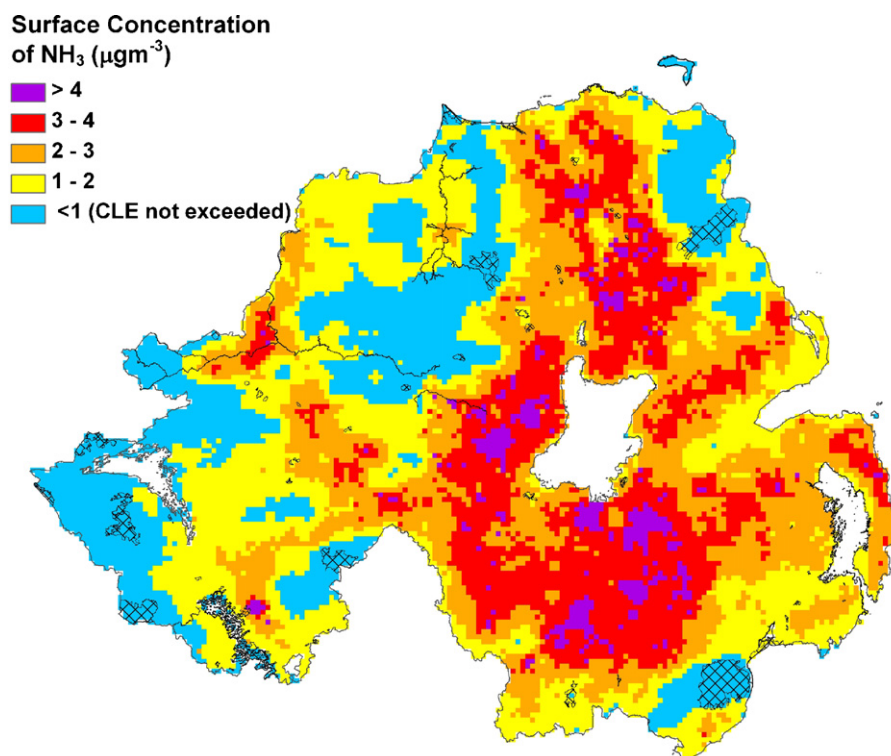


Fig. 9 – Spatial pattern of estimated NH_3 concentration (1 km resolution, calibrated FRAME) and the location of Special Areas of Conservation (SAC) in Northern Ireland. Although many of the largest SAC do not exceed the lowest critical level, substantial exceedance is seen for the smaller sites.

much larger at 59% and 24%, respectively. It is clear that the AWI provides a much less precautionary impression of overall risk than the DWI. The choice between the two approaches comes down to whether each km^2 of the Natura 2000 network is valued equally by society, or whether each designated SAC/SPA is valued equally. Since designation represents an indication of value by society, and since the AWI approach under represents the threat to small, high value Natura 2000 sites in the most threatened areas, we would argue that the DWI is the more suitable indicator.

The high resolution maps show that the larger threat estimated by the DWI approach is explained by (i) an anti-correlation between NH_3 concentration and Natura site area (see Fig. 10) and (ii) the fact that exceedance over part of a Natura site is considered to represent a threat to the integrity of the whole site.

The AWI is similar to the method used to assess the Public Service Agreement target in England for Sites of Special Scientific Interest. In the context of the Habitats Directive, however, there is concern that this does not sufficiently reflect the precautionary principles of the Habitats Directive. An assessment of site condition should aim to show if the critical level is exceeded over any part of a reserve, given that a threat to the integrity of part of a site is considered as representing a threat to the integrity of the whole. Under the terms of the Habitats Directive then, the DWI may be considered as the more appropriate measure.

4.2.3. Comparisons of AWI and DWI statistics with concentrations of ammonia averaged over whole reserves

The importance of variation in ammonia concentrations across individual Natura 2000 sites can be illustrated by the compari-

Table 6 – Summary statistics for the Area Weighted Indicator (AWI): the percentage area of the UK SPA network where NH_3 air concentrations of 1, 2 and 3 $\mu\text{g NH}_3 \text{ m}^{-3}$ are exceeded in England, Wales, Scotland, Northern Ireland and the UK as a whole (uncalibrated 5 km and 1 km models).

FRAME application	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
5 km model	1 $\mu\text{g m}^{-3}$	84.6%	27.6%	4.9%	82.5%	40.9%
5 km model	2 $\mu\text{g m}^{-3}$	21.6%	0.7%	0.8%	54.2%	10.0%
5 km model	3 $\mu\text{g m}^{-3}$	4.7%	0.1%	0.3%	34.7%	2.4%
1 km model	1 $\mu\text{g m}^{-3}$	46.6%	8.5%	2.9%	70.5%	22.3%
1 km model	2 $\mu\text{g m}^{-3}$	6.9%	0.6%	0.5%	39.2%	3.5%
1 km model	3 $\mu\text{g m}^{-3}$	1.7%	0.0%	0.0%	10.1%	0.8%

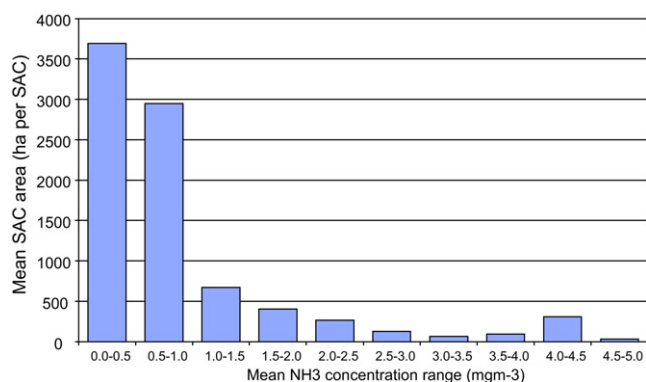


Fig. 10 – Relationship between mean NH_3 concentration over each SAC in the UK and SAC area. The smallest mean NH_3 concentrations are associated with the largest SAC.

son of the DWI (percentage number of sites with some exceedance of the critical level, Table 5) with the percentage number of Natura sites for which the average concentration over the Natura site exceeds the critical level (Table 7). The latter values as shown in Table 5 are lower than the corresponding values in Table 7 because large areas of some reserves are less than the critical level of $1 \mu\text{g NH}_3 \text{ m}^{-3}$ so that the mean concentration for these reserves does not exceed $1 \mu\text{g NH}_3 \text{ m}^{-3}$, 3, even though part of the reserve might be exceeded.

Averaging ammonia concentrations over entire Natura sites before analysing numbers of sites exceeded, is therefore more precautionary than the standard AWI method, but not as precautionary, however, as the application of the DWI approach (Table 5) and does not account for the point that a threat to part of a Natura site, represents a threat to the integrity of the whole site. In this regard, it should be noted that the DWI approach is applied here only down to the resolution of 1 km^2 , and that application of the DWI approach for less than 1 km^2 resolution would be expected to give larger percentage exceedance values.

4.3. Choice of critical level values

The scale of exceedance across the UK, as expressed by the DWI at 1 km (using calibrated NH_3 concentrations, Table 5) illustrates a substantial exceedance of ammonia critical levels, comparable with the exceedance of N critical loads, depending on the choice of critical level value used. Here we do not make any allocation of the different critical level values (1, 2 and

$3 \mu\text{g NH}_3 \text{ m}^{-3}$) to each site, which must be a matter for further analysis at the level of individual sites. For the smallest critical level value of $1 \mu\text{g m}^{-3}$, this has been set for lichens, bryophytes and habitats where these groups are considered to be important for the integrity of the overall ecosystem. In general, habitats like heathlands, shublands, bogs, and Atlantic Oak woods would be included under this heading, given the importance of their bryophyte and lichen communities. Whether this threshold should also apply for other habitats is less clear, and must be a matter for environmental managers based on the local conservation objectives.

In setting the critical ammonia level for higher plants, the UNECE-CLRTAP recognized significant uncertainty, and therefore set best estimate and range ($3 [2-4] \mu\text{g m}^{-3}$). They noted that the application of these values would depend on the regulatory context of assessment. Thus a value of $2 \mu\text{g m}^{-3}$ would be appropriate in the precautionary context, of the Habitats Directive, while a value of $3 \mu\text{g m}^{-3}$ would be appropriate as a value relevant for assessment based on balance of evidence (e.g. UK Sites of Special Scientific Interest). Similarly, the value of $4 \mu\text{g m}^{-3}$ (not analyzed here) could be considered as part of assessments based on balancing local economic development versus environmental protection (e.g. local nature reserves). In taking such an approach for Habitats Directive sites, it may be important to avoid the application of a double level of precaution. In this context, we would therefore recommend the $2 \mu\text{g m}^{-3}$ value together with the calibrated (rather than the uncalibrated) model estimates to be the most appropriate threshold (Table 5).

4.4. Policy implications

The first policy implication arising from this study concerns the choice of biodiversity indicators. The analysis indicates that the Designation Weighted Indicator (DWI) represents a more suitable index than the Area Weighted Indicator (AWI). An anti-correlation between ammonia concentrations and Natura site areas, results in the second approach under-representing the scale of the threat to the Natura 2000 network, especially for small SAC and SPAs located in intensive agricultural landscapes. Based on this analysis we conclude that the DWI, i.e. the percentage number of sites exceeding the thresholds, is the most policy relevant indicator. Accompanied with this is the message that across the UK, the majority of SAC and SPAs exceed the $1 \mu\text{g m}^{-3}$ critical level, with over 90% of sites being exceeded for England. Around a quarter of Natura 2000 sites in the UK exceed the $2 \mu\text{g m}^{-3}$

Table 7 – Summary statistics for the Designation Weighted Indicator (DWI): the percentage number of UK SAC and SPAs where there is some exceedance of 1, 2 and $3 \mu\text{g NH}_3 \text{ m}^{-3}$ in England, Wales, Scotland, Northern Ireland and the UK as a whole calculated in this case using average concentration for each Natura 2000 site (calibrated 1 km statistics only).

Natura 2000 component	Critical level/model	England	Wales	Scotland	Northern Ireland	UK
SAC	$1 \mu\text{g m}^{-3}$	69%	49%	9%	64%	43%
SAC	$2 \mu\text{g m}^{-3}$	15%	9%	1%	19%	9%
SAC	$3 \mu\text{g m}^{-3}$	5%	2%	0%	0%	2%
SPAs	$1 \mu\text{g m}^{-3}$	59%	21%	13%	67%	30%
SPAs	$2 \mu\text{g m}^{-3}$	7%	0%	1%	0%	3%
SPAs	$3 \mu\text{g m}^{-3}$	1%	0%	0%	0%	0%

critical level, with this value being around 40% for England, Wales and Northern Ireland.

A further policy implication of this study is the fine scale nature of ammonia emissions and protection of SAC and SPAs. The analysis shows clearly how high ammonia concentrations result from local variation in sources, which are mainly of agricultural origin. The result is that even small scale relocation of farm point and area sources (over 1–2 km, cf. Dragosits et al., 2006), can have a major impact in reducing ammonia concentrations on Natura 2000 sites. This indicates the potential for local spatial planning of ammonia emissions as a means to achieve protection targets. For example, if national target value were set for the DWI indicator applied to Natura 2000 sites, the degree of exceedance could be substantially reduced by actions taken immediately adjacent to the Natura 2000 sites themselves. Currently, national ammonia abatement policy focuses on application of the UNECE Gothenburg Protocol and the EC National Emissions Ceilings Directive. While such agreements are needed to reduce total ammonia emissions and deposition (especially in reducing wet deposition in remote areas), the analysis indicates that, for Natura 2000 sites adjacent to agricultural activities, local spatial policies could help substantially.

A further option that should be considered is the possibility to extend Local Air Quality Management approaches in relation to the protection of Natura 2000 sites. At present Local Air Quality Management focuses on the improvement of air quality in relation to human health, considering mainly industrial and vehicular emissions, especially in urban areas. As Europe's flagship nature conservation network, Natura 2000 would be a logical focus for extending this approach to protection of the natural environment. For example, an air quality target could be considered that should not be exceeded over any part of the domain of all SAC. The setting of such a value would logically be informed by the critical level values, but depending on ambition, might be different to the critical levels. National scale modelling such as that shown here is able to provide a basis for screening, with targeted ammonia measurements and Local Air Quality Management plans only being required in the main areas identified as being at risk. The present study demonstrates that the necessary tools are available for both the modelling and atmospheric monitoring.

Acknowledgements

We are grateful for financial support of this study from DEFRA and from the Centre for Ecology and Hydrology, together with travel support from the NitroEurope IP and the COST 729 and NinE research networks.

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