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The influence of model grid resolution on estimation of national scale nitrogen deposition and exceedance of critical levels

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Abstract

The Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) has been applied to model the spatial distribution of nitrogen deposition and air concentration over the UK at a 1 km spatial resolution. The modelled deposition and concentration data were gridded at resolutions of 1 km, 5 km and 50 km to test the sensitivity of calculations of the exceedance of critical loads for nitrogen deposition to the deposition data resolution. The modelled concentrations of NO_2 were validated by comparison with measurements from the rural sites in the national monitoring network and were found to achieve better agreement with the high resolution 1 km data.

High resolution plots were found to represent a more physically realistic distribution of nitrogen air concentrations and deposition resulting from use of 1 km resolution precipitation and emissions data as compared to 5 km resolution data. Summary statistics for national scale exceedance of the critical load for nitrogen deposition were not highly sensitive to the grid resolution of the deposition data but did show greater area exceedance with coarser grid resolution due to spatial averaging of high nitrogen deposition hot spots. Local scale deposition at individual Sites of Special Scientific Interest and high precipitation upland sites was sensitive to choice of grid resolution of deposition data. Use of high resolution data tended to generate lower deposition values in sink areas for nitrogen dry deposition (Sites of Scientific Interest) and higher values in high precipitation upland areas. In areas with generally low exceedance (Scotland) and for certain vegetation types (montane), the exceedance statistics were more sensitive to model data resolution.

1 Introduction

Emissions of reactive nitrogen to the atmosphere occur in the form of reduced nitrogen (ammonia gas) and oxidised nitrogen (NO and NO_2). The former is emitted predominantly from agricultural activities including both direct emissions from farm animals and from fertiliser application to farm land whilst the latter is generated from fuel combustion, notably power generation and road transport. NO_2 and NH_3 gas can be efficiently dry deposited to vegetation and, in the case of ammonia, deposition rates to acid grassland and forest are known to be particularly high due to low canopy resistance for these vegetation types. Subsequent atmospheric oxidation and chemical reaction can lead to the formation of nitric acid as well as ammonium and nitrate aerosols. Particulates are associated with long range transport on scales of hundreds or even thousands of kilometres. However the efficiency of nitrate and ammonium aerosols to act as cloud condensation nuclei in the formation of rain clouds results in nitrogen being washed out of the atmosphere and wet deposited to the surface. Whilst the input of nitrogen to agricultural land is generally considered to be a desirable stimulus to productivity, the cycle of emissions and deposition of nitrogen to natural ecosystems has important consequences for changes to biodiversity and can lead to a decreasing incidence of nitrogen intolerant plant species which become replaced by nitrogen-loving plants. Heath-land communities are highly sensitive to N deposition. In areas with high N deposition, extensive loss of heather and conversion to grassland has been detected (Bobbink et al., 1998). Nitrogen deposition is also an important pathway leading to acidification of terrestrial and freshwater ecosystems. Together with habitat conversion and climate change, atmospheric deposition of reactive nitrogen has been recognised as one of the most significant threats to global biodiversity (Sala et al., 2000). Atmospheric nitrogen deposition also poses a serious threat to sensitive semi-natural habitats in the UK (Hall et al., 2006; RoTAP, 2011).

Critical loads of nutrient nitrogen can be defined as "the highest deposition of nitrogen (as NH_x and/or NO_y) below which harmful effects in ecosystem structure and function do not occur according to present knowledge" (UBA, 2004). Two approaches for assigning nutrient nitrogen critical loads are currently in use in the UK:

1. Empirical critical loads based on experimental data or field studies on observed changes in the structure or function of ecosystems in relation to nitrogen deposition (Achermann and Bobbink, 2003; Bobbink and Hettelingh, 2011); these critical

loads have been applied to unmanaged coniferous and broadleaved woodland categories, grassland (acid and calcareous), dwarf shrub heath, bog, montane, and dune grassland (Hall et al., 2003, 2011). The critical loads are expressed as ranges; the UK "mapping value" within each range is the value used for the calculation of critical load exceedances. The critical load mapping values applied in this study are given in Table 1.

2. Mass balance critical loads based on an equation which balances all significant long-term inputs and outputs of nitrogen to terrestrial systems; this equation is used to calculate critical loads for managed coniferous and broadleaved woodland habitats.

Nitrogen deposition for 2006–2008 exceeds the critical loads for nutrient N for 58 % of the sensitive habitat areas of the UK; this is expected to decline to 48 % by 2020. Empirical critical loads have recently been reviewed and updated (Bobbink and Hettelingh, 2011; Hall et al., 2011). Use of the updated critical loads resulted in larger areas of sensitive habitats with critical load exceedance. The range of empirical critical loads for different ecosystem types is illustrated in Table 1.

Critical levels are the atmospheric concentrations of pollutants (e.g. ammonia) above which adverse effects on receptors (e.g. vegetation) may occur according to present knowledge (UBA, 2004).

The level of nitrogen deposition to natural ecosystems is assessed in many countries by national monitoring programmes to measure the concentrations of gases (ammonia and nitrogen di-oxide) and aerosols (ammonium and nitrate) in air as well as the chemistry of precipitation. The long term maintenance of monitoring networks and chemical analysis of samples requires high levels of funding and inevitably imposes restrictions on the number of sites and therefore the spatial density at which measurements can be undertaken. Atmospheric transport models therefore represent an alternative and complimentary means of assessing nitrogen deposition. Despite the fact that models require simple parameterisation of highly complex meteorological, physical and

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chemical processes in the atmosphere, they are able to provide reasonable estimates of nitrogen deposition to natural ecosystems. Models have the added advantage that calculations are made at a large number of model grid cells, invariably with much higher spatial density than that which can be achieved by measurement alone.

The spatial resolution at which calculations can be made with an atmospheric transport model depends on a number of factors. These include the size of the model domain, available computational power and the degree of complexity of the model. Inevitably, highly complex model simulations over large spatial domains will have limits imposed on grid resolution by the available computer facilities. Furthermore, fine spatial scale assessment of nitrogen deposition requires high resolution input data for meteorology as well as maps of land use and reactive nitrogen emissions.

Most atmospheric transport models use grid nesting to pass information from a large scale (i.e. continental) simulation with a coarse grid spacing to a regional scale (typically national) with finer grid spacing. This process can be cascaded in multiple stages. However, plume models are the typical choice for local scale studies as Eulerian models are generally not suitable for implemention with very fine grid horizontal spacing. The OPS model represents a combination of a Gaussian plume model for local-scale application and a trajectory model for long-range transport operating on grid scales of 5 km and 500 m (Van Pul et al., 2004). The model was used to simulate concentrations, deposition and budgets of NH_3 gas and NH_4^+ aerosol for the Netherlands. The Danish Ammonia Modelling system (DAMOS) uses a combination of a long range transport model and a Gaussian local scale transport-deposition model for dry deposition. The model operates on a variety of scales with two-way nesting, from 150 km for the Northern Hemisphere, 50 km for Europe and 16.7 km for Denmark. Ammonia emissions are computed with high spatial and temporal resolution at a single farm and field level (Gyldenkaerne et al., 2005). Vogt (2011) calculated ammonia concentrations and deposition at a 25 m resolution in an agricultural landscape. Fine resolution model simulation was demonstrated to be necessary to reproduce measured ammonia concentrations. A detailed discussion of modelling nitrogen deposition at a local scale is

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presented in Hertel et al. (2006). The high resolution in the inventories was shown to be important for the model performance.

The choice of model grid resolution can influence results in different ways:

1. Spatial Averaging: spatial averaging of both input data and output modelled air concentrations and surface deposition can have important implications. In the real atmosphere, gas concentrations of nitrogen in air may vary significantly on the scale of a few tens of metres in the vicinity of a strong emissions source such as a road or poultry shed. In reality however, national and continental scale modelling assessments are typically practical at grid resolutions in the range of 1 km to 50 km. Finer resolution data, averaged over smaller areas can more effectively capture hot spots of high concentrations and deposition, leading to different results for assessment of the exceedance of critical loads and levels.

2. Process Rate: the processes parameterised in the model simulation may behave differently depending on grid resolution. One example of this is the representation of orographic precipitation in meteorological models (Richard et al., 2007). It has been observed that with a larger grid resolution of 50 km, the influence of hills in generating orographically enhanced precipitation in the UK was not effectively captured. A 5 km simulation generated significantly higher precipitation in hill areas (RoTAP, 2011). A second example is the rate of chemical transformation in the atmosphere. Larger model grid sizes result in more instantaneous mixing of emitted acid gases (NO_x and SO_2) with ammonia (NH_3). This may lead to enhanced chemical reaction rates and more rapid formation of ammonium nitrate and ammonium sulphate aerosol particles.

The Fine Resolution Atmospheric Multi-pollutant Exchange model is a Lagrangian Atmospheric Transport Model. Its relatively simple dynamic framework and chemical schemes result in fast run times. The model is therefore well suited to high resolution national scale simulations, which are currently too computationally demanding for more complex Eulerian models. The aim of this work is to investigate the influence of

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spatial averaging of modelled air concentrations and deposition (effect 1 above) on the exceedance of critical loads for nitrogen deposition.

The results of a 1 km model simulation have been mapped at resolutions of 1 km, 5 km and 50 km. This ensures that total national scale deposition is conserved. The three data sets have been compared spatially and assessed by validation with measurements of NO_2 gas concentrations. The different resolution data have been used to assess the sensitivity of model grid resolution on the exceedance of critical loads for nitrogen deposition in the UK. The influence of model grid resolution on NO_2 concentrations near a Site of Special Scientific Interest (SSSI) and on wet deposition of nitrate in a high precipitation upland region is considered.

1.1 Model description

The FRAME model was initially developed to calculate spatially distributed deposition of reduced nitrogen for the UK (Singles et al., 1998). It was subsequently improved to include a detailed representation of oxidised nitrogen and sulphur deposition (Fournier et al., 2004; Dore et al., 2007; Vieno et al., 2009) to be used for assessment of the exceedance of critical loads for nitrogen deposition and acid deposition (Matejko et al., 2009). Other regional scale applications of the model include estimation of ammonia concentrations and deposition of reduced nitrogen in Poland (Kryza et al., 2010). The relative simplicity of the model corresponds to a fast simulation time (of approximately 12 h for a 1 km resolution using 24 processors on a High Performance Computational Linux cluster).

A summary of the main features of the FRAME model is given below:

- 5 × 5 km or 1 × 1 km resolution over the British Isles (incorporating the Republic of Ireland).

- Input gas and aerosol concentrations at the edge of the model domain are calculated with FRAME-Europe, using European emissions and run on the EMEP 50 km scale grid. (Cooperative Programme for Monitoring and Evaluation of the

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Long-range Transmission of Air Pollutants in Europe http://www.emep.int/UniDoc/index.html).

- Air column divided into 33 layers moving along straight-line trajectories in a Lagrangian framework with a 1° angular resolution. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sonde measurements (Dore et al., 2006a). Variable layer thickness from 1 m at the surface to 100 m at the top of the mixing layer.
- Emissions are gridded separately by SNAP (Selected Nomenclature for Air Pollution) sector for SO_2 and NO_x using emissions data from the National Atmospheric Emissions Inventory (http://naei.defra.gov.uk/) for the year 2007, gridded at a 1 km resolution. NH_3 emissions were input by agricultural sector and injected into vertical model layers which depend on the sector.
- Vertical diffusion in the air column is calculated using K-theory eddy diffusivity and solved with the Finite Volume Method.
- Wet deposition is calculated using a scavenging coefficient and a "constant drizzle" approximation driven by an annual rainfall map. A precipitation model is used to calculate wind-direction-dependent orographic enhancement of wet deposition (Fournier et al., 2005).
- Five land classes: forest, moorland, improved grassland, arable, urban and water are considered. A vegetation specific canopy resistance parameterisation is employed to calculate dry deposition of SO_2, NO_x and NH_3.
- 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.

Complex Eulerian models employing dynamical meteorological drivers such as CMAQ (Chemel et al., 2011) and EMEP4UK (RoTAP, 2011) have also been employed

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to calculate national scale nitrogen deposition in the UK. Such models can calculate temporally resolved deposition and concentration and their dependence on variable dynamic meteorology. Validations of different models against annually averaged measurements have been undertaken by Chemel et al. (2011) and Carslaw (2011). Despite its relatively simple approach, FRAME still offers certain advantages in calculating surface gas concentrations due to its fine vertical grid spacing (1 m at the surface) and detailed treatment of plume rise for point sources as well as in wet deposition due to use of a precipitation map generated from measurements. This resulted in good correlation with measurements of gas concentrations (NH_3, NO_2, SO_2) and an overall reasonable performance when compared to more complex models.

Previous national scale applications of the FRAME model (Dore et al., 2007; Vieno et al., 2009; Matejko et al., 2010; Kryza et al., 2010) were undertaken with a 5 km grid resolution. The development of the FRAME model at a 1 km resolution over the UK and application to assess exceedance of the critical level for ammonia concentration in air over Natura 2000 sites (Special Protection Areas and Special Areas of Conservation) is described in Hallsworth et al. (2010). The study showed that a 1 km model simulation generated significantly lower values for the percentage of land surface area in nature sites with concentrations of ammonia in air exceeding the critical levels of 1 and 3 $\mu g\,m^{-3}$ when compared to 5 km resolution model data. This was attributed to the better spatial separation of agricultural source emissions areas for ammonia from sink nature reserve areas with the fine resolution study.

2 Input data

2.1 Emissions

The UK National Atmospheric Emissions Inventory is updated on an annual basis and provides spatially disaggregated maps of a range of pollutant emissions at a 1 km resolution. Whilst atmospheric transport models have the potential for improved performance by use of high spatial resolution studies, it should be noted that the benefits depend strongly on the accuracy of high resolution input data, in particular emissions.

In the case of NO_x emissions, the location of emissions (from vehicles travelling along roads and from major point source combustion sources) is well known. However due to the large number of different vehicles using the roads, there is uncertainty in emissions factors for NO_x from vehicles as well as how this may change in time for older vehicles. Road traffic is monitored at many locations throughout the country. However in rural areas, there may be greater uncertainty in mapping emissions in the vicinity of minor roads.

Emissions of ammonia are predominantly from agriculture and in particular due to direct emissions from livestock. Total emissions are estimated based on census data of farm animals and estimates of emissions factors per animal. The spatial distribution of agricultural ammonia emissions uses census data at a parish level and distributes emissions within each parish according to land use category and its suitability for agriculture. In general the spatial distribution of ammonia emissions from agriculture are associated with higher uncertainty than NO_x emissions. The concentration and deposition of ammonia has been demonstrated to vary significantly over short distances (Fowler et al., 1998).

2.2 Precipitation

An annual precipitation map of the UK at a 1 km resolution was generated using data from 6000 daily measurement sites and 13 100 monthly measurement sites in the UK Met Office national precipitation monitoring network. (Keller et al., 2006). The triangular planes methodology (Jones, 1983) was used to generate daily 1 km^2 rainfall grids using a weighted average based upon the inverse distance of the three nearest rain gauges. The gridded rainfall data was then normalised and the daily rainfall totals were summed to give annual precipitation. The technique was successfully validated by comparison

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of the calculated rainfall data with site measurements (not included in the calculation) both for specific precipitation events and for monthly totals.

In this study we focus on nitrogen deposition in the UK as a whole as well as a region of highly variable precipitation in Snowdonia in North Wales. Figure 1a and b illustrate the topography of the region and gridded precipitation data at a resolution of 1 km.

The Snowdonia region is located near the west coast of North Wales and contains steep peaks of altitude exceeding 1000 m separated by broad valleys. The orography in the region has a strong influence on annual precipitation which varies from 1200 m near the west coast to approximately 4000 m in the region of Mt. Snowdon and varies significantly at a 1 km resolution scale. Precipitation in this region is strongly influenced by the seeder-feeder effect (Fowler et al., 1988). The ascent and cooling of moist boundary layer air in prevailing winds frequently leads to the formation of low-level hill clouds. Whilst these clouds are generally too short lived to form into rain, their cloud droplets can be efficiently washed out by rain drops falling from higher level frontal rain clouds. A study of precipitation and wet deposition in this region using a simple model of the seeder-feeder effect is described in Dore et al. (2006b). The high precipitation resulted in high levels of deposition of sulphur and nitrogen and exceedance of critical loads for acid deposition and nitrogen deposition.

3 Results and discussion

3.1 Nitrogen concentration and deposition

The concentrations of nitrogen compounds in air as well as nitrogen deposition to the surface were calculated with FRAME at a 1 km resolution for the UK for the year 2007. The model output was aggregated by calculating the average of the 1 km gridded data at spatial resolutions of 5 km and 50 km. This data was then used to investigate the influence of model grid resolution on the exceedance of critical loads for nitrogen deposition.

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Figure 2 illustrates the deposition of nitrogen for the UK calculated by FRAME at a 1 km resolution. A general trend in the deposition of nitrogen deposition, decreasing from south to north is apparent. This is caused by more intense industrial and agricultural activity in the south of the country as well as greater proximity to major sources of emissions of reactive nitrogen from other European countries and from international shipping in the English Channel. Local areas of high nitrogen deposition are also evident in the vicinity of major cities (i.e. London, Birmingham, Manchester) and along major highways as well as in intensive agricultural areas (due to cattle farming in western England and pig and poultry farming particularly in East Anglia) and in upland regions with high precipitation (North Wales, the Pennines, the Lake District).

Figure 3 shows comparisons of NO_x concentrations generated with FRAME and gridded at both 1 km and 5 km resolutions at Stanford Park Site of Special Scientific Interest which is located approximately one km away from a major road. With the 1 km data, the NO_x concentrations are more closely correlated with the locations of the roads, in a more physically realistic manner. This resulted in lower NO_x concentrations being assigned to the grid square containing the SSSI. The 1 km gridded data therefore represents an improved spatial distribution of air pollutant concentrations. However, even with 1 km data, strong spatial gradients in air concentrations may occur as a result of the physical limitations of the specified model grid. For focused local scale studies, dedicated local scale dispersion models are preferable. However the 1 km resolution simulation of nitrogen deposition data represents an improved reference national data set for sites where data from local scale dispersion studies is not available.

Figure 4 illustrates NO_y wet deposition in Snowdonia gridded at resolutions of both 1 km and 5 km. Precipitation in hill areas is known to vary significantly over relatively small distances. This can lead to hot spots in wet deposition which are captured by 1 km resolution data but less evident with 5 km resolution data due to spatial smoothing over wider areas incorporating both hill peaks and lowlands.

3.2 Comparison with measurements of NO_2

Model performance may be assessed by validation with measurements from national monitoring networks. A comparison of model correlation with measurements of ammonia concentrations using both 1 km and 5 km resolution data has been undertaken by Hallsworth et al. (2010). An overview of model performance for gas and aerosol concentrations of sulphur and nitrogen compounds as well as precipitation concentrations is given in Dore et al. (2007) and Carslaw et al. (2011). Here we consider model correlation with measurements of NO_2 gas concentrations. In the UK, monitoring of NO_2 concentrations is undertaken with both the rural monitoring network using diffusion tubes (20 sites) and the Automated Urban and Rural Network (AURN). The 12 rural sites in the AURN are considered here for validation of the model.

Figure 5 illustrates the correlation of the model with measurements of NO_2 concentrations. The statistical analysis in Table 2 demonstrates clearly that there is an improved agreement with measurements of NO_2 concentrations from both the rural monitoring network and the rural sites in the AURN using 1 km resolution gridded data as compared to 5 km or 50 km resolution. With the 1 km data, the lowest values for normalised mean bias, root mean square error and mean average gross error are achieved. However there is only a relatively small improvement in some of these statistics for the 1 km resolution data when compared to the 5 km resolution data. Indeed, as illustrated in Fig. 5, at some sites, the NO_2 concentrations with the 1 km resolution data are almost identical to those with the 5 km resolution data. This result can be explained by considering the criteria for defining a "rural" site which are that it should be located at least 2.5 km from a road. This means that the process of averaging 1 × 1 km gridded concentration data over 25 squares to a 5 × 5 km grid which contains a rural monitoring site is quite likely not to include any elevated NO_2 concentrations from local traffic. As such, the design of the rural monitoring networks are not ideal for demonstrating the benefits of changing model grid resolution from 5 × 5 km to 1 × 1 km which are illustrated in Fig. 3a for Stanford Park SSSI.

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3.3 Exceedance of critical loads

The calculation of the exceedance of critical loads for nitrogen deposition is described in detail in Hall et al. (2006) and RoTAP (2011). Exceedances were calculated separately for each habitat type using 1 km critical loads data and ecosystem-specific deposition (i.e. moorland deposition for the grassland, heath, bog and montane habitats, and woodland deposition for the woodland habitats). However, rather than include separate exceedance maps for each habitat, the exceedance maps presented here (Fig. 6a and b) are based on 5th-percentile critical loads that combine data for all habitats. The 5th-percentile critical load is set to protect 95 % of the total sensitive habitat area in each 1 km square. The exceedance was calculated using grid-average deposition because different habitats may define the percentile critical loads for nitrogen deposition in each grid square.

The results of calculating exceedance of the critical load for nutrient nitrogen deposition for each habitat across the UK are summarised in Table 3a and b. The influence of aggregating 1 km deposition data to a 5 km resolution grid is not manifested in a very large change in the percentage habitat area with exceedance of the critical load. The reason for this is the quasi-random distribution of source areas (i.e. industry, roads and agriculture) and sink areas (sensitive ecosystems). However, overall the exceedance of critical loads for nitrogen deposition is higher using the 5 km resolution data due to mixing of source areas (agriculture for reduced ammonia and road transport for oxidised nitrogen) with sensitive ecosystems in the same model grid square. With deposition data gridded at a 50 km resolution, the national scale area of ecosystem with exceedance is higher (35.4 %) than for 1 km data (31.5 %). This occurs because at the coarse 50 km resolution, nitrogen sources emitted from major industrial and urban areas are effectively co-located with natural ecosystems. Although the area exceeded was higher when using the 50 km deposition data, the magnitude of exceedance was lower. Use of 50 km resolution deposition resulted in spreading the exceedance wider, but with smaller resulting magnitude.

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For regions with lower percentage area exceedance (Scotland and Northern Ireland), the influence of spatial averaging of nitrogen deposition over the larger 50 km grid cells resulted in more significant changes in total area with exceedance (i.e. from 5.1 % for 1 km data to 7.5 % for 50 km data for Scotland). For countries with higher percentage area exceedance (England and Wales) spatial averaging of deposition resulted in relatively small changes in the percentage in total area with exceedance (i.e. from 71.4 % for 1 km data to 73.8 % for 50 km data for England).

When percentage area exceedance is considered for the UK as a whole according to habitat type (Table 3b), the general trend for increased percentage area exceedance using data with larger grid spacing is apparent. However for certain habitats (notably montane) the opposite is true. For this vegetation type, lower exceedances were obtained with the 50 km data. This is due to the fact that this habitat is associated with upland high precipitation areas located almost predominantly in the Scottish Highlands and that spatial averaging over 50 km in these regions leads to reduced wet deposition of nitrogen at the high elevation sites.

Furthermore it should be noted that the importance of spatial resolution inevitably depends on the nature of the landscape and the level of spatial mixing between source areas of nitrogen emissions (agriculture, roads and urban areas) and sink areas (natural ecosystems). In regions where emissions are densely concentrated in areas of intense agriculture and urban agglomerations which are distinctly separated from natural ecosystems, high grid resolution is of lower importance. However for regions such as that illustrated in Fig. 3 (a site of Special Scientific Interest with a nearby strong source of active nitrogen emissions), spatial resolution will be of greater significance.

The exceedance of the 5th percentile critical load for nitrogen deposition for data gridded at 1 km, 5 km and 50 km resolution is illustrated as mapped data for the UK in Fig. 6a and for Snowdonia national park in Fig. 6b. A general tendency is for the larger grid resolution data (i.e. 50 km) to lead to greater areas with exceedance of the critical load but lower values of exceedance. For Snowdonia, the use of 50 km resolution data results in no exceedances greater than 0.6 keq Ha^{-1} whereas with both 1 km and 5 km

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resolution data, exceedances were higher than 1.0 keq Ha^{-1} in many high precipitation upland regions.

4 Conclusions

An atmospheric transport model has been run at a resolution of 1 km over the UK. The data on concentrations of nitrogen compounds in air as well as wet and dry deposition of nitrogen was gridded at three resolutions: 1 km, 5 km and 50 km.

The high resolution 1 km data was found to be an improvement for investigating nitrogen deposition and exceedance of critical loads in areas with high emissions of ammonia from agriculture (i.e. Hallsworth et al., 2011) and high emissions of NO_x from road transport as well as in high precipitation upland areas (this study). Validation of the modelled NO_2 concentrations by comparison with measurements from rural sites demonstrated an improved correlation using the higher resolution 1 km data.

National scale statistics for exceedance of the critical load for nitrogen deposition were not highly sensitive to the resolution of the modelled data. However, the effect of spatial averaging resulted in greater areas with exceedance using the coarser 50 km resolution data and the lowest areas with exceedance using the 1 km data (but with lower values of exceedance at the higher resolution). For certain ecosystems (i.e. montane), the national scale exceedance statistics were particularly sensitive to the grid resolution of the modelled data. For regions with generally low area exceedance (i.e. Scotland) the area exceedance was more sensitive to model data resolution than regions with higher area exceedance (i.e. England).

In conclusion, the 1 km resolution data set provides an improved reference data set for local studies of nitrogen and acid deposition, in particular when data from specific local dispersion models is absent. High resolution data is of particular importance for upland regions with high precipitation or for Sites of Special Scientific Interest located near to major sources of emissions such as road transport or agriculture. Less high

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resolution data (i.e. 5 km) was found to be adequate for calculating national scale summary statistics on the exceedance of critical loads.

Accurate assessment of nitrogen deposition at fine resolution relies on detailed emissions maps of both oxidised and reduced nitrogen. Improvements in information on the spatial distribution of livestock numbers and agricultural practice are necessary to achieve better estimates of nitrogen dry deposition. In upland areas, containing sensitive ecosystems, wet deposition is an important pathway for nitrogen input. Widespread monitoring and accurate modelling and mapping of precipitation in hill regions are required to improve estimates of nitrogen deposition in these regions.

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Table 1. Ranges of empirical critical loads of nutrient nitrogen and UK mapping values.

Ecosystem type	EUNIS code[a]	CLnutN range 2003[b] (kg N ha^{-1} yr^{-1})	UK CLnutN mapping value 2003[c] (kg N ha^{-1} yr^{-1})
Coastal habitats			
Shifting coastal dunes	B1.3	10–20[f]	15
Stable dune grasslands	B1.4	10–20[e]	15
Mire, bog & fen habitats			
Raised and blanket bogs	D1	5–10[d]	10
Grasslands & tall forb habitats			
Semi-dry calcareous grassland	E1.26	15–25[d]	20
Dry acid grassland	E1.7	10–20[e]	15
Moist/wet oligotrophic grassland	E3.52	10–20[e]	15
Moss & lichen dominated montane	E4.2	5–10[e]	7
Heathland, scrub & tundra			
Upland *Calluna*-dominated wet heath	F4.11(U)	10–20[f]	15
Lowland *Erica tetralix*-dominated wet heath	F4.11 (L)	10–25[f]	15
Dry heath	F4.2	10–20[d]	12
Forest habitats			
Unmanaged woodland (to protect ground flora)	G	10–15[e]	12
Unmanaged woodland (to protect epiphytic lichens and algae)	G	10–15[f]	10

[a] Habitat class according to the European Nature Information System (EUNIS) (Davies et al., 2004).
[b] Ranges and reliability scores of critical loads (Achermann and Bobbink, 2003); these have been updated since the analysis for this paper was undertaken, see Bobbink and Hettelingh (2011).
[c] UK mapping values (Hall et al., 2003); these have been updated since the analysis for this paper was undertaken, see Hall et al. (2011).

Reliability scores:
[d] Reliable: when a number of published papers of various studies showed comparable results.
[e] Quite reliable: when the results of some studies were comparable.
[f] Expert judgement: when no empirical data were available for this type of ecosystem. For this, the nitrogen critical load was based upon expert judgement and knowledge of ecosystems which were likely to be comparable with this ecosystem.

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Table 2. Statistics for the model correlation with measurements of NO_2 concentrations ($\mu g\, m^{-3}$) from the rural monitoring and Automated Urban and Rural monitoring Networks (rural sites only) with data gridded at 1 km, 5 km and 50 km resolution.

	rural network			AURN		
	1 km	5 km	50 km	1 km	5 km	50 km
maximum value	1.373	1.352	0.798	6.881	6.640	6.632
minimum value	−5.504	−5.513	−8.075	−4.339	−6.733	−8.992
mean bias	0.197	0.256	0.772	0.183	0.640	2.192
mean average gross error	0.983	1.019	1.053	1.669	2.021	3.356
root mean square error	1.620	1.627	2.015	2.527	2.975	4.478
normalised mean bias	0.039	0.050	0.152	0.018	0.064	0.220
normalised average mean error	0.193	0.200	0.207	0.167	0.203	0.337
correlation coefficient	0.937	0.947	0.897	0.817	0.806	0.804

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Table 3a. National scale summary statistics for habitat areas with exceedance of the critical load for nutrient nitrogen deposition by region.

Country	Habitat Area (km^2)	Percentage area habitats exceeded using FRAME deposition for 2007 at the following resolutions:		
		1 km	5 km	50 km
England	20 299	71.4	72.5	73.8
Wales	7101	76.8	79.9	84.9
Scotland	43 530	5.1	5.6	7.5
Northern Ireland	3500	37.3	40.3	58.0
UK	74 430	31.5	32.6	35.4

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Table 3b. National scale summary statistics for habitat areas with exceedance of the critical load for nutrient nitrogen deposition by habitat type.

Broad Habitat	Habitat Area (km^2)	Percentage area habitats exceeded using FRAME deposition for 2007 at the following resolutions: 1 km	5 km	50 km
Acid grassland	15 247	27.8	29.3	32.0
Calcareous grassland	3578	24.1	23.9	18.2
Dwarf shrub heath	24 826	9.6	10.2	11.6
Bog	5537	25.7	26.9	35.8
Montane	3129	7.3	5.8	0.6
Coniferous woodland (managed)	8383	44.4	48.0	60.7
Broadleaved woodland (managed)	7482	89.7	89.9	89.1
Unmanaged woods (ground flora)	3297	85.8	86.0	86.2
Atlantic oak (epiphytic lichens)	822	40.9	40.9	45.3
Supralittoral sediment	2129	34.3	37.8	43.8
All habitats	74 430	31.5	32.6	35.4

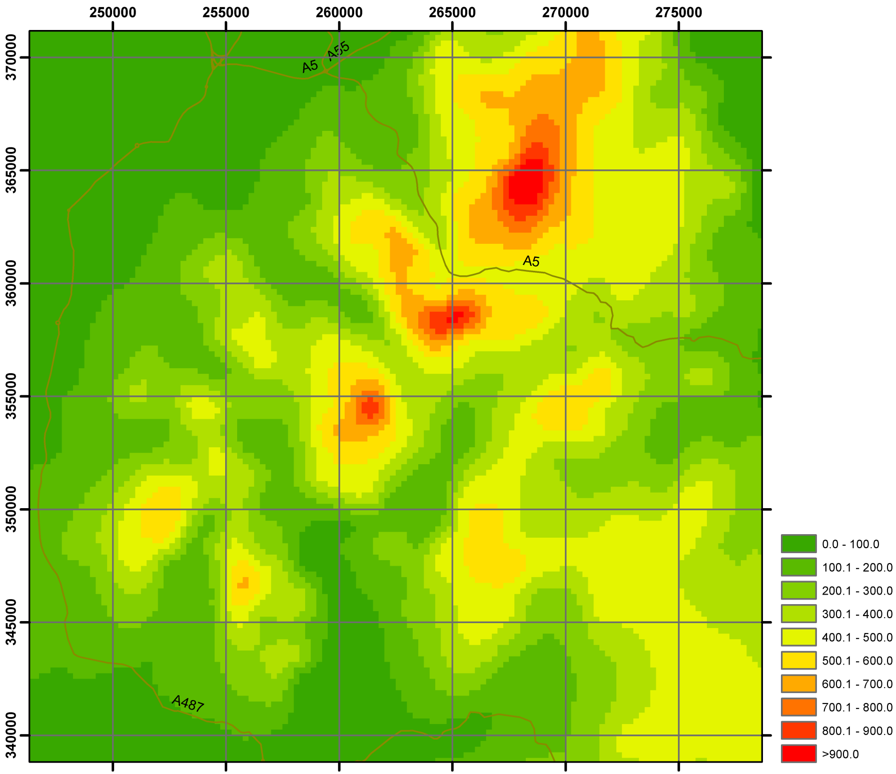

Fig. 1a. Terrain elevation in the region of Snowdonia, North Wales (m a.s.l.).

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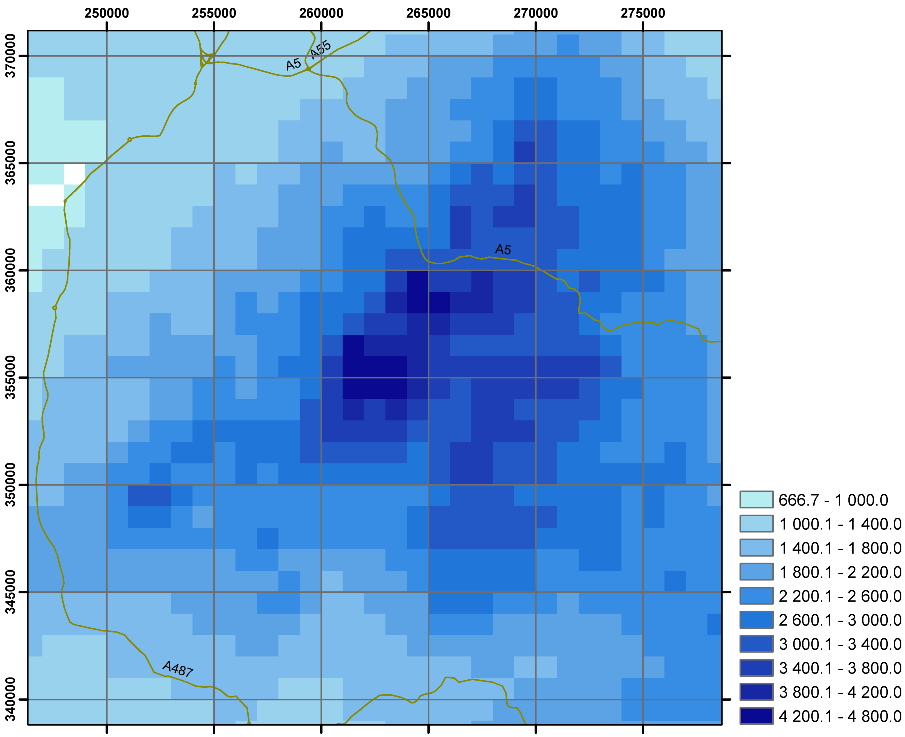

Fig. 1b. Annual precipitation (mm) gridded at a 1 km resolution for Snowdonia.

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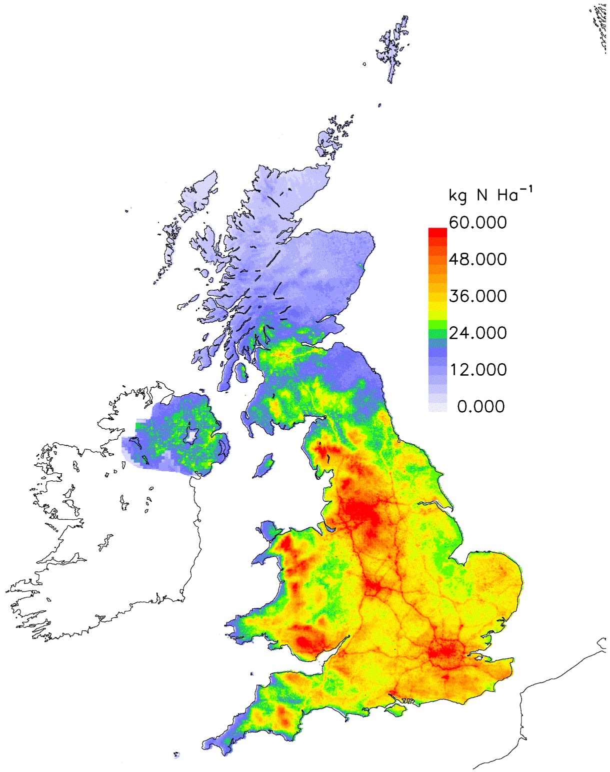

Fig. 2. Total nitrogen deposition in the UK for the year 2007 calculated with the FRAME model at a 1 km resolution.

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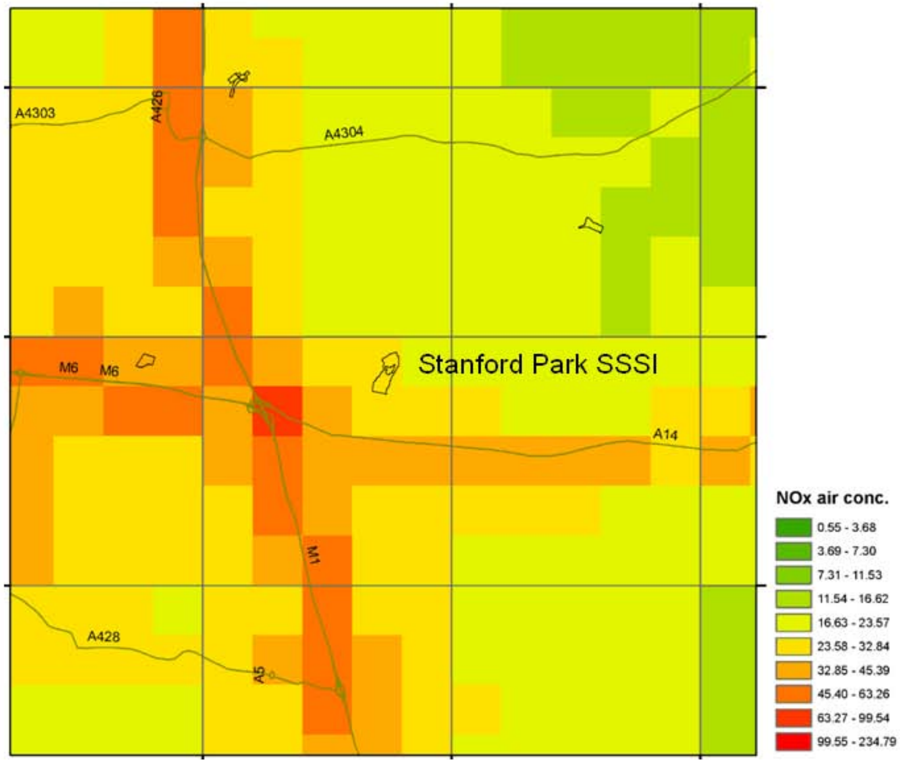

Fig. 3a. NO_x air concentrations at Stanford Park SSSI ($\mu g\, m^{-3}$) with 1 km grid resolution.

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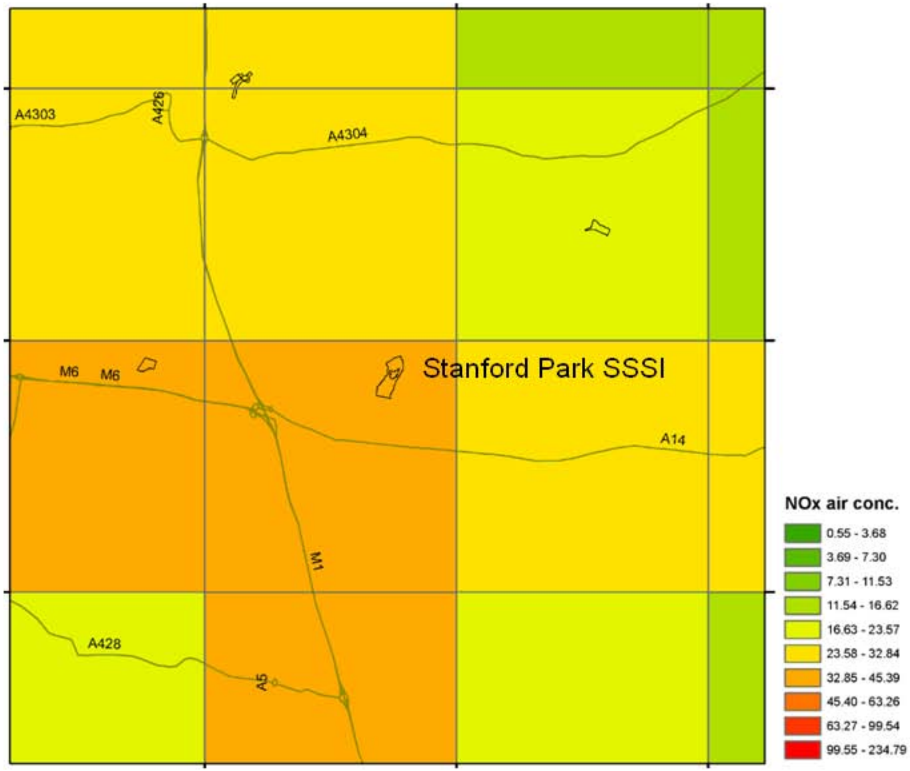

Fig. 3b. NO_x air concentrations at Stanford Park SSSI ($\mu g\,m^{-3}$) with 5 km grid resolution.

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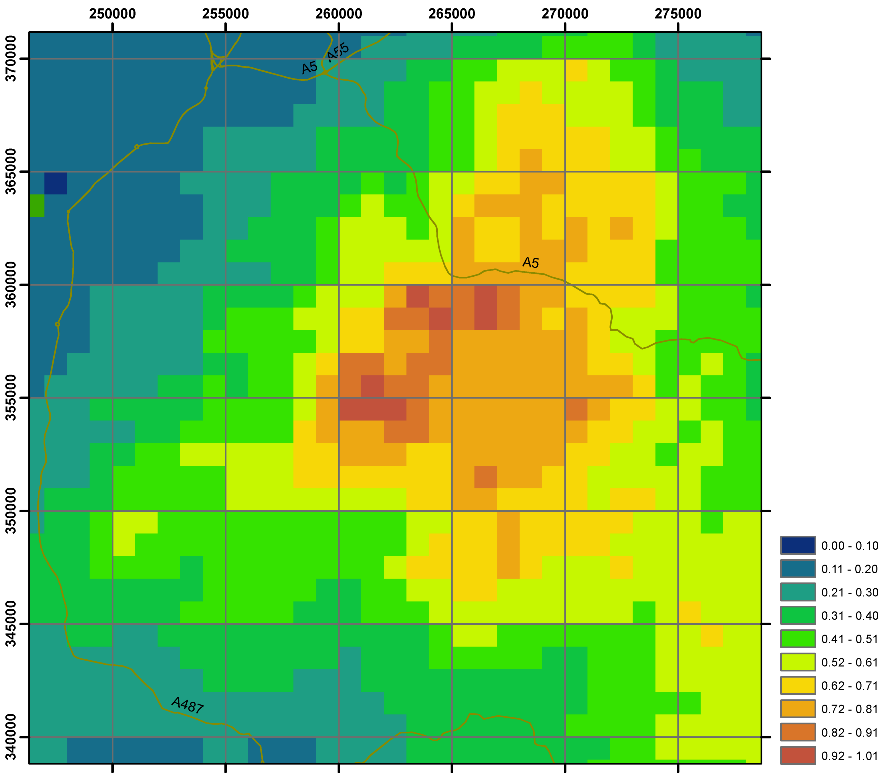

Fig. 4a. NO_y wet deposition in Snowdonia (keq Ha^{-1}) with 1 km grid resolution.

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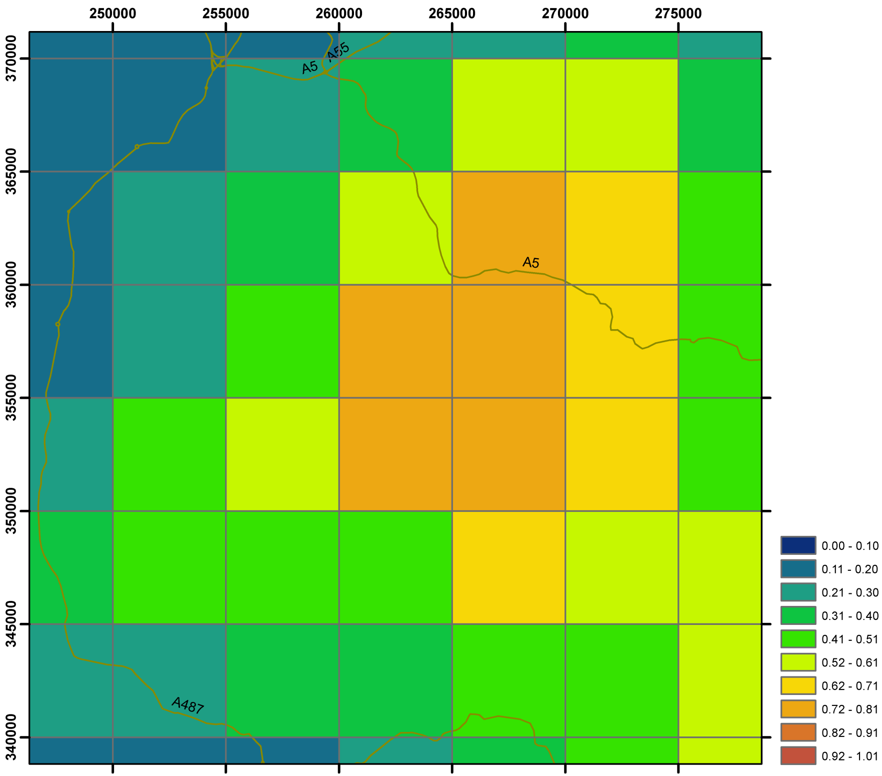

Fig. 4b. NO_y wet deposition in Snowdonia (keq Ha^{-1}) with 5 km grid resolution.

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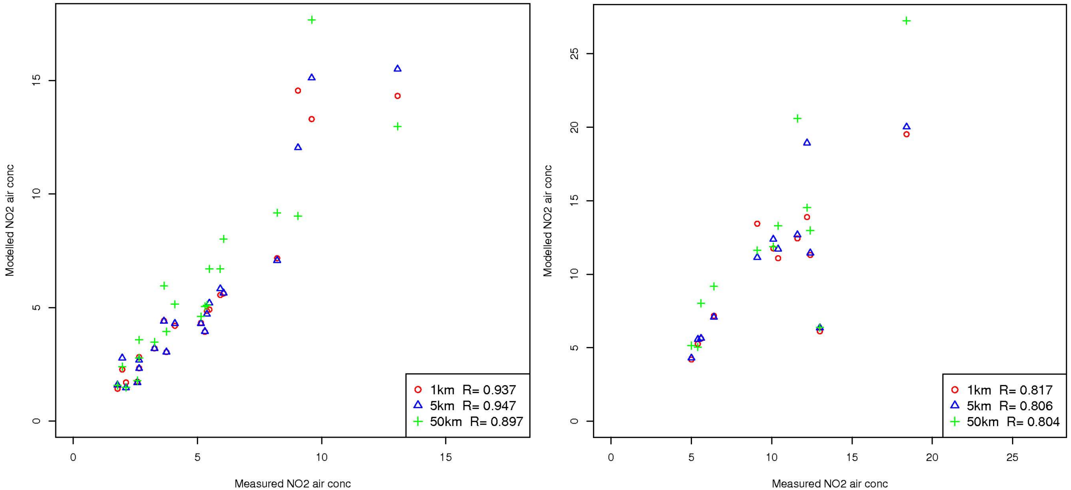

Fig. 5. Correlation with measurements of NO_2 ($\mu g\, m^{-3}$) from the rural monitoring network (left) and rural sites in the Automatic Urban and Rural monitoring Network (right) for 1 km, 5 km and 50 km model resolution data.

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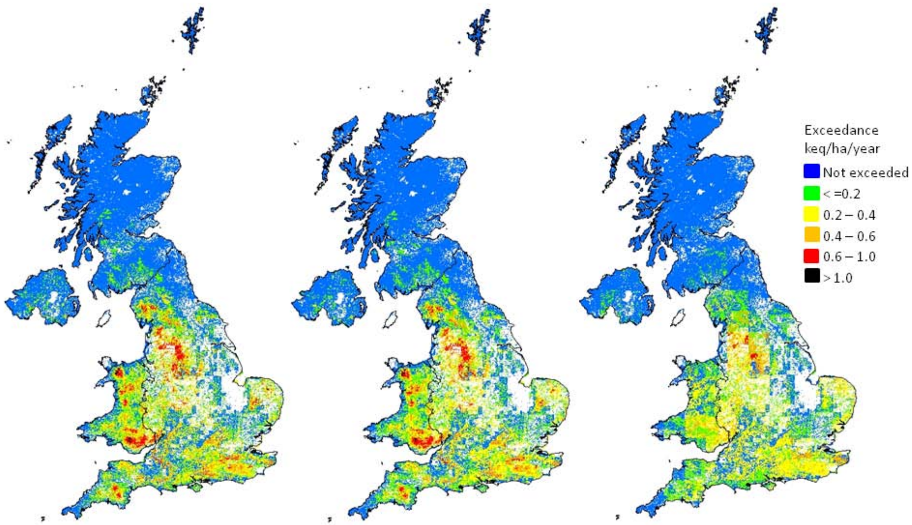

Fig. 6a. Exceedance of 5th percentile nutrient nitrogen critical loads for the UK using modelled deposition data with resolutions of: 1 km (left); 5 km (centre); 50 km (right).

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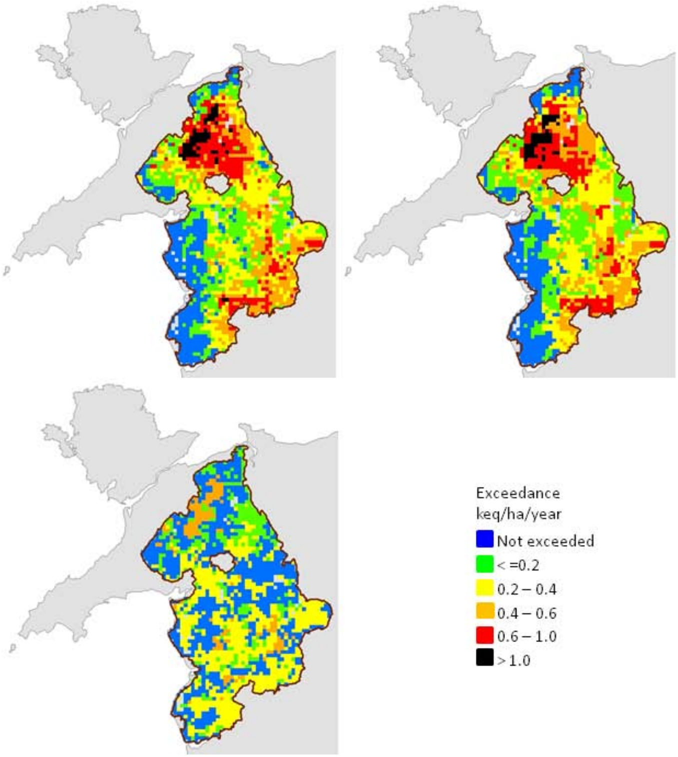

Fig. 6b. Exceedance of 5th percentile nutrient nitrogen critical loads for Snowdonia using model deposition data with resolutions of: 1 km (upper left); 5 km (upper right); 50 km (lower left).

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