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# **Heterogeneity of atmospheric ammonia at the landscape scale and consequences for environmental impact assessment**

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## **Abstract**

We examined the consequences of the spatial heterogeneity of atmospheric ammonia (NH<sub>3</sub>) by measuring and modelling NH<sub>3</sub> concentrations and deposition at 25 m grid resolution for a rural landscape containing intensive poultry farming, agricultural grassland, woodland and moorland. The emission pattern gave rise to a high spatial variability of modelled mean annual NH<sub>3</sub> concentrations and dry deposition. Largest impacts were predicted for woodland patches located within the agricultural area, while larger moorland areas were at low risk, due to atmospheric dispersion, prevailing wind direction and low NH<sub>3</sub> background. These high resolution spatial details are lost in national scale estimates at 1 km resolution due to less detailed emission input maps. The results demonstrate how the spatial arrangement of sources and sinks is critical to defining the NH<sub>3</sub> risk to semi-natural ecosystems. These spatial relationships provide the foundation for local spatial planning approaches to reduce environmental impacts of atmospheric NH<sub>3</sub>.

## **Capsule:**

Fine scale resolution modelling to reproduce the spatial heterogeneity of atmospheric NH<sub>3</sub> concentrations and deposition is critical for NH<sub>3</sub> risk assessment on sensitive ecosystems.

## **Highlights:**

- Local farm inventory provided field-level emissions for high resolution modelling
- Model-derived concentrations were compared against intensive spatial measurements
- Spatial arrangement of NH<sub>3</sub> sources and sinks is critical to environmental impact
- Average national emission factors were not appropriate for an NH<sub>3</sub> risk assessment
- Modelling at 1 km resolution did not capture the full spatial variability of NH<sub>3</sub>

**Keywords:** *ammonia, critical level, landscape scale, dispersion modelling, spatial planning*

## 1 Introduction

Most atmospheric ammonia ( $\text{NH}_3$ ) originates from agricultural activities (Misselbrook et al., 2000; Van der Hoek, 1998). Intensive livestock farming, i.e. large pig and poultry units, represent substantial  $\text{NH}_3$  point sources, due to their high stocking density. Other  $\text{NH}_3$  sources include biomass burning, fuel combustion and industrial processes such as the production of nitrogen (N) fertilisers (Bouwman et al., 1997). High atmospheric  $\text{NH}_3$  concentrations are directly toxic to plants through stomatal uptake as soon as the uptake exceeds the detoxification capacity (Fangmeier et al., 1994). Ammonia deposition (and deposition of other forms of reactive N) can lead to eutrophication and acidification of sensitive ecosystems, causing changes in biodiversity (Cape et al., 2009b; Cellier et al., 2009; Krupa, 2003; Pitcairn et al., 2009). Studies have been conducted to quantify the effect of  $\text{NH}_3$  emission sources on surrounding ecosystems, e.g. Fowler et al. (1998) quantified concentrations and deposition fluxes within 300 m of a poultry farm in Scotland using measurements and deposition modelling, showing rapidly decreasing concentrations with distance from the source. Pitcairn et al. (1998; 2002) analysed the impact of such deposition fluxes on woodland flora and Frati et al. (2007) studied the effect of pig farm emissions on sensitive vegetation (lichens). Sutton et al. (1998) compared deposition estimates based on different scales, ranging from field to landscape to national scale and concluded that, due to the spatial variability of  $\text{NH}_3$ , the quality of an environmental impact assessment is dependent on the spatial resolution of the deposition data used. Dragosits et al. (2002) provide a more detailed analysis of the landscape study in Sutton et al. (1998): Emission, transport and deposition were modelled within a 5 km x 5 km landscape in England at a 50 m grid resolution; however, no  $\text{NH}_3$  measurements were made to verify the estimates. Theobald et al. (2001) and Dragosits et al. (2006) focused on strategies to reduce the effect of emission hotspots on ecosystems by locating tree belts around the sources, indicating the importance of relative spatial location of sources and sinks, and assessed possible landscape planning measures to decrease potential effects on sensitive habitats.

As an approach to assess the risk of environmental impacts by air pollutants, the United Nations Economic Commission for Europe (UNECE) has developed critical thresholds of pollutant concentrations and deposition fluxes: Critical Levels (CLEs) and Critical Loads (CLs). A CLE is a pollutant concentration in the atmosphere above which plants or ecosystems may be directly negatively affected (Posthumus, 1988). Recently, long term CLEs of  $\text{NH}_3$  were reviewed and new, lower values proposed and adopted by the UNECE (Cape et al., 2009a; Sutton et al., 2009a; UNECE, 2007):  $1 \mu\text{g NH}_3 \text{ m}^{-3}$  for the most sensitive ecosystems, i.e. where lichens and bryophytes are part of the ecosystem

integrity, and  $3 \pm 1 \mu\text{g NH}_3 \text{ m}^{-3}$  for higher plants in other semi-natural ecosystems. A CL is a pollutant deposition below which no significant harmful effects on the environment are expected to occur according to current knowledge (Posthumus, 1988). Nitrogen (N) CLs have been defined for specific ecosystem types (see UNECE, 2010 for most up-to-date CLs). In contrast to the CLE approach, which is specifically defined for gases such as  $\text{NH}_3$ , the CL integrates all forms of reactive N and therefore requires estimates of total N deposition. According to Sutton et al. (2009b) these N deposition estimates are inherently more uncertain, and for assessing the environmental risk imposed by  $\text{NH}_3$ , it is much easier to measure  $\text{NH}_3$  concentrations and examine exceedance of the CLE than to verify CL exceedances by measurement. However, until the recent revision of CLEs, exceedance of CLs has been more commonly used for impact assessments of atmospheric N. For atmospheric  $\text{NH}_3$ , this may reflect that previous long-term  $\text{NH}_3$  CLEs were set at much less precautionary level than associated values of N CLs (e.g. Burkhardt et al., 1998), which was one reason for the revision of new long term  $\text{NH}_3$  CLEs (Sutton et al., 2009b).

For assessing the environmental impact of  $\text{NH}_3$  concentrations and deposition by modelling, it is essential to estimate  $\text{NH}_3$  emissions accurately (Dragosits et al., 2002; Hellsten et al., 2008). Hallsworth et al. (2010) highlighted the problem of modelling  $\text{NH}_3$  dispersion at relatively coarse scales, such as 5 km resolution, due to the high spatial variability of  $\text{NH}_3$  emissions and showed that 5 km modelling underestimated the impact of  $\text{NH}_3$  concentrations on semi-natural areas close to intensive agricultural areas. However, at UK national scale, standard assessments of the impact of N deposition are based on 5 km resolution modelling (Dore et al., 2007; Matejko et al., 2009). Dore et al. (2012) compared CL exceedances in the UK using data at 1 km and 5 km resolution. In contrast to results of Hallsworth et al. (2010) for CLEs, CL exceedances were not highly sensitive to grid resolution. This was attributed to the contribution of N wet deposition (which shows less local variability than dry deposition) and that all ecosystem types were included (not only nature reserves protected under the Habitat Directive, as in Hallsworth et al., 2010). However, for an individual nature reserve located a few kilometres from a major road, the standard 5 km grid data were inadequate to accurately assess local N deposition (Dore et al., 2012).

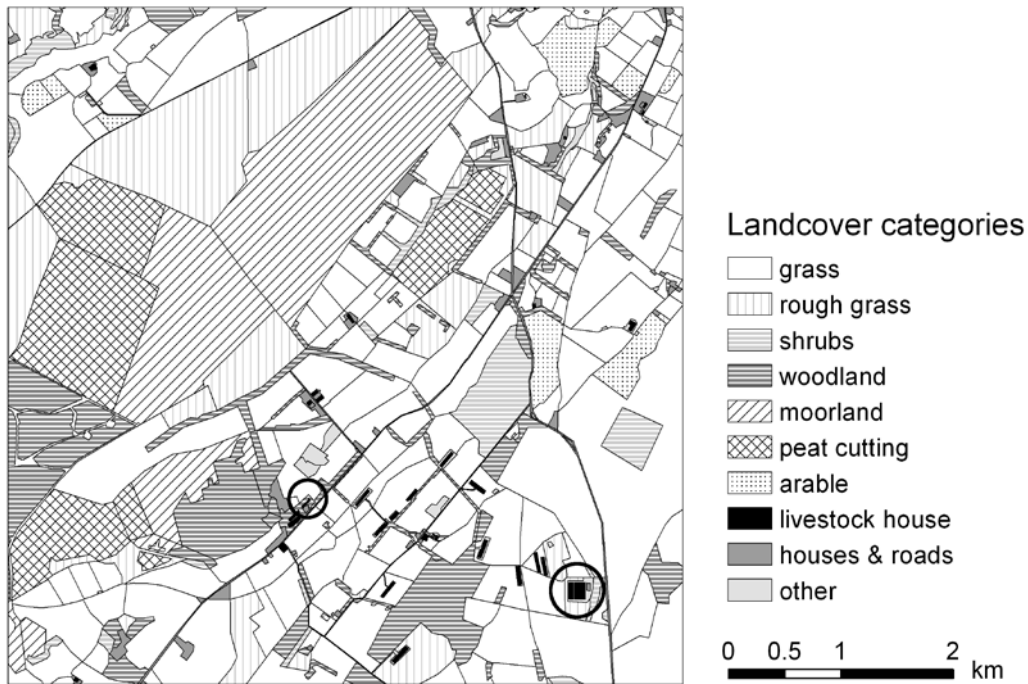
This study provides a contribution to the landscape scale analysis conducted across Europe within the NitroEurope Integrated Project (NEU) (Sutton et al., 2007), in which a landscape is defined as a spatially heterogeneous area covering several square kilometres and contains interacting ecosystems (Forman and Godron, 1981). In rural landscapes, anthropogenic processes in the form of farm management determine to a large extent N dynamics and much of its environmental impact within the landscape

(Cellier et al., 2011). The NEU landscape analysis aimed to quantify N flows at the landscape scale using measurement and modelling approaches. In this study, we analysed NH<sub>3</sub> dispersion and its environmental impact in a 6 km x 6 km rural landscape in southern Scotland. The landscape has a diverse emission pattern with a large number of NH<sub>3</sub> emission hotspots, and large areas of sensitive ecosystems as potential sinks. A detailed landscape inventory of all farms and fields at field-level resolution was conducted to coincide with an intensive spatial monitoring programme of NH<sub>3</sub> concentrations. Ammonia dispersion and deposition was modelled at a 25 m resolution, and the environmental impact of the local NH<sub>3</sub> sources assessed and compared with national 1 km resolution estimates (Hallsworth et al., 2010). The results have general implications for the sustainable management of landscapes that combine both intensive livestock agriculture and ecosystems of relevance for environmental protection.

## **2 Site and methods**

### *2.1 Study area*

The study landscape is situated in southern Scotland, which has a temperate climate, with an annual mean temperature of ~8°C, a typical rainfall of ~1000 mm and predominant southwesterly winds. The 6 km x 6 km area (Figure 1) is dominated by agricultural grassland (48%), followed by moorland (21%), rough grass (13%) and woodland (10%). The moorland area with low NH<sub>3</sub> emissions is in the northwestern part of the landscape and is partially grazed by sheep at very low stocking densities, partly legally protected as a Site of Special Scientific Interest (SSSI), with another part undergoing peat cutting. The southeastern part is dominated by agricultural land, such as sheep and beef cattle pastures interspersed with poultry farming houses containing nearly 1.5 million laying hens. Most of the layers are kept in cage systems with manure removal by belt systems two to three times per week (farm locations circled in black in Figure 1, with other livestock houses shaded in black). However, a number of the houses have deep-pit systems, and in most of them layers are managed as free range birds.



**Figure 1: Land cover types in the 6 km x 6 km study landscape in southern Scotland. The circles highlight poultry houses where manure is cleared at least twice per week. At other animal houses manure is removed less frequently (see section 3.3).**

## 2.2 Landscape inventory and emissions

Detailed land cover/land use and farm activity data were obtained by a local survey carried out by Scotland's Rural College (SRUC) and the Centre for Ecology & Hydrology (CEH). Management activities were recorded for each farm building and agricultural field throughout 2008, including type and numbers of livestock housed and grazed, manure management, ventilation type and emission height, crop type and the application of mineral and organic fertiliser. Land cover/land use and farm activity data were processed with a Geographical Information System (ESRI, ArcGIS) and emissions calculated for each individual field and livestock house. Field emissions were calculated by applying UK average emission factors (EFs) of the UK NH<sub>3</sub> emission inventory to applications of mineral and organic fertiliser and to excreta of grazing livestock (Misselbrook et al., 2009), using fertiliser application rates provided by the farmers. Typical N contents were applied to the different types of organic fertiliser (Defra, 2010). Grazing excreta were calculated using grazing records and daily N excretion rates of the specific type of animal (Misselbrook et al., 2009). Similarly, average NH<sub>3</sub> EFs of the UK emission inventory were applied to calculate housing and manure storage emissions, with housing records on systems and durations provided by the farmers. However, after initial analyses, housing EFs were partly adjusted to account for specific local management practices (see Section

3.3). All spatial datasets were converted to a 25 m grid resolution for atmospheric dispersion modelling (see Section 2.4).

### 2.3 *Spatial NH<sub>3</sub> concentration measurements*

Monthly average concentrations were measured from April 2007 to December 2008 at 31 locations using ALPHA passive diffusion samplers (Tang et al., 2001) at a sampling height of 1.5 m above ground. Measurement locations were distributed across the study area in collaboration with farmers and landowners in the landscape. Sites were selected to cover NH<sub>3</sub> concentrations over different land cover types and farms. More sites were placed in NH<sub>3</sub> emitting areas to capture concentration gradients around emission hotspots and diffuse sources, taking the main wind direction into account. The nearest site to an emission hotspot was located 70 m downwind of a poultry house to avoid saturation of the samplers. To assess measurement precision and uncertainty, samplers were exposed in triplicate at each location. The sampling rate of the ALPHA samplers was calibrated against the DELTA denuder reference system (Sutton et al., 2001b), using the UK National Ammonia Monitoring Network methodology (NAMN, Sutton et al., 2001a). ALPHA samplers were stored in a cold room (4°C) until analysis in the laboratory with an AMFIA NH<sub>3</sub> flow injection analyser, based on analysis by selective ion membrane transfer and subsequent conductivity measurement (Wyers et al., 1993).

### 2.4 *Atmospheric dispersion and deposition modelling*

Atmospheric dispersion and dry deposition of NH<sub>3</sub> within the study landscape was simulated using the LADD (Local Area Dispersion and Deposition) model (Hill, 1998). Loubet et al. (2009) recently reviewed LADD and other models available for simulating NH<sub>3</sub> dispersion. The advantages of LADD are that it operates in 3D (with the atmosphere represented by 44 vertical layers), is computationally fast, and accounts for land cover-specific dispersion and deposition characteristics (Loubet et al., 2009). Input data include land cover and emission data for each grid square (see Section 2.2), wind statistics and NH<sub>3</sub> concentrations at the domain boundaries. Suitable roughness length ( $z_0$ ) and canopy resistance ( $R_c$ ) for each given land cover type were selected and assigned in LADD. The roughness length is used to calculate vertical dispersion and dry deposition rates, while  $R_c$  is used in the calculation of dry deposition velocities within each grid square. Wind statistics were calculated from data collected for 30-minute-intervals during 2008 at a continuous measurement site near the centre of the study area (M. Coyle, CEH, pers. comm. 2010; n.b. exact coordinates not presented for farm data confidentiality purposes). The influence of emission sources outside of the modelling domain was incorporated by setting the atmospheric concentrations for the 44 model layers at the four domain boundaries to values taken from the national FRAME (Fine Resolution Atmospheric Multi-



pollutant Exchange) model run for 2008 at a 5 km x 5 km resolution (Dore et al., 2007). These boundary concentrations were highest at ground level, ranging from 1.34  $\mu\text{g NH}_3 \text{ m}^{-3}$  at the eastern boundary to 1.85  $\mu\text{g NH}_3 \text{ m}^{-3}$  in the south. The FRAME simulations used annual precipitation data for 2008 from the UK Met Office precipitation monitoring network and wind direction frequency data generated from radiosonde data for 2008.

LADD was applied for the year 2008 at 25 m x 25 m grid resolution over an area of 7 km x 7 km with the model domain extended by 500 m on all sides to limit possible edge effects. Annual average  $\text{NH}_3$  concentrations at 1.5 m height above ground level and dry deposition were simulated and subsequently analysed with ArcGIS (ESRI).

## 2.5 Assessment of model performance

Model performance was assessed by comparing modelled with measured annual concentrations at the 31 sampling sites. Statistical metrics used for model evaluation were the fraction of modelled concentrations within a factor of two of observed concentrations (FAC2), the correlation coefficient (R), the geometric mean bias (MG) and the geometric variance (VG) (Chang and Hanna, 2004; Theobald et al., 2009).

$$\text{FAC2} = \text{fraction of data that satisfy } 0.5 \leq C_m/C_o \leq 2.0 \quad (1)$$

$$R = \frac{(\overline{C_o - C_o})(\overline{C_m - C_m})}{\sigma_{C_o} \sigma_{C_m}} \quad (2)$$

$$MG = \exp(\overline{\ln C_o} - \overline{\ln C_m}) \quad (3)$$

$$VG = \exp\left[\overline{(\ln C_o - \ln C_m)^2}\right] \quad (4)$$

where  $C_o$  are the observed (measured) concentrations,  $C_m$  are modelled concentrations,  $\sigma$  is the standard deviation and overlined variables represent mean values. Model performance is considered “acceptable” if FAC2 is 50% or greater, i.e. if  $\text{FAC2} \geq 0.5$ . MG measures the mean relative bias and only indicates systematic errors. It represents the ratio of the geometric mean of  $C_o$  to the geometric mean of  $C_m$ , thus the optimum value is  $MG = 1$ . An “acceptable” model performance is expected to result in a mean relative bias within  $\pm 30\%$ , i.e.  $0.7 < MG < 1.3$ . VG is a measure of mean relative scatter of a log-normal distribution and reflects both systematic and random error. The optimum value is  $VG = 1$ . An “acceptable” model would be expected to have a relative scatter of less than a factor of two (i.e.  $VG < 1.6$ ) or three (i.e.  $VG < 3.3$ ). Overall model performance is evaluated as acceptable when more than 50% of the criteria are met (Hanna and Chang, 2010).

## 2.6 Assessment of potential environmental impacts

Landscape areas with exceeded CLEs and CLs were identified to assess the risk of environmental impact on ecosystems. Analyses of CLE exceedance used modelled NH<sub>3</sub> concentrations at a height of 1.5 m above ground. CL exceedance calculations were based on total N deposition: the LADD estimate of dry NH<sub>3</sub> deposition (see Section 2.4) plus the wet deposition of reduced N and the dry and wet deposition of oxidised N calculated using the UK FRAME model run for 2008 at a 1 km x 1 km resolution. The contribution of particulate ammonium (NH<sub>4</sub><sup>+</sup>) to the dry deposition of reduced N is considered minor compared with NH<sub>3</sub> (e.g. Asman et al., 1998; Duyzer, 1994). Hallsworth et al. (2010) validated the FRAME model at a 1 km resolution for NH<sub>3</sub> concentrations and Dore et al. (2012) for NO<sub>2</sub> concentrations. Dore et al. (2007) validated the FRAME model for aerosol concentrations and wet deposition at 5 km resolution. FRAME gives three different deposition rates for each grid square: a) the average deposition, accounting for land cover mix in the grid square; b) the deposition to woodland in the square; c) the deposition to low semi-natural vegetation in the square. Deposition rates were applied depending on the land cover in each 25 m grid square. The CL exceedance was calculated for woodland, hedgerows, shrubs, moorland and rough grass by subtracting the CL of 10 kg N ha<sup>-1</sup> yr<sup>-1</sup> from the total N deposition. The applied CL is the lower limit of the range shown in Table 1 to protect the most sensitive species of the respective ecosystems. Although these CL values were used in this study, current science indicates that they may need to be revised further to adequately protect sensitive species (Payne et al., 2013).

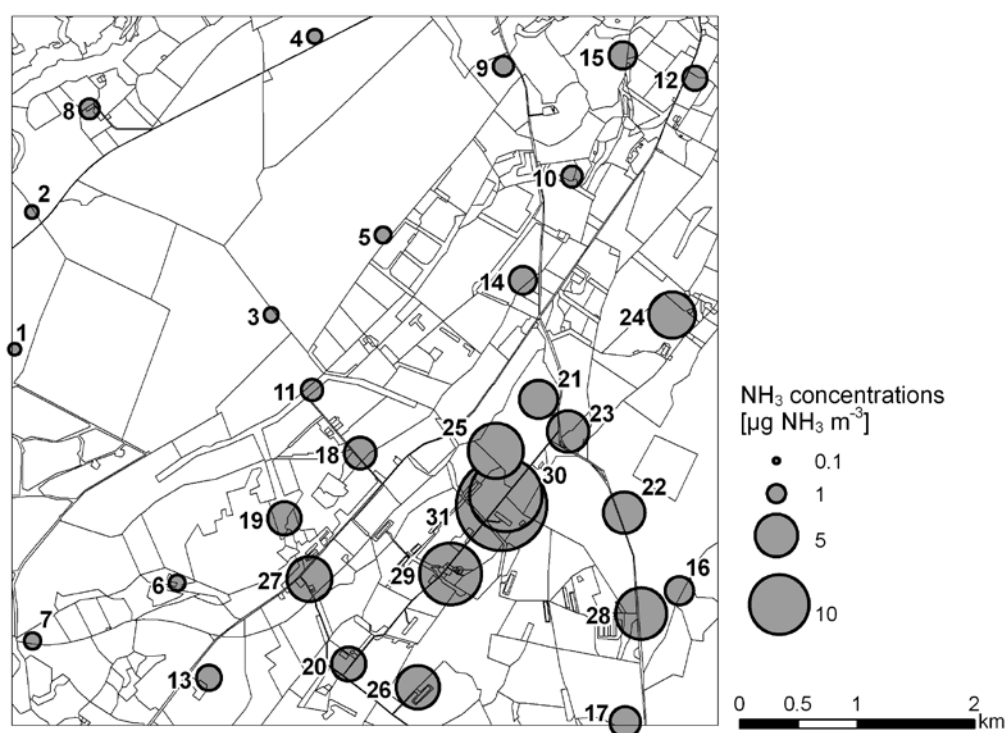
**Table 1: Land cover categories of the study landscape, the associated ecosystem types with the corresponding critical loads for N deposition (UNECE, 2010).**

Land cover category	Ecosystem type	Critical Load [kg N ha <sup>-1</sup> yr <sup>-1</sup> ]
Woodland, hedgerows	Broadleaved deciduous woodland	10-20
Shrubs	<i>Calluna</i> dominated wet heath (upland moorland)	10-20
Moorland, rough grass	Heath ( <i>Juncus</i> ) meadows and humid ( <i>Nardus stricta</i> ) swards	10-20

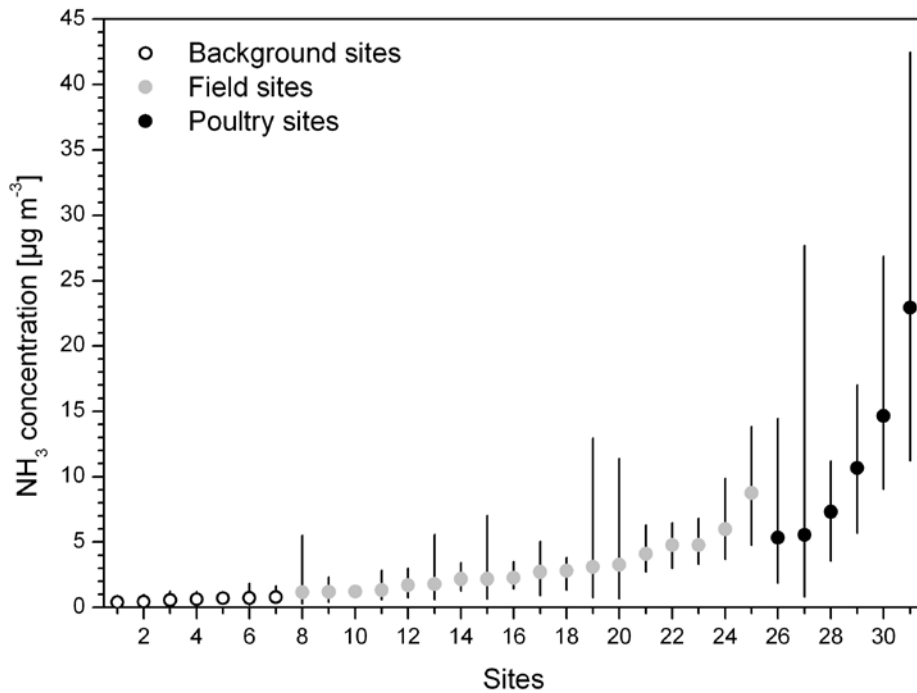
### 3 Results and discussion

#### 3.1 Spatial variability of measured $\text{NH}_3$ concentrations

The spatial variability of  $\text{NH}_3$  concentrations in the landscape was large, with monthly  $\text{NH}_3$  concentrations during 2008 varying from 0.2 to 42.5  $\mu\text{g m}^{-3}$  between the measurement sites. Monthly coefficients of variation of replicate samplers was <24%, with values >15% only occurring at sites with monthly mean concentrations <1  $\mu\text{g m}^{-3}$ . The spatial variability of the measured  $\text{NH}_3$  concentrations is attributed to land use, as shown by classifying the sites into three categories: a) “Background sites” are located away from agricultural  $\text{NH}_3$  sources and have mean annual concentrations of <1  $\mu\text{g m}^{-3}$ , b) “Field sites” are influenced by agricultural  $\text{NH}_3$  sources such as grazing or fertiliser applications, but are >300 m away from large point sources, and c) “Poultry sites” within 300 m of large point sources, i.e. the poultry houses. Annual mean  $\text{NH}_3$  concentrations in 2008 ranged between 0.40 and 22.9  $\mu\text{g NH}_3 \text{m}^{-3}$  (Figure 2) and generally increased from Background to Field to Poultry sites (Figure 3). Two Field sites were exceptions with higher concentrations: Site 24 was close to an open cattle shed and an intensively used field, and site 25, which was only 320 m from a poultry house, i.e. just outside the distance for Poultry site classification.

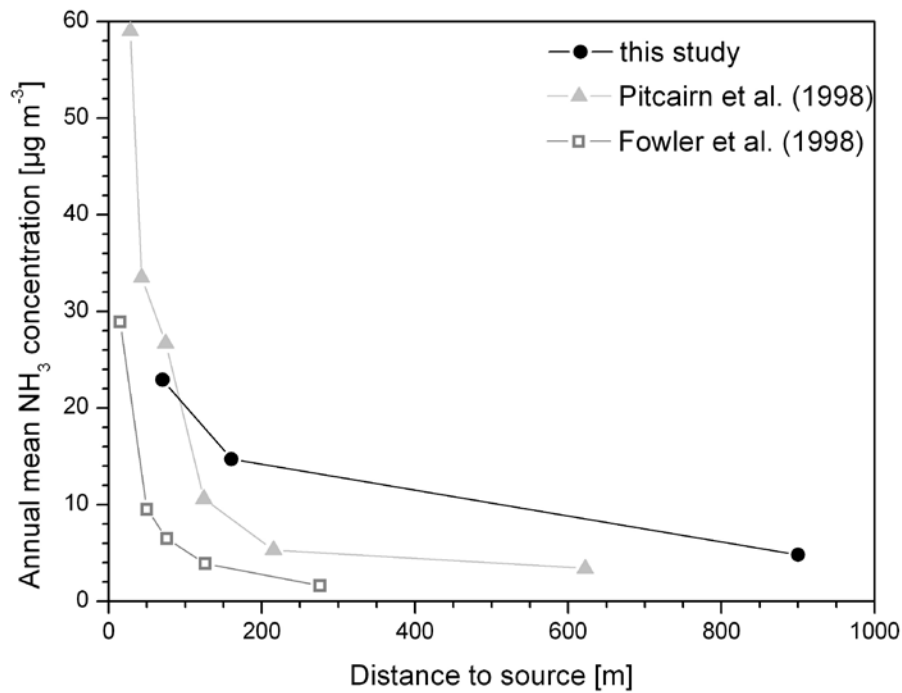


**Figure 2:** Map of numbered measurement site locations showing annual mean  $\text{NH}_3$  concentrations by proportionally sized circles. The sites are numbered in rank order of their mean  $\text{NH}_3$  concentration within their site category.



**Figure 3: Annual measured mean NH<sub>3</sub> concentrations and monthly minima and maxima in 2008 for Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles). The site numbers ranked by site category and mean concentration are those shown in Figure 2.**

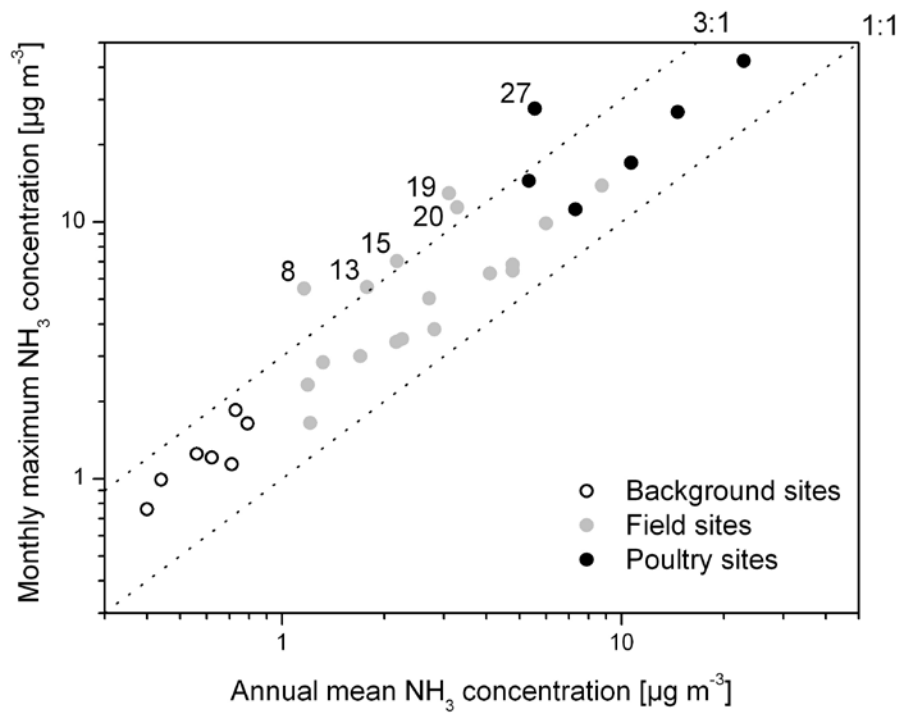
The highest annual mean NH<sub>3</sub> concentrations were measured 70 m downwind (northeast) of a poultry house with an estimated NH<sub>3</sub> emission strength of 5,900 kg N yr<sup>-1</sup> (site 31). A measurement transect of three sites downwind of this house illustrates the concentration gradient with distance from large sources. Measured annual concentrations were 22.9 µg m<sup>-3</sup>, 14.7 µg m<sup>-3</sup> and 4.8 µg m<sup>-3</sup> at distances of 70 m, 160 m and 900 m from the house (sites 31, 30, 23, respectively). Figure 4 compares these results to concentration decreases with distance found by Fowler et al. (1998) and Pitcairn et al. (1998) for poultry houses emitting an estimated 4,800 kg N yr<sup>-1</sup> and 14,000 kg N yr<sup>-1</sup>, respectively. All three studies were conducted in agricultural areas; however, the concentration decrease with distance in this study is more gradual, possibly due to high concentrations in the surrounding area resulting from the large number of emission hotspots.



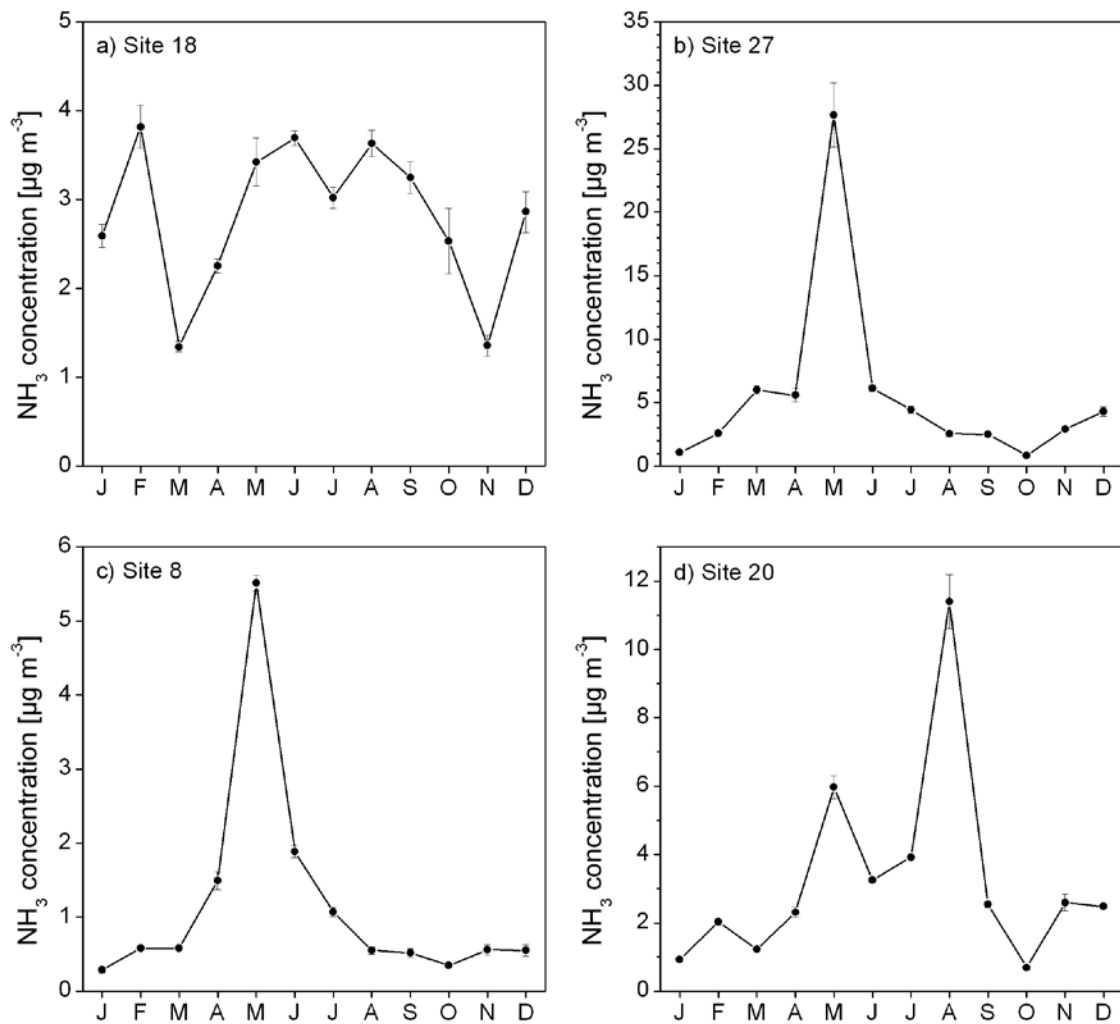
**Figure 4: Ammonia concentration decrease with distance from the source of this study (sites 31, 30, 23) compared with results of Pitcairn et al. (1998) and Fowler et al. (1998).**

### 3.2 Temporal variability in measured NH<sub>3</sub> concentrations

A strong correlation was found between annual mean NH<sub>3</sub> concentrations of all sites in 2007 and 2008 ( $R^2 = 0.98$ ,  $n = 31$ ), using data from April to December for both years. This strong correlation indicates that the surrounding land use is the main driver of variation in the annual concentration. The ratio of monthly concentration maxima to annual mean concentrations can be used as an indicator of temporal variability on an intra-annual basis (Figure 5). Most sites show a ratio below 3:1, which seems to represent a typical temporal variation about a mean of a relatively constant NH<sub>3</sub> concentration (e.g. Figure 6a). This was also shown by Tang et al. (2009) for the UK at the national scale. Monthly maximum concentrations of those sites with larger ratios (up to 5:1) occurred in spring or summer 2008. For example, site 27 (Figure 6b) was located around 200 m south of four poultry houses, but it was also located close to a field where manure was applied in May 2008. Manure heaps and manure applications also accounted for monthly maxima at sites 8 (Figure 6c), 13, 15, 19 and 20 (Figure 6d).



**Figure 5: Relationship between monthly maximum and annual mean NH<sub>3</sub> concentrations in 2008 for Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles). Site numbers are shown for sites with ratios of the monthly maximum to the annual mean higher than 3:1.**



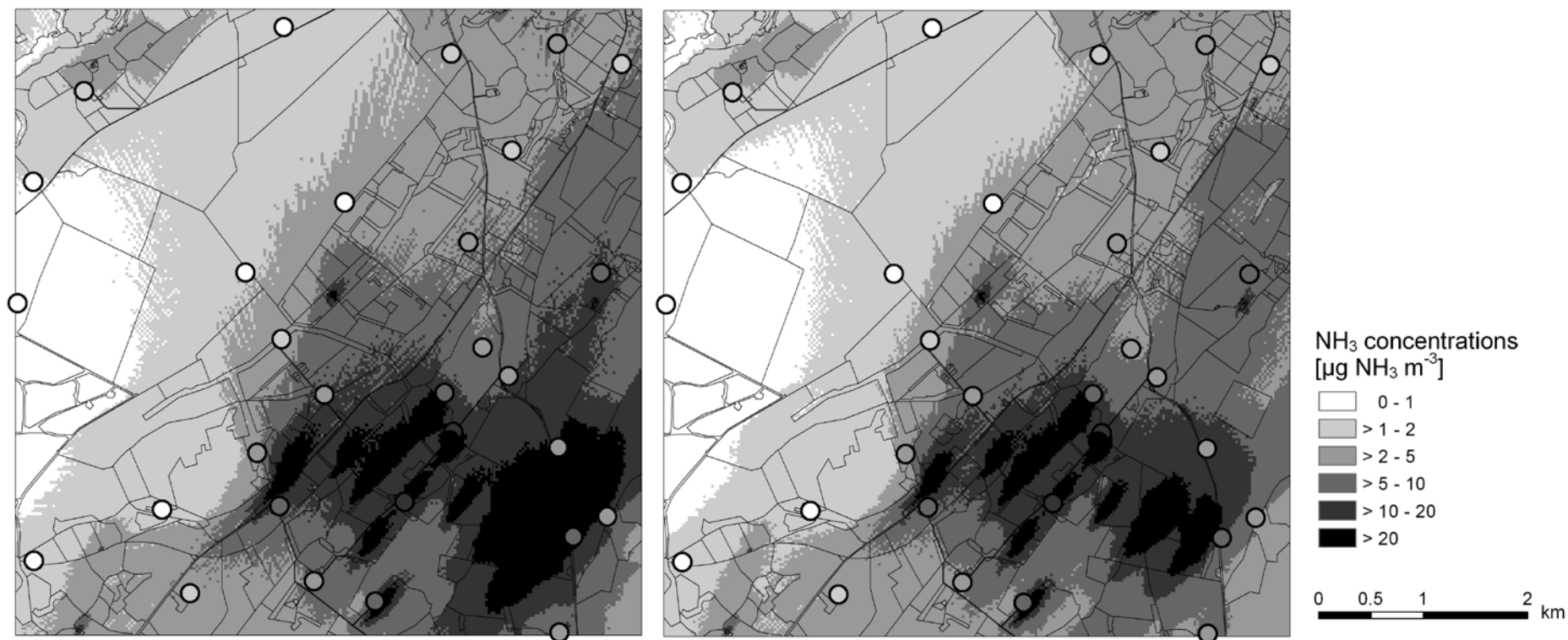
**Figure 6: Temporal variation of monthly  $\text{NH}_3$  concentrations ( $\pm 2$  standard deviations) during 2008 at four sites: a) Site 18 with a ratio of max/mean below 3:1 and b), c) and d) showing sites with ratios higher than 3:1.**

### 3.3 Modelled atmospheric concentration and deposition

The LADD model was initially run using emission factors (EFs) from the UK inventory of  $\text{NH}_3$  emissions and resulted in the general pattern of  $\text{NH}_3$  concentrations being reproduced (Figure 7, left). However there was a significant overestimation of concentration in the landscape, especially in the southeastern quarter. This overestimation was attributed to the emissions from six of the poultry houses (circled houses in Figure 1) which contained  $\sim \frac{3}{4}$  million layers in cage systems. These houses had frequently cleaned belt-systems ( $\geq 2$  times  $\text{week}^{-1}$ ). The EF for a UK average caged layer is calculated assuming that 40% of these birds are housed in deep-pit houses and 60% in belt-system houses with a cleaning frequency of  $\leq 1$  times  $\text{week}^{-1}$  (Misselbrook et al., 2009). Belt-

systems with less frequent cleaning ( $EF = 0.092 \text{ kg NH}_3\text{-N bird}^{-1} \text{ yr}^{-1}$ ) are considered to reduce emissions by 56% compared to deep-pit systems ( $EF = 0.164 \text{ kg NH}_3\text{-N bird}^{-1} \text{ yr}^{-1}$ ), resulting in an average UK caged layer EF of  $0.121 \text{ kg NH}_3\text{-N bird}^{-1} \text{ yr}^{-1}$  (Misselbrook et al., 2009). The European Commission (2003) reported an EF of  $0.029 \text{ kg NH}_3\text{-N bird}^{-1} \text{ yr}^{-1}$  for frequently cleaned belt-systems, more than four times lower than that used in the UK  $\text{NH}_3$  inventory for the average caged layer. LADD runs were repeated using this EF for the six poultry houses concerned and modelled concentrations decreased considerably (Figure 7, right), and matched measured concentrations more closely.

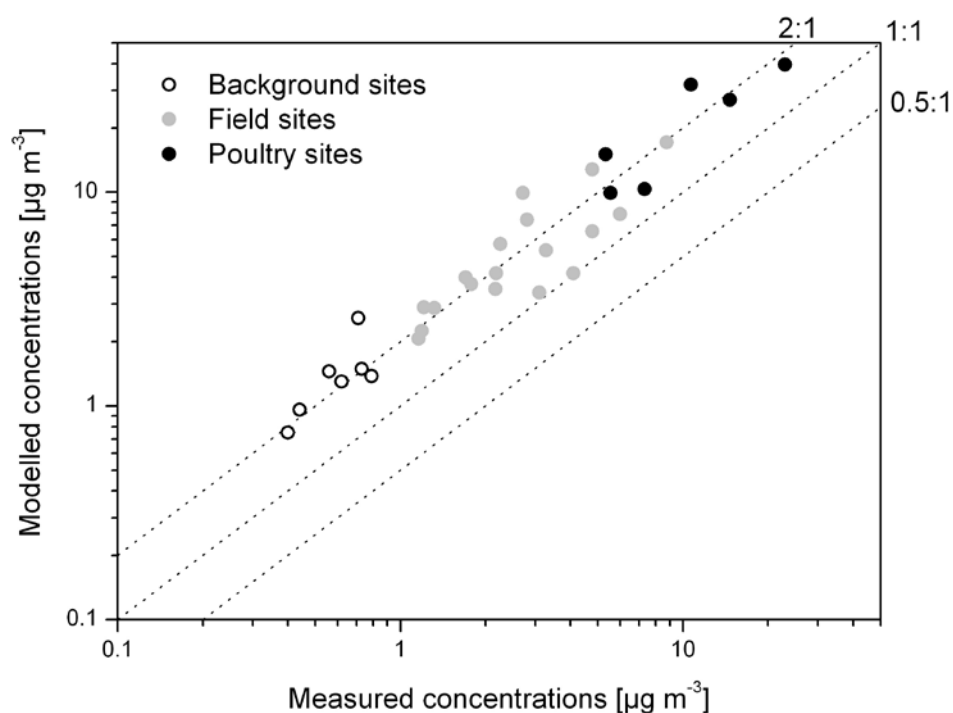




**Figure 7: Measured (circles) and modelled (background colours)  $\text{NH}_3$  concentrations within the landscape. Left map: UK inventory emission factors were applied to all  $\text{NH}_3$  sources; Right map: the European Commission (2003) EF for frequently cleaned belt-systems was applied to the six poultry houses that had that system.**

Figure 8 shows a scatter plot between modelled and measured concentrations and Table 2 summarises the statistical metrics for the model run with system specific EFs applied for the poultry houses instead of UK average EFs. Overall, model performance is evaluated as acceptable, as the FAC2 and VG metrics indicate acceptable model performance when compared with measurements at all sites. However, the MG is lower than recommended for acceptable model performance, reflecting a systematic overestimation by the model, which is apparent at all distances from sources (Figure 8).

Recent work by Theobald et al. (2012) suggests that LADD overestimates concentrations around elevated sources (> 5 m) with high exit velocities, as LADD does not include treatment of plume rise after leaving the source. However, poultry houses in this study area predominantly have emission heights of 4 to 5 m, and most vents are located on the building walls, i.e. most plumes are not expected to exit vertically. Thus this is unlikely to explain the differences shown in Figure 8. For other situations with ground and building emission sources, Theobald et al. (2012) reported acceptable agreement between LADD and measured concentrations.



**Figure 8: Relationship between modelled and measured NH<sub>3</sub> concentrations of Background sites (open circles), Field sites (grey circles) and Poultry sites (black circles) on logarithmic axes.**

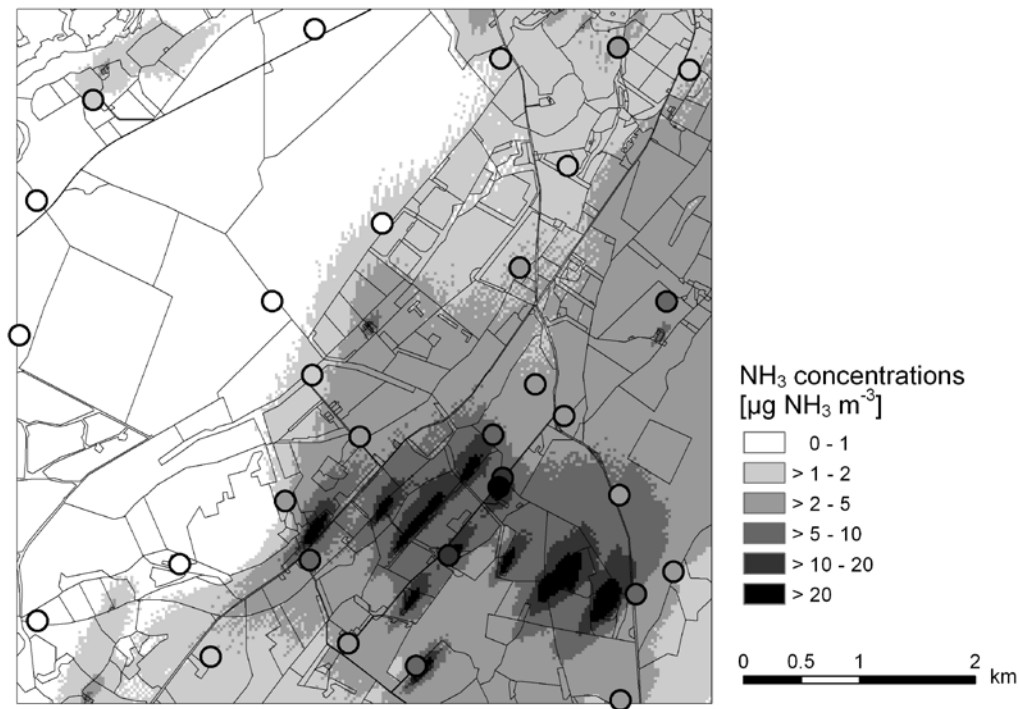
**Table 2: Statistical metrics of model performance comparing measured and modelled NH<sub>3</sub> concentrations for all sites and by separate site categories (see Section 3.1 for category definition).**

	Target performance	All sites	Background sites	Field sites	Poultry sites
FAC2 (%)	≥ 50.0	51.6	28.6	55.6	66.7
R	-	0.95	0.64	0.84	0.89
MG	0.7 – 1.3	0.50	0.45	0.52	0.50
VG	< 3.3	1.77	2.03	1.68	1.76

### 3.4 Model calibration

In order to use modelled concentrations and deposition fluxes for risk assessment of environmental impacts, the systematic overestimation was addressed by calibrating the modelled against the measured concentrations. Modelled concentrations were corrected by the slope of the regression between measured and modelled results ( $[\text{NH}_3]_{\text{meas}} = 0.49 [\text{NH}_3]_{\text{model}} + 0.15$ ,  $R^2 = 0.90$ ). The intercept was not statistically significant, providing the simplified relationship  $[\text{NH}_3]_{\text{meas}} = 0.49 [\text{NH}_3]_{\text{model}}$  ( $R^2 = 0.90$ ) to calibrate modelled concentrations (effectively represented by the 2:1 function in Figure 8).

The calibrated model NH<sub>3</sub> concentrations range from 0.3 to 77.9 µg m<sup>-3</sup> within the study landscape (Figure 9). These results provide the basis to use the model for assessing the risk of environmental impacts in the study landscape at high spatial resolution.



**Figure 9: Measured (circles) and calibrated modelled (background colours)  $\text{NH}_3$  concentrations within the landscape. For all  $\text{NH}_3$  sources, except for six frequently cleaned poultry houses, average emission factors from the UK inventory were used as model input.**

### 3.5 Risk assessment of environmental impacts

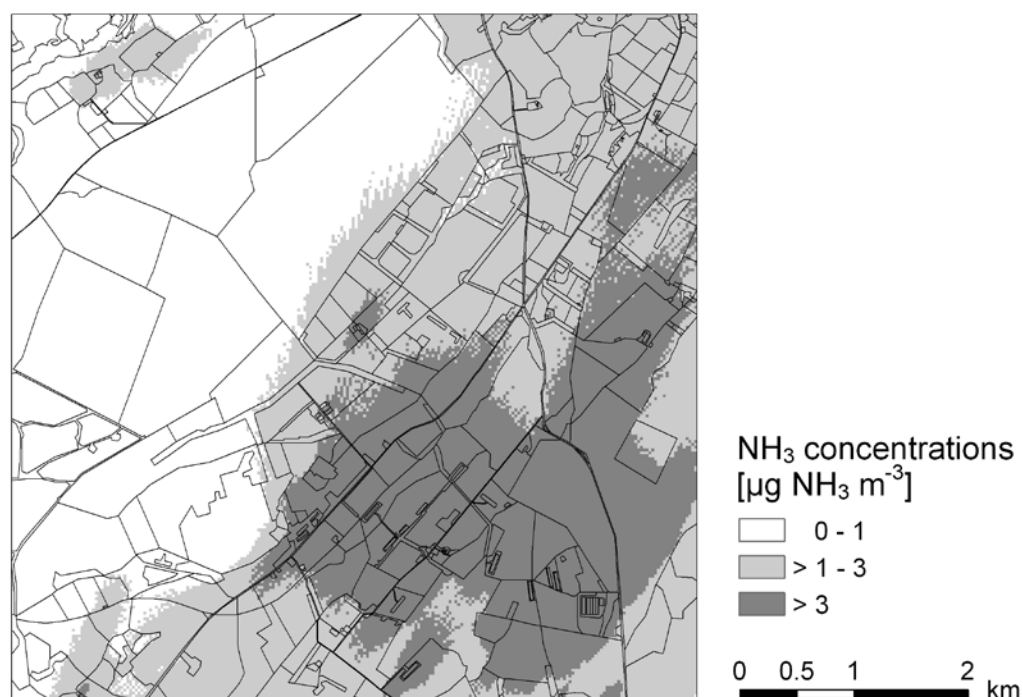
The risk assessment presented is based on comparison of the  $\text{NH}_3$  concentrations with the CLE and N deposition with the CL focussing on the extent of CLE and CL exceedance. For this purpose, the high resolution 25 m calibrated model output data of  $\text{NH}_3$  concentration and dry  $\text{NH}_3$  deposition were supplemented with the 1 km national deposition estimates for oxidised N and wet-deposited reduced N for the study domain. In addition, landscape scale and national scale assessments were compared to investigate fitness-for purpose at the different spatial resolutions.

#### 3.5.1 Concentrations and critical level (CLE) exceedance

For sensitive vegetation (regardless of habitat type), i.e. lichens and bryophytes, the long term CLE for  $\text{NH}_3$  of  $1 \mu\text{g m}^{-3}$  is exceeded in 60% of the landscape (Figure 10). Moorland habitats are naturally low N ecosystems and sensitive to  $\text{NH}_3$ . Within the study area, the CLE is exceeded for 8% of the moorland areas. Such ecosystems could thus be expected to show long term effects of local  $\text{NH}_3$  sources. Although this affects a considerable area (39 ha moorland), it is still a relatively modest fraction considering the extremely high emission fluxes in the vicinity. This is due to most of the moorland in the

study area being located northwest of the poultry houses, in a region with frequent southwesterly winds and low  $\text{NH}_3$  background concentration ( $0.40 \mu\text{g m}^{-3}$  at site 1).

The CLE of  $3 \mu\text{g m}^{-3}$  for higher plants is exceeded in 25% of the landscape. Most of this area is agricultural land: 81% is grass or arable land, and the risk of species composition change is not an issue in arable crops or improved grassland with already substantial fertiliser N input. However, semi-natural vegetation and woodland areas in the landscape with  $\text{NH}_3$  concentrations  $>3 \mu\text{g m}^{-3}$  are at risk, with 7% of such habitats showing an exceedance of the CLE. However, these habitats exceeding the CLE of  $3 \mu\text{g m}^{-3}$  are restricted to relatively small patches within the agricultural area.



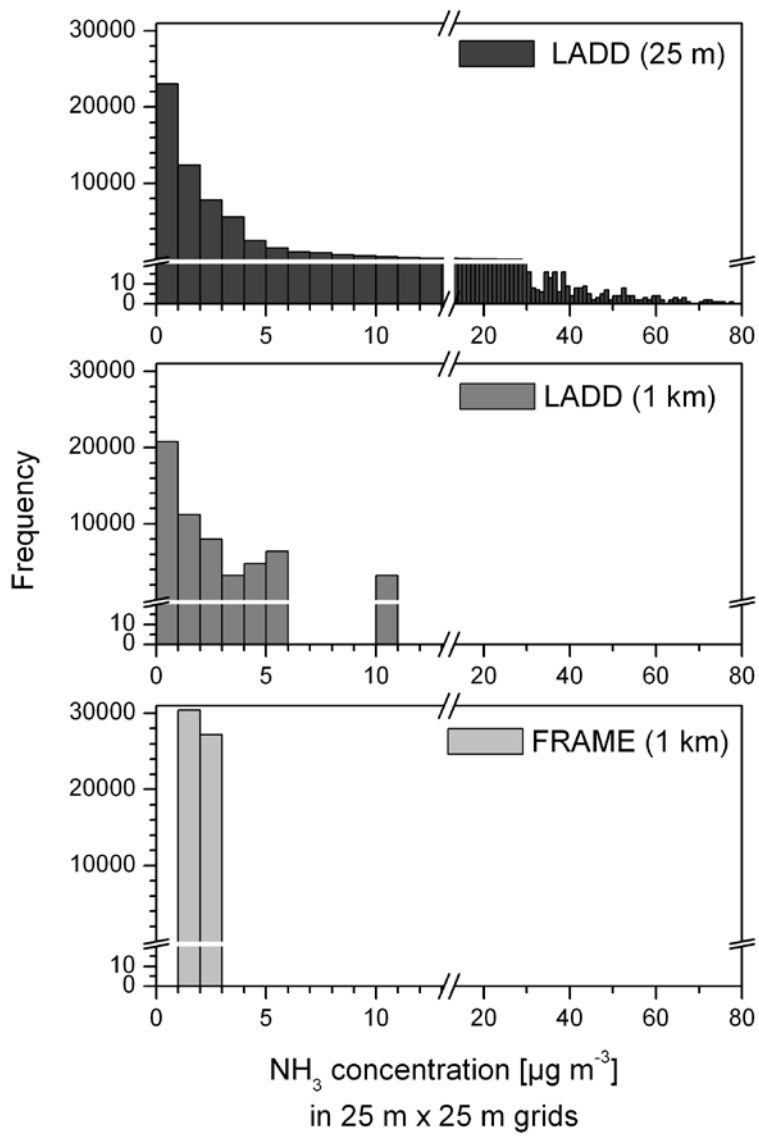
**Figure 10: Modelled  $\text{NH}_3$  concentrations (calibrated) within the study landscape. In white areas, vegetation is not expected to be at risk through  $\text{NH}_3$  concentrations. In light and dark grey areas,  $\text{NH}_3$  concentrations pose a risk to sensitive vegetation, such as lichens and bryophytes. In dark grey areas, all plants of semi-natural ecosystems are at risk.**

These datasets were aggregated from a 25 m resolution to 1 km x 1 km for comparison with concentrations modelled by the UK scale FRAME model at a 1 km resolution using national emission factors (Table 3). FRAME predicts exceedances of the  $1 \mu\text{g m}^{-3}$  CLE for the whole landscape, and conversely no exceedances for the  $3 \mu\text{g m}^{-3}$  CLE. In other words, at the coarser resolution, the impact to the sensitive moorland area northwest of the emission hotspots is overestimated, while the impact downwind of the hotspots is substantially underestimated. Thus, FRAME smoothes out the full spatial

variability of NH<sub>3</sub> concentrations at 1 km resolution to a large degree. By contrast, LADD NH<sub>3</sub> concentrations aggregated to a 1 km resolution capture a much higher level of spatial heterogeneity in CLE exceedances (Table 3, Figure 11 and Figure 12). This suggests that the smoothing out of NH<sub>3</sub> concentrations across a landscape in a national scale model such as FRAME is largely due to coarser scale input data, i.e. during the emission inventory processing. FRAME uses spatial patterns of UK NH<sub>3</sub> emissions calculated in the AENEID model (Dragosits et al., 1998; Hellsten et al., 2008). The model combines parish-level farm statistics with weighted component NH<sub>3</sub> sources according to land cover at 1 km level. Hallsworth et al. (2010) have shown that this approach provides encouraging agreement with NH<sub>3</sub> concentrations modelled at the national scale (model-measurement comparison: R<sup>2</sup> = 0.83), due to 1 km model simulations more effectively separating source (agricultural) areas from sink (semi-natural/nature reserve) areas than the 5 km model. The present study illustrates the limitation of applying a national scale approach at high resolution to a specific landscape, as UK emission mapping is based on general suitability of different land classes for agriculture, but does not include detailed mapping of agricultural point source emissions.

**Table 3: Comparison of the range of modelled NH<sub>3</sub> concentrations within the study landscape and the percentage of CLE exceedance at different resolutions: LADD (25 m, 1 km) and FRAME (1 km).**

	LADD – 25 m	LADD – 1 km	FRAME – 1 km
Min (µg m <sup>-3</sup> )	0.3	0.4	1.1
Max (µg m <sup>-3</sup> )	77.9	10.7	2.9
Mean (µg m <sup>-3</sup> )	2.6	2.6	1.9
% CLE exceedance 1 µg m <sup>-3</sup>	60	64	100
% CLE exceedance 3 µg m <sup>-3</sup>	25	31	0



**Figure 11: Histograms of modelled  $\text{NH}_3$  concentrations in  $25\text{ m} \times 25\text{ m}$  grids in the landscape comparing results at different resolutions: LADD (25 m, 1 km) and FRAME (1 km).**

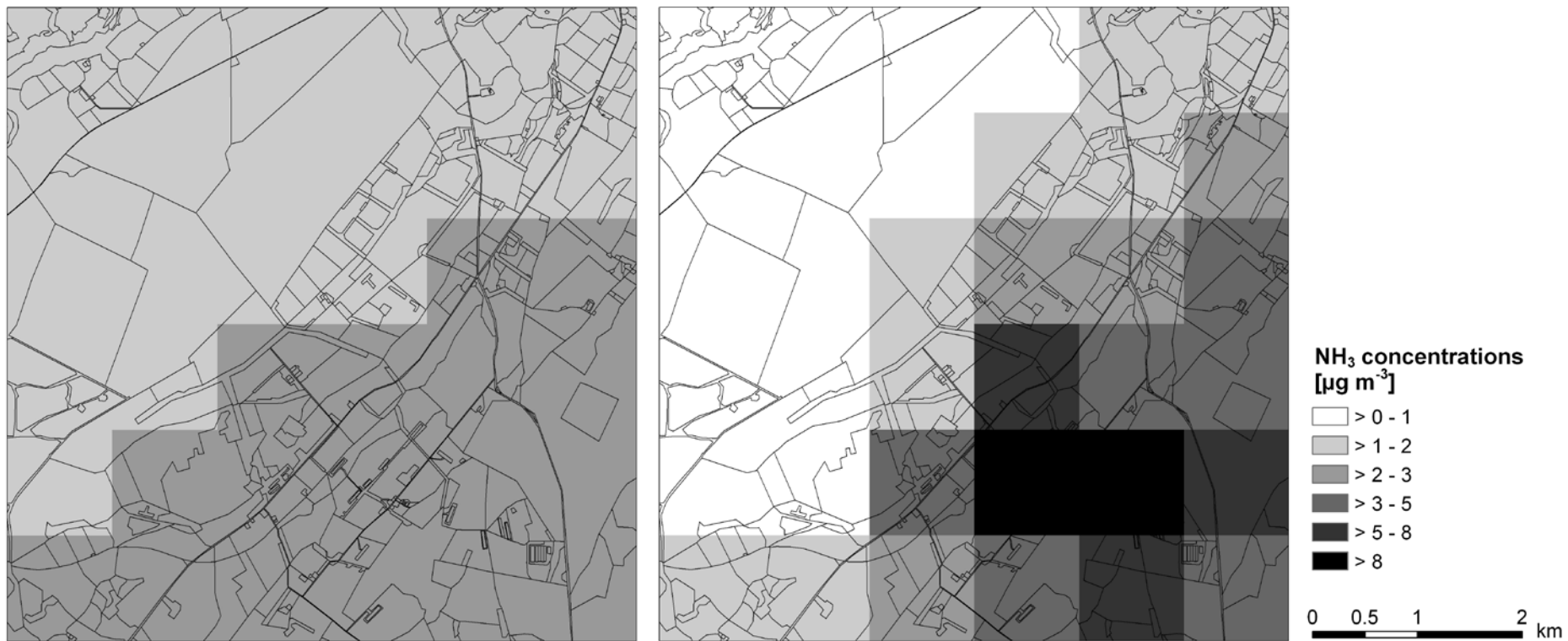


Figure 12: NH<sub>3</sub> concentrations at a 1 km resolution within the study landscape, using FRAME (left map) and LADD (right map).

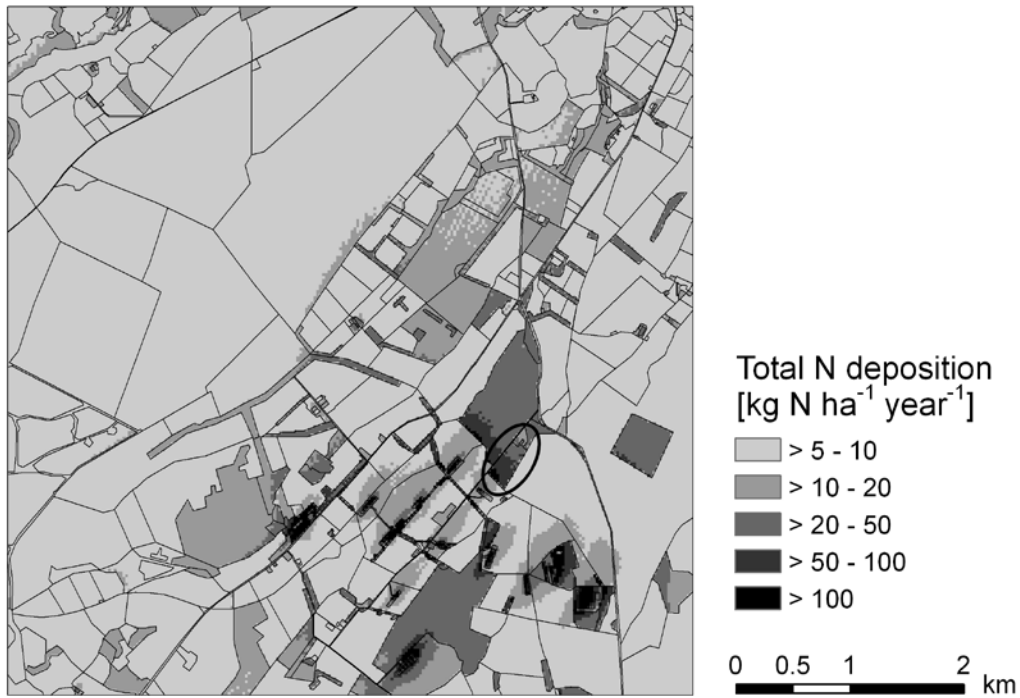


A scenario with all poultry emissions removed was tested for CLE exceedance. In this no-poultry scenario only 12% of the study area exceeded the  $1 \mu\text{g m}^{-3}$  CLE, and 0.2% exceeded the  $3 \mu\text{g m}^{-3}$  CLE, compared with 60% and 25%, respectively, when poultry house emissions were included. This highlights the large contribution of emission hotspots to atmospheric  $\text{NH}_3$  concentrations in mixed landscapes such as this.

### 3.5.2 Deposition and critical load (CL) exceedance

Modelled dry deposition of  $\text{NH}_3$  within the landscape has a high spatial variability ranging from  $0.1$  to  $1200 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$ , with a value  $>1000 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  occurring in only a single  $25 \text{ m} \times 25 \text{ m}$  grid square between closely located poultry houses. Such high dry deposition values can be considered theoretical estimates, as the deposition rate may be expected to be reduced close to large sources due to saturation of the absorbing surfaces (Jones et al., 2007). In most cases, the deposition decreases to  $<100 \text{ kg NH}_3\text{-N ha}^{-1} \text{ yr}^{-1}$  within  $100 \text{ m}$  distance from a source, depending on the absorbing surfaces. To illustrate the importance of capturing the spatial variability, the deposition flux to coniferous woodland downwind of a poultry house was compared with estimates by FRAME (circled area in Figure 13). The woodland of  $6.5 \text{ ha}$  is situated between  $150 \text{ m}$  and  $500 \text{ m}$  from the house. The  $\text{NH}_3$  dry deposition flux to the woodland modelled by LADD varies spatially between  $31$  and  $172 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  and amounts to a total of  $394 \text{ kg N yr}^{-1}$ . This is equivalent to  $6.7\%$  of the  $\text{NH}_3$  emitted from the poultry house near site 31, though other poultry houses would have also contributed to this total. FRAME at  $1 \text{ km}$  estimates a woodland specific dry deposition flux to this area of  $10.8 - 11.9 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (total  $\text{NH}_x$  dry deposition of  $74 \text{ kg N yr}^{-1}$ ). This illustrates how FRAME underestimates the impact of  $\text{NH}_x$  dry deposition in the immediate vicinity of sources, compared with LADD.

Total N deposition (LADD  $\text{NH}_3$  dry deposition + FRAME  $\text{NH}_x$  wet &  $\text{NO}_y$  deposition) ranges from  $5.6$  to  $1206 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (Figure 13). The CL only applies to the land cover categories woodland, hedgerows, shrubs, moorland and rough grass in the landscape, i.e. CL exceedances were calculated only for these categories, which cover  $38\%$  of the study area. In  $34\%$  of this area (or  $13\%$  of the total landscape area) the CL is exceeded, on average by  $17.6 \text{ kg N ha}^{-1} \text{ yr}^{-1}$ , with a median CL exceedance of  $6.5 \text{ kg N ha}^{-1} \text{ yr}^{-1}$  (Figure 14 and Table 4).



**Figure 13: Total N deposition calculated by combining dry deposition of NH<sub>3</sub> simulated by LADD (calibrated) with the remaining components of N deposition from FRAME. The circled area shows a patch of woodland analysed in more detail.**

**Table 4: Land cover specific statistics\* for critical load (CL) exceedance (kg N ha<sup>-1</sup> yr<sup>-1</sup>).**

	Woodland	Shrubs	Rough grass	Moorland
Mean	20.1	21.6	11.6	1.9
Median	7.4	17.6	2.7	0.7
Maximum	1195.6	401.9	406.5	10.5
% exceeding CL	74.2	97.0	28.0	1.7

\*Land cover category hedgerows covered only a very small area and was therefore not considered for these statistics



**Figure 14: Critical load (CL) exceedance calculated using land cover data and CL estimates at 25 m resolution and land cover specific atmospheric deposition estimates at differing spatial resolutions. Left: 1 km N deposition modelled by FRAME; Right: combining 25 m  $\text{NH}_3$  dry deposition simulated by LADD (calibrated) with remaining components of N deposition from FRAME.**

When combining the land cover specific 1 km FRAME results with the 25 m grid resolution land cover data (see Section 2.6), FRAME predicts a CL exceedance in 51% of the area to which a CL applies, compared with 34% simulated by LADD (Table 5). Due to FRAME not fully capturing the spatial variability of NH<sub>3</sub> dry deposition, areas exceeding CL in the whole study landscape are overestimated whereas the extent of CL exceedance in areas close to sources is underestimated (Figure 14).

**Table 5: Comparison of CL exceedances (kg N ha<sup>-1</sup> yr<sup>-1</sup>) within the study landscape between LADD (25 m resolution) and FRAME (1 km resolution).**

	LADD	FRAME
Mean	17.6	3.2
Median	6.5	2.4
Maximum	1195.6	10.8
% exceeding CL	34	51

#### 4 Conclusions

A detailed landscape inventory of all farm activities in the study year 2008 provided data to estimate NH<sub>3</sub> emissions at 25 m resolution. This is essential for studying the actual spatial variability of NH<sub>3</sub> at the landscape scale. The combination of a large number of long term NH<sub>3</sub> concentration measurements across the landscape and the high resolution model output allowed a spatially precise assessment of NH<sub>3</sub> concentrations, which was applied to estimate NH<sub>3</sub> dry deposition. Measured and modelled NH<sub>3</sub> were highly correlated ( $R^2 = 0.90$ ), but model estimates needed to be calibrated by approximately a factor of two to the measurements for environmental risk assessment. This highlights the importance of always including verification measurements in such an assessment. This is also highlighted by Theobald et al. (2012), who showed that the performance of models such as LADD, ADMS and AERMOD, can vary between study sites, for example depending on specific meteorological and emission source characteristics. For robust risk assessment of environmental impacts, models therefore should be appropriately verified with measurements at multiple locations across the study area.

In the present study area, it was also found that standard national emission factors (EFs) were not appropriate for all main NH<sub>3</sub> sources. Ammonia EFs for several poultry houses had to be adjusted to account for the specific manure management practices of frequent litter removal, which resulted in a 75% decrease in emissions compared to the UK average EFs. Thus, for the environmental impact assessment of large livestock houses, it is important to use EFs appropriate to local husbandry systems and manure management.

In this study area, frequent southwesterly winds cause most of the poultry house emissions to disperse to the northeast. As the most sensitive ecosystems in the study area are located northwest of the poultry houses, only a relatively small area is affected by the nearby poultry emissions, despite total poultry  $\text{NH}_3$  emissions exceeding  $100 \text{ t N yr}^{-1}$ , in addition to  $\sim 10 \text{ t N yr}^{-1}$  of  $\text{NH}_3$  emissions from other agricultural sources in the landscape. The ecosystems most at risk from high  $\text{NH}_3$  concentrations are patches of woodland, shrubs and rough grass situated within the agricultural area downwind of the sources. Impact assessment using the critical level (CLE) approach suggested that 8% of the semi-natural moorland may be adversely affected by  $\text{NH}_3$  concentrations above  $1 \mu\text{g m}^{-3}$  (= long term CLE for lichens and bryophytes). By comparison, only 2% of the moorland area is under threat from critical load (CL) exceedance. This relatively small difference between two complementary environmental indicator approaches shows that the present value of the  $\text{NH}_3$  CLE is in reasonably close agreement with the CL values in this upland landscape.

The comparison of the UK national model FRAME at a 1 km resolution with the 25 m resolution LADD estimates showed that FRAME did not capture the full spatial variability of  $\text{NH}_3$  within the study landscape. Furthermore, a comparison of LADD  $\text{NH}_3$  concentrations averaged to 1 km with FRAME 1 km concentrations showed the much higher potential to represent the spatial heterogeneity of  $\text{NH}_3$  in the landscape framework. While the 1 km resolution version from FRAME performs well at the national scale (Hallsworth et al., 2010), this comparison emphasises the need for high resolution emission data obtained at a farm and field level for assessments of environmental impacts from  $\text{NH}_3$ .

This study highlights the importance of the spatial arrangement of  $\text{NH}_3$  sources and sinks within a landscape that is the cause of fine scale heterogeneity in  $\text{NH}_3$  concentrations and N deposition and in the resulting environmental risks. In the study landscape, most sensitive ecosystems are located upwind of the large  $\text{NH}_3$  sources nearby and thus are considered to be at relatively modest environmental risk according to current values of CLEs and CLs. This shows how landscape planning could be used to reduce the impact of intensive agriculture on sensitive ecosystems: Careful planning of the location of the farm point and area sources, considering both distance and the direction in relation to prevailing winds, provides a practical way of avoiding adverse impacts on nearby semi-natural areas.

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