MODELLING OF AMMONIA CONCENTRATIONS AND DEPOSITION OF REDUCED NITROGEN IN THE UNITED KINGDOM

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INTRODUCTION

Emissions of NH_3 in the UK have fallen by 19% since 1990. Further decreases of 10% are forecast by the year 2010. Much larger decreases in emissions of SO_2 and NO_x have occurred in the UK (by 88% and 43% respectively in the last 35 years) Further decreases of 55% and 38% are forecast by the year 2020. As a result of these changes, levels of acid deposition and nitrogen deposition have decreased. However the relative contribution of ammonia to nitrogen deposition and to acid deposition (resulting from in-soil oxidation of ammonia) is increasing. In addition to efforts to nationally monitor the levels of ammonia concentration (Sutton *et al.*, 2001; Tang *et al.*, 2001) and deposition of reduced nitrogen (Fowler *et al.*, 2005), numerical models have been developed to estimate nationally the concentrations of ammonia and ammonium aerosol and the deposition of reduced nitrogen. Models have the advantage that they are able to give good spatial coverage, where measurement data may not be available, as well as the ability to simulate future emissions scenarios. Below we present the results of the Fine Resolution Atmospheric Multi-pollutant Exchange model (FRAME) developed by Singles *et al.* (1998), Fournier *et al.* (2005a, 2005b), Vieno (2005) and Dore *et al.* (2007).

OVERVIEW OF THE FRAME MODEL

The main features of the model can be summarised as:

- 5 x 5 km^2 resolution over the British Isles (incorporating the Republic of Ireland); grid dimensions: 244 x 172 with a 1^o angular resolution in the trajectories.
- 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 150 km scale grid.
- Air column divided into 33 layers moving along straight-line trajectories in a Lagrangian framework with a 1^o angular resolution. The air column advection speed and frequency for a given wind direction is statistically derived from radio-sonde measurements. Variable layer thickness from 1 m at the surface to 100 m at the top of the mixing layer.
- Emissions of NH_3 are gridded separately for cattle, pigs, poultry, sheep, fertiliser and non-agricultural sources and mixed into the lowest surface layers with a source-dependent emissions height.
- 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 diurnally varying scavenging coefficient depending on mixing layer depth and a 'constant drizzle' approximation. A precipitation model is used to calculate wind-direction-dependent orographic enhancement of wet deposition.
- Dry deposition for NH_3 is ecosystem specific and includes five land classes: forest, moorland, grassland, arable, urban & water. A canopy resistance parameterisation is employed including an optional canopy compensation point module for representation of bi-directional exchange of 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.
- The modelled chemical species treated include: NH_3, NH_4^+ aerosol, NO, NO_2, HNO_3, PAN, NO_3^- aerosol, SO_2, H_2SO_4 and SO_4^{2-} aerosol.

- Current model run time: 25 minutes on CEH Edinburgh Beowulf cluster using 100 processors.

RESULTS OF THE MODEL

The output from the model includes maps of annual average surface concentration of NH_3 (Fig. 1-a) which may be used to assess exceedance of the critical level. Maps of annual vegetation-specific dry deposition and wet deposition of reduced nitrogen (Fig. 1-b and Fig. 1-c) are used for calculation of exceedance of critical loads for acid deposition and nitrogen deposition.

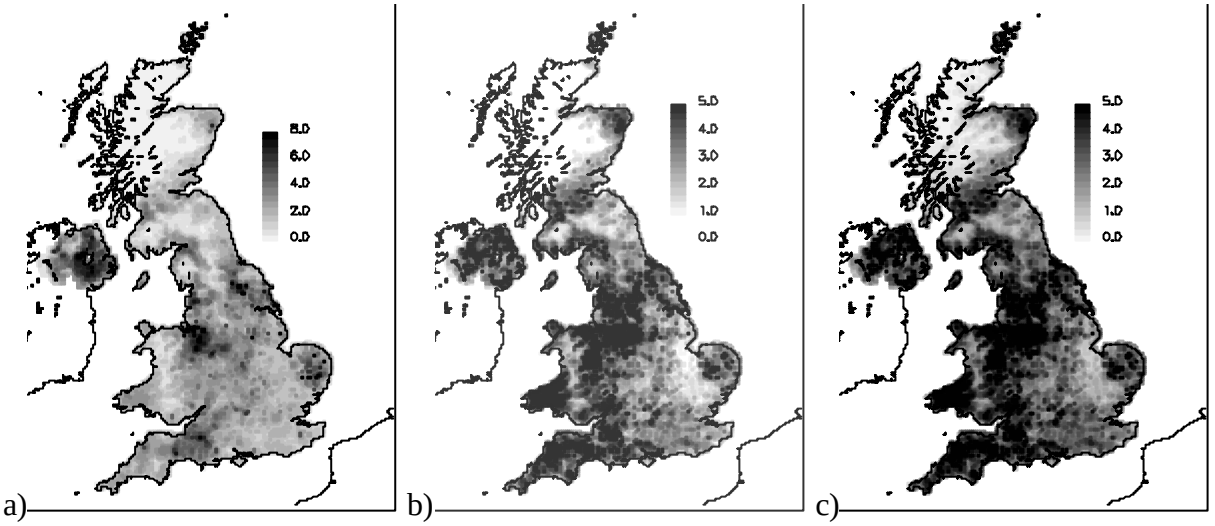


Fig. 1; UK FRAME model prediction for 2002: a) NH_3 surface concentration (µg m^{-3}), b) NH_x dry deposition (kg N ha^{-1}) and c) NH_x wet deposition (kg N ha^{-1}).

Assessment of the accuracy of FRAME in estimating atmospheric concentrations and deposition rates of reduced nitrogen was made by comparison with measurements. For this purpose, data from the UK national ammonia monitoring network was employed comprising over 100 DELTA samplers and ALPHA samplers (http://www.cara.ceh.ac.uk/nh3network). The network uses monthly sampling from the CEH **DELTA** system, (**DE**nuder for **L**ong **T**erm **A**tmospheric sampling; Sutton *et al.*, 2001). ALPHA samplers are passive diffusion samplers, developed for long term monitoring and suitable for use in remote areas with low ammonia concentrations (Tang *et al.*, 2001). Wet deposition data were obtained from the secondary acid precipitation monitoring network, comprising fortnightly collections of precipitation from 38 sites with ion concentrations analysed by ion chromatography (NEGTAP, 2001).

Fig. 2-a, Fig. 2-b and Fig. 2-c illustrate the correlation of the model with measurements. The correlation of modelled concentrations of NH_3 with measurements (Fig. 2-a) shows considerable scatter. The principal reason for this is the highly localised nature of NH_3 emissions, such that the modelled average concentration from a 5 x 5 km^2 model grid cell may differ significantly from that measured at a specific location within the grid cell (Dragosits *et al.*, 2002). The graph shows evidence that, particularly at low concentrations, the model overestimates NH_3 surface concentrations. There is a need for finer scale national modelling of ammonia concentrations, preferably at a 1 km resolution, in order to perform a more accurate model-measurement comparison. A better correlation is observed between modelled and measured NH_4^+ concentrations (Fig. 2-b) and wet deposition (Fig. 2-c). This is

due to the more slowly changing pattern in NH_4^+ aerosol concentrations, which are not expected to vary on a scale smaller than the 5 km model grid resolution. Fig. 2-b shows that the model generally underestimates NH_4^+ aerosol concentrations which may indicate either an underestimate in the rate of production of NH_4^+ aerosol from NH_3 gas or in the import of aerosol at the model boundaries.

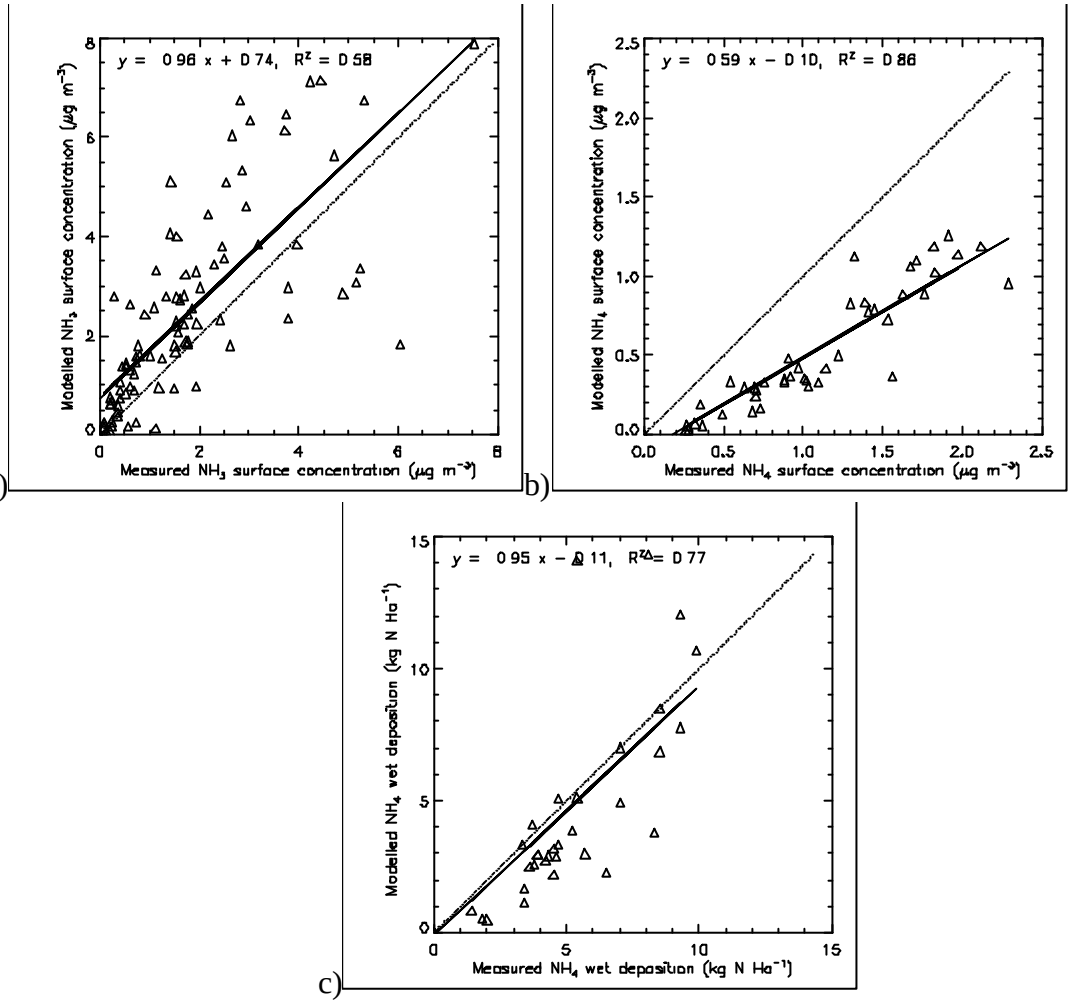


Fig. 2; Correlation of modelled: a) NH3, b) NH4+ aerosol concentrations and c) NH4+ wet deposition with measurements from the national monitoring network for the year 2002. The continuous line shows the linear regression. The dashed line represents a 1:1 relationship.

COMPARISON WITH A LOCAL DIPERSION MODEL

FRAME was run with the model set up to consider only emissions of NH_3 from a theoretical typical poultry farm. The poultry unit was assumed to contain 40,000 birds, each with an annual emission of 0.05 kg NH_3, comprising a total of 2 Mg NH_3 per year. The unit was assumed to be side ventilated with emissions in the height range 1-2 m. The simulation was reduced to one of simple transport, diffusion and dry deposition by switching off both the model chemical scheme and washout from precipitation. A neutral atmospheric thermal stratification was assumed. The results from FRAME were compared with those obtained from ADMS, a local dispersion model. Two model runs were undertaken with both FRAME and ADMS, firstly with local land cover assumed to be grassland and secondly with land cover assumed to be forest. Both models assumed a deposition velocity of 5 mm s^{-1} for grassland and 40 mm s^{-1} for forest. For the ADMS simulation representing grassland, the model was run both with emissions evenly distributed across a 5km x 5km area, similar to

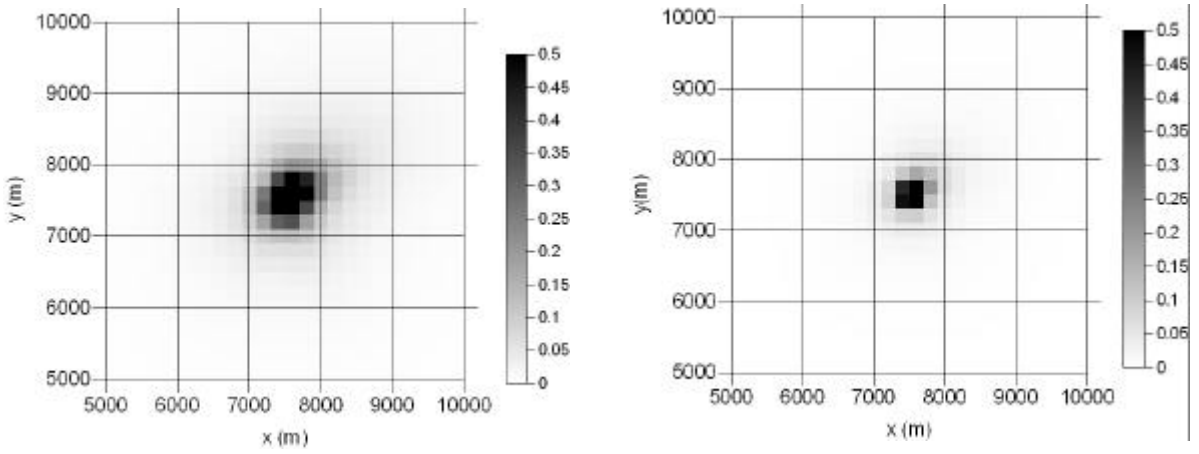
FRAME, and with emissions located in a single 200 m grid square, more typical of a real poultry unit. The results of comparing FRAME with ADMS for evenly distributed emissions are given in Table 1.

Table 1. Comparison of ammonia concentration and NH_x dry deposition modelled with FRAME for a single 5km grid square and for ADMS with a distributed 5km x 5km source.

	FRAME	ADMS
Average concentration ($\mu g\ m^{-3}$)	0.044	0.039
Average deposition (kg N ha^{-1})	0.056	0.050

Close agreement in estimates of concentration and deposition between the two models was found despite the very different approaches adopted in calculating vertical diffusion. For poultry farms and other intensive farming techniques, the even distribution of NH_3 emissions over a 25 km^2 area is clearly physically unrealistic. In reality, emissions may be confined to a single building or group of buildings. This is better represented with the local dispersion model by allocating emissions to a single 200 m by 200 m grid square as illustrated in Figures 3-a and b for grass and forest land cover respectively.

The use of the fine scale local dispersion model shows that the areas of high concentration are restricted mostly to the 1x1 km square at the centre of which is located the point of emissions. Higher concentrations are located to the north east of emissions source due to the predominance of south-westerly winds. The presence of forest land cover (Figure 3-b) and its associated higher deposition velocity is clearly seen to restrict the area of high concentrations to a smaller area. Across the 5 x 5 km^2 domain, in the presence of forested vegetation, average concentrations with ADMS were found to 3.3 times lower and NH_x deposition 4.3 times higher than with the grassland scenario.



(a) (b)

Fig. 3; Ammonia concentration from a single point source (mg m^{-3}) modelled with ADMS: (a) grassland ; (b) forest.

With the local dispersion model, it is clearly seen that ammonia concentrations associated with a single point source emitter vary by over an order of magnitude on the scale associated with a single 5 km FRAME grid cell. This gives the clear message that the current 5 km resolution of national scale assessment of nitrogen deposition will have major uncertainties associated with it in certain areas, depending on the nature of the emissions source. This may result in an overestimation of ammonia concentrations at sites away from point sources. This

emphasises the need to develop national modelling capabilities (i.e. with FRAME) at a finer 1 km resolution.

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