NERC Open Research Archive sharing our research

Centre for Ecology & Hydrology
NATURAL ENVIRONMENT RESEARCH COUNCIL

Chapter (non-refereed)

Tang, Y. S.; Dragosits, U.; Theobald, M. R.; Fowler, D.; Sutton, M. A.. 2001 Sub-grid variability in ammonia concentrations and dry deposition in an upland landscape. In: Fowler, D.; Pitcairn, C., (eds.) *Air-surface exchange of gases and particles: poster proceedings.* Edinburgh, CEH, 48-57.

Copyright © 2001 NERC

This version available at http://nora.nerc.ac.uk/4933/

NERC has developed NORA to enable users to access research outputs wholly or partially funded by NERC. Copyright and other rights for material on this site are retained by the authors and/or other rights owners. Users should read the terms and conditions of use of this material at http://nora.nerc.ac.uk/policies.html#access

This document is extracted from the publisher's version of the volume. If you wish to cite this item please use the reference above or cite the NORA entry

Contact CEH NORA team at
nora@ceh.ac.uk

Sub-grid variability in ammonia concentrations and dry deposition in an upland landscape

Y.S. TANG*, U. DRAGOSITS, M.R. THEOBALD, D. FOWLER AND M.A. SUTTON

Centre for Ecology and Hydrolog (Edinburgh), Penicuik, Midlothian, Scotland, UK.

Abstract

Ground level atmospheric ammonia (NH_3) concentrations and dry deposition are spatially very variable, due to the large range of NH_3 emissions in the rural landscape, as well as large NH_3 dry deposition velocities. In the UK National Ammonia Monitoring Network, NH_3 concentrations are compared with estimates from an emission inventory and atmospheric transport model at 5 km grid resolution. However, it is recognised that mapping of NH_3 concentration and deposition at a national scale using monitoring and atmospheric transport models need to consider the extent of sub-grid variability and the representativity of sampling points.

In the present study, spatially detailed NH_3 monitoring is reported across an example upland 5 km grid square containing one of the National Network sites. This is compared with the national inventory and a local-scale dispersion model at a sub-1 km level to assess the extent to which the monitoring location is representative.

The National Network site selected (Glenshee: annual mean 2.6 $\mu g\ m^{-3}$) was located at the side of a valley in the Scottish Highlands. Sampling for 8 months was made at 12 sites over the surrounding 5 km grid, including both valley and hill sampling locations, using a high sensitivity passive sampling method. The results demonstrate concentrations typically a factor of 4 larger in the valley than on the hills; monthly concentrations were in the range 0.15 – 2.01 $\mu g\ m^{-3}$ and were closely correlated with the output from the local transport model. The analysis supports the performance of the 1 km emissions inventory in this landscape and provides a means to identify sampling locations representative of the 5 km grid square.

Introduction

The UK National Ammonia Monitoring Network operated by CEH (formerly ITE) on behalf of the Department of Environment, Transport and the Regions (DETR) has been running since 1996. Data from the network confirm the high spatial variability in NH_3 concentration and dry deposition across the country, reflecting the large regional variability in NH_3 emissions, and rapid rates of NH_3 dry deposition (Sutton *et al.*, 1998b).

The network currently estimates the national UK concentration field for ammonia from 90 sites and is combined with the CEH deposition model (Smith *et al.*, 2000) to provide estimates of NH_3 dry deposition. Monitored concentrations from the network (Sutton *et al.*, 1998b) are also compared with estimates from an atmospheric transport model (FRAME) at a 5 km grid resolution (Singles 1996, Singles *et al.*, 1998). The underlying NH_3 emission inventory is modelled at a 1 km resolution, using agricultural census and land cover data, and is subsequently aggregated to a 5 km grid (Dragosits *et al.*, 1998, Dragosits 1999) for input to the dispersion model.

Substantial variability in NH_3 concentration and deposition is expected even at the sub-5 km level (Dragosits *et al.* 2001). For this reason, it is important that the extent of sub-grid variability and the representativity of sampling sites in the network are considered.

In the present study, a detailed local variability study was conducted across one 5 km x 5 km grid square in an upland landscape (Glenshee in NE Scotland). The results are compared with the 5 km resolution estimates for the UK national model, and a local scale dispersion model at 250 m and 500 m grid resolution (LADD model, Hill 1996, Sutton *et al.*, 1998a). This allows the performance of the national NH_3 emission model of Dragosits *et al.* (1998) to be tested and an assessment of the representativity of the existing monitoring network site to be made.

Methods

Local scale modelling of NH_3 emissions

Ammonia emissions for the 5 km x 5 km study area were modelled at a 1 km resolution, using the CEH Landcover data and agricultural census data from 1988. Although the model of Dragosits *et al.* (1988) is also available for 1996, the 1988 estimates were used due to sensitivity of confidentiality with the data for 1996. Comparison of the two datasets for Glenshee however showed no major difference in emissions between the two years. Sheep and cattle grazing emissions are the dominant NH_3 sources within the grid square, occurring mainly within the more intensively managed valley, whereas most of the 5 km square is upland moorland (Figure 1).

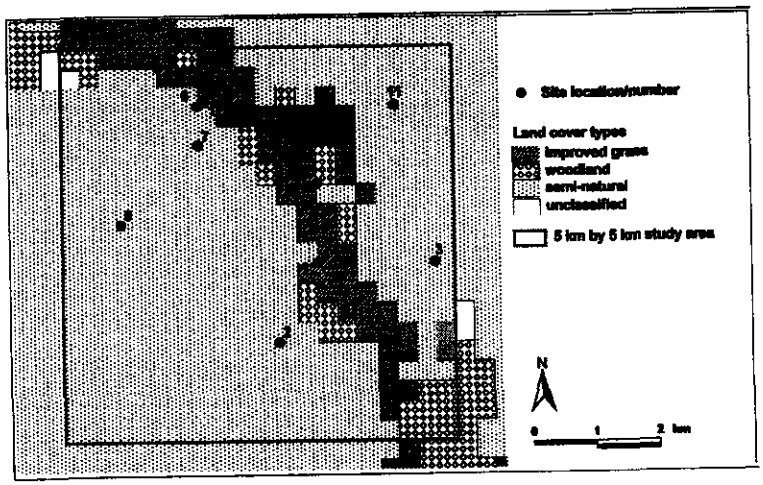

Figure 1: Land cover map of the Glenshee study area at a 250 m grid resolution, showing sampling locations with site numbers.

A model for the spatial distribution of NH_3 emission sources applied to the whole of the UK provided weighted allocations of emissions to different land cover types at Glenshee at the 1 km level (Dragosits *et al.*, 1998). This approach results in a large proportion of the emissions being realistically located in the intensively farmed areas, and only small sources of livestock grazing on the extensive upland areas. In Figure 1, land cover at 250 m grid resolution is shown, which allows a greater precision in the model analysis.

Local scale modelling of NH_3 concentrations and deposition

The LADD model (Hill 1998, Sutton *et al.* 1988a) was used to simulate atmospheric NH_3 concentration and dry deposition at 250 m and 500 m grid resolutions within the 5 km x 5 km grid. LADD is a multi-layer Lagrangian model, which utilises statistical meteorology and straight line trajectories. Information from the local scale emissions inventory at the 1 km level, plus meteorological data (obtained from Aboyne automatic weather station: 57.08 ° latitude, -2.84° longitude) are incorporated as input to the model. The concentration field output is coupled with land-use dependent deposition velocities (V_d) to provide a detailed dry deposition field. Land cover information at matching scales (250 m and 500 m resolution) is included within the model, so that the values of V_d can be varied according to land-use in the grids. Different estimates of ecosystem specific surface roughness (z_0) and canopy resistances (R_c) (Sutton *et al.*, 1998a) similar to those adopted by Singles *et al.* (1998) in FRAME are used to calculate ecosystem specific V_d, which should give improved estimates of deposition, allowing the distinction to be made between the semi-natural upland areas and agricultural lands in the valley.

Field measurements of NH_3 concentrations

The national NH_3 network site selected is located at the side of a valley in the Scottish Highlands (56.81 ° latitude, -3.45° longitude). The local variability measurements began in April 1999 and sampling for 8 months was conducted at 12 sites, including both valley and hill sampling locations (Figure 1). NH_3 concentrations were measured using a new improved high sensitivity diffusion sampler with 3 replicate samplers at each site at 1.5 m above ground (Sutton *et al.*, 1998b, Tang *et al.*, 2001). The average reproducibility of replicate samples in the field was 6 % (RSD) and the detection limit (3 μ of blanks) was 0.02 μg m^3 for a one month exposure period.

Results and Discussion

Ammonia emissions map

A high degree of variability in NH_3 concentrations within the 5 km grid area was expected on the basis of the emission estimates at a 1 km resolution, as shown in Figure 2.

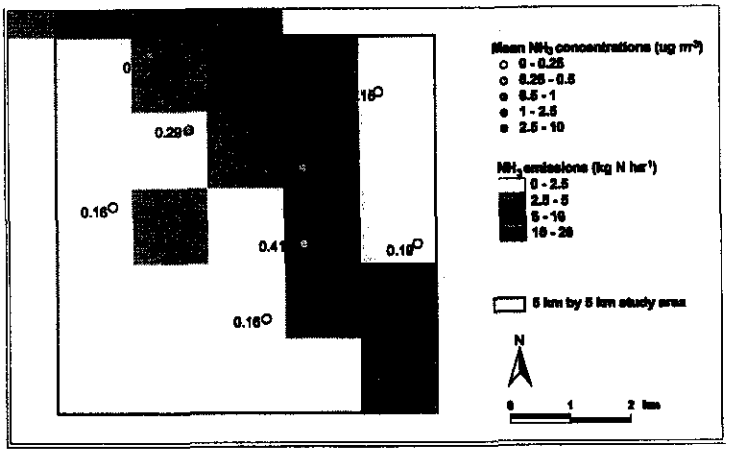

Figure 2: Modelled ammonia emissions over the study area at a 1 km grid resolution (Dragosits et al., 1998), compared with the measured mean NH_3 concentrations at the 12 monitoring sites.

The emissions map shows much higher emissions in the intensively farmed valley areas with both sheep and cattle farming, against the very low background emissions over the extensive upland moorlands. Emissions from the feeding and housing of livestock, and the storage and disposal of their wastes are much larger than from grazing animals. As a result, although animals may graze hill areas at some time (e.g. sheep taken to higher pastures in summer), most of the NH_3 emissions are located within better agricultural land in the valley. The emission model of Dragosits *et al.* (1998) therefore provides realistic spatial NH_3 emissions estimates for the upland landscape, by relocating emission sources from the extensive upland areas to the more intensively farmed lowland areas within the grid square. This is a direct result of reallocation rules introduced into the emission model, which avoided the artificial location of housing and landspreading emissions in hill areas. Confidence in the accuracy of the emissions estimates is vital, since they are used as primary input into the LADD model to predict atmospheric NH_3 concentration and deposition.

Measured air NH_3 concentrations

The measured concentrations from the sampling sites also confirmed the spatial variability in relation to valley and hill locations, with concentrations typically a factor of 4 larger in the valley than on the hills (Figure 2). This provides support for the emission inventory approach developed by Dragosits *et al.* (1998).

Substantial temporal variability in ammonia concentration was observed at all the measurement sites (Figure 3).

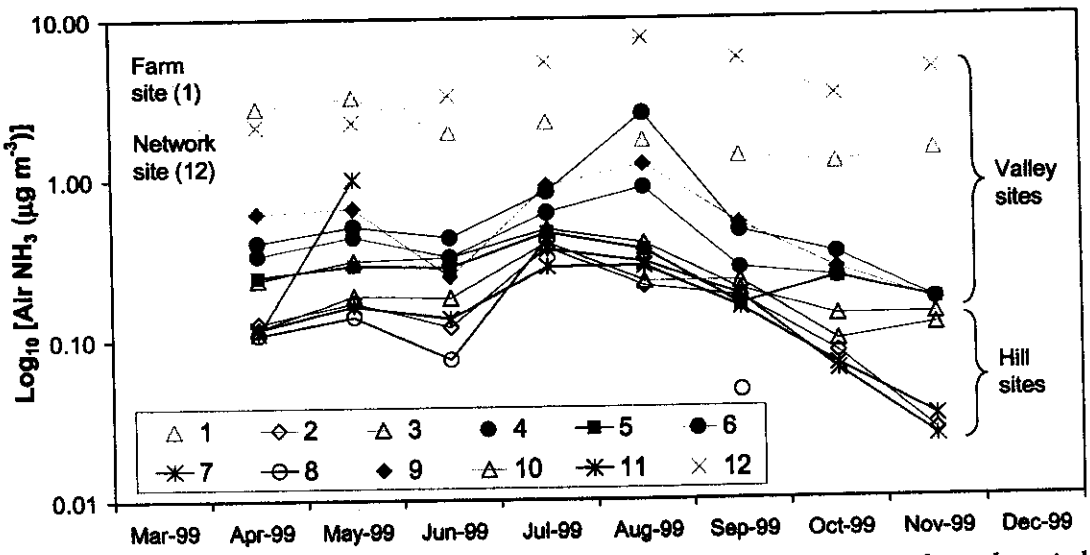

Figure 3: Temporal trends in measured NH_3 air concentrations over the 8 months study period.

At Site 1, peaks in ammonia concentrations were observed in the spring, coinciding with the usual time for land spreading of animal wastes. At all other sites, the highest concentrations occurred during the summer months, with values up to five times higher than during winter. This is due to increased potential for ammonia volatilisation in the warmer and drier conditions during the summer months. Temporal patterns for the hill sites are very similar and are typical of clean background sites (Sutton *et al.*, 1998b), where the variation in NH_3 concentration is mostly influenced by the seasonal effect on NH_3 emissions.

Ammonia concentrations at the network site (site 12) were very high for the study period, even compared with other sites in the valley area (Figure 3). The mean concentration of 4.2 μg NH_3 m^{-3} for the study period is also larger than previous measurements prior to December 1998 (Figure 4).

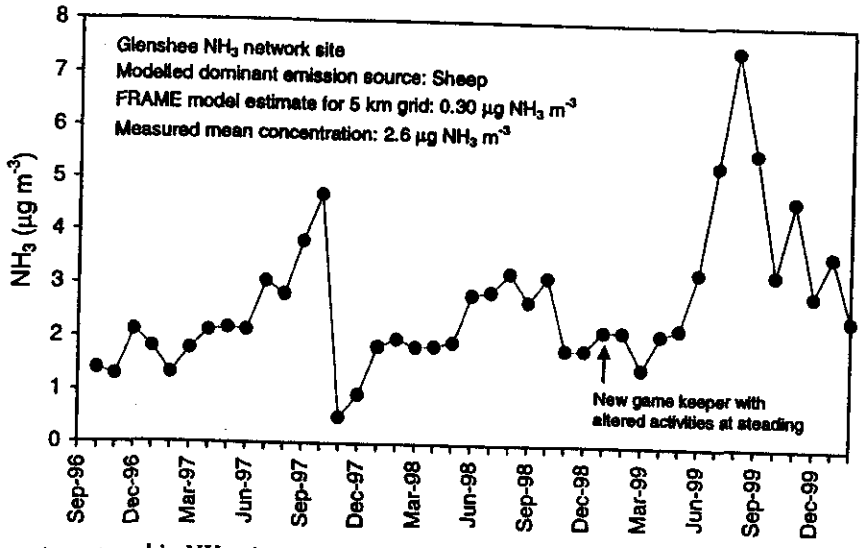

Figure 4: Long-term trend in NH_3 air concentrations at the main NH_3 network site for Glenshee from October 1996.

It transpires that this site is affected by very local sources of NH_3 (e.g. proximity of dog kennels, preparation of game and other game-keeping activities), since the concentrations decreased to 0.71 μg NH_3 m^3 at site 6 over a distance of 200 m. Significant emission sources local to site 12 had not been expected, and the site location had originally been selected on the basis of being at the valley edge with mains electricity available for active sampling of both air NH_3 and aerosol NH_4^+ (Sutton *et al.*, 2001).

Modelled air NH_3 concentrations

The predicted NH_3 concentration field output from LADD at 500 m and 250 m grid resolutions are shown in Figures 5A and 5B.

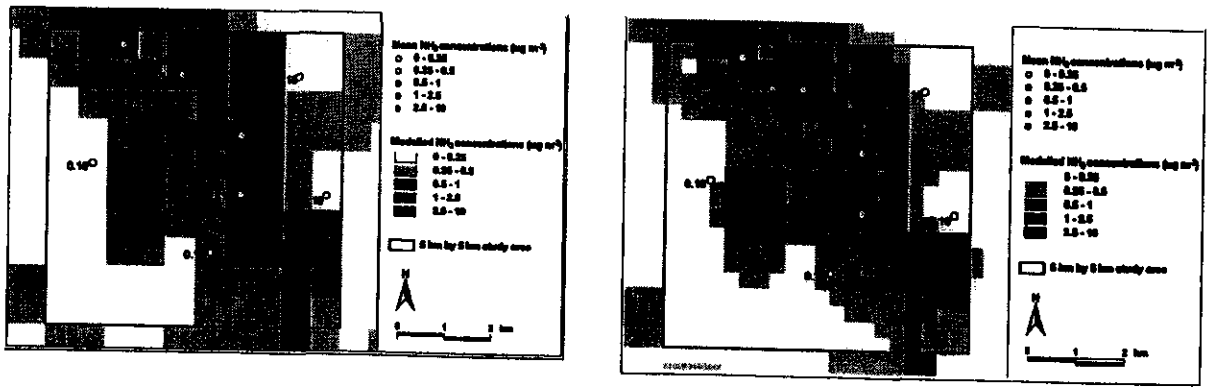

Figure 5: Modelled NH_3 air concentrations from the LADD model for the study area, with land cover data at (A) 500 m grid resolution and (B) 250 m grid resolution. The mean measured NH_3 concentrations for each sampling site are plotted on the map for comparison.

Both 500 m and 250 m estimates were made with land cover data at corresponding resolutions, to compare the difference in LADD model output when using the emissions inventory at the 1 km level. The estimates for both grid resolutions are generally similar (Figure 6), although at site 10, the LADD estimate at 500 m resolution for the grid square is about 60 % greater than at 250 m resolution. The difference between the estimates at site 10 is due to the resolution of the land cover data, which affects values of z_o and V_d.

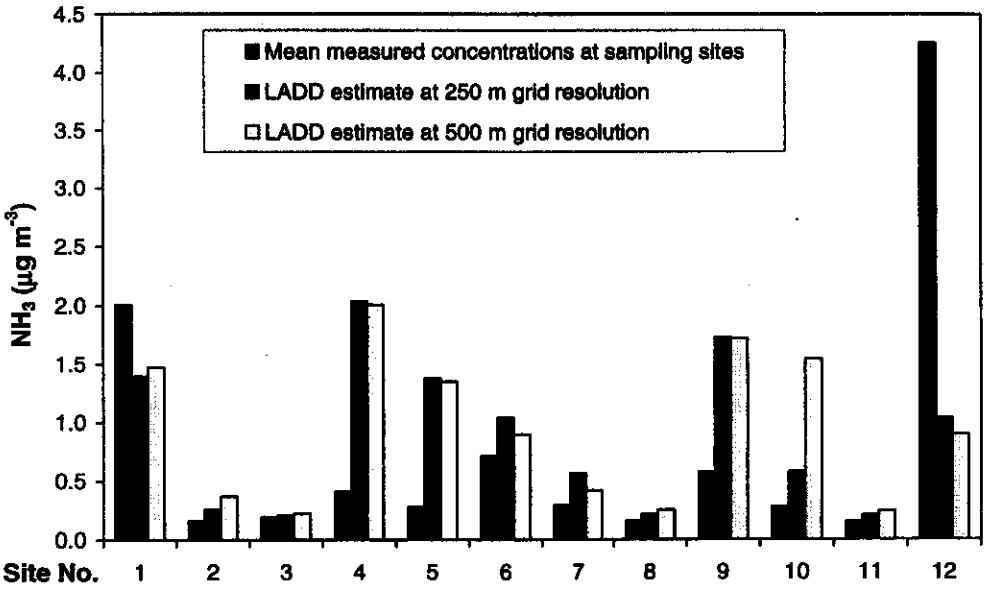

Figure 6: Comparison of measured and modelled air NH_3 concentrations at the 12 sampling locations around Glenshee.

3.4. Comparison of measured and modelled NH_3 concentrations

The local variability study at Glenshee provided data with which to test the performance of the LADD model. Good agreement was shown between the LADD model results (mean = 0.89 µg NH_3 m^{-3}, range 0.21 - 2.04 µg NH_3 m^{-3}) and field measurements for 11 of the 12 sampling sites (mean = 0.48 NH_3 µg m^{-3}, range 0.15 - 2.01 µg NH_3 m^{-3}) (Table 1).

	Mean (µg NH_3 m^{-3})	Min (µg NH_3 m^{-3})	Max (µg NH_3 m^{-3})
FRAME estimate for 5 km grid square	0.30	-	
LADD estimate for 5 km grid square: 250 m resolution	0.60	0.16	2.65
500 m resolution	0.62	0.18	2.46
LADD estimate for grid squares : 250 m resolution	0.89	0.21	2.04
with measurement sites 500 m resolution	0.95	0.22	2.01
Measurement data: All 12 sampling locations	0.79	0.15	4.26
11 sampling locations (excluding site 12)	0.48	0.15	2.01

Table 1: Comparison of measured and modelled air ammonia concentrations in the 5 km x 5km study area.

Overall, LADD is seen to overestimate ammonia concentrations compared with the measurements (Figure 6). This may be because of uncertainties in the diffusion processes, which are sensitive to surface roughness (z_o) and atmospheric stability, as well as the fact that this model does not describe wet deposition scavenging of ammonia. The local emission and

land cover data may also be limited by the resolutions used, which may not be sufficiently detailed to resolve the more complex landscape, especially in grid cells where semi-natural areas are integrated closely with improved grasslands. The locations of sites 4 and 5 are borderline semi-natural / improved grassland, whereas the grid cells in which site 9 is on is classified by the land cover dataset as improved grassland, when in actual fact the immediate area is interspersed with woodlands and semi-natural areas. Ammonia concentrations are also calculated within the 1 m vertical layer, whilst measurements were made at 1.5 m above ground. The vertical concentration profile of NH_3 in source regions (e.g. agricultural land in the valley) can vary by as much as a factor of 2 between heights of 1 m and 2 m, whereas the gradient is smaller in sink areas (e.g. moorland) (Sutton *et al.*, 2000). Work is currently in progress to refine the model to estimate concentrations at different vertical profiles. It is expected that this will result in better agreement with the measurements which are made at 1.5 m above ground, particularly in source areas.

Recognizing the uncertainties in the model, and the dependence of LADD on the emission inventory as a primary input, this comparison demonstrates that the Dragosits *et al.* (1998) model provides sound separation between source and sink areas in this study area. This is important to note, since this model is based on parish agricultural statistics and land cover data across Great Britain, and this study provides the first field testing of the inventory at a sub-5 km level.

The present study also provides information to assess the most representative sampling location in the 5 km grid square for future measurements. The measurements have shown that the original measurement site (Site 12) was unrepresentatively high, even compared with other sites in the valley area. Of the remaining sites, the question arises as to which is the most representative. The simplest estimate would be to calculate the mean concentration from the measurements (= 0.48 μg m^{-3}), but this is potentially biased by having too few measurements in the hill areas of the grid square. Therefore the model was used to calculate the most representative concentration, since by definition this covers the whole grid square.

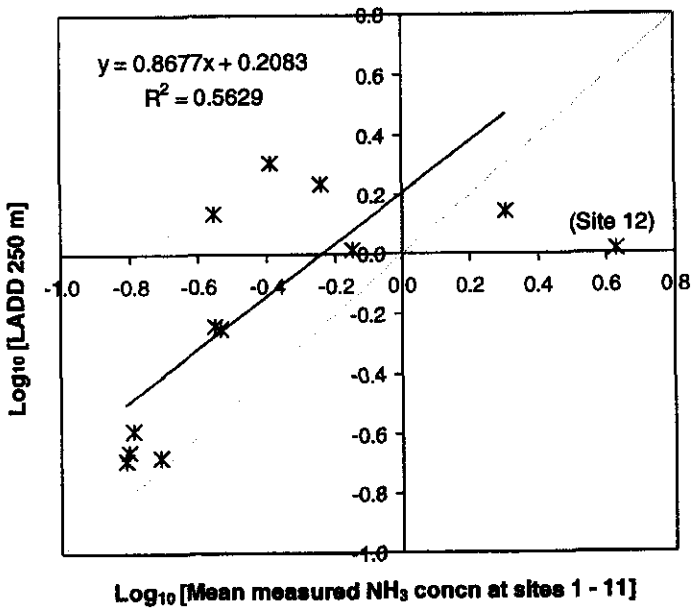

Figure 7: Regression plot of predicted NH_3 concentration (□g NH_3 m^{-3}) from LADD at 250 m grid resolution versus the average of the field measurements from sites 1 to 11.

For this purpose the LADD estimates of NH_3 concentration at a 250 m grid resolution were used to gauge the most typical model concentration. Of the 400 250 m x 250 m estimates in the study area, the mean modelled concentration is 0.49 $\mu g\ m^{-3}$. Applying this value to the regression of LADD using the emissions at a 250 m resolution on the average of the measurements at each monitoring site (Figure 7) would imply a mean measured concentration representative for the 5 km grid square of 0.29 $\mu g\ m^{-3}$. LADD is taken as the independent variable in the regression, since the graph is used to estimate the most representative measured concentration. For the purpose of the regression, Site 12 has not been included, as this was established as being affected by very local sources of NH_3. With this exception, there is a significant correlation between the model and the measurements (R^2 = 0.56, $P<0.01$), once the data are transformed to a more normal distribution by taking the Log_{10} of concentration.

Three of the measurement locations provided measured concentrations close to the calculated representative concentration of 0.29 $\mu g\ m^{-3}$: sites 5, 7 and 10. Of these, site 5 was selected for future long-term monitoring in the grid square because of the availability of a suitable site operator and the presence of electricity nearby to allow monitoring of ammonium aerosol. It may be noted that the national estimate of the FRAME model for the 5km grid square is 0.30 $\mu g\ m^{-3}$, which is very close to the most representative measured concentration.

Modelled dry deposition of NH_3

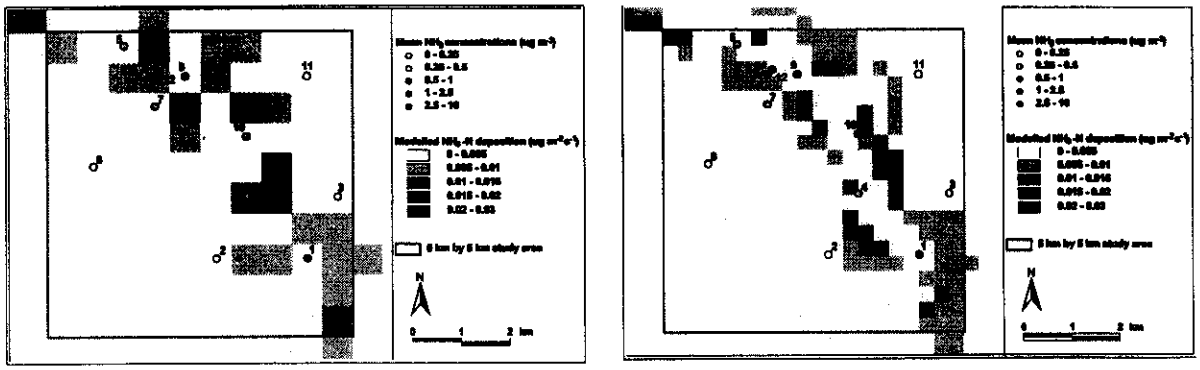

Figure 8: Modelled NH_3-N dry deposition from the LADD model for the study area, with input data at (A) 500 m grid resolution, and (B) 250 m grid resolution.

Large spatial variability in NH_3 dry deposition was estimated within the study area (Figure 8), reflecting both the local patterns of ammonia concentration and the land-cover dependent differences in deposition velocity. Forests and moorland areas downwind of emission sources (wind direction from N / NW) received more NH_3 dry deposition on average, due to greater surface roughness (forests) and small values of R_c. Upland moorland areas further than 1 km from NH_3 sources were estimated to receive less dry deposition than moorlands adjacent to agricultural land. Although concentrations were highest over the agricultural land in the valley, dry deposition was small to these areas due to low deposition velocities. Hence the largest rates of dry deposition were to moorland and forest vegetation located along the edge of the valley bottom. The results highlight the importance of fine scale (sub-5 km) variability of NH_3 deposition in relation to impacts of N deposition.

Conclusions

Substantial variability in NH_3 air concentrations and dry deposition has been demonstrated within a 5 km x 5 km grid study area surrounding an NH_3 monitoring site in the Scottish Highlands. Ammonia concentrations were typically a factor of 4 larger in the valley (mean = 0.72 $\mu g\ m^{-3}$, range = 0.28 – 2.01 $\mu g\ m^{-3}$, excluding one outlier) compared with the hill areas (mean = 0.18 $\mu g\ m^{-3}$, range = 0.13 – 0.29 $\mu g\ m^{-3}$), and are seasonally variable, with peak concentrations in the summer. In the valley, peak concentrations were observed in the spring, coinciding with manure application to fields.

Good agreement was shown between the LADD model results and field measurements, lending support for the performance of the NH_3 emission model of Dragosits *et al.* (1998) for an upland landscape at 1 km resolution. Measured NH_3 concentrations were generally lower than LADD predictions. This may be attributed to limitations of the input data at the grid resolutions used, output of NH_3 concentrations within the 1 m vertical layer and uncertainties within the model. While the study provides measurement support for the 1 km national NH_3 emission inventory of Dragosits *et al.* (1998), further development of this approach is essential.

It has been established that the National Network site was an outlier influenced by very localised emission sources (e.g. gamekeeping activities), and that the monitoring location overestimated NH_3 concentrations compared with the national 5 km grid estimate due to the presence of such local sources. A more representative new sampling location for the area has been identified from this local variability study, by applying the relationship between modelled and measured concentrations for the grid square. While the previous monitoring location provided a long term mean of 2.6 $\mu g\ m^{-3}$ (1996 – 1999), the most representative measured concentration for the grid square is 0.29 $\mu g\ m^{-3}$. The latter compares favourably with the 5 km grid square average value of 0.30 $\mu g\ m^{-3}$ estimated by the UK-scale FRAME dispersion model.

Acknowledgement

This work was carried out with funding from the Department of Environment, Transport and the Regions (DETR) and from supporting CEH programmes. We would like to thank the Invercauld estate, Finegand farm, Glenshee outdoor centre and Spittal of Glenshee hotel for permitting access to field sites for carrying out the monitoring work.

References

Dragosits, U., Sutton, M.A., Place, C.J. & Bayley, A.A. (1998) Modelling the spatial distribution of agricultural ammonia emissions in the UK. Environmental Pollution **102**, S1, 195-203.

Dragosits, U. (1999) *A spatially distributed ammonia emissions inventory for the UK*. Ph.D. thesis. University of Edinburgh.

Dragosits, U., Theobald, M.R., Place, C.J., Lord, E., Webb, J., Hill, J., ApSimon, H.M. & Sutton, M.A. (2001) Ammonia emission, deposition and impact assessment at the field scale: a case study of sub-grid spatial variability. Environmental Pollution (in press).

Hill, J. (1998) *Applications of computational modelling to ammonia dispersion from agricultural sources*. Ph.D. thesis, University of London.

Singles, R.J. (1996) *Fine resolution modelling of ammonia dry deposition over Great Britain.* Ph.D. thesis, University of Edinburgh.

Singles, R.J., Sutton, M.A. & Weston, K.J. (1998) A multi-layer model to describe the atmospheric transport and deposition of ammonia in Great Britain. Atmospheric Environment **32 (3)** (Ammonia Special Issue), 393-399.

Sutton, M.A., Milford, C., Dragosits, U., Place, C.J., Singles, R.J., Smith, R.I., Pitcairn, C.E.R., Fowler, D., Hill, J., ApSimon, H.M., Ross, C., Hill., R., Jarvis, S.C., Pain, B.F., Phillips, R., Harrison, R., Moss, D., Webb, J., Espenhahn, S.E., Lee, D.S., Hornung, M., Ullyett, J., Bull, K.R., Emmett, B.A., Lowe, J. & Wyers G.P. (1998a) Dispersion, deposition and impacts of atmospheric ammonia: quantifying local budgets and spatial variability. Environmental Pollution **102**, S1, 349-361

Sutton, M. A., Nemitz, E., Fowler, D., Wyers, G. P., Otjes, R. P., Schjoerring, J. K., Husted, S., Nielsen, K. H., San Jose, R., Moreno, J., Gallagher, M. W. & Gut, A. (2000) Fluxes of ammonia over oilseed rape - Overview of the EXAMINE experiment. Agricultural and Forest Meteorology. **105**, 327-349.

Sutton, M.A., Tang, Y.S., Miners, B.P., Coyle, M., Smith, R. & Fowler, D. (1998b) Spatial and temporal patterns of ammonia concentration in the UK. Results of the National Ammonia Monitoring Network. Final Report to the Department of the Environment Transport and Regions (DETR), Air and Environmental Quality Division. (Contracts EPG 1/3/58). ITE Edinburgh, Penicuik.

Sutton, M. A., Tang, Y.S., Miners, B. and Fowler, D. (2001) A new diffusion denuder system for long-term, regional monitoring of atmospheric ammonia and ammonium. Water, Air and Soil Pollution (in press).

Tang, Y.S., Cape, J.N. & Sutton, M.A. (2001) Development and types of passive samplers for monitoring NO_2 and NH_3 concentrations. The Scientific World (under review).