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The relationship between NH_3 emissions from a poultry farm and soil NO and N_2O fluxes from a downwind forest

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Abstract. Intensive livestock farms emit large concentrations of NH_3, most of which is deposited very close to the source. The presence of trees enhances the deposition. Rates to downwind forests can exceed 40 kg N ha^{-1} y^{-1}. The steep gradient in large NH_3 concentrations of 34.3±20.4, 47.6±24.9, 21.7±16.8 μg NH_3 m^3 at the edge of a forest 15, 30 and 45 m downwind of the farm to near background concentrations within 270 m downwind (1.15±0.7 μg NH_3 m^3) provides an ideal site to study the effect of different rates of atmospheric NH_3 concentrations and inferred deposition on biological and chemical processes under similar environmental conditions. We have investigated the effect of different NH_3 concentrations and implied deposition rates on the flux of NO and N_2O from soil in a mixed woodland downwind of a large poultry farm (160 000 birds) in Scotland, which has been operating for about 40 years. Measurements were carried out for a 6 month period, with hourly NO flux measurements, daily N_2O fluxes close to the farm and monthly at all sites, and monthly cumulative wet and dry N deposition. The increased NH_3 and NH_4^+ deposition to the woodland increased emissions of NO and N_2O and soil available NH_4^+ and NO_3^- concentrations. Average NO and N_2O fluxes measured 15, 25 and 45 m downwind of the farm were 111.2±41.1, 123.3±40.7, 38.3±28.8 μg NO-N m^{-2} h^{-1} and 9.9±7.5, 34.3±33.3 and 21.2±6.1 μg N_2O-N m^{-2} h^{-1}, respectively. At the background site 270 m downwind the N_2O flux was reduced to 1.75±2.1 μg N_2O-N m^{-2} h^{-1}. NO emissions were significantly influenced by seasonal and daily changes in soil temperature and followed a diurnal pattern with maximum emissions approximately 3 h after noon. For N_2O no consistent diurnal pattern was observed. Changes in soil moisture content had a less clear effect on the NO and N_2O flux. In spite of the large NO and N_2O emissions accounting for >3% of the N deposited to the woodland downwind of the farm, extrapolation to the entire British poultry flock suggests that these NH_3 emissions contribute to less than 0.5% and 0.02%, respectively of the total annual UK NO_x and N_2O emissions.

1 Introduction

The deposition of nitrogen (N) to the soil has increased since the industrial revolution and now ranges from less than 5 kg N ha^{-1} y^{-1} in pristine environments to over 80 kg N ha^{-1}y^{-1} in the most polluted locations of Europe (Fowler et al., 2004a). In many areas N deposition is dominated by reduced N, NH_3 (ammonia) and NH_4^+ (ammonium), which together contribute 60% of the total annual emission rate in the UK (Fowler et al., 2004a). Most of the reduced N is re-deposited over a much shorter distance than oxidised N, and for islands like the UK the export of NH_3 to other countries is negligible. The livestock industry is the single largest source of NH_3 and intensive livestock farms create significant local hotspots of elevated atmospheric NH_3 concentrations (Theobald et al., 2004). The rate of deposition is dependent on the downwind land use and its surface roughness. Deposition rates to a forest can be 2 to 3 fold larger than to shorter vegetation (Fowler et al., 2004b). Fowler et al. (1998) have shown that over 60% of the elevated NH_3 emitted was deposited to a downwind forest within 50 m, but declined to background concentrations within a few hundred meters. The high rates of N deposited to the forest floor increased the N content of the plant tissues and the soil and also soil emissions of N_2O (nitrous oxide) and NO (nitric oxide) (Pitcairn et al., 1998; Skiba et al., 2004). High N deposition rates will eventually result in changes in the forest floor plant communities to nitrogen tolerant species (Pitcairn et al., 1998).

The steep gradient of NH_3 deposition rates does provide an ideal opportunity to study the effect of different rates of N
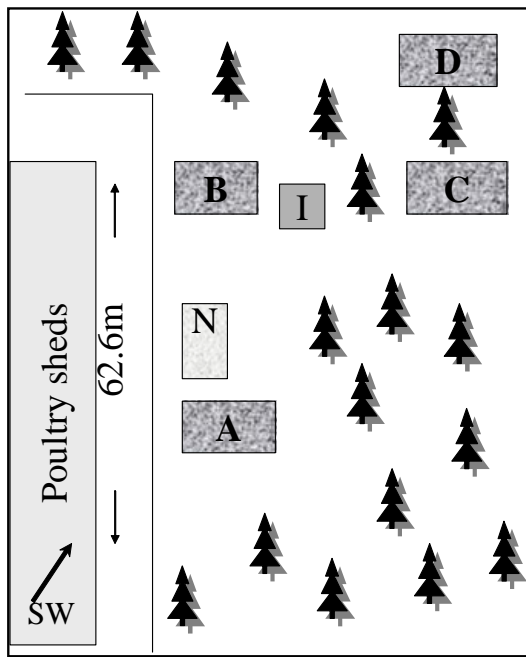
Fig. 1. A schematic diagram, not drawn to scale, of the study site. The prevailing wind direction is southwesterly (SW). Measurements were made 15 m (A), 25 m (B), 45 m (C) and 270 m (D) downwind from the centre of the closest shed. (I) represents the shed housing loggers, instruments and control unit for the autochambers. (N) represents the autochamber for N_2O flux measurements.

deposition on biological and chemical processes under similar climatic conditions. In this paper we have concentrated on the significance of an intensive poultry farm for increased downwind emissions of the atmospheric pollutants NO and N_2O.

2 Site description and methods

Measurements were made in a mixed woodland downwind of a large poultry farm in Scotland, which has been operating for about 40 years, from January to mid July 2004. The poultry farm grows broilers in 8 sheds (63 m long), each containing 20 000 birds on a 40-day cycle. The sheds are arranged in parallel pointing from NNW to SSE. The prevailing wind direction is from the SW. The edge of this farm borders onto the woodland located on freely drained brown earth (pH 3.2 in $CaCl_2$, 9.5% C and 0.44% N), containing a mixture of pine and birch with undergrowth of grasses in the sunnier parts and brambles, ferns and mosses dominant in the shadier parts.

Four sites were established at increasing distance downwind from the centre of the closest of the poultry sheds. The closest site (A) was established at the edge of the woodland at 15 m, directly opposite the centre of the shed. The second

site (B) was installed 17 m away from A and 15 m opposite the downwind edge of the poultry shed, and therefore was 25 m downwind of the centre of the farm building. Site C was located 18 m east of site A and 45 m downwind from the centre of the poultry shed. A "background" site (D) was installed 270 m downwind of the farm (NW) (Fig. 1)

Monthly concentrations of atmospheric NH_3 and NO_2 were monitored using continuous time-integrated sampling with passive diffusion samplers. Triplicate ALPHA samplers for NH_3 and triplicate modified GRADKO diffusion tubes for NO_2 were installed at a height of 1.5 m at all sites and were prepared and analysed within two weeks of collection according to standard protocols developed at CEH; by conductivity for NH_3 and by colorimetric methods for NO_2 (Tang et al., 2001). The ALPHA sampler method was rigorously tested against a reference active diffusion denuder method (Sutton et al., 2001). The GRADKO diffusion tube (Stevenson et al., 2001) was modified by addition of a turbulence damping membrane across the air inlet and validated (Bush et al., 2001).

Throughfall containers (2 per site), containing the biocide thymol, collected rainwater and material that has been dry deposited onto the leaves falling through the tree canopy over a $0.2\,m^2$ area at sites A–C. At the background site D, 3 rainfall collectors ($0.035\,m^2$) were installed at a height of 1.5 m. Throughfall and rainfall samples were bulked to provide a monthly sample analysed for NH_4^+ and NO_3^- concentrations by standard colorimetric methods.

For manual N_2O measurements by the static chamber approach, two small round polypropylene chambers fitted with a 4.5 cm wide outward facing flange (31 l) and covering a surface area of $0.123\,m^2$ were installed at each site for the duration of the field experiment (MacDonald et al., 1997). Fluxes were measured at roughly monthly intervals, by closing the chambers for a 1 h period around midday using lids manufactured from UV permeable polyvinylchloride attached in a dome shape fashion to a second flange. A circle of rubber attached to the underside of the flange acted as a seal when attached to the flange of the chamber during the measurement period. Samples were collected by syringe from a small port at the site of the chamber and were stored in 1 l Tedlar bags until analysis within 3 days of sample collection, by gas chromatography using an electron capture detector (ECD) detector. The detection limit of the ECD for N_2O was <20 ppb and the precision was 2%. Concentrations were calculated against an N_2O standard containing 1 ppm N_2O in a 20% O_2, 80% N_2 mix. The ECD response was linear in the range of 0.3 to >10 ppm N_2O. The bulk samples were analysed for N_2O by gas chromatography at least twice and more often if agreement between duplicates was poor.

In addition to the above described monthly samples at different locations in the forest the temporal variations of N_2O were monitored by collecting one daily sample from an automatically closing static chamber ($1\,m^2$, 130 l) positioned between sites A and B, 20 m downwind of the centre point

of the shed. The design of the autochamber is described in detail by Kitzler et al. (2005). The chamber was closed for one-hour periods every midday and samples were collected immediately after chamber closure and 45 and 60 min later. The samples were pumped at a flowrate of $100 \, ml \, min^{-1}$ into 20 ml glass vials for a period of 10 min. The vials were situated on a rotational fraction collector housed in a weather proof container and were collected for analysis of N_2O by gas chromatography at least every 14 days. Routine tests showed that over the 14-day period concentrations of 1 ppm N_2O did not deteriorate inside the vials. Fluxes were calculated from the difference between N_2O concentrations in ambient air (manual static chambers) or in chamber air immediately after closure (automatic static chamber), the headspace volume, and the surface area enclosed by the chamber and the length of enclosure time. The enclosure time of 1 h was routinely applied; tests showed that the N_2O concentration inside both chamber types increased linearly with time of closure for at least 2 h. There are views that the static enclosure method underestimates the real flux by not considering N_2O stored in the surface layers of the soil. The linear model developed by Conen and Smith (2000) to calculate the fraction of N_2O stored in soil air suggests that the potential underestimation by using static rather than dynamic chambers was 9% for the $1 \, m^2$ autochamber and 16% for the 40 cm diameter round chambers. These underestimates are not trivial and therefore absolute values should be treated with caution. However, as the same static chamber approach was used for all chambers, a comparison between sites receiving different rates of deposition is valid.

NO fluxes were measured by the dynamic chamber approach. At the three closest sites (A–C) 2 square automatically closing dynamic PTFE chambers (18 l) were installed for hourly NO, (NO_x-NO) and O_3 flux measurements per site. The chamber system is described in detail by Pilegaard et al. (1999). Two small fans stirred the air in each chamber. Upon closure ambient air from a single location, and 1 m above the soil surface close, was pushed through the chamber at a rate of $11 \, l \, min^{-1}$. Complete mixing of air inside the chambers was not demonstrated here, but was shown by Pilegaard et al. (1999) under very similar conditions. At the opposite chamber outlet the combined pump vacuum of the NOx and O_3 analysers drew air into the analysers at a rate of $2 \, l \, min^{-1}$. The pressure deficit in the chamber was $<0.5 \, Pa$, therefore not affecting the magnitude of the NOx fluxes measured. One minute average NO, (NO_x-NO) and O_3 concentrations were recorded on a 23x Campbell data logger. These data could be viewed and downloaded online via a modem. The analysers, logger and pumps were housed in a waterproof powered shed receiving mains power from the poultry house. NO and (NO_x-NO) was measured by chemiluminescence fitted with a molybdenum converter, the analyser provides an output of NO, NO_x and (NO_x-NO); the latter are all other nitrogenous species measured by the analyser and include NO_2, HNO_3, HONO, PAN, NH_3 and aerosol NH_4,
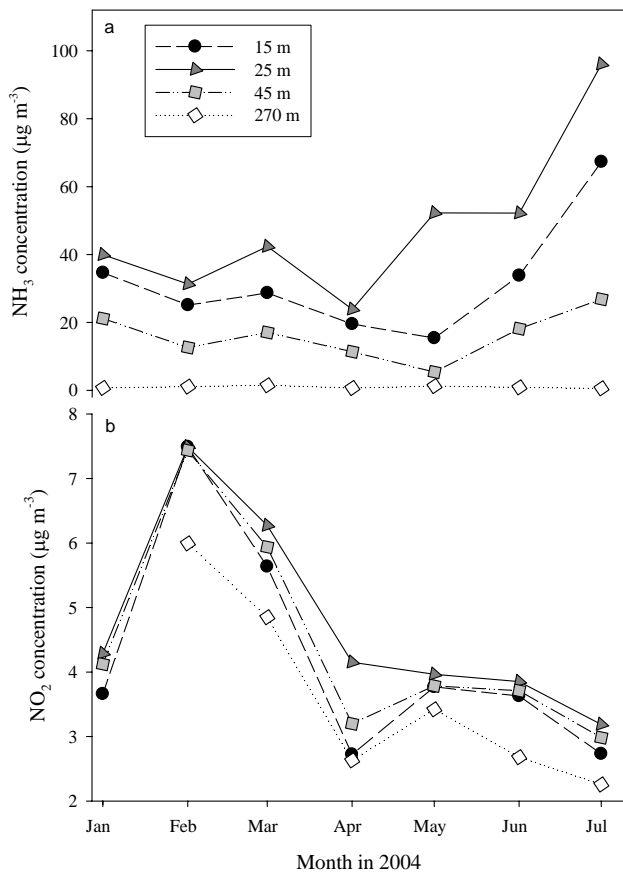
NO_3 and NO_2. The analyser had a detection limit of 0.3 ppb for NO and a response time of 40 s over a 10 s average period. The stability of the analyser was <0.4 ppb zero drift over a 24 h period. Calibration was performed by diluting 10.3 ppm NO in nitrogen by gas phase titration. Due to the excellent stability of the analyser this calibration was only performed at the beginning and end of the flux measurement period, but zero drift was monitored weekly using a portable calibrator. Ozone was measured using a dual channel photometric ozone analyser with a detection limit of 1 ppb.

The chambers closed in sequence for 8 min in the order Site A, Site B, a blank chamber fitted with PTFE bottom adjacent to the two chambers at site B and then site C. In the remaining 4 min of every hour ambient air was analysed for NO, (NO_x-NO) and O_3. In order to avoid long residence times of unstable gases in the supply lines (6 mm bore), these were kept to a maximum, but same length of 20 m. This maximum distance between chambers and analysers also determined the distances between the 3 transect sites. Unfortunately the fourth site was too far to be connected to the automated NO, (NO_x-NO) flux measurement system and the power supply. Fluxes were calculated from the concentration difference between the chamber and blank chamber, the flow rate of air through the chamber and the surface area of the chamber. The chemical reactions of NO with O_3 inside the chambers were taken into account, as described by Butterbach-Bahl et al. (1998).

Measurements of soil temperatures at 2, 5, 7, 10 cm depth using thermocouples and soil moisture by theta probe (Delta T) were made continuously. Fifteen-minute averages were stored on a Campbell 21x data logger. The theta probe voltage measurements were converted to volumetric moisture contents, using conversion factors established for the study soil, rather than using the company's standard calibration factors. Soils were analysed for KCl extractable (1 M) NH_4^+ and NO_3^- concentrations in the surface 10 cm once during the measurement period using established colorimetric methods (Henriksen and Slemer-Olsen, 1970).

3 Results

The monthly NH_3 concentrations measured at the three sites close to the farm were on average 20 to 40 times larger than at the background site, 270 m away (Fig. 2a). Largest NH_3 concentrations were measured at site B, which is further away from the centre of the farm building (25 m downwind of the farm) than site A; but due to its position in relation to the farm building and the prevailing SW wind direction receives the plume from all farm vents, whereas site A only receives the plume from 50% of the farm vents. This difference was reflected in the NH_3 concentration measurements (Fig. 2a). Average concentrations for the three sites 15, 25 and 45 m downwind were 34.3 ± 20.4, 47.6 ± 24.9 and

**Fig. 2.** Concentrations of atmospheric NH$_3$ **(a)** and NO$_2$ **(b)** downwind of the poultry farm.

Table 1. Estimated dry and measured wet N deposition to a mixed forest downwind of an NH$_3$ emitting poultry farm. Average of monthly cumulative samples (January–June 2004).

Distance from farm	NH$_3$-N[a] kg N ha^{-1}y^{-1}		NO$_2$-N[a] kg N ha^{-1}y^{-1}		NH$_4$-N[b] kg N ha^{-1}y^{-1}		NO$_3$-N[b] kg N ha^{-1}y^{-1}	
m	mean	stdev	mean	stdev	mean	stdev	mean	stdev
15	42.62	24.59	0.46	0.06	22.10	11.82	2.96	1.57
25	62.13	32.29	0.54	0.06	27.29	19.65	3.23	2.12
45	23.43	12.53	0.48	0.05	15.05	7.63	4.41	2.36
270	5.26	4.51	0.35	0.08	2.40[c]	1.36	1.43[c]	1.01

[a] estimated using a concentration dependent deposition velocities for NH$_3$ and a deposition velocity of 1.5 mm s^{-1} for NO$_2$;
[b] measured concentrations in throughfall;
[c] concentrations in rainwater
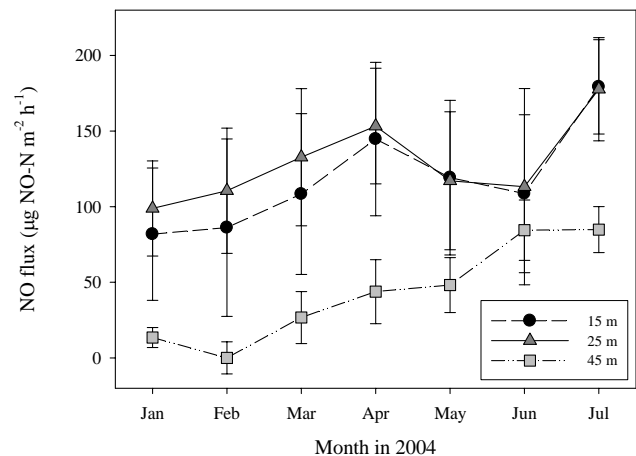
Fig. 3. Average monthly NO fluxes from hourly data of two automatically closing flow through chambers per site. The bars show standard errors of the temporal and spatial variability.

21.7±16.8 μg NH$_3$ m^{-3}. Background concentrations at the site 270 m downwind were 1.15±0.7 μg NH$_3$ m^{-3}.

The NO$_2$ concentrations did not differ between the three sites closest to the farm, but they were all slightly larger compared to the background site (Fig. 2b). This could simply reflect the frequent traffic of vehicles to the farm, which did not occur at the more remote background site. The average annual concentrations at sites A–C were 3.9±1.4, 4.3±1.3, 4.1±1.3 mg NO$_2$ m^{-3} and at the background site 3.1±1.4 mg NO$_2$ m^{-3}.

Rates of NH$_3$ deposition to the relatively open forest floor were estimated using deposition velocities calculated elsewhere, because necessary meteorological variables to calculate deposition velocities from first principles were not measured in this forest. The same concentration dependent deposition velocities as used by Fowler et al. (1998) for the same forest a few years earlier were applied in this study. These NH$_3$ deposition velocities were based on observations that over a moorland vegetation canopy resistance increased from 20 s m^{-1} at 0.3 μg NH$_3$ m^{-3} to 50 s m^{-1} at 2 μg NH$_3$ m^{-3} for example for wet canopy surfaces (Flechard and Fowler, 1998). Average NH$_3$ deposition rates close to the farm may

therefore be 62, 42, 23 kg NH$_3$-N ha^{-1} y^{-1} compared to the background deposition rate of 5 kg NH$_3$-N ha^{-1} y^{-1} (Table 1). The uncertainties in assuming same canopy and aerodynamic resistances for a forest and moorland ecosystem at slightly different locations are high. However, in comparison the deposition of NH$_4^+$, measured by the bulk samplers, do show almost identical fractional increases above background deposition rates in rainwater as calculated for dry NH$_3$ deposition rates (Table 1). Monthly NH$_4^+$ concentrations in the bulk collectors were at least 6 times larger than those in rainwater and mostly followed the same pattern observed for NH$_3$, with largest concentrations measured at site B, 25 m downwind, and smallest at site C, 45 m downwind. The ranking order of NH$_4^+$ concentrations, site B > site A > site C, was observed in most months. Nitrate concentrations in the bulk collectors were at least 2 times larger than in rainfall, however differences between sites were negligible. The relationship between the NH$_4^+$ concentration in the bulk collectors and dry deposition of NH$_3$ alone was linear and highly
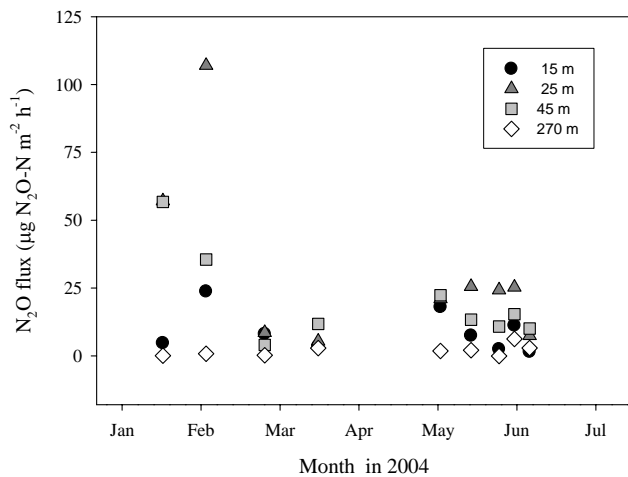
Fig. 4. Nitrous oxide emission downwind of the poultry farm; average fluxes from 2 manual static chambers per site.

Fig. 5. The relationship between monthly average NO flux and NH_3 concentrations at the three transect sites downwind of the poultry farm.

correlated, with $r^2 > 90\%$ for the two sites closest to the farm (A and B).

Assuming that the calculations of dry deposition rate, using the concentration dependent deposition rates, are correct, then in this forest the dry deposition of NH_3 was more important than the wet deposition of NH_4^+.

Rates of NO_2 deposition, assuming a deposition velocity of 1.5 mm s^{-1} (Duyzer, personal communication), where almost two orders of magnitude smaller than NH_3 deposition rates. Close to the farm the bulk deposition of oxidised N, as measured in the throughfall, was at least 7 fold smaller than that of reduced N (Table 1).

The increased rate of N deposition to the woodland was reflected in the soil available NH_4^+ and NO_3^- concentrations, measured on one occasion (31 March 2004). The same pattern as observed for the deposition rates, with largest concentrations at site B (25 m), followed by site A (15 m) and then C (45 m) was followed. The soil extractable NH_4^+ and NO_3^- concentrations were 6.8 ± 6.0, 10.3 ± 4.1, $3.2\pm1.9\,\mu g\ NH_4^+$-N $g_{dry\ soil}^{-1}$ and 3.0 ± 1.5, 9.9 ± 5.3, $0.5\pm0.4\,\mu g\ NO_3^-$-N $g_{dry\ soil}^{-1}$ at site A, B and C with n=6, 9, 6, respectively.

The increased mineral N concentrations along this NH_3 deposition gradient provide the substrate for the nitrifying and denitrifying microbes, and consequently have increased the emissions of NO and to a lesser extent N_2O (Figs. 3 and 4). Average NO fluxes for the entire study period for site A, B and C were 111.2 ± 41.1, 123.3 ± 40.7, $38.3\pm28.8\,\mu g$ NO-N m^{-2} h^{-1}. The differences between the three sites were statistically significant at $p<0.001$. Daily NO emissions at site A and B were larger than those measured at site C on 93% and 95% of the 190 days for which flux measurements could be calculated, respectively. Fluxes measured at site B were larger than those measured at site A on 69% of the 190 days. The relationship between the monthly average NO flux
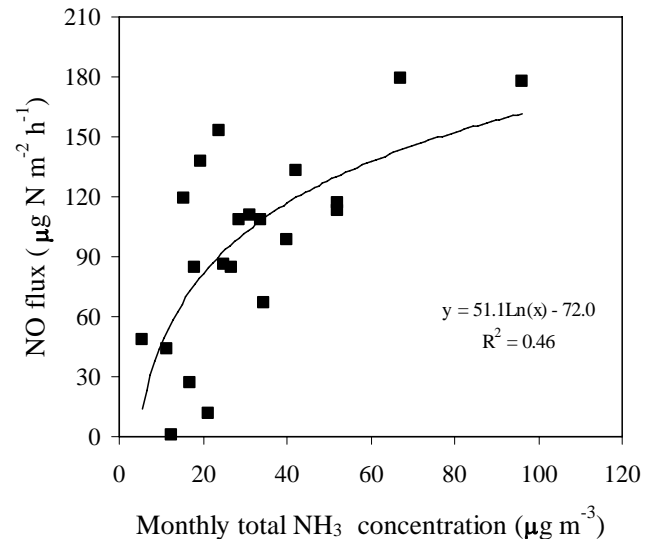
and monthly cumulative NH_3 concentration was log linear and significant ($p<0.001$) (Fig. 5).

For N_2O, average fluxes for the 8 measurements were 9.9 ± 7.5, 34.3 ± 33.3, 21.2 ± 6.1 and $1.8\pm2.5\,\mu g\ N_2O$-N m^{-2} h^{-1} for site A, B, C and the background site D respectively. Standard deviations of the mean were large due to temporal and spatial variability. The autochamber positioned close to sites A was employed to provide 1 daily N_2O flux measurement. The temporal scatter of the N_2O fluxes was normally distributed over the entire measurement period and the average flux ($12.9\pm15\,\mu g\ N_2O$-N m^{-2} h^{-1}, n=136) was in reasonable agreement with the monthly static chamber measurements from site A. At site B largest flux rates were measured and at site C occasional negative fluxes of N_2O, indicating N_2O uptake, were measured (Fig. 4). None of these differences were statistically significant. Fluxes at site C were larger than those at site A, possibly caused by the slightly wetter soil conditions at site C. Average volumetric soil moisture contents for site C were $35.1\pm6.2\%$ and at sites A and B $31.3\pm2.3\%$ and $31.9\pm5.2\%$, respectively.

Nitric oxide fluxes were significantly influenced by daily and seasonal temperature changes. NO emissions followed a diurnal pattern with maximum emissions approximately 3 h after noon at sites A and B (Fig. 6). The oscillation frequency was much larger for locations where large NO fluxes were measured compared to locations where the NO flux was small. Daily average NO fluxes correlated significantly with daily average soil temperatures measured at the soil surface and 2, 5 and 10 cm below the soil surface. The daily average soil temperature ranged between −1 and 13°C at the soil surface and between 4.3 and 11.7°C at a soil depth of 10 cm. For NO the correlations were significant at $p>0.001$ at all
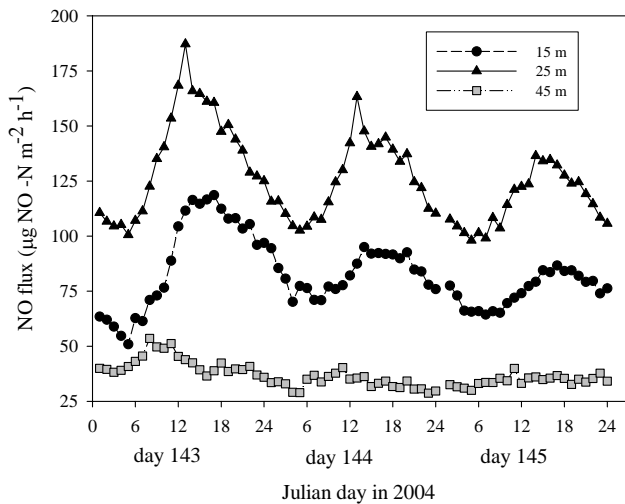
Fig. 6. The diurnal variations in NO flux from 2 chamber each at 15, 25 and 45 m downwind of the poultry farm in May 2004. Data are averages from two chambers.

soil depths. Best linear correlations were observed between NO flux and the soil surface temperature and a depth of 2 cm depth for plots at site A and B. At site C, however, best correlations were obtained with soil temperatures at 5 and 10 cm depth. The linear relationships between the inverse absolute soil temperature and the natural logarithm of the NO flux at site A and B provided r^2 values of 46% and 24% (n=186) and activation energies, calculated from the slope of the relationship, of 59 and 33 kJ mol^{-1}. For site C the r^2 was 39.4% (n=234) and the activation energy was 140 kJ mol^{-1}. For N$_2$O the relationship with soil temperature was established over a 15 day period, when the autochamber was used to measure fluxes every 6 h for 15 days. No consistent diurnal pattern could be established from this work.

The soil moisture content for the entire study period varied between 16 and 63% (v/v). A linear relationship between NO flux and soil moisture content was only observed at site C where NO emissions decreased with increasing soil moisture content (r^2=0.001). For N$_2$O fluxes measured by static and auto chambers no linear relationship with soil moisture was observed. However, fluxes >27 μg N$_2$O-N m^{-2} h^{-1} were only observed at soil moisture contents above 30% (v/v).

4 Discussion

We have shown that point source emissions of NH$_3$ significantly increased atmospheric concentrations of NH$_3$ downwind of the source, implying increased rates of dry deposition of NH$_3$ and wet deposition of NH$_4^+$ to the close by woodland. Ammonia concentrations, measured inside the woodland downwind of the farm, were between 20 and 40 times larger than the background concentrations of 1.15 μg

m^{-3} 270 m downwind. The background concentrations are within the range of concentrations measured and modelled for UK agricultural fields (Dragosits et al., 2002). Also the FRAME (Fine Resolution Atmospheric Multi-species Exchange, Singles et al., 1998) model output for the 5 km grid square, in which the study farm is situated, estimated a grid average NH$_3$ concentration of 1.32 μg m^{-3}. Similar elevated NH$_3$ concentrations and relatively fast decay rates over a short distance (<300 m) have been observed at several woodlands downwind of intensive poultry and pig farms (e.g. Theobald et al., 2004; Spangenberg and Kölling, 2004) and also downwind of an artificial NH$_3$ release source over an ombrotrophic bog (Leith et al., 2004). Thus a tree shelterbelt can be ideal in not only screening an unsightly animal farm from the public, but also by creating a biological filter, which contains the pollution to a small local area. The side effect, however, is that the enhanced N deposition can increase the soil mineral N content (Skiba et al., 2004) and consequently the emission of the atmospheric pollutants N$_2$O and NO and also can increase the risk of loss or change in biodiversity. Pitcairn et al. (1998) have reported a prevalence of N loving species and lack of N sensitive species close to the poultry farm discussed in this paper. Due to the close distance to this very large NH$_3$ point source, the total N deposition appears to be dominated by dry deposition of NH$_3$, rather than wet deposition of NH$_4^+$ and NO$_3^-$, which was the case for the transect of European forests investigated under NOFRETETE (Pilegaard et al., 2006). Both studies have shown strong relationships between N deposition and NO emission. A linear relationship between NO flux and wet deposition of N was observed for the coniferous forests of the European transect studied in the NOFRETETE project (Pilegaard et al., 2006). For this forest downwind of the poultry farm, where environmental variations are minimal in comparison to the European transect, a log linear relationship between NO flux and atmospheric NH$_3$ concentration was demonstrated (Fig. 5). In addition to nitrogen, the most important variables that influenced NO emission rates were soil moisture and soil temperature. The literature supports the observations that NO increased linearly with increased soil temperature, but decreased with increasing soil moisture content once the optimum moisture content has been passed (Kitzler et al., 2005; Davidson et al., 2000; van Dijk and Duyzer, 1999; Skiba et al., 1997). The much larger activation energy calculated for the relationship between soil temperature and NO flux at site C, suggests that conditions were less optimal for NO to be produced and emitted compared to the closer sites receiving larger rates of N deposition (Skiba et al., 1992).

For both studies, the European transect studied in the NOFRETETE project (Pilegaard et al., 2006) and the forest downwind of the poultry farm, relationships between N deposition rate and N$_2$O emission were very weak. However, the trend of decreasing N$_2$O emissions with decreasing NH$_3$ concentrations and distance away from the farm are in

good agreement with previous measurements from the same forest in 1995 (5 dates) and 1997 (3 dates) when the same chambers, this time three replicates, and the same measurement strategy was employed (Skiba et al,, 1998). For the poultry farm also relationships between N_2O flux and soil moisture and soil temperature changes were much weaker than for NO. Possible reasons are that the data set for N_2O in this study is much smaller than for NO (monthly rather than hourly data), but perhaps more importantly, that the environmental variables, particularly the redox potential of the soil, most commonly measured by the surrogates soil moisture or water filled pore space, exert a stronger control on N_2O than NO.

Overall the NO and N_2O emissions downwind of this poultry farm were larger than the maximum annual emissions calculated for the European transect (Pilegaard et al., 2006). Downwind of the poultry farm maximum emissions for the study period were 123 μg NO-N m^{-2} h^{-1} and 34.3 μg N_2O-N m^{-2} h^{-1} compared to 75 and 81 μg NO-N m^{-2} h^{-1} in the Dutch and south German coniferous forests and 20 μg N_2O-N m^{-2} h^{-1} in the Hungarian and Italian forests.

On average the NO emissions expressed as a fraction of the elevated N deposited as throughfall were 39% (at 15 m), 30% (at 25 m) and 17% (at 45 m) downwind of the farm, whereas for N_2O the emissions were 3.5% (at 15 m), 9.8% (at 25 m), 9.5% (at 45 m) and 4.1% (at 270 m) downwind of the farm. Due to high spatial and temporal variability the uncertainty in these emission factors is high, 50% for NO and 80% for N_2O. However these emission fractions exceed the emission factor of 1% ($\pm$50%) advised by the IPCC for N_2O emissions resulting from atmospheric N deposition (Houghton et al., 2001) for some of the sites.

5   Conclusion

This study has shown that increased atmospheric nitrogen deposition rates increased the emissions of NO and N_2O. For N_2O the rate of increase was larger than predicted by the IPCC (Houghton et al., 2001).

Although the emission rates of atmospheric pollutants downwind of intensive NH_3 emitting livestock farms were large, the contribution to the total national NO and N_2O emissions are negligible. If we assume the conditions and emission factors observed in this study are true for the entire British poultry flock (360 million), which emits 28 kt NH_3 y^{-1} (calculated from standard NH_3 emission factors for poultry manure); then intensive poultry farming will contribute an additional 36 t NO-N y^{-1} and 10 t N_2O-N y^{-1}. This is only a small percentage of the total annual NO and N_2O emitted from British soils of 7 kt NO-N y^{-1} and 56 kt N_2O-N y^{-1} (Skiba et al., 2001).

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