

The relationship between ammonia emissions from a poultry farm and soil NO and N2O fluxes from a downwind source

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The relationship between ammonia emissions from a poultry farm and soil NO and N₂O fluxes from a downwind source

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Abstract

Intensive livestock farms emit large concentrations of NH₃, most of which is deposited very close to the source. The presence of trees enhances the deposition. Rates to down wind forests can exceed 40 kg N ha⁻¹. The steep gradient in large NH₃ concentration and deposition at the edge of a downwind forest to background concentrations within a few hundred meters provides an ideal site to study the effect of different rates of N deposition on biological and chemical processes under similar environmental conditions. We have investigated the effect of different rates of NH₃ deposition (62, 45, 24 and 5 kg NH₃-N ha⁻¹ y⁻¹) on the flux of NO and N₂O 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 N2O fluxes close to the farm and monthly at all sites and monthly cumulative wet and dry N deposition. The increased NH₃ and NH₄ deposition to the woodland increased emissions of NO and N₂O and soil available NH₄⁺ and NO₃⁻ concentrations. Average NO and N₂O fluxes measured 15, 25 and 45 m downwind of the farm were 111.2 \pm 41.1, 123.3 \pm 40.7, 38.3 \pm 28.8 μ g NO-N m $^{-2}$ h $^{-1}$ and 9.9 ± 7.5 , 34.3 ± 33.3 and $21.2\pm6.1 \,\mu g \, N_2 \, O - N \, m^{-2} \, h^{-1}$, respectively. At the background site 270 m downwind the N_2O flux was reduced to $1.75\pm2.1\,\mu\mathrm{g}\,N_2O$ -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 3h after noon. For N₂O no consistent diurnal pattern was observed. Changes in soil moisture content had a less clear effect on the NO and N₂O flux. On average the NO emissions expressed as a fraction of the elevated N deposited were 7.1% (at 15 m), 6% (at 25 m) and 2.3% (at 45 m) downwind of the farm, whereas for N₂O the emissions were only 2.8% (at 15 m), 3% (at 25 m) and 3% (at 45 m) downwind. These emission fractions exceed the emission factor of 1% advised by the IPCC for N₂O emissions resulting from atmospheric N deposition.

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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., 2004). In many areas N deposition is dominated by reduced N, NH₃ (ammonia) and NH₄ (ammonium), which together contribute to 60% of the total annual emission rate in the UK (Fowler et al., 2004). Most of the reduced N is deposited over a much shorter distance than oxidised N, and for islands like the UK the export of NH₃ to other countries is negligible. The livestock industry is the single largest source of NH₃ and intensive livestock farms create significant local hotspots of elevated atmospheric NH₃ concentrations (Theobald et al., 2004). The rate of deposition is dependent on the downwind land use and the surface roughness. Deposition rates to a forest can be 2 to 3 fold larger than to shorter vegetation (Fowler et al., 2004). Fowler et al. (1998) have shown that over 60% of the elevated NH₃ 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 N₂O (nitrous oxide) and NO (nitric oxide) (Pitcairn et al., 1998; Skiba et al., 2004). Such high emissions will eventually result in changes in the forest floor plant communities to nitrogen tolerant species (Pitcairn et al., 1998). The steep gradient of NH₃ deposition rates does provide an ideal opportunity to study the effect of different rates of N 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 N₂O.

2. Site description and methods

Measurements were made in mixed woodland downwind of a large poultry farm in Scotland, which has been operating for about 40 years, from January to mid July 2004.

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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 CaCl₂, 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).

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 diffusion tubes for NO_2 were installed at a height of 1.5 m at all sites and were prepared and analysed according to standard protocols developed at CEH (Tang et al., 2001). Throughfall containers (2 per site) containing the biocide thymol collected rainwater falling through the tree canopy over a $0.2\,\text{m}^2$ area at sites A-C. At the background site D 3 rainfall collectors (0.035 m²) 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 (31 l) (MacDonald et al., 1997) were installed. Fluxes were measured at roughly monthly intervals, by closing the chambers for 1 h periods. Samples were collected by syringe and stored in Tedlar bags until analysis within 3 days of sample collection, by gas chromatography using an electron capture detector (ECD). A daily sample of the N_2O flux was obtained from an automatically closing chamber (1 m², 130 l) positioned between site A and B, 20 m downwind of the centre point of the

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shed, and is described in detail by Kitzler et al. (2005). Chambers were closed for one-hour periods every midday and samples were stored in glass vials for up to 14 days prior analysis for N₂O by gas chromatography using an ECD detector. Routine tests showed that over the two-week period concentrations of 1 ppm N₂O did not deteriorate inside the vials.

NO fluxes were measured by the dynamic chamber approach. At the three closest sites (A-C) 2 square automatically closing dynamic PTFE chambers (181) were installed for hourly NO, NO₂ and O₃ flux measurements. 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, was pushed through the chamber at a rate of 111 min⁻¹. At the opposite chamber outlet the combined pump pressure of the NO_x and O₃ analysers drew air into the analysers at a rate of 21 min⁻¹. One minute average NO, NO₂ and O₃ 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.

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_x 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_x 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

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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) NH₄⁺ and NO₃⁻ concentrations in the surface 10 cm twice during the measurement period.

3. Results

The monthly NH₃ 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., 1a). Largest NH₃ 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 NH₃ concentration measurements (Fig. 1a). Average concentrations for the three sites 15, 25 and 45 m downwind were 34.3 ± 20.4 , 47.6 ± 24.9 and $21.7\pm16.8\,\mu g\,NH_3\,m^{-3}$. Background concentrations at the site 270 m downwind were $1.15\pm0.7 \mu g NH_3 m^{-3}$.

The NO₂ concentrations did not differ between the three sites closest to the farm, but they were all slightly larger compared to the background site (Fig. 1b). 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 , $4.1\pm1.3 \mu g NO_2 m^{-3}$ and at the background site $3.1\pm1.4 \mu g NO_2 m^{-3}$.

Rates of NH₃ deposition to the forest floor were calculated using concentration dependent deposition velocities for NH₃ as described by Fowler et al. (1998), and a deposition velocity of 1.5 mm s⁻¹ was applied for NO₂ (Duyzer, pers. comm.). Average NH_3 deposition rates close to the farm were calculated at 62, 42, 23 kg NH_3 -N ha⁻¹ y⁻¹ compared to the background deposition rates of 5 kg NH₃-N ha⁻¹ y⁻¹ (Table 1).

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Cumulative NH₄⁺ concentrations in throughfall were at least 6 times larger than those in rainwater and followed the same pattern observed for NH₃, with largest concentrations measured at site B, 25 m downwind, and smallest at site C, 45 m downwind (Table 1). The ranking order of NH₄ concentrations, site B > site A > site C, was observed in most months. Nitrate concentrations in throughfall were at least 2 times larger than in rainfall, however differences between sites were negligible.

In this forest the dry deposition of NH_3 was more important than the wet deposition of NH_4^+ . A two fold difference between NH_3 deposition and the rate of NH_4^+ deposition was observed at the sites closest to the farm (A & B). This difference was reduced to a 1.5 fold difference at the site C further away from the farm. The relationship between the NH_4^+ concentration in the throughfall and dry deposition of NH_3 was linear and highly correlated, with $r^2 > 90\%$ for the two sites closest to the farm (A & B).

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±1.9 μ g NH $_4^+$ -N g $_{dry\,soil}^{-1}$ and 3.0±1.5, 9.9±5.3, 0.5±0.4 μ g NO $_3^-$ -N g $_{dry\,soil}^{-1}$ at site A, B and C with n=6, 9, 6, respectively.

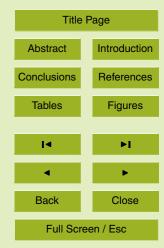
The increased mineral N concentrations along this NH_3 deposition gradient provide the substrate for the nitrifying and denitrifying microbes, and consequently have increased the emissions of NO and to a lesser extent N_2O (Figs. 2 and 3). Average NO concentrations for the entire study period for site 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

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significant (p<0.001) (Fig. 4).

For N₂O average fluxes for the 8 measurements were 9.9 ± 7.5 , 34.3 ± 3.3 , 21.2 ± 6.1 and $1.8\pm3.5\,\mu g\,N_2O$ -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. At site B largest flux rates were measured and at site C occasional negative fluxes of N₂O, indicating N₂O uptake, were measured (Fig. 3). None of these differences were statistically significantly. Fluxes at site C were larger than those at site A, possibly caused by the slightly wetter soil conditions at site C. Average soil moisture contents for site C were $35.1\pm6.2\,g\,g_{dry\,wt}^{-1}$ and at sites A and B $31.3\pm2.3\,g\,g_{dry\,wt}^{-1}$ and $31.9\pm5.2\,g\,g_{dry\,wt}^{-1}$, respectively.

Nitric oxide concentrations were significantly influenced by daily and seasonal temperature changes. NO emissions followed a diurnal pattern with maximum emissions approximately 3 h after noon (Fig. 5). 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 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 r² 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 r² was 39.4% (n=234) and the activation energy was 140 kJ mol⁻¹. For N₂O the relationship with soil temperature at all depth was not significant and no diurnal pattern was observed.

The soil moisture content for the entire study period varied between 16 and 63% (v/v). A linear relationships between NO flux and soil moisture content was only ob-

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served at site C where NO emissions decreased with increasing soil moisture content (r^2 =0.001). For N₂O fluxes measured by static and auto chambers no linear relationship with soil moisture was observed. However, fluxes >27 μ g N₂O-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 NH₃ significantly increased the rate of NH₃ deposition 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 $1.15 \,\mu \mathrm{g}\,\mathrm{m}^{-3}$ at 270 m downwind. The background concentrations are within the range of concentrations measured and modelled for UK agricultural fields (Dragosits et al., 2002); and 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 NH₃ concentration of 1.32 μ g m⁻³. Similar elevated NH₃ 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 NH₃ release source over an ombrotrophic bog (Leith et al., 2004). Thus a 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 increases the emission of the atmospheric pollutants N₂O and NO and increases 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 NH₃ point source, the total N deposition is dominated by dry deposition of NH₃ rather than wet deposition of NH₄ and NO₃, which was the case for the transect of European forests investigated

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under NOFRETETE (Pilegaard et al., 2005¹). 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., 2005¹). For this forest downwind of the poultry farm, where environmental variations are minimal in comparison to the European transect, a log linear relationship was demonstrated (Fig. 4). 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 (Kitzler et al., 2005; van Dijk and Duyzer, 1999; Skiba et al., 1997).

For both studies, the European transect studied in the NOFRETETE project (Pilegaard et al., 2005^1) and the forest downwind of the poultry farm, relationships between N deposition rate and N₂O emission were very weak. For the poultry farm also relationships between N₂O flux and soil moisture and soil temperature changes were much weaker than for NO. Possible reasons are the data set for N₂O in this study is much smaller than for NO (monthly rather than hourly data), but perhaps more importantly, 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 N₂O than NO.

Overall the NO and N₂O emissions downwind of this poultry farm were larger than the maximum annual emissions calculated for the European transect (Pilegaard et al., 2005^1). Downwind of the poultry farm maximum emissions for the study period were $123\,\mu g\,NO-N\,m^{-2}\,h^{-1}$ and $34.3\,\mu g\,N_2O-N\,m^{-2}\,h^{-1}$ compared to 75 and 81 $\mu g\,NO-N\,m^{-2}\,h^{-1}$

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¹Pilegaard, K., Skiba, U., Ambus, P., Beier, C., Bruggemann, N., Butterbach-Bahl, K., Dick, J., Dorsey, J., Duyzer, J., Gallagher, M.,Gasche, R.,Horvath, L., Kitzler, B., Leip, A., Pihlatie, M. K., Rosenkranz, P., Seufert, G., Vesala, T., Westrate, H., and Zechmeister-Boltenstern, S.: Nitrogen load and forest type determine the soil emission of nitrogen oxides (NO and N₂O), Biogeosciences Discuss., in preparation, 2005.

N m⁻² h⁻¹ in the Dutch and south German coniferous forests and 20 μ g N₂O-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 were 7.1% (at 15 m), 6% (at 25 m) and 2.3% (at 45 m) downwind of the farm, whereas for N_2O the emissions were only 2.8% (at 15 m), 3% (at 25 m) and 3% (at 45 m) downwind. These emission fractions greatly exceed the emission factor of 1% advised by the IPCC for N_2O emissions resulting from atmospheric N deposition (Houghton et al., 2001). Even at the background site, 270 m downwind of the farm the N_2O emissions accounted for 1.6% of the N deposition.

5. Conclusions

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 $_2$ O 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 $_2$ O-N y $^{-1}$. This is only a small percentage of the total NO (7 kt NO-N y $^{-1}$) and N $_2$ O (56 kt N $_2$ O-N y $^{-1}$) emitted from British soils (Skiba et al., 2001).

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Table 1. Dry and wet N deposition to a mixed forest downwind of an NH₃ emitting poultry farm. Average of monthly cumulative samples (January–June 2004).

Distance from farm	NH ₃ -N kg N ha ⁻¹ y ⁻¹		NO_2 -N y^{-1} kg N ha ⁻¹ y ⁻¹		NH ₄ -N kg N ha ⁻¹ y ⁻¹		NO ₃ -N 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*	1.36	1.43*	1.01

^{*} concentrations in rainwater

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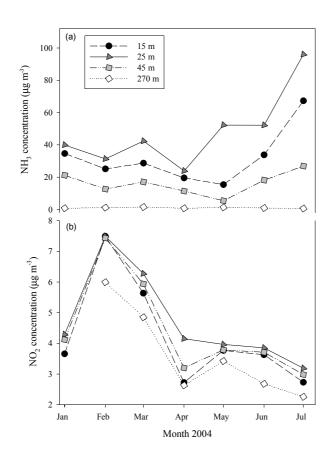


Fig. 1. Concentrations of dry deposited NH_3 (a) and NO_2 (b) downwind of the poultry farm.

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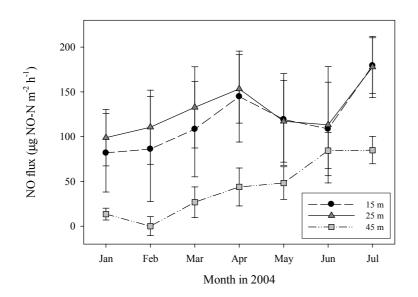


Fig. 2. Average monthly NO fluxes from hourly data of two automatically closing flow through chambers per site. The bars show standard errors of the temporal and spatial variability.

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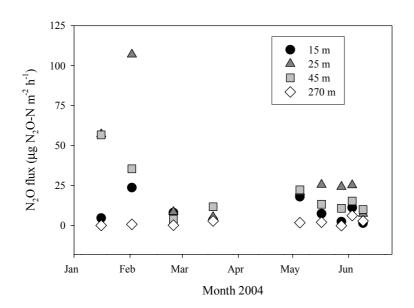
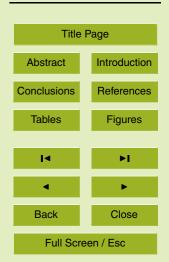


Fig. 3. Nitrous oxide emission downwind of the poultry farm; average fluxes from 2 manual static chambers per site.

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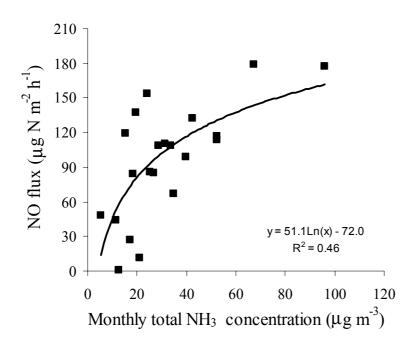


Fig. 4. The relationship between monthly average NO flux and NH₃ concentrations at the three transect sites downwind of the poultry farm.

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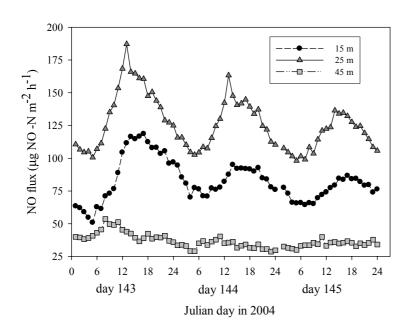


Fig. 5. The diurnal variations in NO flux from 2 chamber each at 15, 25 and 45 m downwind of the poultry farm in May 2004. Data are averages from two chambers.

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