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Ammonia deposition and N trace gas emissions

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The relationship between ammonia emissions from a poultry farm and soil NO and N_2O fluxes from a downwind source

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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 down wind forests can exceed $40\,kg\,N\,ha^{-1}$. The steep gradient in large NH_3 concen-
tration 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 con-
ditions. We have investigated the effect of different rates of NH_3 deposition (62, 45, 24 and $5\,kg\,NH_3\text{-}N\,ha^{-1}\,y^{-1}$) on the flux of NO and N_2O from soil in a mixed wood-
land 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, $38.3\pm28.8\,\mu g\,NO\text{-}N\,m^{-2}\,h^{-1}$ and 9.9 ± 7.5, 34.3 ± 33.3 and $21.2\pm6.1\,\mu g\,N_2O\text{-}N\,m^{-2}\,h^{-1}$, respectively. At the background site 270 m downwind the N_2O flux was reduced to $1.75\pm2.1\,\mu g\,N_2O\text{-}N\,m^{-2}\,h^{-1}$. NO emissions were significantly influenced by seasonal and daily changes in soil temper-
ature 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. 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 exceed the emission factor of 1% advised by the IPCC for N_2O 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 $5\,\text{kg}\,\text{N}\,\text{ha}^{-1}\,\text{y}^{-1}$ in pristine environments to over $80\,\text{kg}\,\text{N}\,\text{ha}^{-1}\,\text{y}^{-1}$ in the most polluted locations of Europe (Fowler et al., 2004). In many
5   areas N deposition is dominated by reduced N, NH_3 (ammonia) and NH_4^+ (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 NH_3 to other countries is negligible. The livestock industry is the single largest source of NH_3 and intensive livestock farms
10 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 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 NH_3 emitted was deposited to a downwind forest within 50 m, but declined to
15 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). 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
20 NH_3 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 N_2O.

2. Site description and methods

25 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 $CaCl_2$, 9.5% C and 0.44%

5 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

10 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 con-

15 tinuous 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 $0.2\,m^2$ area

20 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 (31 l) (MacDonald et al., 1997) were installed. Fluxes were

25 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 ($1\,m^2$, 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 N_2O by gas chromatography using an ECD detector. Routine tests showed that over the two-week period concentrations of 1 ppm N_2O did not deteriorate
5 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 (18 l) were installed for hourly NO, NO_2 and O_3 flux measurements. The chamber system is described in detail by Pilegaard et al. (1999). Two small fans stirred the air in each
10 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 11 l min^{-1}. At the opposite chamber outlet the combined pump pressure of the NO_x and O_3 analysers drew air into the analysers at a rate of 2 l min^{-1}. One minute average NO, NO_2 and O_3 concentrations were recorded on a 23x Campbell data logger. These data could be viewed and downloaded
15 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
20 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 concen-
25 tration 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 twice during the measurement period.

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,. 1a). 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. 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±16.8 $\mu g\, NH_3\, m^{-3}$. Background concentrations at the site 270 m downwind were 1.15±0.7 $\mu g\, NH_3\, m^{-3}$.

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. 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±1.3 $\mu g\, NO_2\, m^{-3}$ and at the background site 3.1±1.4 $\mu g\, NO_2\, m^{-3}$.

Rates of NH_3 deposition to the forest floor were calculated using concentration dependent deposition velocities for NH_3 as described by Fowler et al. (1998), and a deposition velocity of 1.5 mm s^{-1} was applied for NO_2 (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 NH_3-N ha^{-1} y^{-1} (Table 1).

Cumulative NH_4^+ concentrations in throughfall were at least 6 times larger than those in rainwater and 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 (Table 1). The ranking order of NH_4 concentrations, site B > site A > site C, was

5 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

10 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

15 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 $\mu g\,NH_4^+\text{-N}\,g^{-1}_{dry\,soil}$ and 3.0±1.5, 9.9±5.3, 0.5±0.4 $\mu g\,NO_3^-\text{-N}\,g^{-1}_{dry\,soil}$ at site A, B and C with n=6, 9, 6, respectively.

20 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±28.8 $\mu g\,NO\text{-N}\,m^{-2}\,h^{-1}$. The differences between the three sites

25 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 N_2O average fluxes for the 8 measurements were 9.9 ± 7.5, 34.3 ± 33.3, 21.2 ± 6.1 and $1.8\pm3.5\,\mu g\,N_2O\text{-}N\,m^{-2}\,h^{-1}$ 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 N_2O, indicating N_2O 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 $35.1\pm6.2\,g\,g^{-1}_{dry\,wt}$ and at sites A and B $31.3\pm2.3\,g\,g^{-1}_{dry\,wt}$ and $31.9\pm5.2\,g\,g^{-1}_{dry\,wt}$, 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 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_2O 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 N_2O fluxes measured by static and auto chambers no linear relationship with soil moisture was observed. However, fluxes >27 $\mu g\,N_2O$-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 the rate of NH_3 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 $\mu g\,m^{-3}$ 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 NH_3 concentration of 1.32 $\mu g\,m^{-3}$. 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 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 N_2O 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 NH_3 point source, the total N deposition is 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

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under NOFRETETE (Pilegaard et al., 2005[1]). Both studies have shown strong rela-
tionships 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., 2005[1]). For this forest
downwind of the poultry farm, where environmental variations are minimal in compar-
ison 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 (Pile-
gaard et al., 2005[1]) and the forest downwind of the poultry farm, relationships between
N deposition rate and N_2O emission were very weak. For the poultry farm also rela-
tionships between N_2O flux and soil moisture and soil temperature changes were much
weaker than for NO. Possible reasons are the data set for N_2O 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 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., 2005[1]). 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-

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[1]Pilegaard, 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,
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$N m^{-2} h^{-1}$ in the Dutch and south German coniferous forests and $20 \, \mu g \, N_2O$-$N \, m^{-2} \, h^{-1}$ in the Hungarian and Italian forests.

On average the NO emissions expressed as a fraction of the elevated N deposited 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_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 NO ($7 \, kt \, NO$-$N \, y^{-1}$) and N_2O ($56 \, kt \, N_2O$-$N \, y^{-1}$) 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 NH_3 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 $kg\,N\,ha^{-1}\,y^{-1}$		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

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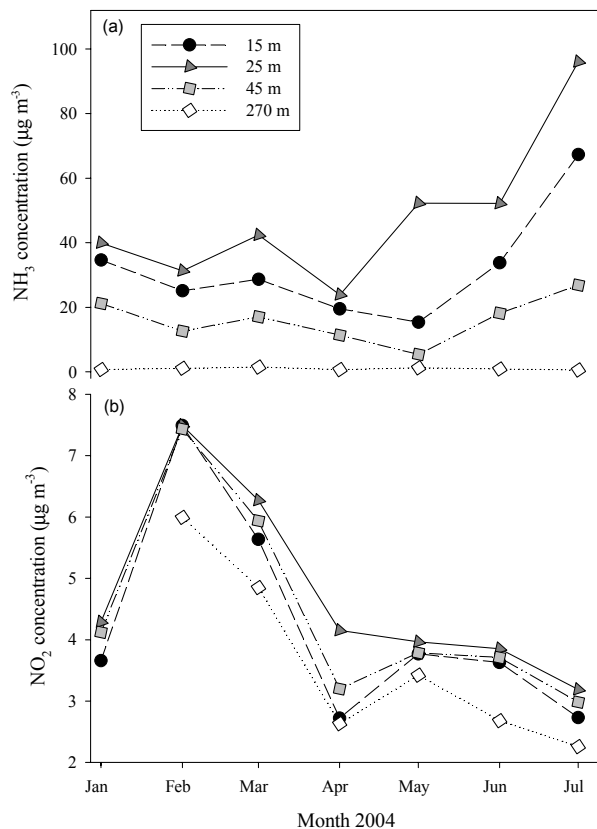
Fig. 1. Concentrations of dry deposited NH_3 **(a)** and NO_2 **(b)** downwind of the poultry farm.

EGU
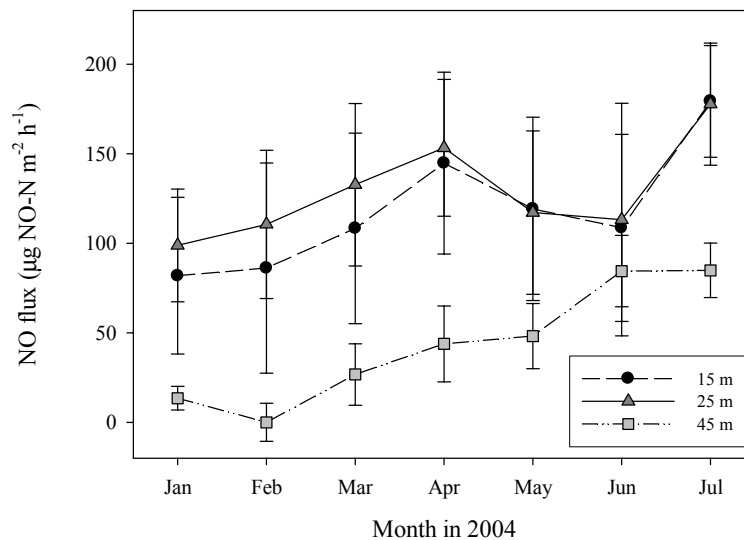
Fig. 2. Average monthly NO fluxes from hourly data of two automatically closing flow through chambers per site. The bars show standard errors of the temporal and spatial variability.
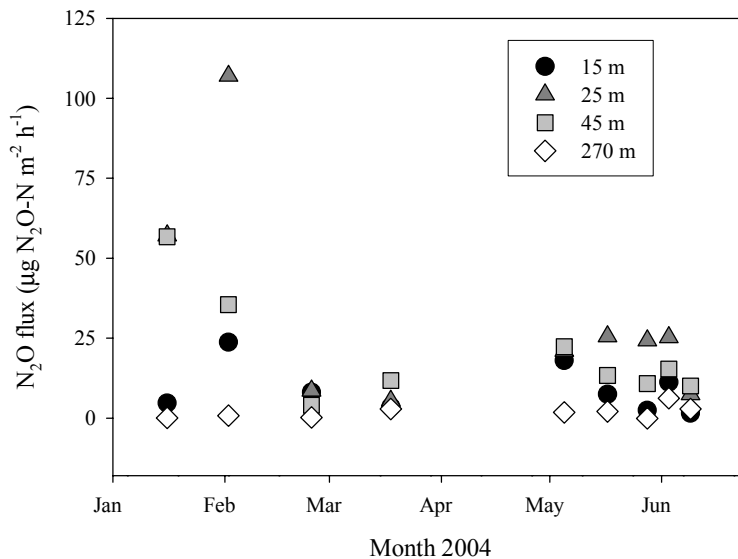
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Fig. 3. Nitrous oxide emission downwind of the poultry farm; average fluxes from 2 manual static chambers per site.

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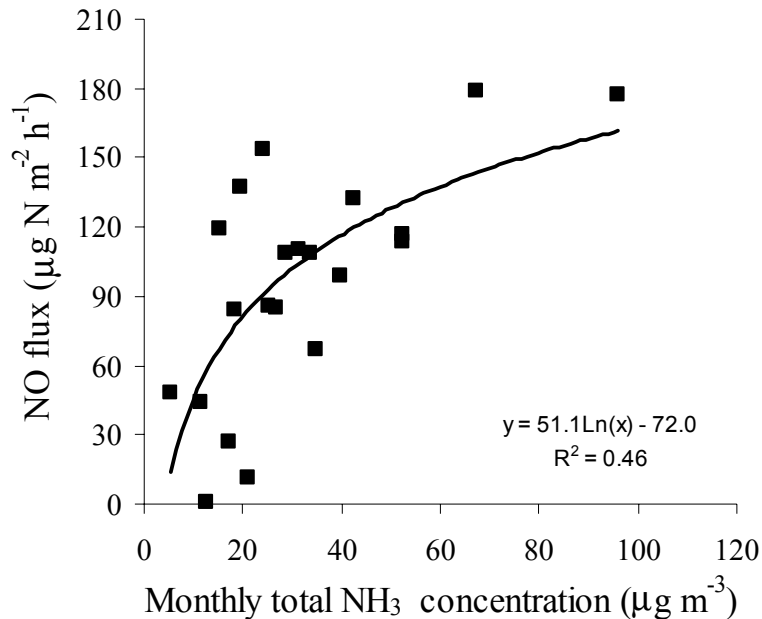
Fig. 4. The relationship between monthly average NO flux and NH_3 concentrations at the three transect sites downwind of the poultry farm.
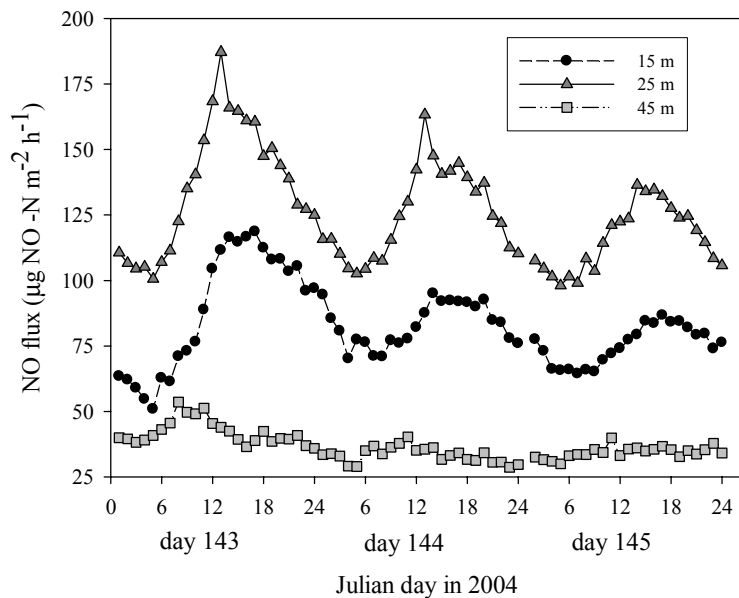
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Fig. 5. The diurnal variations in NO flux from 2 chamber each at 15, 25 and 45 m downwind of the poultry farm in May 2004. Data are averages from two chambers.