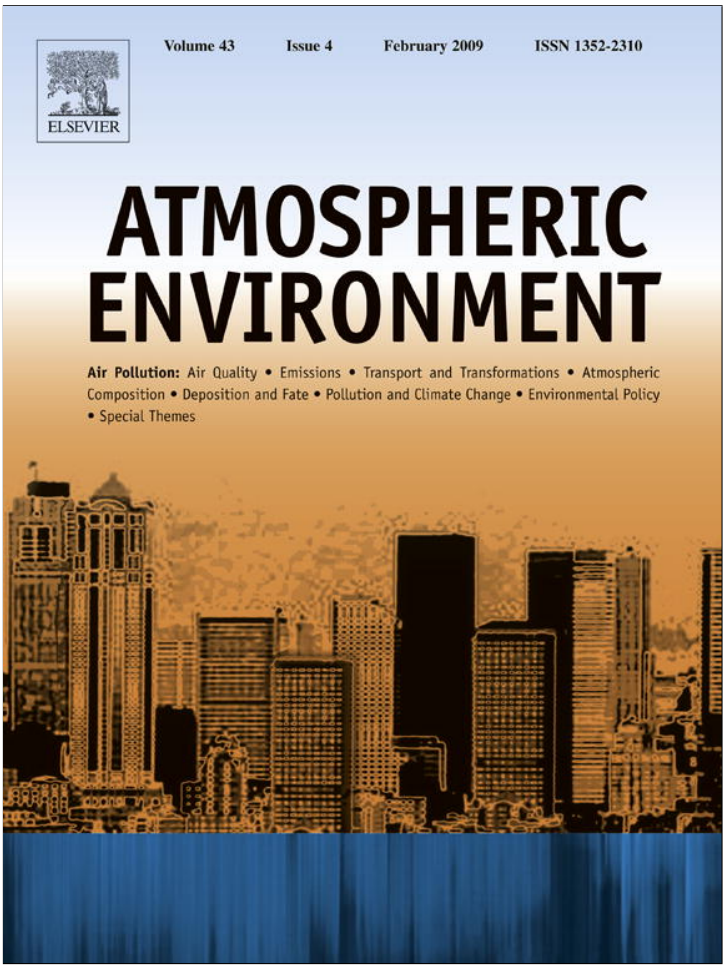

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Validation of model calculation of ammonia deposition in the neighbourhood of a poultry farm using measured NH_3 concentrations and N deposition

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ABSTRACT

Substantial emission of ammonia (NH_3) from animal houses and the related high local deposition of NH_3-N are a threat to semi-natural nitrogen-deficient ecosystems situated near the NH_3 source. In Denmark, there are regulations limiting the level of NH_3 emission from livestock houses near N-deficient ecosystems that are likely to change due to nitrogen (N) enrichment caused by NH_3 deposition. The models used for assessing NH_3 emission from livestock production, therefore, need to be precise, as the regulation will affect both the nature of the ecosystem and the economy of the farmer. Therefore a study was carried out with the objective of validating the Danish model used to monitor NH_3 transport, dispersion and deposition from and in the neighbourhood of a chicken farm. In the study we measured NH_3 emission with standard flux measuring methods, NH_3 concentrations at increasing distances from the chicken houses using passive diffusion samplers and deposition using ^{15}N-enriched biomonitors and field plot studies. The dispersion and deposition of NH_3 were modelled using the Danish OML-DEP model. It was also shown that model calculations clearly reflect the measured NH_3 concentration and N deposition. Deposition of N measured by biomonitors clearly reflected the variation in NH_3 concentrations and showed that deposition was not significantly different from zero ($P < 0.05$) at distances greater than 150–200 m from these chicken houses. Calculations confirmed this, as calculated N deposition 320 m away from the chicken farm was only marginally affected by the NH_3 emission from the farm. There was agreement between calculated and measured deposition showing that the model gives true estimates of the deposition in the neighbourhood of a livestock house emitting NH_3.

1. Introduction

Livestock production is the most important source of ammonia (NH_3) in the atmosphere (Bouwman et al., 1997; Hutchings et al., 2001) and, in Europe, manure in animal houses may account for more than one-third of the NH_3 emitted from livestock manure (ECETOC, 1994; Hutchings et al., 2001). Close to the source, NH_3 is deposited on vegetation, soil and water (Asman and van Jaarsveld, 1991). This deposition may cause acidification and eutrophication of natural ecosystems (Fangmeier et al., 1994).

In addition, the NH_3 may be transported over long distances as ammonium (NH_4^+), mainly in the form of particles. The long-distance transport of ammonium (NH_4^+) has been regulated by the EU and the UN by the inclusion of this emission type in the Gothenburg Protocol on long-range transboundary air pollution (United Nations, 2004) and the EU National Emissions Ceilings Directive—NECD (EEA, 1999) regulations.

The risk of high local deposition of NH_3 from livestock operations is regulated by national legislation. In Denmark the regulation is based on a zoning principle. Natural or semi-natural ecosystems designated as being worthy of protection are classified according to their critical load capacity (Fisher et al., 2007), i.e. their capacity to assimilate nitrogen (N) without concurrent changes in the flora and fauna. Up to 300 m from N-vulnerable ecosystems, livestock producers are not permitted to increase their NH_3 emissions when changing their production system. It is expected that, with time, few—if any—livestock producers will be situated in this zone (Anonymous, 2006). At distances between 300 and 1000 m from these ecosystems, farmers who wish to change or increase their livestock production have to comply with strict rules and emission thresholds. Obviously there is a need for precise and accurate models for assessing how much the livestock production will affect N deposition in nearby natural ecosystems.

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Models for assessing the dispersion of NH_3 from animal housing have been tested on data from measurements of dispersion of NH_3 from a synthetic source (Asman et al., 1989), but few studies have examined and tested the models by calculating the deposition of NH_3 near the animal houses of actual livestock production farms or cattle feedlots (Hao et al., 2006). In the 1980s and 1990s, the deposition of NH_3 near a dairy farm was examined using biomonitors (Sommer, 1988; Sommer and Jensen, 1991), showing that this method can be used to provide information on local NH_3 deposition. However, no measurements of emissions from animal houses and no dispersion studies were carried out, and therefore these measurements can not be used to verify recently developed models for assessing local deposition of NH_3.

The objective of the current study was to provide data needed to validate the Danish OML-DEP model that is used to assess transport, dispersion and deposition of N in the neighbourhood of livestock farms. Further, the study provided measurement of total N deposition and NH_3 gradients near a chicken farm as affected by the emission of NH_3 from the chicken houses, which was also measured.

2. Materials and methods

The NH_3 deposition in the neighbourhood of the chicken house was measured using plant biomonitors enriched with ^{15}N placed at different distances and directions from the farm (Fig. 1). The deposition measurements were carried out from 1 September to 25 October 2005 (i.e. for 54 days). Passive diffusion samplers were used to provide long-term mean values (weeks) of the NH_3 concentration in different directions and distances from the farm in the period (Fig. 1). The NH_3 concentration was measured at three periods in intervals of 2–3 weeks in the period from 5 September until 17 October 2005. Meteorological data were combined with the estimates of emission from the animal houses to assess the local NH_3 concentration and deposition of N. It was decided to carry out the deposition measurements within a 300 m zone for the purpose of getting a significant response when measuring NH_3 concentration and deposition gradients. To determine the crop response to NH_3 deposition, a winter wheat fertilizer response trial was established at distances 60–200 m from the chicken house (Fig. 1). The winter wheat was drilled in the soil in September 2004 and harvested in July 2005.

2.1. Ammonia emission

The dispersion and deposition of NH_3 was examined around two chicken houses; 14, 400 chickens were transferred to the house 1 on 30 June 2005, and 12, 700 chickens were transferred to the house 2 on 4 July. The chicken houses were emptied on 3–5 November 2005. The initial weight of the chickens was approx. 45 g and their final weight was approx. 1500 g.

Ammonia emission from the chicken houses was estimated by measuring the air exchange rate through the animal house and the NH_3 concentrations to and from the animal house. The air exchange rate was measured by use of a calibrated measuring wing (Fancom AT(M) unit 80) and the NH_3 concentration in air to and from the farm was measured using an electrochemical sensor (Dräger Polytron 1). The concentrations were measured at every second outlet and the intermediate concentrations were interpolated. The amount of emission (E_x mg NH_3-N s^{-1}) was calculated using the following equation:

$$E_x = V(C_{x,\mathrm{out}} - C_{x,\mathrm{in}}) \quad (1)$$

where $C_{x.\mathrm{out}}$ and $C_{x.\mathrm{in}}$ are the concentrations of NH_3 (mg N m^{-3}) in air collected in the ventilation outlet and in incoming air, respectively, and V is the air exchange rate (m^3 s^{-1}).

2.2. Biomonitors

Exchange of atmospheric NH_3 was estimated by measuring the dilution of ^{15}N in ryegrass (*Lolium multiflorum* Lam.) grown in pots standing on the soil. The pots had a surface area of 0.0577 m^2 (volume 10 l) and contained 13.5 kg of N-free sand. On the sand surface of each pot 0.6 g seed was spread evenly and covered with a 1 cm layer of sand. Initially the pots were watered with N-free nutrient solution (Sommer, 1988); 10 and 25 days after sowing, 0.26 g N as KNO_3 with 2.786% ^{15}N excess was supplied to each pot with the nutrient solution. Until placement in the field, N-free nutrient solution was supplied as required. Surplus rain water draining through the sand was collected and recycled automatically.

The pots were placed in a glasshouse at sowing on 1 August 2005, and on 1 September the pots with plants were placed in groups of three replicates in the field at 15–320 m distance from the farm buildings in south-west, north-west, north-east and south-east directions (Fig. 1). The biomonitors were irrigated according to demand; i.e. they were supplied with demineralized water on 6 and 16 September. The plants were harvested on 25 October after having been exposed in the field for 54 days. The roots were recovered by gently washing the sand from the roots. The harvested plant material—tops (above-ground leaves and stems) and roots

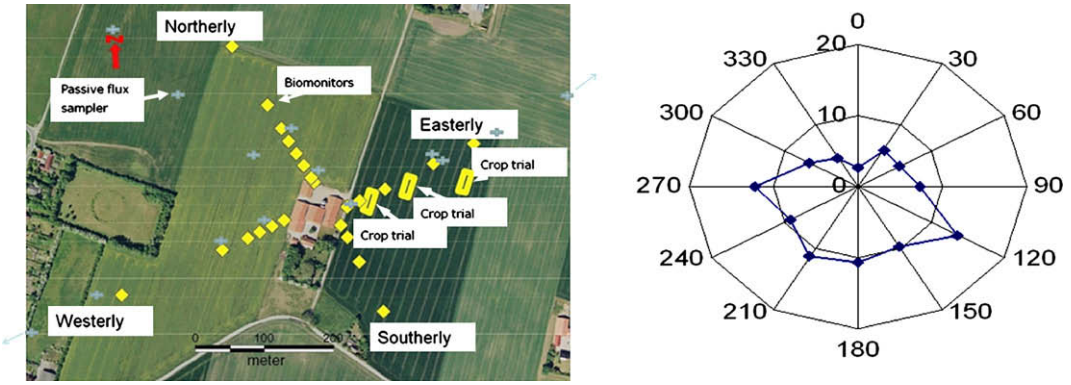

Fig. 1. (Left) Aerial photograph of the poultry farm showing the position of the ryegrass biomonitors (diamonds) located at increasing distances north-west (Northerly), north-east (Easterly), south-east (Southerly) and south-west (Westerly) of the chicken farm, the positions of the three nitrogen fertilizer trials (rectangles) and of the passive flux samplers for measuring NH_3 concentrations (blue cross). The two outermost positions of the passive denuders, which are marked with an arrow, were positioned at the following distances from the farm: (515 m west and 214 m south) and (539 m north and 215 m east). (Right) Wind rosette showing the prevailing wind directions during the experimental period.

(below ground) separately—was oven-dried at 80 °C for 24 hours, weighed and finely ground.

Analysis for total N and ^{15}N enrichment in the dried and ground plant material was carried out using a isotope ratio mass spectrometer (IRMS type 20-20) coupled to an elemental analyser sample preparation module (ANCA-SL; both instruments from Europe Scientific, Crewe, UK).

N exchange (g N pot^{-1}) was calculated using the following equation:

$$\text{N exchange} = N_{\text{total}} \times \text{DM}\left(\frac{\text{atom\% }^{15}\text{N excess (control)} - \text{atom\% }^{15}\text{N excess (plant)}}{\text{atom\% }^{15}\text{N excess (control)}}\right) \quad (2)$$

where N_{total} (g N DM^{-1}) is the concentration of N in the harvested and dried plant material, and DM (g pot^{-1}) is the dry matter harvested in tops or roots. N exchange was calculated separately for tops and roots and then added together for each pot. The ^{15}N excess of control plants from pots placed 320 m west of the farm was used to provide control data. Thus the results of the calculation provide estimates of the surplus N deposition caused by the NH_3 emitted from the farm in the zone nearest to the chicken houses. This strategy for measuring the control ensured that all plants were influenced by similar environmental conditions.

2.3. Winter wheat N response experiment

The fertilizer value of the increase in deposited N near the chicken houses was examined by measuring the uptake of N by winter wheat in plots (2.5 m wide × 14 m long) at 60, 120 and 200 m north-east of the chicken houses. The area surrounding the trial area was supplied with 40 kg N ha^{-1}, and the individual plots were supplied with three N levels (60, 90 and 120 kg N ha^{-1}) in four replications; thus there were 36 observations in all. Data are therefore available for each observation with regard to the amount of N applied (kg N ha^{-1}) and distance from the animal house (m), as the distance between the chicken house and each plot was measured. The wheat was sown in September 2004 in the winter wheat fields surrounding the farm (distance between rows 11 cm) and harvested in July 2005. The yield of seed was measured, and the nitrogen content of the seed was determined with the Kjeldahl method.

2.4. NH_3 concentration measurements

The ammonia concentrations were measured with passive diffusion tube samplers, consisting of a permeable outer tube with an absorbing rod inside. The outer tube is 4.6 cm long, 1.6 cm in diameter, and made of polyethylene. Inside the tube is a 4.6- cm-long rod with a diameter of 0.6 cm. The rod is impregnated with phosphoric acid, which absorbs the NH_3 that diffuses through the wall of the outer tube. Two or three samplers were placed under a rain-cover on a stick at a height of 2 m (i.e. 2 or 3 replicated measurements at each position). After exposure, the rods were extracted in 5 ml of deionized water and the ammonium content was determined by spectrophotometric detection using flow injection (ISO 11732). The mean concentration of ammonia during the exposure period was calculated by using the exposure time, ammonium content and a constant factor, which was defined in an earlier test of the method (Andersen et al., 2006).

Long-term mean values of NH_3 concentrations were measured in three periods of 2–3 weeks duration from 5 September until 17 October 2005 (Fig. 1).

2.5. NH_3 dispersion model

Two models were used for the calculations. The Danish Eulerian long-range transport model DEHM (Christensen, 1997; Frohn et al., 2002) was used to estimate the background ammonia concentration and the wet and dry depositions of N (Ellermann et al., 2006) except for NH_3 dry deposition. The local-scale transport deposition model OML-DEP was used to estimate local NH_3 dry deposition.

The DEHM calculations were performed using a 16.67 × 16.67 km resolution for Denmark and nearby areas. These calculations were based on meteorological data generated by MM5 (Grell et al., 1994). The local NH_3 emissions in DEHM were computed using the parameterizations with high spatial and temporal resolution (Skjoth et al., 2004; Gyldenkaerne et al., 2005).

OML-DEP is based on the Gaussian OML model (Olesen et al., 2007) for point and area sources. It has been further developed for ammonia dry deposition by making use of the surface depletion principle of Horst (1977). The model converts the deposition to an area (source) into a negative surface emission from the same area. The chemical transformation of NH_3 to NH_4^+ is simulated by a pseudo-first-order reaction velocity (Asman et al., 1989), which is shown to be unimportant at short transport distances.

OML-DEP calculations were performed for a 1200 × 1200 m local domain split into 1600 area segments each of 30 × 30 m. The dry deposition velocities (in both DEHM and OML-DEP) are calculated with the same module, which is based on the methodology used in the EMEP model (Simpson et al., 2003). OML-DEP uses hourly background concentrations of ammonia and sulphur dioxide from DEHM.

Meteorological data (temperature, heat flux, global radiation, humidity, wind speed and direction, speed of wind friction, and derived atmospheric stability parameters such as Monin–Obukhov Length) were measured at a local meteorological station set up for this study.

3. Results and discussion

The study was carried out in the area surrounding a chicken farm consisting of about 24, 000 chickens contained in two animal houses (Fig. 1). With the exception of the trees in a garden south–south-west of the farm there were no protruding landscape elements within a distance of 250–300 m from the animal houses. During the period with measurements of N deposition, the total emission of NH_3 was 356 kg (227 kg N from House 1 and 129 kg N from House 2) and the emission rate varied from 0.0176 to 0.227 g NH_3-N s^{-1}, with mean daily emissions ranging from 2.59 to 11.8 kg NH_3-N day^{-1}. During the exposure of biomonitors the average temperature was 12 °C and wind speed was 3.7 m s^{-1}. Prevailing wind directions were south-east, south and south-west (Fig. 1).

3.1. Ammonia concentration

The net- NH_3 concentrations, i.e. the measured concentrations minus the background concentration, measured in three periods during the exposure of the biomonitors varied from 0 up to ca. 13.01 μg NH_3-N m^{-3} (Fig. 2A). The background concentration is estimated from the upwind measurements at the farthest distances

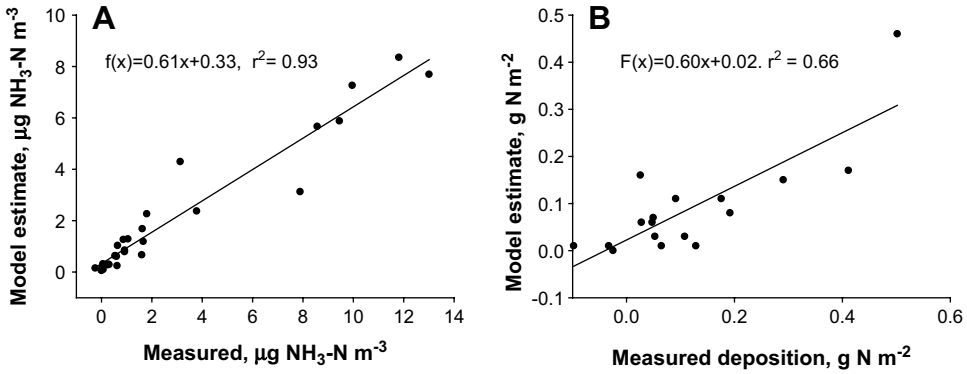

Fig. 2. (A) The contribution of the poultry farm to ammonia concentrations (2-week averages) measured with passive diffusion samplers and calculated with OML-DEP during the period of biomonitor exposure. The measurement of NH_3 concentration south-east of the farm was not included because a source of NH_3 to the south affected the concentrations. (B) Deposition measured and calculated for all directions except for the outliers indicated with parentheses in Fig. 3 and data from the southeast, where a local source probably interfered. The line is the regression line for model versus measured deposition.

from the chicken houses. In the measuring period the average background level was estimated to be 1.4 µg NH_3-N m^{-3}.

There was a clear tendency for decreasing concentrations with increasing distance from the farm. This pattern was also measured by Fowler et al. (1998), who showed a sharp decline in NH_3 concentrations measured at increasing distances from livestock buildings in the prevailing wind direction. They observed a yearly mean NH_3 concentration close to background NH_3 concentrations 276 m from the source with emission strength of 4500 kg NH_3-N $year^{-1}$.

3.2. Nitrogen deposition

Measured and model-calculated NH_3 deposition is presented in Fig. 3. The measurements show high standard errors (SEs) for some of the deposition estimates, probably due to inter-pot variability in plant dry matter yield, total N concentration and ^{15}N enrichment. Because each of these measurements is subject to a small amount of error, they can all contribute to the SE of the calculated mass balance, which in this study is expressed as the exchange of N between the biomonitors and the atmosphere in excess of the background concentration. The exchange of N measured 320 m west (240°) of the farm was set to 0 because the biomonitors at this location were used as the control (see Eq. (1)). These calculations are supported by the wind direction measurement, showing that during the study there were few occasions when the wind was blowing from the farm towards these biomonitors, i.e. from the 60° direction (see Fig. 1). Therefore, the biomonitors at this position were affected mainly by 'background' deposition of NH_4^+ and NO_3^-. When using the concentration of ^{15}N in the biomass of these biomonitors as a control, the dilution of ^{15}N in the biomonitors caused by the deposition of NH_3 emanating from the chicken houses is determined. The exchange varies from a deposition of 0.5 g N m^{-2}

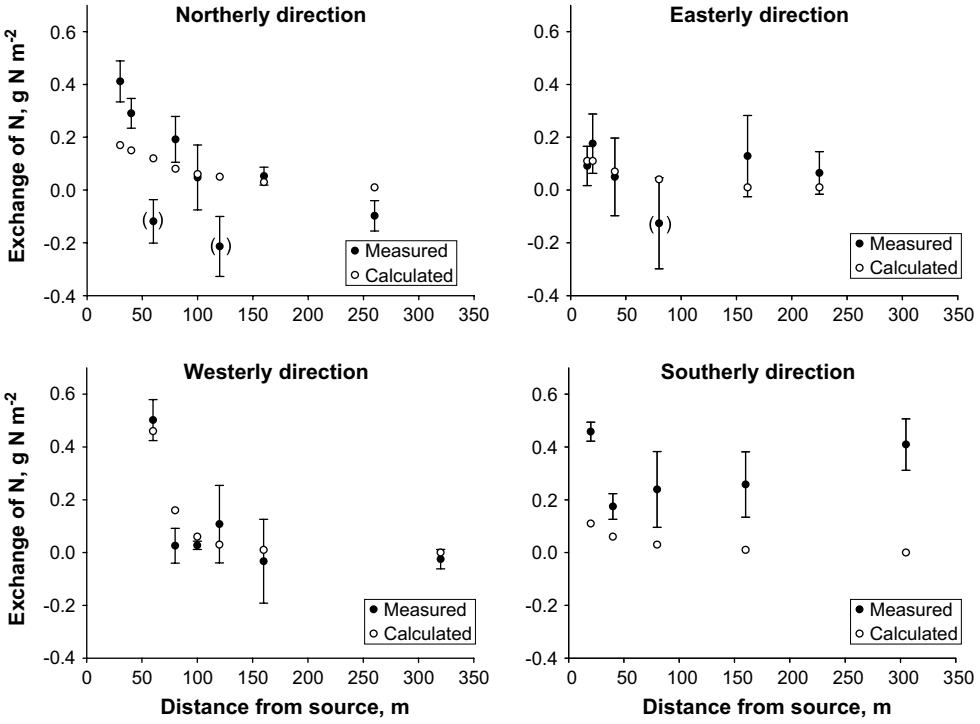

Fig. 3. Exchange of N between biomonitors (ryegrass in pots; area 0.0577 m^2) exposed to the atmosphere for 54 days at increasing distances from a chicken house and the deposition of N calculated using the OML-DEP model. Vertical bars depict standard errors (SE) of the mean. Outliers with markedly negative values are indicated with parentheses. These observations of loss of N from the biomonitors may be considered as outliers, since the NH_3 concentrations are high and above the compensation points of healthy plants (Sutton et al., 1993).

(SE 0.08 g N m^{-2}) at 60 m west, to a loss of N of 0.2 g N m^{-2} (SE 0.11 g N m^{-2}) at 120 m north of the chicken houses. These observations of loss of N from the biomonitors is here considered as outliers, since the atmospheric NH_3 concentrations are high and above the compensation points of healthy plants (Sutton et al., 1993). Thus we speculate that the loss could have been caused either by plant disease or by damage from wild animals eating the plants. The deposition is related to distance from the farm and prevailing wind direction (Figs. 1 and 3). Thus, 60 m from the chicken houses, the daily deposition of N was up to 0.09 kg N ha^{-1} day^{-1}. The deposition declined with distance from the farm and the deposition measured was not significantly different from zero ($P < 0.05$) at distances greater than 150–200 m from the chicken houses. For comparison, about 0.017–0.085 g N ha^{-1} day^{-1} was deposited on biomonitors situated at a distance of 10–130 m from a dairy cattle farm with 70 Jersey dairy cattle (Sommer and Jensen, 1991).

3.3. Validation of the deposition model

A comparison with the OML-DEP-modelled NH_3 concentrations is shown in Fig. 2A. The modelled NH_3 concentrations only included estimates of NH_3 from the chicken houses therefore the background concentration was subtracted from the measured concentrations. The background concentration was estimated as the average of the 3–5 lowest measurements obtained at the measuring points at the greatest distance (600 m) from the chicken houses. This explains the small negative concentrations. There is a reasonable correlation between measured and calculated concentrations, but it also appears that the model underestimates the concentrations close to the source. There could be two explanations for this. First, the slope of the fitted line is greatly influenced by a few very high concentrations that were found at the monitors closest to the farm buildings, and the buildings themselves have a relatively large influence on the dispersion at short distances. Therefore, the model in this case may underestimate the influence of the buildings at short distances. Secondly, the total emission rate may have uncertainty because emission only has been monitored at half of the outlets. The uncertainty is accentuated due to the measured 'total' emissions from the two identical poultry houses varied by almost a factor of two. Therefore, it can not be excluded that the emission rate has been underestimated.

On average over the period, the modelled deposition velocity—calculated as mean deposition divided by mean concentration at the biomonitors—varied between 0.31 and 0.73 cm s^{-1} for the individual locations, with an overall mean of 0.53 cm s^{-1}, which is in agreement with the deposition velocities measured earlier (Sommer and Jensen, 1991; Krupa, 2003).

The model calculations estimate the local and regional deposition of N to be 0.18 g N m^{-2} at 320 m west of the chicken houses. This is 0.01 g N m^{-2} or 6% higher than the modelled regional deposition of N, which is 0.17 g N m^{-2} for the period of the measurements carried out in this experiment (Table 1). It can be seen that this estimate fits with the assumption that deposition to the biomonitors of NH_3-N emitted from the chicken houses is near zero at 320 m west of the chicken farm. The slope of the fitted line of modelled versus measured deposition is <1 (Fig. 2B). The model's underestimation of the deposition could be due to an underestimation of the emission of NH_3, and thus underestimation of the NH_3 concentration (Fig. 2A). The amount of underestimation of the deposition (Fig. 2B) is almost equal to the underestimation of the NH_3 concentration (Fig. 2A) indicating that the mean deposition velocity in the model is accurate.

Table 1
Deposition of oxidized and reduced nitrogen onto grass in an area not affected by local sources of atmospheric nitrogen.

	g N m^{-2}			
	NH_3	NH_4^+	NO_2+NO_3^-	Sum
Dry deposition	0.057	0.004	0.060	0.120
Wet deposition	0.009	0.018	0.025	0.051
Total	0.066	0.022	0.085	0.172

The deposition was calculated for 54 days using the regional DEHM model (Christensen, 1997; Frohn et al., 2002).

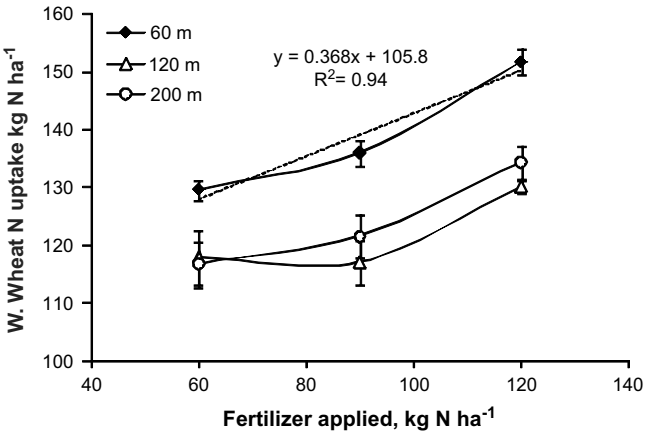

Fig. 4. Uptake of N by winter wheat in plots situated 60, 120 and 200 m north-east of the chicken houses. At each position, experimental plots were established and fertilized with 60, 90 or 120 kg N ha^{-1} each with four replicates (SE in brackets).

3.4. N uptake in field trials

The field plot trials reflecting the effect of nitrogen deposition on the winter wheat crop were established in autumn 2004 and continued until the end of nitrogen uptake the following growing season in 2005; which was a much longer time than the biomonitor experiment in August–September 2005. At all distances from the chicken houses (60, 120 and 200 m) there was a curvilinear increase in wheat N uptake with increasing fertilizer N applied (Fig. 4). The N uptake and N response were not significantly different between plots at 120 and 200 m, but the N uptake was significantly higher closest to the farm at 60 m from the chicken houses. The wheat response (N uptake) to additional fertilizer N at 60 m distance showed a crop uptake efficiency of 0.368 (slope of regression line) for the applied N, meaning that only 37% of the N applied was taken up by the crop. The increase in wheat N uptake from 200 m to 60 m distance from the chicken house was 14.8 kg N taken up ha^{-1} on average across all fertilizer N levels and, with a crop uptake efficiency of 0.368 as described above, this corresponds to an increase in N deposition from 200 m distance to 60 m distance equivalent to approx. 40 kg N ha^{-1} (14.8 divided by 0.368). For comparison the deposition during the same 8–9 month period corresponds to approx. 22 kg N ha^{-1} if the deposition to the biomonitor 60 m east of the chicken houses is used (i.e. a deposition rate of 0.05 g N m^{-2} over 54 days), and not taking into consideration the fact that uptake in winter (with no plant growth) will be lower than during summer .

Furthermore, the uptake of emitted NH_3 was higher when crops were fertilized at 90 kg N ha^{-1} and 120 kg N ha^{-1} than when they were fertilized at 60 kg N ha^{-1}. There may be several reasons for this pattern—i.e. better plant growth in general—but also a higher leaf area may contribute to a larger uptake of atmospheric NH_3 at higher fertilizer levels.

4. Conclusions

It is shown that OML-DEP model calculations reflect measured NH_3 concentration and N deposition in the neighbourhood of

a chicken farm. Calculations using the model indicated that N deposition at 320 m distance from the chicken farm was only marginally affected by the NH_3 emission from the farm. This was also confirmed by N deposition measurements using bio monitor measurements and winter wheat field trials. Thus, within the uncertainties of the measurements, this field study shows that the OML-DEP model gives valid estimates of dispersion and deposition of NH_3 emitted from a livestock farm.

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