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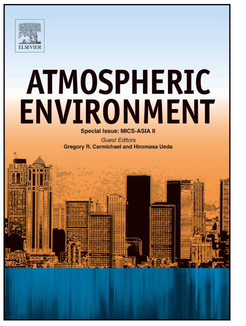

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Authors: Alberto Sanz, Thomas Misselbrook, Maria José Sanz, Antonio Vallejo

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Use of an inverse dispersion technique for estimating ammonia emission from surface applied slurry in Central Spain

Alberto Sanz[a], Thomas Misselbrook[b], Maria José Sanz[c], Antonio Vallejo[a]

[a]*ETSI Agrónomos, Technical University of Madrid, Ciudad Universitaria. 28040 Madrid, Spain.* [b]*North Wyke Research, North Wyke, Okehampton, Devon EX20 2SB, UK.*, [c]*Fundación CEAM, Charles R. Darwin 14, 46980 Paterna, Spain.*

Abstract

Ammonia (NH_3) emission from land application of manure is typically measured using the integrated horizontal flux (IHF) micrometeorological method. However, there are some situations in which alternative techniques (such as an inverse dispersion modelling technique) might be preferable, for example when measuring from large or irregularly shaped source areas. In this study, an inverse dispersion technique using the backward Lagrangian Stochastic model (bLS), with 2 different experimental configurations, was compared with the Integrated Horizontal Flux method (i.e. IHF), which was used as reference technique. Pig slurry was surface-applied at 125 kg N ha^{-1} to bare soil on a large plot (80 x 125 m). Cumulative emissions were 19.3, 21.2 and 18.4 kg N ha^{-1} from the IHF and the bLS technique (experimental configurations I and II), respectively. Mean flux within each sampling period as estimated by the two techniques compared extremely well, with a slope not significantly different from 1 and r^2 of 0.99. Although limited in extent, this dataset agree with a previous study in demonstrating the use of the bLS technique with longer period time-averaged concentration measurements.

Key words: Ammonia emission, micrometeorological technique, Backward Lagrangian stochastic method, dispersion model.

1. Introduction

Ammonia (NH_3) emitted to the atmosphere from agricultural sources readily reacts with sulphate (SO_4^{2-}) and nitrate (NO_3^-) to form particulates (Ansari and Pandis, 1998), which can be a health hazard, and may cause acidification and nutrient-N enrichment of natural ecosystems when deposited to land or water (Erisman et al., 2007).

Intensive agriculture is estimated to produce around 90% of NH_3 emissions in Europe (CORINAIR, 2007). From those, between 80 and 90% arise from the excreta produced by livestock (ECETOC, 1994). In Spain, diluted slurries are typically surface applied to land via high-rate irrigation systems, potentially associated with high NH_3 losses via volatilisation, particularly if the application rate exceeds the infiltration capacity of the soil, because of the large emitting surface area (compared with slurry injection or band spreading, e.g. Misselbrook et al., 2002), thereby reducing the N fertiliser efficiency.

A number of techniques have been developed to quantify NH_3 emission, to provide data for national emission inventories so that impacts on the environment can be assessed and the effectiveness of abatement strategies evaluated. Measurement techniques may be categorised according to the scale of measurement (e.g. laboratory, small plots, field scale) or the measurement principle (soil N balance, closed chamber, open chamber, micrometeorological) and a review of most current techniques is provided by McGinn and Janzen (1998). Micrometeorological techniques, such as the mass-balance Integrated Horizontal Flux (IHF) method, are often preferred for measuring from manure-treated plots at the medium-scale (i.e. approximately 0.1 ha) since they do not disturb the surface environment (e.g. Misselbrook et al., 2005). However, there are some circumstances in which the IHF method cannot be satisfactorily used, e.g. when measuring from large source areas which would require the use of very tall measurement masts, or estimating emissions from irregularly shaped source areas. The use of an inverse dispersion modelling technique may be an effective alternative in such situations. Among these, the backward Lagrangian Stochastic (bLS) dispersion model has been shown to effectively infer the NH_3 emission rate of surface area sources based on single-height measurements of wind speed, NH_3 concentration and wind statistics (Flesch et al., 2007). This technique (like other micrometeorological techniques), is designed for use with concentration and wind measurement averaging times of the order of 30 min. For many types of problems and gases, such short measurements intervals would be a considerable challenge. However, Sommer et al. (2005) demonstrated that the bLS technique could be effectively used for inferring the NH_3 emission with much longer averaging times (e.g. 5-26 h).

The objective of this study was to assess the use of the bLS technique, using time-averaged concentration measurements, for estimating NH_3 emission from surface-applied pig slurry to bare soil in Central Spain. The IHF technique was used as reference technique in the study. Obtained data would also add to the relatively sparse database of emissions from land applied manures under Mediterranean conditions.

2. Materials and methods

2.1. Experimental set-up

The experiment was conducted between the 26th September and the 2nd October 2006 in a Typic Haploxeralf soil (USDA-SSS, 1999) in Valverde del Majano (Segovia), central Spain (40º 95´ N, -4º 23´ W). Pig slurry was evenly surface-applied at a rate of 125 kg

N ha^{-1} to a rectangular plot (80 x 125 m) using a vacuum tanker fitted with a splash-plate. Slurry characteristics were pH of 6.8, dry matter content of 4.6% and total nitrogen content of 2.1 g kg^{-1} of which 76% was in the ammoniacal form. Wind speed and wind direction were monitored by means of three anemometers at heights of 0.5, 1.0 and 5.0 m, respectively. Rainfall and air temperature data were provided by a meteorological station located in the experimental area. There were no wind-disturbing elements, e.g. trees, houses or any other protruding landscape element, within approximately 800 m of the experimental plot.

2.2. Measurement of ammonia emissions

Ammonia emission was estimated using two different techniques: the micrometeorological mass balance integrated horizontal flux (IHF) method (Denmead et al., 1977) and a backward Lagrangian Stochastic (bLS) modelling approach (Flesch et al., 2005).

2.2.1. Integrated Horizontal Flux method

The mass balance IHF method equates the vertical flux of NH_3 from a treated area of limited upwind extent with the net integrated horizontal flux at a known downwind distance (Denmead et al., 1977). Vertical masts were placed towards the centre of the slurry-applied plot and upwind of the treated area (CM and BM, respectively, Fig. 1). Each mast supported 5 passive flux samplers (PFS) coated with oxalic acid (Leuning et al., 1985) mounted at 0.25, 0.5, 1.00, 2.05 and 3.05 m height (z) above the soil surface. Passive flux samplers were replaced seven times during the whole experiment at 4.5, 11.5, 23.75, 35.5, 47.5, 71.5, 143.5 h after slurry application. At the end of each sampling period and immediately following exposure, samplers were transported to the laboratory where they were leached with 40 ml of deionised water and the solution analysed for NH_4^+-N by spectrophotometry using the nitroprussiate method (Searle, 1984).

The mean horizontal flux, $\overline{uc}$ (mg N m^2 s^{-1}), at each sampling height was calculated from

$$\overline{uc} = M/At \tag{1}$$

where, *M* is the mass of NH_3-N collected (mg) in the sampler during sampling period, *A* is the effective cross-sectional area of the sampler (m^2) as determined in wind tunnel calibrations and *t* the duration of the sampling period (s). The net flux of NH_3 (*F*, μg m^{-2} s^{-1}) from the treated area was then calculated according to:

$$F = \frac{1}{x}\left[\int_0^z \left(\overline{uc}\right)_{cm} dz - \int_0^z \left(\overline{uc}\right)_{bm} dz\right] \tag{2}$$

where *x* (m) is the mean fetch length (i.e. distance from the measurement mast to the upwind boundary of the treated area), *z* (m) the height of the uppermost sampler and $\overline{uc}$ the mean horizontal flux measured by each sampler at the centrally-located downwind (cm) or upwind background (bm) mast. Equation 2 assumes the plot extends infinitely in the across-wind direction so that net lateral flux of NH_3 is zero; a circular treated area is often used to approach conditions nearing this assumption. In the case of a rectangular plot, as in the present study, flux estimates associated with wind directions from the corner of the plot will potentially be associated with errors due to net lateral flux of NH_3.

2.2.2. Backward Lagrangian Stochastic dispersion technique

The bLS approach enables estimation of the emission rate from a downwind concentration measurement (Flesch et al., 2007). The emission rate (F, µg m^{-2} s^{-1}) is derived according to

$$F = \frac{c - c_b}{\left(c/F \right)_{sim}} \qquad (3)$$

where c-c_b is the time average gas concentration above background, and the ratio $(c/F)_{sim}$ is simulated by means of a dispersion model (Flesch et al., 2007). Several authors (McBain and Desjardins, 2005; Flesch et al., 2007) have demonstrated that, for an homogeneous surface layer and under conditions of stationarity (atmospheric invariance), the principles of the Monin-Obukhov similarity theory (MOST) can describe the wind near the ground using four variables: the calculated friction velocity, u^*; the Obukhov stability length, L; the surface roughness length, z_o and wind direction according to North, β.

The software “WindTrax” (Thunder Beach Scientific, Nova Scotia, Canada), which combines an interface where sources and sensors are mapped with the bLS model described by Flesch et al. (2004, 2005), was used in this study for the estimation of NH_3 emission rate by the bLS technique (F_{bLS}). Inputs were average wind speed at heights of 0.5, 1.0 and 5.0 m height, the default Obukhov stability length (L) (or optionally input as stability class), the default surface roughness length for bare soils (z_0=0.01 m), the average wind direction (β) determined at 1.0 m height and ammonia concentration (µg NH_3 m^{-3}) derived from the measured NH_3 horizontal flux (µg NH_3 m^{-2} s^{-1}) at 1 m height and wind speed monitored at the same height. For long sampling intervals, Sommer et al. (2005) recommended that neutral atmospheric stability conditions are assumed and showed that the sensitivity of the flux estimate was lowest at a concentration measurement height of 1 m (with little change in sensitivity for ±0.5 m) for plot dimensions up to 100 m. Wind direction in the present study was predicted to be parallel to the 80 m plot dimension, so the measurement height of 1 m was considered appropriate.

Two different experimental configurations were used in the calculation of F_{bLS} in order to test the flexibility in plot design (e.g. measurement location) and number of samplers of this modelling approach. In the first configuration (Experimental Configuration I), NH_3 concentrations at 1 m height derived from the central (CM) and background (BM) masts (Fig. 1) were used, corresponding with the measurement positions for the IHF technique. In the second configuration (Experimental Configuration II), NH_3 concentration at 1 m height derived from masts U1, D1, D2 and D3 (Fig. 1) were also included in the model calculations ("overdetermination").

3. Results and discussion

3.1. Environmental data

Average temperature during the experimental period was 18.9 ºC, with daily mean maximum and minimum temperatures of 26.8 and 4.9 ºC, respectively. Wind speed at 1.0 m height ranged between 0.07 and 3.86 m s^{-1} for the entire period, with a mean value of 1.35 m s^{-1}. The prevailing wind direction measured over the experimental period was NW. No rainfall occurred during the experimental period.

3.2. Ammonia emissions

Estimated pattern of NH_3 emission was similar for the IHF technique and the two configurations of the bLS technique. Ammonia emission peaked only once during the measuring period, within the first 5 h after slurry application. Highest emission rates were 2.00, 2.07 and 1.83 kg N ha^{-1} h^{-1} for the IHF technique and the bLS configurations I and II, respectively (Fig. 2a). Mean flux within each sampling period, as estimated by the techniques, compared extremely well with a slope not significantly different from 1 and r^2 of 0.99 (Fig. 3).

Similarities between NH_3 fluxes calculated with the IHF and bLS-Configuration I technique (Normalised Mean Square Error of 0.025) might be expected because they were both based on the same 'single point' measurements and, therefore, should have had a similar 'footprint'. The observed small differences between derived NH_3 flux estimates from the IHF and the bLS-Configuration II method (Normalised Mean Square Error of 0.031) suggested that a better plot coverage than that achieved with the experimental configuration I (6 and 19% of touchdown values from Windtrax for configurations I and II, respectively) did not lead to a significantly different result, suggesting an uniform emission rate across the slurry-applied plot. In cases where a uniform emission rate is not expected then multiple down wind concentration measurements, or the use of a long-path laser instrument to provide an integrated down wind concentration measurement, would provide a more accurate estimate of emission from the treated area than single point IHF or bLS techniques. In this study we treat the IHF measurement of volatilization rates as "truth", ignoring the possibility of measurement errors. For example, with short-averaging intervals the most commonly applied IHF formula is known to over-estimate surface emissions due to the neglect of turbulent fluxes (e.g. Gao et al., 2009). However, the potential for IHF errors when applied to our long-averaging times and our passive samplers is uncertain, and at present we will assume the error is suitably small.

Cumulative NH_3 emissions were 19.3, 21.2 and 18.4 kg N ha^{-1} for IHF and bLS configurations I and II, respectively (Fig. 2b), representing 20.1, 22.0 and 19.1 %, respectively, of the Total Ammonium Nitrogen (TAN) applied with the slurry. Emission within the first 24 h after slurry application accounted for 63-75% of the total NH_3-N volatilised over the entire measurement period, which is typical for surface applied slurries (e.g. Pain et al., 1989).

Cumulative NH_3 flux derived using the bLS technique, with assumed neutral stability, gave values 10% greater and 5% less, for experimental configurations I and II, than that calculated using IHF (Fig. 2b). These differences were smaller than those considered as acceptable in previous studies (Sommer et al., 2005), also using long sampling intervals (i.e. 5-26 h) and assuming neutral stability in which the bLS technique underestimated NH_3 emission by 16-24% in comparison with IHF. However, it should be noted that in the present study the cumulative accuracy of the two measurement techniques was essentially determined by the accuracy of the first observation, which took place during the shortest measurement interval (4.5 h).

Both the IHF and bLS techniques will be subject to uncertainties. Denmead et al (1977) estimated the uncertainty in the IHF technique to be of the order of 20% and Misselbrook et al. (2005) reported coefficients of variation of 10% (across 6 tests of paired samples) for concentration measurement using PFS and 23-52% (triplicate plots, 4 tests) for the IHF technique as a whole. For the bLS technique, similar measurement errors will exist for ammonia concentration, wind speed and direction measurements, so a similar level of uncertainty might be assumed. However, in addition to this is the assumption regarding atmospheric stability class, so an uncertainty analysis for this parameter was performed on the data from the present study by changing from neutral to stable or unstable conditions. For bLS experimental configuration I, the estimated cumulative emission of NH_3 was 21.8 and 19.8 kg N ha^{-1} for stability class unstable and stable, respectively (i.e. 3% overestimation and 7% underestimation compared with neutral stability). For bLS experimental configuration II, NH_3 emission estimates were 20.9 and 18.6 kg N ha^{-1} for stability class unstable and stable, respectively (i.e. 13% and 1% overestimation compared with neutral stability). The range in F_{bLS}/F_{IHF} was therefore 1.03–1.13 and 0.95–1.08 for bLS configurations I and II, respectively, across the different stability classes. Uncertainty due to the assumption of neutral conditions in the present study can therefore be regarded as relatively small.

The results of the present study provide further confirmation of the findings of Sommer et al. (2005) that the bLS technique can be used with longer sampling averages provided that concentrations are measured at a height least affected by atmospheric stability.

4. Conclusion

Cumulative NH_3 emission after surface application of pig slurry were 20.1 and 22.0% of the Total Ammonium Nitrogen (TAN) applied according to the estimated by the IHF and the bLS technique, respectively. Using the bLS inverse dispersion model technique to estimate NH_3 emissions following pig slurry application to land gave values to within 5-10% of those estimated using the reference (IHF) technique. The results of this study have demonstrated that the bLS dispersion model can be effectively used, even in a non-ideal way (i.e. long sampling intervals), for estimating NH_3 emission from field sources.

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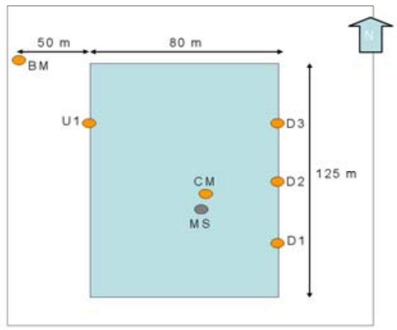
Figure captions

Figure 1. Experimental layout at Valverde del Majano (Segovia, Spain). Orange coloured dots represent the measurement masts. Central Mast (CM) and Background Mast (BM) were equipped within 5 passive flux samplers at heights of 0.25, 0.5, 1.25, 2.05 and 3.05 m. Whereas, Downwind Masts (D1, D2, D3) and Upwind Mast (U1), equipped with one PFS at 1 m of height, were placed in the downwind and upwind edges of the plot, respectively. The grey dot represents the Meteorological Station (MS) used during the experiment.

Figure 2. Ammonia emission rate (a) and cumulative NH_3 emission (b) measured with the IHF method and bLS method for configuration I and II.

Figure 3. Ammonia emission estimated with the bLS technique, configuration I (a) and II (b), respectively, using average wind speed at 1 m height, surface roughness length (Z_0) of 0.01 m, average wind direction (β) determined at 1 m height, scalar NH_3 concentration measured at 1 m and assuming neutral atmospheric stability vs. IHF reference estimates (1:1 line is included).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BM
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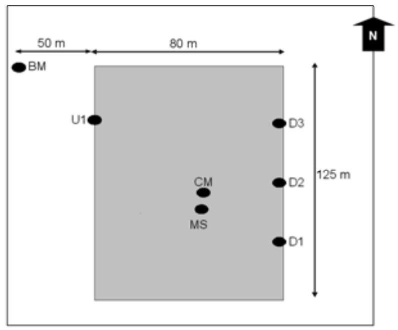
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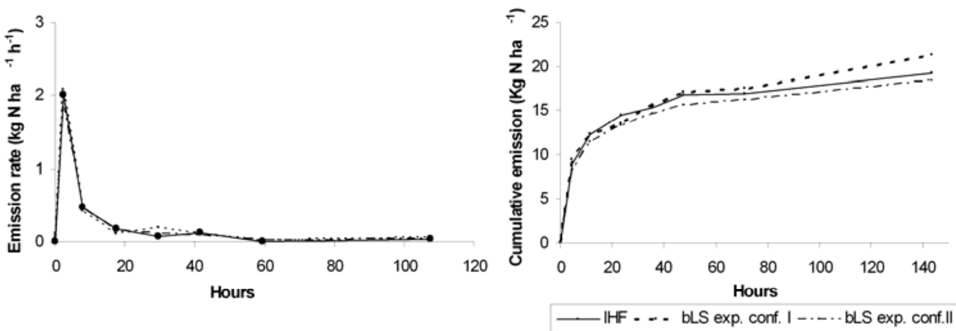
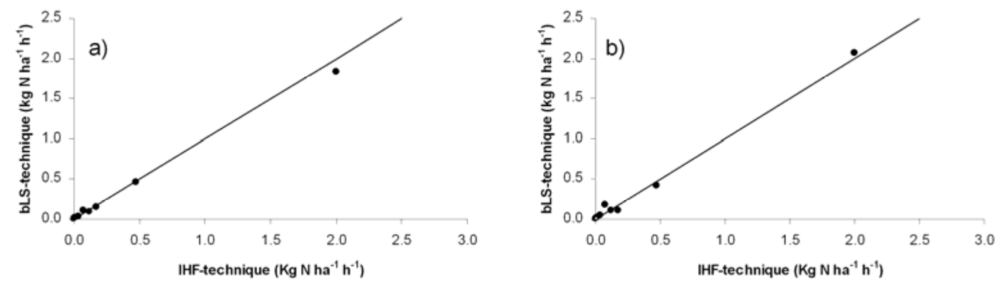

Emission rate (kg N ha-1 h-1)
3
2
1
0
0
20
40
60
80
100
120
Hours
Cumulative emission (Kg N ha-1)
25
20
15
10
5
0
0
20
40
60
80
100
120
140
Hours
IHF
bLS exp. conf. I
bLS exp. conf.II



a)
b)
bLS-technique (kg N ha^{-1} h^{-1})
IHF-technique (Kg N ha^{-1} h^{-1})
0.0
0.5
1.0
1.5
2.0
2.5
3.0