Agricultural and Forest Meteorology 234–235 (2017) 196–202

Contents lists available at ScienceDirect

Agricultural and Forest Meteorology

journal homepage: www.elsevier.com/locate/agrformet

Evaluation of a backward Lagrangian stochastic model for determining surface ammonia emissions

Wenliang Yang, Anning Zhu*, Jiabao Zhang, Xiuli Xin, Xianfeng Zhang

Fengqiu Agro-ecological Experimental Station, State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, 71 East Beijing Road, Nanjing 210008, China

ARTICLE INFO

Article history:
Received 22 July 2016
Received in revised form
26 December 2016
Accepted 2 January 2017
Available online 9 January 2017

Keywords:
Ammonia emission
Backward Lagrangian stochastic model
Open-path tunable diode laser
Integrated horizontal flux method
Static chamber method

ABSTRACT

The rate of ammonia emissions from a small circular plot of maize was estimated by three procedures: (i) by inverse dispersion (based on upwind and downwind laser concentration-detectors interpreted using a backward Lagrangian stochastic model); (ii) by the height-integrated horizontal flux measured by point flux-detectors arrayed along the axis of the plot (i.e., the integrated horizontal flux method, IHF); and (iii) by extrapolation from static flux chambers (SC). The results indicated that the estimates made by the open-path tunable diode laser (OPTDL) system combined with the backward Lagrangian stochastic (BLS) model were statistically equivalent to those made by the IHF method. The ammonia fluxes estimated by the OPTDL-BLS technique were only 2.3% higher than those from the IHF method. Although the OPTDL technique failed to monitor concentration differences at low ammonia fluxes due to its detection limit, the OPTDL-BLS technique estimated the total ammonia loss to be only 10.9% less than the IHF results. The SC method was found to underestimate ammonia emissions and cumulative ammonia loss significantly compared with both the OPTDL-BLS and IHF methods Although the ammonia emissions estimated by the OPTDL-BLS technique showed a similar emission pattern to those estimated by the IHF method, the former provided an opportunity to estimate the diurnal pattern of ammonia emissions and to understanding the primary driving factors. A clear diurnal cycle and a dominant net solar radiation dependence in ammonia emissions were found.

1. Introduction

Ammonia (NH_3) is the most abundant alkaline gas in the atmosphere. It has many negative effects on ecosystem function and health, and on air quality. Deposition of NH_3 and NH_4^+ ions is an important contributor to soil acidification, eutrophication of natural ecosystems, and nitrate leaching (Binkley and Richter, 1987; Schulze et al., 1989). Further, NH_3 is a chemically active gas and readily reacts with sulfates and nitrates to form particulates of mean aerodynamic diameter smaller than 2.5 μm (PM2.5) (Asman et al., 1998) which have been implicated in human respiratory problems. Agriculture is the main source of atmospheric NH_3, accounting for some 55–56% of global NH_3 emissi ons (Bouwman et al., 1997; Schlesinger and Hartley, 1992). The volatilization of NH_3 following fertilizer application is recognized as an important path of nitrogen movement to the atmosphere. This not only reduces the efficiency of applied nitrogen-based fertilizers, but it also results in financial loss to farmers. Accurate measurement of NH_3 emissions is therefore pertinent and necessary.

There are many methods of measuring ammonia emissions: the dynamic chamber method, the static chamber method, the micrometeorological mass balance method, the eddy covariance method, the relaxed eddy accumulation method, the flux gradient method, the wind tunnel method, and so on. Depending on the purpose of gathering this information, these methods may be grouped into two categories: chamber methods and micrometeorological methods. Due to the modification of the environment by the chamber, chamber methods are mainly used to compare NH_3 losses from different treatments; whereas micrometeorological methods provide reliable estimates of ammonia emissions and are often used for the quantitative determination of ammonia losses.

Flesch et al. (1995) described a backward Lagrangian stochastic (BLS) model that calculates emission rates from measurements of wind speed and upwind and downwind concentrations at a single height and is independent of the size and shape of the source. With gas release experiments, Flesch et al. (2004), Gao et al. (2009), McBain and Desjardins (2005) and Ro et al. (2013) evaluated the accuracy of the BLS model and offered some guidelines for its use.

* Corresponding author.
E-mail address: anzhu@issas.ac.cn (A. Zhu).

http://dx.doi.org/10.1016/j.agrformet.2017.01.001
0168-1923/

This technique has been successfully used to estimate ammonia (Flesch et al., 2007; McGinn et al., 2007) and methane emissions (Loh et al., 2008). However, few reports are available on use of the BLS model for estimating NH_3 emissions from fertilized cropland.

This present study compares estimated NH_3 emissions from a circular plot treated with urea using the BLS model against estimates using the integrated horizontal flux (IHF) method and the static chamber (SC) method. The objective was to assess the accuracy and capability of the BLS technique for estimating NH_3 emissions from farmland.

2. Materials and methods

2.1. Experimental site and set-up

The experiment was conducted in a maize field adjacent to the Fengqiu Agro-Ecological Experimental Station of the Chinese Academy of Sciences in Fengqiu County, Henan Province, China (114°24′E, 35°00′N). The experimental site was flat, with no significant disturbing elements for several hundred meters in all directions. Commercial granular urea (total N ≥ 46.4%) was homogeneously applied by hand to the soil surface of a circular plot (radius 20 m) at a rate of 174 kg N ha^{-1} between 5:15 and 5:45 p.m. on June 18, 2012, during the seedling stage of maize. The experimental areas were irrigated with approximately 60 mm of water immediately after fertilizer application. The fields surrounding the experimental areas did not receive any N fertilizer during the experimental period to avoid the influence of NH_3 emitted from the surrounding area.

2.2. Measurement of ammonia emissions

Ammonia emissions were estimated using three methods: the BLS model, the IHF method and the SC method.

2.2.1. Backward Lagrangian stochastic model

The free software WindTrax2.0 (Thunder Beach Scientific, Halifax, Canada) based on the BLS model was used to calculate ammonia emission rates. The emission rate was estimated by simulating the upwind transport trajectories of the gas particles from the concentration measurement location in the downwind emission plume back to the source area (Flesch et al., 1995, 2004). The BLS model is based on Monin–Obukhov similarity theory (MOST), which states that over short time intervals (e.g., 15–60 min) the wind properties in a horizontally homogenous surface layer (height $z \leq 50$ m, but above a plant canopy) can be specified by four variables (Garratt, 1992): the atmospheric friction velocity u_*, the Obukhov stability length L, the surface roughness length z_0, and the wind direction β. Following Flesch et al. (2004), the line-averaged concentration (C_L) is assumed to be the average of P point concentrations spaced evenly along the measurement line, with the link to emission rate modeled by computing an ensemble of particle trajectories backward from each point. The BLS model then simulates the ratio of the concentration rise over the background to the emission rate, $(C/Q)_{sim}$:

$$(C/Q)_{sim} = \frac{1}{P}\sum_{i=1}^{P}\left(\frac{1}{N}\sum\left|\frac{2}{w_o}\right|\right), \quad (1)$$

where N is the number of particles released at each point, P is the number of specific release points, and w_0 is the vertical 'touchdown' velocity (i.e., the velocity of the gas particles as they reach the ground). The inner summation refers only to touchdowns within the emission source. The emission rate Q_{BLS} (kg N ha^{-1} d^{-1}) can therefore be calculated as:

$$Q_{BLS} = \frac{(C_L - C_b)}{(C/Q)_{sim}} \quad (2)$$

where C_b is the background concentration.

In this study, the number of particles released $N = 50,000$ and the number of release points $P = 50$ were used in the software. Each particle was followed horizontally for 500 m upwind.

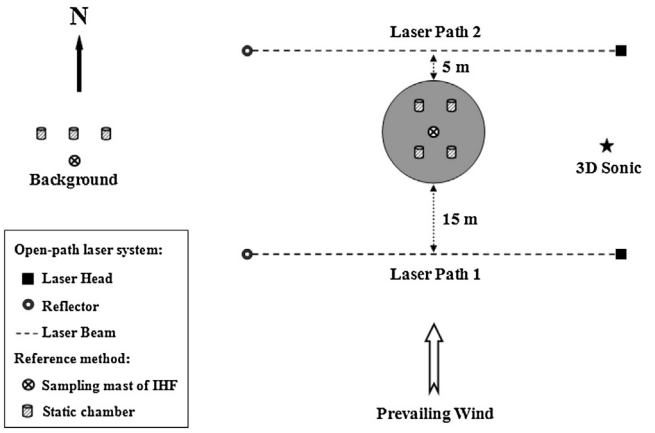

Fig. 1. Illustration of the source-laser configuration. The ammonia emission source is shown by the shaded area.

2.2.2. Integrated horizontal flux method

In this study, the IHF method, which is also known as the micrometeorological mass balance method (Denmead, 1983), was used as the standard reference method to estimate NH_3 emissions. It equates the vertical flux of NH_3 (i.e., the ammonia emissions) with the integrated horizontal flux (minus background flux) at a known downwind distance. As described by Denmead (1983) and Wilson et al. (1982), the net vertical flux of NH_3, Q_{IHF} (mg N m^{-2} s^{-1}), was calculated using the trapezoidal rule:

$$Q_{IHF} = \frac{1}{X}\int_0^z (F - F_b)\,dz = \frac{1}{X}\left[\int_0^{z_1}(F - F_b)\,dz + \int_{z_1}^{z_2}(F - F_b)\,dz + \cdots \int_{z_{n-1}}^{z_n}(F - F_b)\,dz\right], \quad (3)$$

where X (m) is the distance traveled by the wind over the fertilized area (i.e. the radius of the treated circle, 20 m in this case), z (m) is the height of the uppermost sampler, F (mg N m^{-2} s^{-1}) is the mean horizontal flux at each sampling height and F_b is the background horizontal flux. Ammonia flux rates Q_{IHF} were reported in units of kg N ha^{-1} d^{-1} to be comparable with Q_{BLS}.

The average horizontal flux of NH_3 was measured by passive flux samplers coated internally with oxalic acid, which were described in detail by Leuning et al. (1985). In this study, six passive flux samplers at $z = 0.4$, 0.8, 1.2, 1.6, 2.0, and 2.5 m were mounted on a mast placed at the center of the circular plot. A sampling mast was placed 200 m west of the plot to make background measurements at the same height (Fig. 1). After exposure, ammonium oxalate in the sampler was eluted with 100 mL deionized water, and the solution was analyzed for NH_4^+–N content by spectrophotometer (UV1601, Shimadzu, Japan) using the indophenol blue colorimetric method (Lu, 2000). The mean horizontal flux F was calculated as:

$$F = \frac{M}{At}, \quad (4)$$