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Assessing the backward Lagrangian stochastic model for determining ammonia emissions using a synthetic source

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ABSTRACT

The backward Lagrangian stochastic (BLS) model for estimating ammonia emissions from agricultural sources was assessed in an ammonia recovery experiment. The open-path tunable diode laser spectrometer was used to measure atmospheric ammonia concentrations. The results indicated that the ratio of estimated to actual emission (Q_{BLS}/Q) decreased with increasing concentration measurement height, and the optimum measurement height increased as fetch (F) increased. The accuracy of Q_{BLS} was insensitive to fetch with a range of 15–60 m. The optimum ratio of concentration measurement height to fetch decreased with increasing fetch. In this study, the optimum ratio of concentration measurement height to fetch was 0.083 and 0.045 for F = 15 m and F = 30 m, respectively. It is recommended that the laser sensor should be placed as close to the source as possible to reduce concentration measurement uncertainty for measurement of ammonia emission from farmland. The wheat canopy had a significant effect on Q_{BLS}/Q when the concentration measurement height above the wheat canopy was no more than 0.38 m. The laser path should be placed at least 0.58 m above the wheat canopy to minimize the effect of wheat canopy on the accuracy of the BLS model.

1. Introduction

Many micrometeorological techniques are available for estimating ammonia fluxes: the micrometeorological mass balance method, the integrated horizontal flux method, the eddy covariance method, the relaxed eddy accumulation method, the flux gradient method, inverse dispersion method, and so on (Harper et al., 2011). But requirement of most techniques for complex and expensive instrumentations and strict environmental conditions may also be prohibitive. The backward Lagrangian stochastic model (BLS) is an inverse dispersion method for inferring ground-to-air emissions. The BLS model has several advantages compared with other micrometeorological methods: insensitivity to the physical structure of the source, simple field observations, and flexibility in location of the concentration measurement (Flesch et al., 2004). The main advantage of this technique is that gas emissions can be determined with concentration measurements at just one height. A disadvantage of the BLS model is that its accuracy is inhibited by parameterization of turbulent transport (Denmead, 2008). But many field tests have demonstrated that with appropriate quality assurance and control, the BLS model can be used to determine gas emissions with acceptable accuracy (Flesch et al., 2004; McBain and Desjardins, 2005; Gao et al., 2009; Ro et al., 2013).

In theory, the BLS model can accurately simulate emission rates wherever the concentration measurement location may be within the emission plume. However, in practical usage, if the concentration measurement lies at the edge of the plume, the BLS model cannot accurately mimic atmospheric transport, and the concentration rise above background at the plume edge is too small to accurately detect (Flesch et al., 2004). Further, the modeling errors caused by wind and source complexity can be minimized by selecting an appropriate position for concentration measurement (Flesch et al., 2005). So placement of the downwind sensor for measuring gas concentration emitted from the source is of crucial importance in applying the BLS technique (Harper et al., 2011). Flesch et al. (2004) found that concentration measurements should be made at least 10H (H being the height of concentration measurement) downwind of the upwind source edge to avoid making measurements near the edge of the tracer plume. McBain and Desjardins (2005) found that the optimum measurement height was estimated to be 1.35 m with the average fetch of 22 m (fetch being the distance between the source and the position of concentration measurement, F). The two studies tested the model in

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homogenous surface layer, but neglected the influence of surface roughness. Ro et al. (2013) obtained the poor estimates by the BLS model when the adjacent corn crop grew more than 2 m in height, compared to that when the surrounding vegetation was short. Laubach (2010), Gao et al. (2010) and Grant et al. (2013) also assessed the influence of concentration or wind measurement location on the accuracy of the BLS model. But these evaluation experiments were made under the non-ideal site conditions (animal barn, pen or waste lagoon). When using the BLS model to estimate gas emissions from fertilized cropland which is the ideal source with spatially homogeneous configuration and evenly distributed emission rate, these guidelines may be unsuitable or complex.

This paper describes a field experiment in which the combined use of open-path tunable diode laser spectrometer and BLS model was employed to measure the release rate of ammonia from a synthetic tracer source. The objective of this study was to assess the influence of concentration measurement height, fetch and surface roughness on the accuracy of the BLS model.

2. Materials and methods

2.1. Backward Lagrangian stochastic model

A commercial software WindTrax2.0 (Thunder Beach Scientific, Nanaimo, Canada) based on the BLS model was used to calculate gas emissions. The WindTrax software calculates the emission rate Q_{BLS} by simulating the ratio of the concentration rise over background at the concentration measurement position to the source emission rate $(C/Q)_{sim}$. So the emission rate can be calculated as:

$$Q_{BLS} = \frac{(C - C_b)}{(C/Q)_{sim}}, \quad (1)$$

where C is the concentration within the downwind plume, and C_b is the background concentration. The detailed description of the BLS model is given in Flesch et al. (2004).

In the WindTrax software, particle release number $N = 50000$ and release point number $P = 50$ were used in the software. Each particle was followed horizontally for 500 m upwind.

2.2. Experimental set-up

The assessment experiment was conducted on a wheat field adjacent to the Fengqiu State Key Agro-Ecological Experimental Station of the Chinese Academy of Sciences in Fengqiu County, Henan Province, China (114°24′ E, 35°00′ N), in March–April 2012 and March 2013. The experimental site was flat with no significant obstructions for several hundred meters. There was only a row of about 15 m tall trees within about 100 m east of it. But measurements were suspended when the wind was easterly. Initially, the wheat was approximately 15 cm tall (re-greening stage), but by the end of the experiment it reached a maximum height of 45 cm (jointing stage).

2.2.1. Ammonia emission source

Ammonia emission source was a square area grid (6 m × 6 m) at ground level constructed from 18 mm (inner diameter) ethylene-propylene random copolymer (PPR) pipe with 112 holes (0.8 mm diameter) spacing 0.4 m according to Flesh et al. (2004). Ammonia (99.9% purity) released from holes was supplied from a high pressure ammonia cylinder coupled to the grid through a pressure regulator and rotameter. Release rates were varied from 5 to 40 L min^{-1} over the course of the experiment. Each release lasted from 30 to 120 min and was adjusted manually every minute or two to maintain a steady release rate. The holes were adjusted to provide uniform emissions, as the gas flow rate across the hole was much higher than that through the pipe (Flesch et al., 2004). Flesch et al. (2004) found that by a distance of 10 m, the inhomogeneity of the source can be neglected. To allow the downwind concentrations to reach equilibrium, the NH_3 gas release started 5 min before the measurements began. An electronic scale (precision = 5 g) was used to weigh cylinder before and after each release to determine the mass of the released ammonia. Over the course of the experiment, the minimum weight of the released ammonia was 0.195 kg, indicating that uncertainty in actual emission (Q) was no more than 2.6%.

2.2.2. Ammonia concentration measurements

Ammonia concentrations were measured using the tunable diode laser system (developed by Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, China) over open path. The system comprises three transmitter heads, three retroreflectors and a central unit which contains the laser source, a reference cell and the control and processing electronics. A laser beam of light (an NH_3 absorption line centered at 1544 nm) produced by a tunable infrared diode laser is divided into two beams by a beam splitter. The weaker beam passes through the reference cell to provide a continuous calibration update. The other beam emitted from the transmitter head is directed toward a retroreflector through the atmosphere and then returns to the transmitter head. The returning signal and the signal from the reference cell are processed by the central unit to determine the ammonia concentration along the measurement path. Details of the system, field data collection and analytical methods are summarized in Xia et al. (2008).

The performance of the system was evaluated through multi-point calibrations and single-point calibrations in the lab before the field observation period, using a 1 m calibration tube flooded with standard ammonia. Multi-point calibrations used three concentrations (20, 50 and 100 ppm) and conducted 6 times at every concentration. Single-point calibrations was conducted at 20 ppm every 30 min, and repeated 8 times. The precision of the measurement can be described by the relative error. The relative error of multi-point calibrations and single-point calibrations were less than 1.1% and 1.6%, respectively. The resolution of the system given by the manufacturer is 5 ppm-m for NH_3. The length of the laser paths was 100 m throughout the experiment period, corresponding to a resolution of 0.05 ppm. Ammonia concentrations were recorded every 7 s and averaged into 30-min values.

2.2.3. Test #1: various concentration measurement heights

To assess the accuracy of the BLS technique in estimating ammonia emissions at different concentration measurement heights and determine the optimum measurement height for the BLS model, the laser paths were mounted at a combination of heights (0.8, 1.0, 1.2, 1.5, 1.7 and 2.0 m above the ground). Owing to the limited number of sensors, the laser paths were set at 0.8, 1.2 and 1.7 m above the ground in early releases, and then mounted at heights of 1.0, 1.5 and 2.0 m. The three laser paths were aligned along a fixed direction of the east-west (Fig. 1). The gas release grid was placed 15 m or 30 m upwind of the center of the laser path (the distance between the downwind source edge and the mid-point of the laser path), and repositioned to be north or south of the laser path based on the direction of the wind. The experiment was conducted when the wheat was 15–20 cm (re-greening stage) and 40–45 cm (jointing stage) tall to evaluate the influence of the plant canopy on the ideal concentration measurement height. The background ammonia concentration at each measurement height was determined by calculating the average concentrations 30 min before and after each gas release. In order to minimize error, the volumetric