Atmospheric Environment 104 (2015) 1-10



Contents lists available at ScienceDirect

## Atmospheric Environment

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

## Estimating ammonia emissions from a winter wheat cropland in North China Plain with field experiments and inverse dispersion modeling



ATMOSPHERIC ENVIRONMENT



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#### HIGHLIGHTS

• Heterogeneous NH<sub>3</sub> emissions from a cropland are common due to patched fertilization.

• We estimate these heterogeneous NH<sub>3</sub> emissions via a detailed field-scale experiment.

• An inverse dispersion method is adopted with two downwind concentration measurements.

• Representative NH<sub>3</sub> emission factors of synthesis fertilizers in China are obtained.

### A R T I C L E I N F O

Article history: Received 30 September 2014 Received in revised form 30 December 2014 Accepted 2 January 2015 Available online 2 January 2015

Keywords: NH<sub>3</sub> emission factor Synthetic fertilizer Field-scale experiment Inverse dispersion method North China Plain

#### ABSTRACT

A field-scale experiment was conducted in the spring of 2012 at a winter wheat cropland, aiming to quantify ammonia (NH<sub>3</sub>) emissions from surface fertilization under realistic cultivation conditions. Since the fertilization lasted about 20 days for hundreds of divided plots and three types of fertilizers were used (i.e., urea, ammonium sulfate and compound nitrogen-phosphorous-potassium fertilizer), the heterogeneity was one of the significant characteristics of the cropland NH<sub>3</sub> emissions during the experiment, which is a great challenge for the classical micrometeorological methods to calculate NH<sub>3</sub> fluxes. Based on continuous measurements of NH<sub>3</sub> concentrations at two heights (2.5 m and 8 m) and detailed records of the fertilization plot by plot, an inverse dispersion method was employed to derive the heterogeneous NH<sub>3</sub> emissions and the corresponding emission factors (EFs). The EFs derived from this experiment for urea, ammonium sulfate and compound fertilizer were 12.0%  $\pm$  3.1%, 8.5%  $\pm$  1.6% and 4.5%  $\pm$  1.7%, respectively. The EF of urea we obtained was lower than most of other domestic measurements and those used in the NH<sub>3</sub> emission inventories in China. Measurements on EFs of ammonium sulfate and compound fertilizer were the torse of the state of the state and compound fertilizer are not available in China. However, the EFs of ammonium sulfate and compound fertilizer were Ltd. All rights reserved.

#### 1. Introduction

Ammonia (NH<sub>3</sub>) is the dominant alkaline species in the atmosphere and can react readily with acids to form fine particles (Seinfeld and Pandis, 2006). These fine particles can cause several environmental issues, such as visibility degradation (Battye et al., 2003; Ye et al., 2011) and climate change by radiative forcing effects (Martin et al., 2004; Deandreis et al., 2012). These particles are also great threats to human health (Pope and Dockery, 2006). When the formed ammonium compounds  $(NH_4^+)$  deposit to the soil, they can cause soil acidification through the process of nitrification and roots uptakes (Sutton et al., 1993).

Since modeling has been increasingly utilized to study these various effects of NH<sub>3</sub>, a robust emission inventory is essential (Zheng et al., 2012). There have been lots of NH<sub>3</sub> emission inventories around the world at different spatial scales (Sutton et al., 2008). However, great uncertainties still exist in these inventories, mainly due to the uncertainties in the emission factors (EFs) and

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the unrecognized sources (Sutton et al., 2008; Zheng et al., 2012). Kim et al. (2006) showed that NH<sub>3</sub> emissions may be overestimated in Asia. Due to the lack of robust field experiments, EFs are even more uncertain in the developing regions, which are the main NH<sub>3</sub> emission contributors globally (Bouwman et al., 1997). China, as the largest developing country, contributes about 20% of the global NH<sub>3</sub> emissions and 55% of Asian emissions (Olivier et al., 1998). Therefore, quantifying the NH<sub>3</sub> emissions from China is important for the understanding of the global NH<sub>3</sub> budget. Known as the second main NH<sub>3</sub> emitter, synthetic fertilizer application contributes about 20% of the global NH<sub>3</sub> emissions (Bouwman et al., 1997; Beusen et al., 2008) and up to 45% in Asia (Olivier et al., 1998; Streets et al., 2003). In China, NH<sub>3</sub> volatilized from synthetic fertilizer amounts to 30%–50% of the total emissions (Zhao and Wang, 1994; Streets et al., 2003; Huang et al., 2012).

Campaigns on quantifying NH<sub>3</sub> exchange of various ecosystems were intensively carried out in developed regions in the world (e.g., Flechard and Fowler, 1998; Sutton et al., 2000, 2009), in which various techniques on NH<sub>3</sub> flux measurements were applied, including wind tunnel (e.g., Sanz-Cobena et al., 2011), integrated horizontal flux method (e.g., Laubach et al., 2012), aerodynamic gradient method (e.g., Milford et al., 2009; Spirig et al., 2010) and relaxed eddy accumulation method (Hensen et al., 2009). Some state-of-art techniques have enabled NH3 flux measurements, such as the eddy covariance method (Whitehead et al., 2008; Sintermann et al., 2011b) and the inverse dispersion method (Flesch et al., 2004). In contrary, the NH<sub>3</sub> flux measurements in China are scarce (Dong et al., 2009; Zhang et al., 2010, 2011; Huang et al., 2012; Gong et al., 2013), and a major limitation is that the enclosure and integrated horizontal flux method are most taken, both of which are suitable for spatial scales less than several tens of meters. Sintermann et al. (2012) pointed out a scale effect that NH<sub>3</sub> emissions of fertilization measured from small-scale plots are higher than that from field-scale measurements (>100 m). Moreover, experiments with the enclosure and integrated horizontal flux method are usually implemented under controlled conditions, which cannot represent the actual emissions for the realistic cultivation condition. It is acknowledged that field-scale measurements can be more representative of the real emission process.

In field-scale measurements under realistic cultivation conditions, since the fertilization for the whole farmland cannot be accomplished simultaneously and usually different types of fertilizer are used, heterogeneity is one of the significant characteristics of the cropland NH<sub>3</sub> emissions, which is a great challenge for the classic micrometeorological methods to calculate NH<sub>3</sub> fluxes. For example, the conventional gradient method may suffer from large uncertainties, not only for the inaccurate parameterization of the eddy diffusivity (Huo et al., 2014), but also for the advection errors (Loubet et al., 2009).

In this paper, we describe an experiment conducted in the spring of 2012 in North China Plain, aiming to quantify the fieldscale NH<sub>3</sub> emissions from surface fertilization. The experiment was designed to estimate the NH<sub>3</sub> emissions under realistic cultivation conditions. That is, different plots of the cropland were cultivated by different farmers and the fertilization practices were conducted plot by plot. Therefore, estimation of the NH<sub>3</sub> emission is challenged by the heterogeneous sources in the field-scale: temporal and spatial variations of NH<sub>3</sub> emissions. For such heterogeneous emissions, a method similar to Huo et al. (2014) was employed, which is based on the principle of the inverse dispersion method (IDM). They inferred the heterogeneous cropland evapotranspiration of the same experiment via IDM with two-level downwind water vapor measurements. The objectives of this paper are two folds: one is to extend the IDM method to the estimation of heterogeneous NH<sub>3</sub> emissions; the other is to obtain the realistic  $NH_3$  EFs for the major emission period of winter wheat cropland in North China Plain.

This paper is organized as follows: Section 2 provides an overview of the experiment site and details on the field measurements. Section 3 describes the implementations for the inference of  $NH_3$  emission rates using IDM. Then in Section 4, results and uncertainty analysis are given. And a summary is provided in Section 5.

#### 2. Site and observations

#### 2.1. Study site

Our experiment was conducted in a representative farmland in the North China Plain (37°32'04"N, 115°54'51"E, 16 m a.s.l.) in Guangchuan town, Hebei province. Huo et al. (2014) described the study site in detail. Fig. 1 presents the illustrations of the study area. The main Characteristics of the soil in the experimental field are given in Table 1. The experiment was carried out from 25 March to 6 May 2012, which encompassed the local spring topdressing procedures. During the experiment, most of the plots were planted with winter wheat, while others were left as bare soil. Plots were irrigated immediately (within 30 min) after fertilization. The fertilization and irrigation lasted for more than 20 days across the whole study area, extending from 28 March for the first plot until 19 April for the last one. Detailed information on fertilization dates, fertilizer types and dosage were recorded plot by plot. The approximate fertilization sequence is illustrated in Fig. 1. During the experiment, three types of fertilizers were used, i.e., urea (UR, N content 246.3%, particle size: 0.85-2.80 mm in diameter), ammonium sulfate (AS, N content  $\geq$ 20.5%, S content  $\geq$ 24%) and compound (nitrogen-phosphorous-potassium) fertilizer (CF, N: P<sub>2</sub>O<sub>5</sub>:  $K_2O = 30: 5: 5$ ). Fig. 2 illustrates the spatial distributions of fertilizer types. The total input N for UR, CF and AS are averaged as 140 kg N ha<sup>-1</sup>, 117 kg N ha<sup>-1</sup> and 122 kg N ha<sup>-1</sup>, respectively.



**Fig. 1.** Illustrations of the study area. The center of the study area locates at (1000, 1000). The approximate fertilization sequence is indicated by arrows and dates (month-day).Wind rose during the experiment is shown at the upper right corner.

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