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High-resolution measurement of ammonia emissions from fertilization of vegetable and rice crops in the Pearl River Delta Region, China

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HIGHLIGHTS

► In-situ measurement on ammonia emissions were carried out by self-designed system.
► NH_3 emission fluxes with 30-min data resolution from vegetable and rice fields were obtained.
► The real-time meteorological conditions of the experiment area were monitored.
► Ammonia emission flux was positively correlated with temperature in most experiments.

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ABSTRACT

Loss of ammonia (NH_3) as a result of intensive N fertilization, especially due to agronomic practices in South China, is not well characterized. To investigate mechanisms and characteristics of NH_3 volatilization after urea application, an on-line monitoring system, with 30-min data resolution, was used to study vegetable and rice fields from January 2009 to September 2010. Ammonia emissions and concurrent meteorological conditions were monitored for up to 20 days after fertilization in 12 experiments. Standard recovery test results indicated that the on-line measurement system was both stable and accurate. The NH_3 emission factors (EFs) related to broadcast (soil surface) basal dressing and top dressing to *Brassica rapa* L. were 23.6% and 21.3%, respectively. The NH_3 EFs from holing basal dressing and broadcasting top dressing for lettuce were 17.6% and 24.0%, respectively. The NH_3 EFs for early rice in parallel broadcast basal dressing process were 10.7% and 14.2%, while in parallel top dressing process were 24.0% and 22.6%, respectively. The NH_3 EFs for late rice were 15.4% and 21.0% in parallel broadcasting basal dressing process, while 13.2% and 17.6% in parallel top dressing process. Emission of NH_3 from vegetable and rice fields occurred mainly in the first 2–3 weeks after fertilization. Ammonia emission flux was positively correlated with air temperature and soil temperature in the majority of the experiments. Relationships between NH_3 emissions and humidity, soil moisture or wind speed were explored, which were not consistent among all tests. Ammonia emission in vegetable and rice fields was primarily associated with temperature. High-resolution data, such as those gathered in the current investigation, will contribute to a more thorough quantitative understanding of the relationship between fertilizer application, environmental conditions, and NH_3 volatilization which, in turn, will improve the accuracy of atmospheric modeling on local, regional and global scales.

1. Introduction

As a natural component and a dominant atmospheric alkaline gas, NH_3 plays an important role in atmospheric chemistry and ambient aerosol formation (Erisman and Schaap, 2004; Quan and Zhang, 2008; Zhang et al., 2010, 2011b; Ye et al., 2011; Langridge et al., 2012). Ammonia is a major component in atmospheric particles and in precipitation, which is a key precursor to neutralization of H_2SO_4 and HNO_3 in the air, forming $(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3 (Anderson et al., 2003; Erisman and Schaap, 2004;

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Walker et al., 2004; Olszyan et al., 2005; Sharma et al., 2007; Renner and Wolke, 2010; Behera and Sharma, 2012). The resulting particles may contribute to degraded visibility, regional haze and health impacts associated with fine particulate matter (Kim et al., 2006; Pinder et al., 2007; Langridge et al., 2012).

Utilizing approximately 9% of the world's arable land, and feeding 22% of the human population, China has consumed more than 30% of the world's nitrogen (N) fertilizer over the last decade (http://www.stats.gov.cn/; IFA, 2011). A portion of the field-applied nitrogen fertilizer is converted to ammonium ions and, eventually, to non-ionized NH_3, a large portion of which escapes in gaseous form to the atmosphere (Lin et al., 2007). Studies of NH_3 emission inventories suggested that approximately 20% of global NH_3 emissions originated from China (Zhao and Wang, 1994; Klimont et al., 2001; Yamaji et al., 2004), in particular, from intensive N-fertilizer application to agricultural areas (Yan et al., 2003; Wang et al., 2005). In 2006, the total emissions for ammonia in China was 9.8 Tg, of which nitrogen fertilizer application and livestock manure management were the largest contributors, at 3.2 Tg and 5.3 Tg, respectively (Huang et al., 2012). In China, the greatest contribution to NH_3 emissions after application of synthetic N fertilizer came from urea (Zhang et al., 2011a).

Several field studies in China have explored NH_3 emissions related to fertilizer application, most of which occurred in the Taihu Lake region and used relatively low resolution metrics (Cai et al., 1985; Freney et al., 1987; Cao et al., 2000; Tian et al., 2001; Deng et al., 2006; Fan et al., 2006; Huang et al., 2006; Tian et al., 2007; Li et al., 2008a). Results from these studies indicated that NH_3 volatilization during rice planting was highly variable and affected by several factors (Fan et al., 2006). Reported NH_3 emission rates (as net N) from applied nitrogen fertilizer ranged from 10% to 50% (Anderson et al., 2003; Streets et al., 2003). Despite the fact that total acreage dedicated to vegetable production in China has increased sharply in recent years, there have been few studies on NH_3 emission related to fertilization of vegetable fields (Yan, 2008). Limited investigations have indicated that NH_3 emissions from vegetable croplands were likely to be significant (He et al., 2005; Zhang et al., 2009).

The Pearl River Delta (PRD) region of southeastern China, one of the most densely populated and economically developed regions in the world, faces serious, and regionally complex, air pollution problems (OECD, 2010; Zheng et al., 2009). Cities in the region have suffered from a high concentration of fine airborne particles, frequent haze and acid rain events (Zhang et al., 2008). The NH_3 emitted from synthetic nitrogenous fertilizers was considered to be an important, though poorly studied, contributor to local atmospheric pollutants in the PRD region (Zhang et al., 2008). Uncertainty analysis for a high-resolution NH_3 emission inventory of the PRD region revealed that a relative error of N fertilizer application was high which mainly depended on adopting foreign NH_3 emission factors (EFs) (Zheng et al., 2011). The questionable reliability of EFs and high-resolution emission profiles are two of the major obstacles in more fully understanding the role of NH_3. Previous field investigations, designed to better elucidate NH_3 emission characteristics in China, yielded variable EFs, indicating the calculation of these factors by extrapolation alone may result in significant uncertainty that failed to contribute to a more accurate understanding of farming practices and subsequent NH_3 contributions (Zhang et al., 2011a). Obtaining high-resolution (e.g. hourly) measurements would reduce uncertainty and enhance the development of robust quantitative estimates.

Regardless of the qualitative nature of many previous studies, the extant data do show that NH_3 emitted from agricultural applications is significant and suggest the necessity for a better description of emission profiles from fertilizer sources. The information on NH_3 emission characteristics is also useful in tabulating reliable emission inventories and evaluating regional atmospheric chemistry. In the current investigation, the emission characteristics of NH_3 from typical vegetable (*Brassica rapa* L. and lettuce) and rice (early rice and late rice) after urea application were measured by a self-designed online system with a relatively high resolution of 30 min. The correlation between NH_3 emission and management and metrological conditions were also explored.

2. Experiments and methodologies

2.1. Site description

The PRD region, located along the southeastern coast of China, is an important agricultural area, producing several crops, including a variety of vegetables. Large amounts of nitrogenous fertilizers were applied in order to reach high productivity goals (EBASYG, 2009). The area dedicated to vegetable production has increased from 243,451 hm^2 in 1990 to 464,777 hm^2 in 2009, accounting for 40.8% of the total vegetable production area in Guangdong province. During the same period, the rice acreage reduced to 524,264 hm^2, accounting for 26.8% of the total rice planting area in Guangdong province. Urea is one of the most commonly used synthetic fertilizers in the cultivation of vegetables and rice.

An experimental field (22°35′32″N, 113°58′21″E) was selected in Shenzhen, an eastern city of the PRD (Fig. 1). The altitude of the experimental site is 36 m, with an annual average air temperature of 22.5 °C, and an annual average wind speed of 2.7 m s^{-1}. The annual mean rainfall was 1966.5 mm, concentrated mainly during the rainy season from April to September. Soils in the experimental fields were somewhat acidic (Table 1). Soil from the rice field was less acidic than that of vegetable field, and also had relatively higher concentrations of total organic carbon (TOC), N, P and K.

2.2. Fertilizer treatment

B. .rapa L. (often referred to as turnip mustard or field mustard) and lettuce were the vegetables selected for the study. In addition, rice (*Oryza sativa* L.), both early rice and late rice, was studied. The dose of applied fertilizer represented a typical amount used in traditional methods for growing vegetables and rice. Vegetable seeds and rice seedlings were planted a few days prior to fertilizer application. All local dominant agricultural management practices in terms of crop type, rotation regime, tillage, irrigation and fertilization (including timing, rate, and source) followed traditional methods. The crop-growing process was managed and maintained by local farmers.

B.. rapa L. and lettuce were treated separately, with one round of holing basal dressing into a hole and one round of top dressing. Early rice and late rice in parallel tests were treated in exactly the same manner at the same time. Samples were taken continuously, and test duration ranged from 15 to 20 day post-fertilization. A common urea fertilizer (Trademark: ZhiHua; N content $\geq$ 46.3%; particle size: 0.85–2.80 mm in diameter; Standard: GB2440-2001) was used (Table 2).

As prior researches had suggested, NH_3 emission from nitrogen fertilizers occured approximately two or three weeks after fertilizer application (Bouwman et al., 2002; Zhang et al., 2005; Su et al., 2007; Li et al., 2008b,c). The samples in test 1 were collected continually for 19 days, revealing an increase of 8% in total NH_3 emission when compared to the emission in a 15 days test in a vegetable field. These data indicated a 15-day cycle was suitable to adequately record NH_3 emission in a vegetable field. 92% of the total experiments' duration on monitoring ammonia emission after application of synthetic N fertilizer before 2002 was less than