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Changes in fertilizer categories significantly altered the estimates of ammonia volatilizations induced from increased synthetic fertilizer application to Chinese rice fields

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ABSTRACT

As the primary rice-producing country in the world, China has applied increasing amounts of synthetic fertilizers on rice fields, which has a large impact on environmental pollution and human health. In this study, a comprehensive inventory of fertilizer application to rice fields was compiled for China from 1979–2015. In 2015, fertilizer application was estimated to be 0.96×10^9 kg for early rice, 2.67×10^9 kg for single rice, and 1.11×10^9 kg for late rice. Based on the fertilizer application and region-specific emissions factors, ammonia (NH_3) volatilizations from growing-season rice fields were estimated over the same period. We found that the total NH_3 emissions increased during 1979–1998, while decreased during 1998–2015 with fluctuations. The decreasing trend of NH_3 emission was likely attributed to changes of application proportions of ammonium bicarbonate (ABC) and other fertilizers. ABC and urea were the two dominant contributors, and contributed over 90% of the total NH_3 emissions. Spatially, high emissions were identified in the Middle-lower Yangtze River Plain, Huaihe River Basin, Taihu Lake region, Pearl River delta and Sichuan basin. Monthly emissions patterns were estimated according to rice calendars. The damage from NH_3 emissions was estimated as 26.79 billion yuan and accounted for 0.04% of the Gross Domestic Production of China in 2015. Finally, we discussed the NH_3 reduction strategies from the perspective of both the government and farmers and suggested that an incentive scheme should be established to guide farmers to optimize traditional agricultural management.

1. Introduction

China is the major rice-producing country in the world. The annual rice yield and harvested area were 208.24 million tons and 30.57 million hectares, or 28.08% and 18.79% of the global rice yield and harvested area in 2014, respectively (FAO, 2014). The high rice yields in China mainly results from the massive amount of synthetic nitrogen (N) fertilizer application (Xia and Yan, 2012). As the largest consumer in the world, China applied 31.07 million tons of chemical fertilizers, accounting for 28.53% of the worldwide total in 2014 (FAO, 2014). However, the excessive application of fertilizer has resulted in low utilization efficiency. The nitrate use efficiency in China is 30 − 40%, whereas in developed countries, it is 70 − 80% (Raun and Johnson, 1999; Zhang and Li, 2003). About 10 − 50% of N applied to farmland is released through volatilization (Xu et al., 2012). Ammonium bicarbonate (ABC) and urea are two common synthetic N fertilizers in China. Nonetheless, these fertilizers, because of the high ammonia (NH_3) volatilization rate, can easily emit ammoniacal nitrogen after being applied to farmlands (Yan et al., 2003). The total NH_3 emissions in China accounted for approximately 55% and 21% of the Asian and global NH_3 emissions, respectively (Zhao and Wang, 1994). Compared to uplands, rice fields have a higher potential for NH_3 volatilization (Xing and Zhu, 2000). However, there is limited knowledge about the detailed spatiotemporal variability of NH_3 emission of rice fields in China.

Gaseous NH_3 is a primary atmospheric alkaline trace gas that is a key atmospheric pollutant with various kinds of impacts. Due to its high reactive property, NH_3 usually reacts with atmospheric acidic gases (SO_2 and NO_X) to form ammonium-containing aerosols ($(NH_4)_2SO_4$, NH_4HSO_4 and NH_4NO_3), which are the secondary inorganic

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components in $PM_{2.5}$ (fine particulate matter smaller than 2.5 micrometers in aerodynamic diameter) (Lee et al., 2003; Ouyang et al., 2015). Such aerosols can have detrimental consequences on air visibility, climate change, and human health (Wang et al., 2013; Zhao et al., 2016). Moreover, particulate ammonium (NH_4^+) can exist longer than NH_3 in the atmosphere, and can be transported over a long distance (Asman et al., 2010; Zhao et al., 2017). Excessive deposition of atmospheric NH_3 and NH_4^+ may fertilize the oligotrophic ecosystems on the planet's surface, which may contribute significantly to soil acidification, water eutrophication, and loss of biodiversity (Kramer et al., 1990; Sprague et al., 2017). Furthermore, NH_3 can be transformed to nitrous oxide (N_2O), which is one type of greenhouse gases (Zaman et al., 2009). Therefore, a reduction in NH_3 emissions palys an important role in preventing regional air pollution, mitigating global climate change and protecting human health (Galloway et al., 2008).

Based on emissions factors (EFs) and agricultural activity data, a number of previous studies have estimated NH_3 emissions from farmlands in China (Beusen et al., 2008; Zhang et al., 2011). In these studies, NH_3 emissions from different base years were calculated using identical or EFs from developed countries throughout China without consideration of differences in typical local environmental conditions and NH_3 volatilization potential of various crop categories, which amplified the degree of uncertainty in these inventories (Yan et al., 2003). Furthermore, EFs were derived from studies in early years without consideration of up-to-date EFs from in situ measurements (Huang et al., 2012). Finally, estimates at the national- (Olivier et al., 1998), provincial- (Huang et al., 2012), and city- (Xu et al., 2015) level may hide regional trends based on a corresponding level of agricultural activity data. High-resolution agricultural activity data were needed to identify NH_3 emissions hotspots. Therefore, a comprehensive quantification of the NH_3 emissions of rice fields in China remains a challenge.

In 2015, the Chinese government issued a "Zero-growth Action Plan" for chemical fertilizer application by 2020 with the aim to decrease the environmental and economic costs from food production (MOA, 2015). Therefore, instead of just estimating an NH_3 emissions inventory for crop fields from an environmental perspective, combining this with an economic assessment is beneficial for translating scientific results into essential information for policy makers (Amann et al., 2011). Meanwhile, the integration of NH_3 emissions estimation and economic assessment may provide assistance to famers to determine both the environmental and economic effects from rice production systems (Xia and Yan, 2012). Furthermore, it provides useful guidelines for advanced technologies and integrated agricultural management strategies to improve fertilizer use efficiency and to decrease NH_3 emissions, especially in hotspot regions (Tian et al., 2014).

To improve the understanding of NH_3 emissions from rice fields, we analysed the spatiotemporal variability of synthetic N fertilizer from 1979 to 2015 in China in this study. Then, high-resolution estimates of NH_3 emissions were generated based on various categories of synthetic N fertilizers and their EFs from rice fields and the rice-cropping calendar. The historical patterns and spatial dynamic structure of NH_3 emissions, as well as the monthly variation, from 1979 − 2015 in the Chinese mainland (Hong Kong, Macao and Taiwan were not included), are discussed in this study. Furthermore, an economic assessment of Chinese rice production was performed. Finally, uncertainty analysis was conducted to evaluate the accuracy and feasibility of these estimates.

2. Materials and methods

2.1. Calculation of synthetic fertilizer use amounts

In this study, synthetic N fertilizer use amounts were calculated by considering three rice crops (early rice, single rice and late rice). Synthetic N fertilizers were classified into five categories, which included urea, ABC, ammonium nitrate (AN), ammonium sulfate (AS), and other N. The synthetic fertilizer use for these five types were estimated using a bottom-up method and rice paddy planting structure database (Dangal et al., 2017). The basic equation for synthetic fertilizer application is as follows:

$$U_{Total} = \sum_i U_{i,j} = \sum_i \sum_j \sum_m \sum_n p_{i,j,m,n} \times A_{i,j} \times \eta_j \times 10^{-9} \quad (1)$$

where U is the synthetic fertilizer use amount for a given rice crop in China (Tg yr^{-1}); p is the synthetic fertilizer use intensity for rice (kg ha^{-1}); i is the fertilizer type; j is the rice category; m is the different fertilizer application allotment (basal, tillering, and panicle fertilization); n is the county; A is the annual rice land area in each county (ha); and η_j is the cropping index for various rice crops.

There are different fertilizer application rates in different rice-growing seasons in different regions. We surveyed the proportion of fertilizer applications in 31 provinces from the corresponding local Academy of Agricultural Sciences and previous studies (Table S1). Then, the synthetic fertilizer use intensities in different application splits were estimated using Eq. (2):

$$p_{i,j,k} = p_k \times \varepsilon_{i,j,k} \quad (2)$$

where ε is the fertilizer application proportion; i is the different fertilizer application splits; j is the fertilizer category; and k is the province.

Due to the lack of the cultivated area data for three rice crops at the county level, we assumed that the cropping index at the county level for each kind of rice was equivalent to the provincial level. The cropping index was calculated using Eq. (3):

$$\eta_{i,j} = \frac{AS_{i,j}}{AP_{i,j}} \quad (3)$$

where AS and AP are the cultivated area and rice land area (ha); i is the province; and j is the rice crop.

Synthetic N fertilizer use intensity data and different fertilizer type application proportions during the rice-growing season in each province from 1979 to 2015 were acquired from the National Agricultural Production Cost and Revenue Information Summary (http://data.cnki.net/), Xu et al. (2015) and Zhang et al. (2011). Rice cultivated area data, and rice land area data at the province and county levels over the same period were provided by the China Statistical Yearbook (http://data.cnki.net/yearbook/) and the Chinese Rural Statistical Yearbook (http://tongji.cnki.net/). The rice calendar was obtained from the Chinese Ministry of Agriculture (http://www.zzys.moa.gov.cn/) and is shown in Table S2.

2.2. Estimation of NH_3 emissions from rice fields

The NH_3 emissions from Chinese rice fields were estimated using a bottom-up method (Beusen et al., 2008; Ouyang et al., 2016a). The NH_3 emissions were calculated using Eq. (4):

$$E_{NH_3} = \sum_i \sum_j \sum_k U_{i,j,k} \times EF_{i,j} \times 10^3 \quad (4)$$

where E_{NH_3} is the NH_3 emissions from rice fields in China (Gg NH_3 yr^{-1}); i is the synthetic fertilizer category; j is the month; k is the county; and EF is the emission factor.

In order to obtain a specific emission factor for a given scenario, six important variables (fertilization method and rate, soil pH, water input, wind speed and ambient temperature) were used to adjust the reference emissions factor (EF_{φ}), which was derived from in situ measurement data and literature-reported data in China (Freney and Denmead, 1992; Webb et al., 2006). The equation is expressed as follows:

$$EF_{i,j} = EF_{\phi i} \times IF_{method} \times IF_{soilpH} \times IF_{waterinput} \times IF_{temperature} \times IF_{wind} \times IF_{rate} \quad (5)$$

where $EF_{i,j}$ is the emissions factor for synthetic fertilizer of type i for a specific condition; $EF_{\varphi i}$ is the in situ measurement and literature-