Environmental Pollution 235 (2018) 482–488

Contents lists available at ScienceDirect

Environmental Pollution

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

Ammonia emissions from paddy fields are underestimated in China☆

Hongyuan Wang [a, 1], Dan Zhang [a, 1], Yitao Zhang [a], Limei Zhai [a], Bin Yin [b], Feng Zhou [c], Yucong Geng [a], Junting Pan [a], Jiafa Luo [d], Baojing Gu [e, *], Hongbin Liu [a, **]

[a] Key Laboratory of Nonpoint Source Pollution Control, Ministry of Agriculture, Institute of Agricultural Resources and Regional Planning, Chinese Academy of Agricultural Sciences, Beijing 100081, China
[b] State Key Laboratory of Soil and Sustainable Agriculture, Institute of Soil Science, Chinese Academy of Sciences, Nanjing 210008, China
[c] Sino-France Institute of Earth Systems Science, Institute of Integrated Watershed Management, Laboratory for Earth Surface Processes, College of Urban and Environmental Sciences, Peking University, Beijing 100871, China
[d] AgResearch, Ruakura Research Centre, 10 Bisley Road, Hamilton 3214, New Zealand
[e] Department of Land Management, Zhejiang University, Hangzhou 310058, China

A R T I C L E I N F O

Article history:
Received 7 September 2017
Received in revised form
11 December 2017
Accepted 25 December 2017

Keywords:
Nitrogen rate
Ammonia loss proportion
Non-linear response
Air pollution
Climate factor

A B S T R A C T

Excessive nitrogen (N) fertilizers are often used in China, and a large proportion of the N can be lost as ammonia (NH_3). However, quantifying the NH_3 emission from paddy fields is always affected by large uncertainties due to different measuring methods and other factors such as climate. In this study, using a standardized method, we measured the NH_3 emissions in three typical annual rice cropping systems: single rice, double rice and rotation with other crops. The measurements were conducted for 2 years with a total of 3131 observations across China. Results showed that NH_3 emissions accounted for 17.7% (14.4–21.0%) of the N applied under current farm practice, which was 33.1% (10.6–52.6%) higher than previous estimates. Nitrogen application rate was the dominant factor influencing NH_3 emission rate, which exponentially increased with the N fertilizer rate ($p < .001$). Total NH_3 emissions from paddy fields were estimated at 1.7 Tg N yr^{-1} in 2013 in China, several times the amount of N lost through leaching or runoff. This suggests that mitigation measures for non-point source pollution from cropland should take into account not only the N lost to water, but also to air, thereby improving air quality.

1. Introduction

China produced 208.2 million tons of rice in 2014, accounting for 28.1% of global production (FAO, 2017). However, excessive amounts of nitrogen (N) fertilizers are being used in paddy fields, with an average N fertilizer rate of over 300 kg N ha^{-1} yr^{-1} (Deng et al., 2011), and over 50% of this input N is lost to the environment through multiple pathways. Ammonia volatilization is one of the dominant pathways of N loss in paddy fields (Yan et al., 2011; Soares et al., 2012; Xu et al., 2012). NH_3 volatilization increases farmers' production costs and causes environmental degradation (Xu et al., 2015). NH_3 is a major atmospheric pollutant that plays an important role in the formation of secondary inorganic aerosols, leading to poor air quality and adverse impacts on human health (Behera et al., 2013; Gu et al., 2014). The emitted NH_3 can also return to land and surface water through deposition, resulting in soil and water acidification, eutrophication and biodiversity loss (Hellsten et al., 2008; Guo et al., 2010).

NH_3 emission is affected by a number of factors, such as fertilizer application rate (Dattamudi et al., 2016; Huang et al., 2016; Jiang et al., 2017), climate conditions (e.g. temperature, wind speed) (Fan et al., 2011; Louro et al., 2013), and soil properties (e.g. pH, soil type) (Fan et al., 2011; Zhang et al., 2013; Webb et al., 2014). Inventories of NH_3 emissions from paddy soils in China have been conducted (Zhang et al., 2011; Chen et al., 2014). However, large uncertainties exist in these inventories due to the variations in emission factors derived from different measuring methods (Hayashi et al., 2011; Zhao et al., 2012). For example, Chen et al. (2014) calculated that NH_3 lost from paddy fields accounted for

☆ This paper has been recommended for acceptance by Dr. Hageman Kimberly Jill.

* Corresponding author. Department of Land Management, Zhejiang University, Zijingang Campus, 866 Yuhangtang Road, Hangzhou 310058, China.
** Corresponding author. Key Laboratory of Nonpoint Source Pollution Control, Ministry of Agriculture, Institute of Agricultural Resources and Regional Planning, Chinese Academy of Agricultural Sciences, No. 12 Zhongguancun South Street, Haidian district, Beijing 100081, China.
E-mail addresses: bjgu@zju.edu.cn (B. Gu), liuhongbin@caas.cn (H. Liu).
[1] These authors contributed equally to this work.

https://doi.org/10.1016/j.envpol.2017.12.103
0269-7491/

approximately 16% of N input by compiling over 265 measurements across China, while the results derived from Zhou et al. (2016), with an even bigger dataset, showed that the average NH_3 loss accounted for only 12% of N input for paddy soils. However, the methods of measuring these NH_3 emissions were quite different, and whether the results of these measurements could be comparable for a meaningful inventory is unclear. Therefore, refining the understanding of these variations is crucial for reducing the uncertainty in NH_3 emissions from paddy fields.

We established a Nationwide NH_3 Emission Monitoring Network (NNEMN) for paddy fields in 2012–2013, and measured NH_3 emissions for 2 years continuously, by using a standardized measuring method. The network included 12 field sites, which covered three types of rice cultivation (single, double and rotation with wheat or potato or a vegetable crop) located in the main rice planting regions across China (Northeast China, Southeast China and Yangtze River Basin). The objectives of this study were to (i) quantify NH_3 loss and its uncertainties from paddy fields in different types of rice cultivation, (ii) explore the influence factors affecting NH_3 emission rate, including N application rate, soil properties and climate factors and (iii) estimate total NH_3 volatilization from paddy fields in China.

2. Materials and methods

2.1. Study sites

The distribution of the 12 field sites used in this study is shown in Fig. 1. There are great regional differences in NH_3 emissions resulting from the differences in environmental factors and management practices. In order to determine the spatial variation in NH_3 emissions from paddy soils, we divided the 12 sites into three types according to the environmental conditions, cropping system and cultivation history: (i) Single-cropping rice; (ii) Double-cropping rice and (iii) Rice-upland crop (wheat/potato/vegetable rotation). Details for the rice types are given in supplementary information. The number of study sites in each region was mainly determined by the total planting area of rice and heterogeneity of the environmental factors and management practices, and 1, 3 and 8 field sites were set up for single rice, double rice and rice-upland rotation system, respectively. The sowing areas of the above systems in China were 4.6, 10.9 and 11.1 million ha, respectively (NBS, 2014). Compared with the latter two systems, the single rice system is commonly planted over smaller areas with little variation in climatic conditions and soil type. Therefore, only one typical field site was chosen for the study. In view of the large variations in climatic conditions in the rice-upland growing regions and different crops used for rotation, 8 field sites were chosen for the study.

2.2. Experiment design

Basic information about climate, soil properties, and fertilization for each site is shown in Tables S1–3. The experiments were performed during 2012–2013, and urea, superphosphate, and potassium sulfate were used. Prior to rice transplantation, soil was irrigated and plowed, followed by basal fertilization. Based on local farmers' practices, some sites applied tillering topdressing and anthesis topdressing fertilization. Six fertilization rates were used: zero N-fertilizer (CK), local farmers' practice (FT), and another four treatments with 50, 66.7, 83.3, and 133.3% of FT. Although each site had 6 treatments, the local farmers' practice treatment (FT) included a range of rates due to the variations in local practice in the various regions. Consequently, the treatments with 50, 66.7, 83.3, and 133.3% of the local FT rates also varied (Table S2). At each experiment site, the treatment plots (20–40 m^2 in area) were arranged by a randomized complete block experimental design with three replicates. There were bunds between each plot.

2.3. Field measurements

A standardized dynamic chamber method, with a continuous

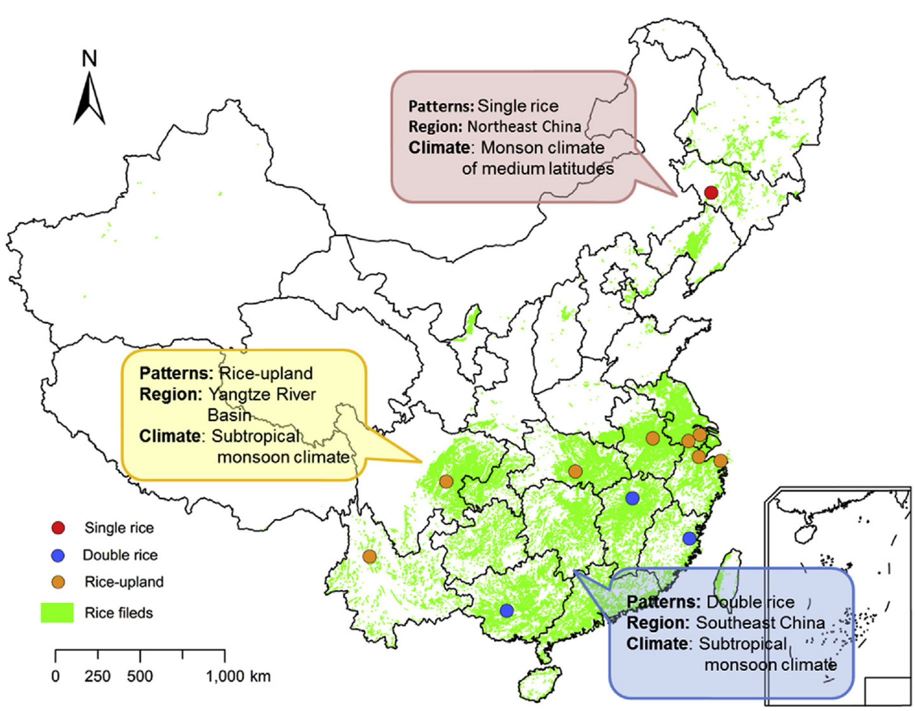

Fig. 1. Geographical distribution of the 12 monitoring sites in China.

airflow enclosure (CAE), was utilized to measure NH_3 volatilization from the paddy fields in all the treatment plots at the 12 sites. The NH_3 volatilization collection device (Fig. S1) consisted of a chamber, a vent pipe, a chemical trap bottle and a vacuum pump linked by plastic pipes to form a confined space (Cao et al., 2013). A chemical trap bottle filled with 60 mL of 20 g L^{-1} boric acid (H_3BO_3) connected with another hole was used to collect NH_3 gas from paddy field. The air exchange rate was set to 15–20 headspace volumes min^{-1}. The NH_3 volatilization rate was measured twice daily: morning (9:00–11:00) and afternoon (16:00–18:00) (Hou et al., 2007; Zhao et al., 2015). The average hourly NH_3 flux observed during the 4 h was directly converted into the average hourly flux per day (Zhao et al., 2015). The NH_3 trap solutions were brought to the laboratory and the ammonium (NH_4^+) nitrogen content of the traps was titrated with 0.005 M sulfuric acid (H_2SO_4). The NH_3 volatilization was continually monitored for 10–15 days following each fertilization event until volatilization became negligible (Chen et al., 2015a). The daily NH_3 volatilization rate was calculated from the average of the rates measured each day. The NH_3 volatilization flux was calculated as following equation:

$$F = \frac{2 \times \mathrm{C} \times \mathrm{V} \times 14 \times 10^{-2} \times 24}{t \times \mathrm{S}} \qquad (1)$$

where F is the flux of NH_3 volatilization (kg N ha^{-1} d^{-1}), C is the concentration of H_2SO_4 (M), V is the consumption volume of H_2SO_4 (mL), t is the duration of collection (h), and S is the chamber area (m^2). The cumulative NH_3 losses were the sum of NH_3 volatilization fluxes on sampling days.

The NH_3 loss proportion was calculated as following equation:

$$E_N = \frac{C_{NH_3} - C_0}{N} \times 100\% \qquad (2)$$

where E_N is the NH_3 loss proportion (%), C_{NH_3} is the NH_3 volatilization at every non-zero N application rate (kg N ha^{-1}), C_0 is the NH_3 volatilization at the zero N application rate (kg N ha^{-1}), and N is N fertilizer application rate (kg N ha^{-1}).

The total amount of NH_3 volatilization for rice fields across China was calculated as following equation:

$$T_N = \sum_{n=1}^{n} \left(C_{F_{iNH_3}} \times S_i \right) \qquad (3)$$

where T_N is the total amount of NH_3 volatilization (kg N), i represents an individual province, $C_{F_{iNH_3}}$ is the NH_3 volatilization of ith province at farm's N practice (kg N ha^{-1}), S_i is the growing rice area of the ith province (ha). If there was no monitor site in a certain province, the total NH_3 volatilization in this province was calculated using the monitored data of neighboring provinces.

The air temperature was determined using a mercury thermometer or a potentiometer when the NH_3 volatilization was measured. The precipitation and wind speed were obtained from the China Meteorological Forcing Dataset (CMFD).

2.4. Sample analysis

Soil samples at a depth of 0–20 cm were collected at the beginning of the trial using a 5-cm internal diameter auger. For each site, five replicate soil samples were randomly taken and mixed together to produce a composite sample. Average soil texture was determined for all fields using a mixed bulk sample from all samples across a field. Bulk density of soil samples was measured after being dried in an oven at 105 °C for 24 h. The soil pH (1:5 soil/water) and floodwater pH were determined using a potentiometer. The soil organic matter (SOM) was analyzed with dichromate oxidation, while total N (TN) was determined using the Kjeldahl digestion method (KDY-9830, Beijing) (Peng et al., 2011).

2.5. Statistical analysis

Spearman's correlation coefficients and multiple linear regression were used to test for significant correlations between NH_3 volatilization rates and influencing factors using SPSS 19.0 statistical software (SPSS Inc., Chicago, IL). A p values less than 0.05 was considered statistically significant.

3. Results

3.1. Temporal and spatial variations

The temporal variations indicated that NH_3 volatilization rate peaked 1–3 days after fertilization, followed by a rapid decline (Fig. 2a). For the three types of rice cultivation, the trends of NH_3 volatilization exhibited similar patterns, but the peak values for the single-rice were far lower than those for the double-rice and rice-upland rotation (Fig. S2), which had a lower N application rate. The average peak values for the single, double and rice-upland after basal fertilizer application were 4.2, 11.5 and 8.1 kg N ha^{-1} d^{-1}, respectively, for which the N application rates were 108, 149.2 and 117.9 kg N ha^{-1}. NH_3 volatilization rates dropped to a low level (<2 kg N ha^{-1} d^{-1}) from the peak value after 7–10 days.

The NH_3 volatilization for single rice, double cropping rice and rice-upland were 40.1, 54.9, and 57.9 kg N ha^{-1}, respectively, for typical farmers' practice (average rate of 269.0 kg N ha^{-1}). The average cumulative NH_3 volatilization was 56.0 kg N ha^{-1}, which represented 17.7% (14.4–21.0%) of the total N input. The average NH_3 volatilization in 2013 was 57.2 kg N ha^{-1}, slightly higher than that found in 2012 (55.1 kg N ha^{-1}) with no significant difference ($p > .05$). The proportion of the applied N lost as NH_3 from the double cropping rice was the highest, up to 19.1% (Fig. 2b). In this system, the NH_3 loss from the late rice was 1.4 times that of the early rice (Fig. S3). The NH_3 emission per unit of grain N production (NPGN) for the double cropping rice was the highest, followed by rice-upland and single rice (Fig. 2c).

3.2. Influencing factors

The NH_3 volatilization was positively correlated with N input rate ($p < .01$, $R^2 = 0.52$) increasing with N application rate following a quadratic or exponent function (Fig. 3a and Figs. S4–5). This suggests that the NH_3 loss proportion was larger under higher N input compared to that under the lower N input (Fig. S6). It seems that soil properties (soil organic matter, soil total N, and soil bulk density), climate conditions (air temperature, precipitation, and wind speed) and water pH did not have a significant impact on NH_3 volatilization ($p > .05$, Fig. 3b–h).

Interestingly, multiple linear regression indicated that soil organic matter had a significant negative effect while wind speed had a significant positive effect on total NH_3 volatilization ($p < .05$, Table 1 and Table S4). However, the different rice cropping systems responded differently to these influencing factors. For example, soil properties and temperature were more important for the double rice and rice-upland rotation cropping systems compared to the overall regression between the emissions and all factors combined.

3.3. Total amount of NH_3 volatilization

We estimated that the total amount of NH_3 volatilization from paddy soils in China, was 1.7 Tg N yr^{-1} in 2013, using the typical

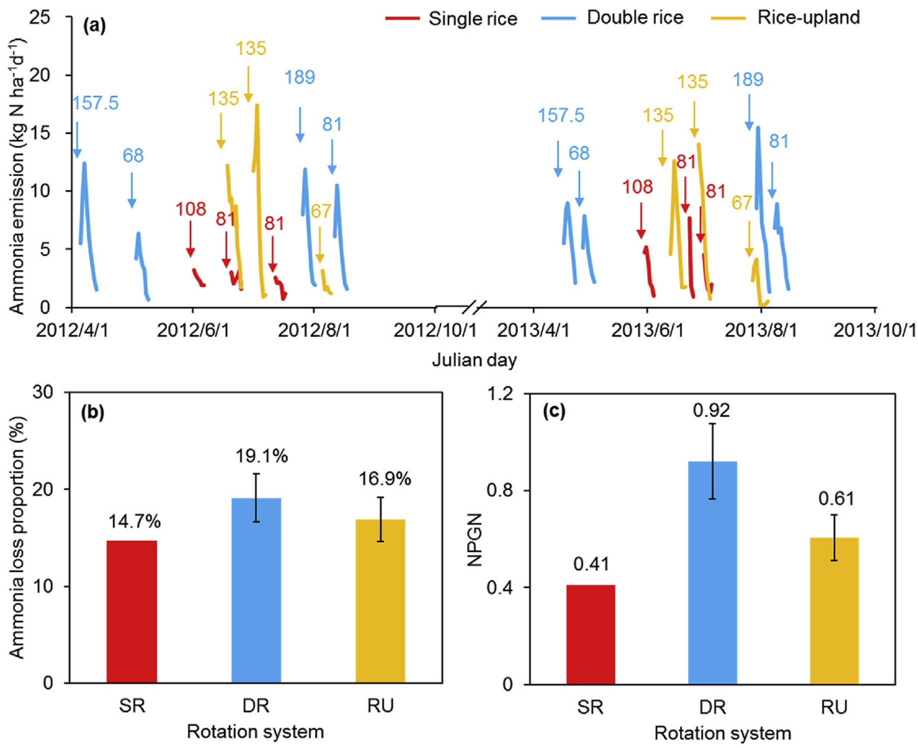

Fig. 2. NH_3 loss from paddy soil. (a) The temporal variation of NH_3 volatilization in three typical monitoring sites of single rice, double rice and upland rice crop rotation with wheat: Jilin (43.9°N, 124.3°E), Fujian (26.2°N, 119.1°E) and Anhui (31.7°N, 117.7°E), respectively; (b) the total NH_3 loss proportion in the different rice cropping systems; (c) the NH_3 emission per unit of grain nitrogen production. The arrows in (a) represent fertilizer application, and the number above represents the N application rate (kg N ha^{-1}). SR, DR and RU in (b) and (c) represent single rice, double rice, and rice-upland, respectively, and NPGN in (c) represents the NH_3 emission per unit of grain N production. Data are shown as mean ± SEM (standard error of mean).

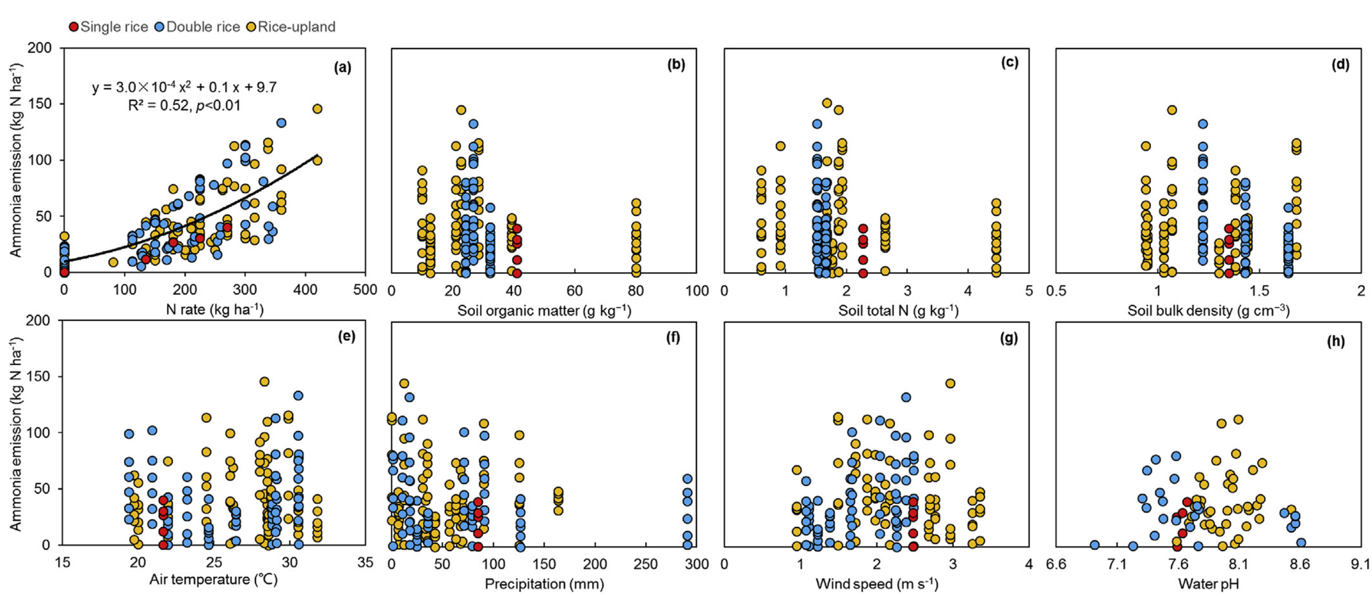

Fig. 3. Relationship between NH_3 volatilization and associated influencing factors.

farmer's N application rate. The spatial pattern of NH_3 emission was illustrated in Fig. 4. High emission rates were found in Hunan, Anhui and Jiangxi provinces, all of which have large growing areas, accounting for 16.5%, 14% and 13.3% of the total NH_3 emission in China, respectively. For Liaoning, Jilin, Guizhou and Hainan provinces, the NH_3 emissions were all less than 30 Gg N yr^{-1}.

4. Discussion

4.1. Non-linear response of NH_3 emissions to N rate

N fertilizer rate directly impacts NH_3 volatilization, and an elevated N concentration in soil accelerates the loss of NH_3 to the

Table 1
Model coefficients of multiple linear regression on NH_3 emission and associated influencing factors.

Influencing factors	Variable	Total	Single rice	Double rice	Rice-upland
N fertilizer[2 #]	x_1^2	**$2.4\times10^{-4*}$**	3.0×10^{-4}	2.3×10^{-4}	**$2.7\times10^{-4*}$**
N fertilizer	x_1	**0.12****	0.07	**0.13****	**0.10***
Soil organic matter	x_2	**−0.3****	–	0.6	**−0.4***
Soil bulk density	x_3	−2.8	–	**−103.9****	**49.3****
Temperature	x_4	−1.2	–	**1.0***	**−2.4***
Precipitation	x_5	−0.06	–	-5.0×10^{-3}	−0.08
Wind speed	x_6	**8.1****	–	1.5	−6.0
Constant	a	**38.8***	0.06	111.5	41.8

Note: Values in bold indicate statistically significant, $^*p < .05$; $^{**}p < .01$. [#] The values of N fertilizer^2 coefficients were small, because the square of N fertilizer rate was usually very large. A serious multicollinearity problem existed between TN and other variables (such as SOM, Fig. S7), therefore, Soil TN was omitted from the multiple linear regression.

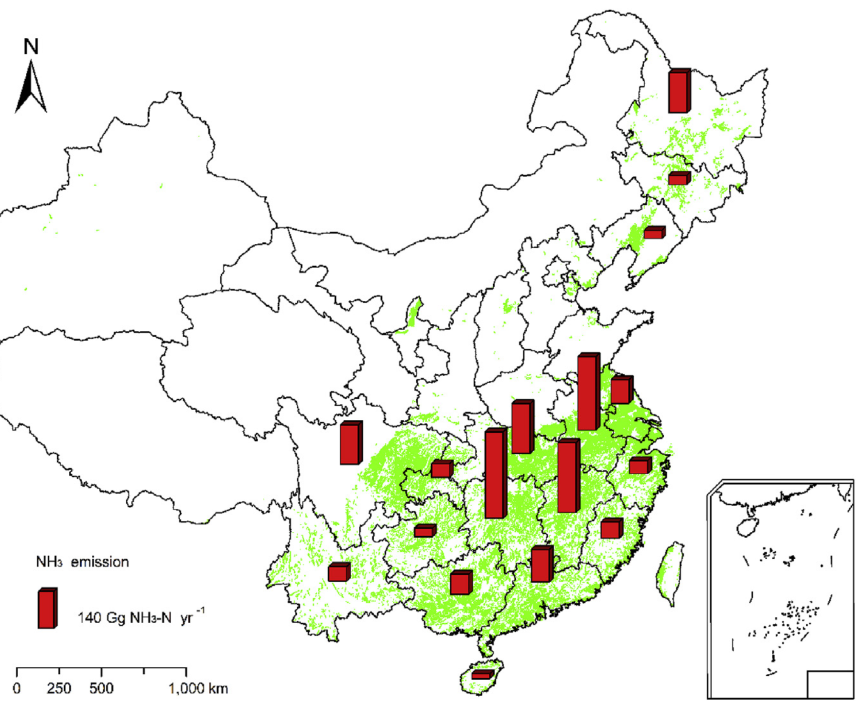

Fig. 4. Total annual amounts of NH_3 volatilization in the rice-growing regions in 2013.

atmosphere (Ju et al., 2009; Jiang et al., 2017). Most previous estimates have found that the response of NH_3 emission to N application rates is linear (Cui et al., 2013, 2014; Chen et al., 2014). Recently, several studies using data synthesis analysis have suggested that NH_3 emission response to N addition could be non-linear (Zhou et al., 2016; Jiang et al., 2017). By controlling for the differences resulting from different methodologies and measurement timing, our study suggests that NH_3 volatilization increases with N application rate following a quadratic function, rather than a linear relationship. There could be several reasons for the observed non-linear response. Firstly, as a dominant factor affecting NH_3 emissions in paddy fields, NH_4^+ concentration in the surface water of paddy fields increases with N inputs (Sommer et al., 2004; Rochette et al., 2013; Shang et al., 2014), and the instantaneous rate of NH_3 emission is exponentially correlated with NH_4^+ concentration (Chen et al., 2015a). Secondly, crop N use efficiency decreases dramatically with N application rate, particularly when N input exceeds crop needs (Peng et al., 2006). Therefore, more NH_4^+ will be produced once excess N fertilizer is used. Thirdly, high N input may inhibit the growth and activity of nitrifying microbes because of the increase in salinity with NH_4^+, thereby reducing NH_4^+ consumption (Shen et al., 2010; Norman and Barrett, 2014). Thus, NH_3 emissions increase exponentially with N application rates, especially when the N rate reaches a high level (>300 kg N ha^{-1}).

Non-linear regression indicated that the amount NH_3 emitted as a proportion of N applied was not constant, but increased with N application rate. The estimates of proportion for a linear model was close to that of the quadratic model at N application rates between 160 and 180 kg N ha^{-1} (Fig. S6). This implies that there would be a higher proportion of NH_3 emitted in regions with high N inputs while there would be a lower proportion of NH_3 emitted in regions with low N inputs.

4.2. Underestimate of NH_3 volatilization

Chemical fertilizer is the second largest global emission source of NH_3 (Xu et al., 2015). In this study, we found that 17.7% of N fertilizer input was lost though NH_3 emission in paddy fields, which was 33.1% (10.6–52.6%) higher than previous studies (Fig. 5). Recently, a top-down estimate also showed that the NH_3 loss proportion of mineral fertilizer was 16.2–18.4%, based on a mass balance approach, and validated by N deposition monitoring and

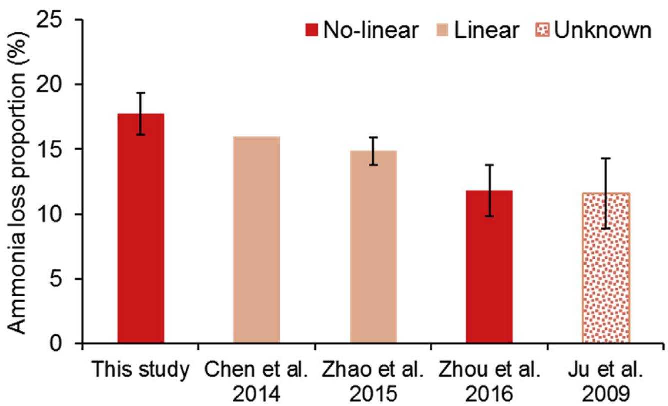

Fig. 5. Comparisons with NH_3 estimates from paddy soil in other studies.

satellite observations, which implies that NH_3 emissions may be substantially underestimated in China (Zhang et al., 2017). The underestimation of NH_3 emissions could be an integrated result of the following reasons: (i) variation in methodology; (ii) lower N application rate used by farmers in the past; and (iii) non-linear response of NH_3 emissions to N rate.

In this study, we used a continuous airflow enclosure (CAE) method to estimate NH_3 emission, which is widely used in field scale studies (Cao et al., 2013; Chen et al., 2015b; Zhao et al., 2015). Besides the CAE method, another commonly used method for the determination of NH_3 emission is a high-resolution micrometeorological (HRM) method. Compared to the open HRM method, the closed CAE method may result in uncertainty due to the fact that the chamber can't accurately simulate the natural wind conditions above the soil surface (Li, 2013). However, it is still the most suitable method for comparing multiple treatments at field scale (Jantalia et al., 2012; Yang et al., 2013). The deviation of CAE from the HRM method has been found to be less than 5% (Pacholski et al., 2006).

Previous national scale studies have normally collected data on NH_3 emission factors that were determined using different measuring methods at different times (Chen et al., 2014; Pan et al., 2016; Zhou et al., 2016). Different methods (e.g., dynamic chamber, closed chamber and micrometeorological method) may result in systematic variations in the measured NH_3 emissions, which cannot be accounted for in an inventory study. Additionally, many experiments were conducted decades ago (e.g. in 1990s), when the typical N application rate used by farmers was much lower and soil N level was also lower. Based on our non-linear regression, a low N application rate would suggest a low NH_3 emission proportion. With an increase in N fertilizer application rate, the NH_3 emission proportions are likely to be higher. Zhou et al. (2016) suggested that NH_3 emission response to N addition could be non-linear using data synthesis analysis of historical measurements. Chen et al. (2014) calculated that NH_3 lost from paddy fields accounted for approximately 16% of N input (209 kg N ha^{-1}), and non-linear response was the main reasons causing underestimation.

A “zero increase of fertilizer use” policy has been introduced in China, however, the total amount of N fertilizer use will be still be increasing until 2020. Therefore, the total NH_3 emission from paddy soil in China previously calculated is likely to be an underestimate. China consumed 31.1 Mt of synthetic N fertilizers in 2014, accounting for 28.5% of global total (FAO, 2017). The underestimate of NH_3 emission factors would result in a large underestimate of the total NH_3 emissions in China. Thus, refining the estimate of NH_3 emission factors, as found in this study, is vital for the accuracy of the national scale inventory of total NH_3 emissions, which is important for developing strategies to improve the air quality in China.

4.3. Policy implications

Approximately 20% of global NH_3 emissions originate from China (Klimont et al., 2001; Yamaji et al., 2004), in particular from intensive N fertilizer application to croplands (Yan et al., 2003; Wang et al., 2005). Compared to reactive N (Nr) loss through other pathways, such as N leaching and runoff (Gao et al., 2016; Hou et al., 2016), the amount of Nr loss through NH_3 volatilization is much larger and plays a more important role in the environment. Previous policy regulations mainly focus on the reduction of Nr loss from agricultural non-point-source pollution to water bodies to mitigate eutrophication. For example, to balance food security and environmental protection, several policies have been implemented, including “Water Pollution Prevention and Control Action Plan” (WPPCAP), “Soil Pollution Prevention and Control Action Plan” (SPPCAP) and “Opinions on Taking Measures to Prevent and Control Agricultural Nonpoint Source Pollution”. In particular, the first document issued annually by the Central Committee of the Communist Party of China (The No.1 Document) made special regulations for strengthening agricultural ecological governance, with an emphasis on increasing efforts to control agricultural nonpoint source pollution to water bodies. Measures include ecological ditches, runoff storage facilities, treatment of farmland drainage and surface runoff and encouragement of farmers to increase organic fertilizer and reduce chemical fertilizer use. To reduce chemical fertilizer use, the Ministry of Agriculture implemented the “Zero Increase of Fertilizer Use Towards 2020” policy. These policies and actions all focus on prevention and control of water and soil pollution and less on air pollution.

Air pollution has become a serious problem in China, with fine particulate matter ($PM_{2.5}$) increasingly important (Zhang and Cao, 2015; Maji et al., 2017). The $PM_{2.5}$ concentrations are strongly affected by NH_3 emissions during severe haze episodes, and improved agricultural N management is critical for reducing NH_3 emissions to mitigate production of $PM_{2.5}$ (Wu et al., 2016). However, the “Air Pollution Prevention and Control Action Plan” (APPCAP) only focused on the reduction of air pollutants from industries and transportation, and has not considered the air pollutants from agricultural sources. Currently, there is no regulation or incentive program to tackle the challenge of NH_3 volatilization from agriculture (Velthof et al., 2012). Therefore, the mitigation policies and measures for non-point-source pollution from cropland should consider not only the N lost to water, but also to the air, which is severely polluted in China.

Acknowledgements

This research was partially supported by the National Key Research and Development Program (2016YFD0800500, 2016YFC0207906), Special Fund for Agro-scientific Research in the Public Interest from the Ministry of Agriculture, China (201303089), Newton Fund (Grant Ref: BB/N013484/1) and the National Natural Science Foundation of China (41773068, 41671464).

Appendix A. Supplementary data

Supplementary data related to this article can be found at https://doi.org/10.1016/j.envpol.2017.12.103.

References

Behera, S.N., Sharma, M., Aneja, V.P., Balasubramanian, R., 2013. Ammonia in the

atmosphere: a review on emission sources, atmospheric chemistry and deposition on terrestrial bodies. Environ. Sci. Pollut. R 20, 8092–8131.
Cao, Y., Tian, Y., Yin, B., Zhu, Z., 2013. Assessment of ammonia volatilization from paddy fields under crop management practices aimed to increase grain yield and N efficiency. Field Crop. Res. 147, 23–31.
Chen, A., Lei, B., Hu, W., Lu, Y., Mao, Y., Duan, Z., Shi, Z., 2015a. Characteristics of ammonia volatilization on rice grown under different nitrogen application rates and its quantitative predictions in Erhai Lake Watershed, China. Nutr. Cycl. Agroecosys 101, 139–152.
Chen, G., Chen, Y., Zhao, G., Cheng, W., Guo, S., Zhang, H., Shi, W., 2015b. Do high nitrogen use efficiency rice cultivars reduce nitrogen losses from paddy fields? Agric. Ecosyst. Environ. 209, 26–33.
Chen, X., Cui, Z., Fan, M., Vitousek, P., Zhao, M., Ma, W., Wang, Z., Zhang, W., Yan, X., Yang, J., Deng, X., Gao, Q., Zhang, Q., Guo, S., Ren, J., Li, S., Ye, Y., Wang, Z., Huang, J., Tang, Q., Sun, Y., Peng, X., Zhang, J., He, M., Zhu, Y., Xue, J., Wang, G., Wu, L., An, N., Wu, L., Ma, L., Zhang, W., Zhang, F., 2014. Producing more grain with lower environmental costs. Nature 514, 486–489.
Cui, Z., Wang, G., Yue, S., Wu, L., Zhang, W., Zhang, F., Chen, X., 2014. Closing the N-use efficiency gap to achieve food and environmental security. Environ. Sci. Technol. 48, 5780–5787.
Cui, Z., Yue, S., Wang, G., Meng, Q., Wu, L., Yang, Z., Zhang, Q., Li, S., Zhang, F., Chen, X., 2013. Closing the yield gap could reduce projected greenhouse gas emissions: a case study of maize production in China. Global Change Biol. 19, 2467–2477.
Dattamudi, S., Wang, J.J., Dodla, S.K., Arceneaux, A., Viator, H.P., 2016. Effect of nitrogen fertilization and residue management practices on ammonia emissions from subtropical sugarcane production. Atmos. Environ. 139, 122–130.
Deng, J., Zhou, Z., Zhu, B., Zheng, X., Li, C., Wang, X., Jian, Z., 2011. Modeling nitrogen loading in a small watershed in southwest China using a DNDC model with hydrological enhancements. Biogeosciences 8, 2999–3009.
Fan, X.H., Li, Y.C., Alva, A.K., 2011. Effects of temperature and soil type on ammonia volatilization from slow-release nitrogen fertilizers. Commun. Soil Sci. Plan 42, 1111–1122.
FAO (Food and Agriculture Organization of the United Nations), 2017. FAOSTAT: FAO Statistical Databases. http://faostat.fao.org/default.aspx. (Accessed 30 June 2017).
Gao, S., Xu, P., Zhou, F., Yang, H., Zheng, C., Cao, W., Tao, S., Piao, S., Zhao, Y., Ji, X., Shang, Z., Chen, M., 2016. Quantifying nitrogen leaching response to fertilizer additions in China's cropland. Environ. Pollut. 211, 241–251.
Gu, B., Sutton, M.A., Chang, S.X., Ge, Y., Chang, J., 2014. Agricultural ammonia emissions contribute to China's urban air pollution. Front. Ecol. Environ. 12, 265–266.
Guo, J.H., Liu, X.J., Zhang, Y., Shen, J.L., Han, W.X., Zhang, W.F., Christie, P., Goulding, K.W.T., Vitousek, P.M., Zhang, F.S., 2010. Significant acidification in major Chinese croplands. Science 327, 1008–1010.
Hayashi, K., Koga, N., Fueki, N., 2011. Limited ammonia volatilization loss from upland fields of Andosols following fertilizer applications. Agric. Ecosyst. Environ. 140, 534–538.
Hellsten, S., Dragosits, U., Place, C.J., Vieno, M., Dore, A.J., Misselbrook, T.H., Tang, Y.S., Sutton, M.A., 2008. Modelling the spatial distribution of ammonia emissions in the UK. Environ. Pollut. 154, 370–379.
Hou, H., Zhou, S., Hosomi, M., Toyota, K., Yosimura, K., Mutou, Y., Nisimura, T., Takayanagi, M., Motobayashi, T., 2007. Ammonia emissions from anaerobically-digested slurry and chemical fertilizer applied to flooded forage rice. Water Air Soil Pollut. 183, 37–48.
Hou, X., Zhou, F., Leip, A., Fu, B., Yang, H., Chen, Y., Gao, S., Shang, Z., Ma, L., 2016. Spatial patterns of nitrogen runoff from Chinese paddy fields. Agric. Ecosyst. Environ. 231, 246–254.
Huang, S., Lv, W., Bloszies, S., Shi, Q., Pan, X., Zeng, Y., 2016. Effects of fertilizer management practices on yield-scaled ammonia emissions from croplands in China: a meta-analysis. Field Crop. Res. 192, 118–125.
Jantalia, C.P., Halvorson, A.D., Follett, R.F., Rodrigues Alves, B.J., Polidoro, J.C., Urquiaga, S., 2012. Nitrogen source effects on ammonia volatilization as measured with semi-static chambers. Agron. J. 104, 1595–1603.
Jiang, Y., Deng, A., Bloszies, S., Huang, S., Zhang, W., 2017. Nonlinear response of soil ammonia emissions to fertilizer nitrogen. Biol. Fertil. Soils 53, 269–274.
Ju, X., Xing, G., Chen, X., Zhang, S., Zhang, L., Liu, X., Cui, Z., Yin, B., Christie, P., Zhu, Z., Zhang, F., 2009. Reducing environmental risk by improving N management in intensive Chinese agricultural systems. P. Natl. Acad. Sci. USA 106, 3041–3046.
Klimont, Z., Cofala, J., Schopp, W., Amann, M., Streets, D.G., Ichikawa, Y., Fujita, S., 2001. Projections of SO2, NOx, NH3 and VOC emissions in East Asia up to 2030. Water Air Soil Pollut. 130, 193–198.
Li, D., 2013. Emissions of NO and NH3 from a typical vegetable-land soil after the application of chemical N fertilizers in the Pearl River Delta. PLoS One 8, 1–8.
Louro, A., Sawamoto, T., Chadwick, D., Pezzolla, D., Bol, R., Baez, D., Cardenas, L., 2013. Effect of slurry and ammonium nitrate application on greenhouse gas fluxes of a grassland soil under atypical South West England weather conditions. Agric. Ecosyst. Environ. 181, 1–11.
Maji, K.J., Arora, M., Dikshit, A.K., 2017. Burden of disease attributed to ambient PM2.5 and PM10 exposure in 190 cities in China. Environ. Sci. Pollut. R 24, 11559–11572.
NBS (National Bureau of Statistics), 2014. China Agricultural Statistical Yearbook. China Statistics Press, Beijing.
Norman, J.S., Barrett, J.E., 2014. Substrate and nutrient limitation of ammonia-oxidizing bacteria and archaea in temperate forest soil. Soil Biol. Biochem. 69, 141–146.
Pacholski, A., Cai, G., Nieder, R., Richter, J., Fan, X., Zhu, Z., Roelcke, M., 2006. Calibration of a simple method for determining ammonia volatilization in the field - comparative measurements in Henan Province, China. Nutr. Cycl. Agroecosys 74, 259–273.
Pan, B., Lam, S.K., Mosier, A., Luo, Y., Chen, D., 2016. Ammonia volatilization from synthetic fertilizers and its mitigation strategies: a global synthesis. Agric. Ecosyst. Environ. 232, 283–289.
Peng, S., Yang, S., Xu, J., Luo, Y., Hou, H., 2011. Nitrogen and phosphorus leaching losses from paddy fields with different water and nitrogen managements. Paddy Water Environ. 9, 333–342.
Peng, S.B., Buresh, R.J., Huang, J.L., Yang, J.C., Zou, Y.B., Zhong, X.H., Wang, G.H., Zhang, F.S., 2006. Strategies for overcoming low agronomic nitrogen use efficiency in irrigated rice systems in China. Field Crop. Res. 96, 37–47.
Rochette, P., Angers, D.A., Chantigny, M.H., Gasser, M., MacDonald, J.D., Pelster, D.E., Bertrand, N., 2013. NH3 volatilization, soil NH4+ concentration and soil pH following subsurface banding of urea at increasing rates. Can. J. Soil Sci. 93, 261–268.
Shang, Q., Gao, C., Yang, X., Wu, P., Ling, N., Shen, Q., Guo, S., 2014. Ammonia volatilization in Chinese double rice-cropping systems: a 3-year field measurement in long-term fertilizer experiments. Biol. Fertil. Soils 50, 715–725.
Shen, W., Lin, X., Shi, W., Min, J., Gao, N., Zhang, H., Yin, R., He, X., 2010. Higher rates of nitrogen fertilization decrease soil enzyme activities, microbial functional diversity and nitrification capacity in a Chinese polytunnel greenhouse vegetable land. Plant Soil 337, 137–150.
Soares, J.R., Cantarella, H., de Campos Menegale, M.L., 2012. Ammonia volatilization losses from surface-applied urea with urease and nitrification inhibitors. Soil Biol. Biochem. 52, 82–89.
Sommer, S.G., Schjoerring, J.K., Denmead, O.T., 2004. Ammonia emission from mineral fertilizers and fertilized crops. Adv. Agron. 82, 557–622.
Velthof, G.L., van Bruggen, C., Groenestein, C.M., de Haan, B.J., Hoogeveen, M.W., Huijsmans, J.F.M., 2012. A model for inventory of ammonia emissions from agriculture in The Netherlands. Atmos. Environ. 46, 248–255.
Wang, X.P., Mauzerall, D.L., Hu, Y.T., Russell, A.G., Larson, E.D., Woo, J.H., Streets, D.G., Guenther, A., 2005. A high-resolution emission inventory for eastern China in 2000 and three scenarios for 2020. Atmos. Environ. 39, 5917–5933.
Webb, J., Thorman, R.E., Fernanda-Aller, M., Jackson, D.R., 2014. Emission factors for ammonia and nitrous oxide emissions following immediate manure incorporation on two contrasting soil types. Atmos. Environ. 82, 280–287.
Wu, Y., Gu, B., Erisman, J.W., Reis, S., Fang, Y., Lu, X., Zhang, X., 2016. PM2.5 pollution is substantially affected by ammonia emissions in China. Environ. Pollut. 218, 86–94.
Xu, J., Peng, S., Yang, S., Wang, W., 2012. Ammonia volatilization losses from a rice paddy with different irrigation and nitrogen managements. Agric. Water Manag. 104, 184–192.
Xu, P., Zhang, Y., Gong, W., Hou, X., Kroeze, C., Gao, W., Luan, S., 2015. An inventory of the emission of ammonia from agricultural fertilizer application in China for 2010 and its high-resolution spatial distribution. Atmos. Environ. 115, 141–148.
Yamaji, K., Ohara, T., Akimoto, H., 2004. Regional-specific emission inventory for NH3, N2O, and CH4 via animal farming in south, southeast, and East Asia. Atmos. Environ. 38, 7111–7121.
Yan, X., Cai, Z., Yang, R., Ti, C., Xia, Y., Li, F., Wang, J., Ma, A., 2011. Nitrogen budget and riverine nitrogen output in a rice paddy dominated agricultural watershed in eastern China. Biogeochemistry 106, 489–501.
Yan, X.Y., Akimoto, H., Ohara, T., 2003. Estimation of nitrous oxide, nitric oxide and ammonia emissions from croplands in East, Southeast and South Asia. Global Change Biol. 9, 1080–1096.
Yang, W., Zhu, A., Zhang, J., Zhang, Y., Chen, X., He, Y., Wang, L., 2013. An inverse dispersion technique for the determination of ammonia emissions from urea-applied farmland. Atmos. Environ. 79, 217–224.
Zhang, J., Cai, Z., Zhu, T., Yang, W., Mueller, C., 2013. Mechanisms for the retention of inorganic N in acidic forest soils of southern China. Sci. Rep.-UK 3, 1–8.
Zhang, J., Zhang, F., Yang, J., Wang, J., Cai, M., Li, C., Cao, C., 2011. Emissions of N2O and NH3, and nitrogen leaching from direct seeded rice under different tillage practices in central China. Agric. Ecosyst. Environ. 140, 164–173.
Zhang, X., Wu, Y., Liu, X., Reis, S., Jin, J., Dragosits, U., Van Damme, M., Clarisse, L., Whitburn, S., Coheur, P., Gu, B., 2017. Ammonia emissions may be substantially underestimated in China. Environ. Sci. Technol. 51, 12089–12096.
Zhang, Y., Cao, F., 2015. Fine particulate matter (PM2.5) in China at a city level. Sci. Rep.-UK 5, 1–12.
Zhao, M., Tian, Y., Ma, Y., Zhang, M., Yao, Y., Xiong, Z., Yin, B., Zhu, Z., 2015. Mitigating gaseous nitrogen emissions intensity from a Chinese rice cropping system through an improved management practice aimed to close the yield gap. Agric. Ecosyst. Environ. 203, 36–45.
Zhao, X., Zhou, Y., Wang, S., Xing, G., Shi, W., Xu, R., Zhu, Z., 2012. Nitrogen balance in a highly fertilized rice-wheat double-cropping system in Southern China. Soil Sci. Soc. Am. J. 76, 1068–1078.
Zhou, F., Ciais, P., Hayashi, K., Galloway, J., Kim, D., Yang, C., Li, S., Liu, B., Shang, Z., Gao, S., 2016. Re-estimating NH3 emissions from Chinese cropland by a new nonlinear model. Environ. Sci. Technol. 50, 564–572.