**Crop-specific ammonia volatilization rates and key influencing factors in China - A data synthesis**

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**Abstract** Ammonia (NH3) is an important alkaline reactive nitrogen (Nr) species which is involved in global nitrogen (N) biogeochemical cycling, but which has negative impacts on the environment and human health. In order to better understand and control the NH3 loss potential in soil-crop systems in China, an integrated data analysis including 2002 observations from 344 published articles between 1980 and 2021 was conducted. The typical NH3 volatilization rate (AVR) and the main factors influencing AVR in the major Chinese crops (rice, maize, wheat, field-grown vegetables and greenhouse vegetables) were estimated and analyzed. The mean AVR for all crops was 9.7%, with rice having the highest AVR (16.0%) and greenhouse vegetables the lowest (1.8%). The most important influencing factors were fertilizer placement, meteorological conditions (especially temperature and rainfall) and soil properties (SOM). Subsurface N application produced a significantly lower AVR compared to surface application. High crop yield and N use efficiency were generally associated with low AVRs. In conclusion, high N application rates, inefficient application methods and the use of loss-prone N fertilizer types are the main factors responsible for high AVRs in major Chinese croplands.

**Key words:** Soil-crop systems; ammonia volatilization loss; machine learning model; influencing factors

**1 Introduction**

With the development of ammonia (NH3) synthesis for nitrogen (N) fertilizer production, the effective application of the fertilizer has become important for ensuring the productivity and efficiency of agricultural systems (Erisman et al., 2008). However, the continuous increase of N inputs and the extensive use of N in crop production have led to the inefficient utilization of this resource, resulting in large reactive N (Nr) losses, causing a series of ecological and environmental issues (Canfield et al., 2010; Galloway et al., 2008). NH3 volatilization is one of the main pathways of N loss from fertilizers, and its emission is a major determinant of air quality, impacting on public health (Gu et al., 2021). In addition, NH3 can return to land and water through atmospheric deposition, disrupting terrestrial ecosystem functions (Bergstrom and Jansson, 2006; Clark and Tilman, 2008; Liu et al., 2013).

China is the largest user of N fertilizer, accounting for 1/3 of the world’s consumption. An NH3 emission inventory suggests that about 3.0-6.2 Tg NH3 is being lost from China’s crop production systems, accounting for 29.9-57.5% of total NH3 emissions from China (Sha et al., 2021). The NH3 volatilization rate (AVR, the N lost by NH3 volatilization after fertilization (and deducting the soil background emission) as a percentage of the N fertilizer applied, is an important indicator for evaluating the NH3 emission potential of farmland (Xu et al., 2019; Zhan et al., 2021). In order to evaluate the contribution of deposited NH3 to indirect greenhouse gas emissions, the IPCC set the AVR as a constant 10%. However, Jiang et al. (2017) showed that the AVR has a nonlinear response to N application rate, so using the IPCC constant would overestimate emissions at low N applications but underestimate them at high N applications (Jiang et al., 2017). Also, due to differences in field management practices and the heterogeneity of soils and climate, NH3 emissions vary greatly at a range of spatiotemporal scales. An increasing number of studies have taken into account the effects of soil properties, climate factors and management on NH3 losses in order to obtain region-specific emission factors, increasing the robustness of emission inventories or model estimates (Kang et al., 2016; Zhou et al., 2016).

The factors influencing NH3 emissions from soil-crop systems are complex and diverse (Sha et al., 2021). Analysis of the potential contributions of each factor to AVR can not only deliver more accurate NH3 volatilization loss rates, such as the AVR from specific crops, fertilizers or fertilization methods, but also provide strategies for NH3 emission mitigation. The traditional linear regression model has significant limitations in analysis or prediction, in particular because of the non-linear response of AVR to some explanatory variables (Zhou et al., 2016). The Boosted Regression Trees (BRT) model is a method for machine learning that can solve the complex nonlinear relationships between predictors and response variables, and also deal with the interactions between predictors, eliminating colinearity interference (Elith et al., 2008). In contrast to traditional regression analysis (fitting a single model), BRT continuously fits a single model by sampling (Bagging), evaluates the performance of the added model using the prediction deviation and eliminates the model with the weakest fit. In this stagewise simulation process, thousands of simple models will eventually be found to describe the relationship between explanatory variables and response variable. Finally the “Boosted” method summarizes the results of multiple single tree models in order to optimize and improve the performance and predictive ability of the overall model, and avoid over-fitting (Carslaw and Taylor, 2009; Leathwick et al., 2006).

We integrated data from published literature to analyze the characteristics of NH3 emissions after fertilizer application in China, summarized the AVR of different crops and used the BRT model to analyze the effects of different climatic conditions and farmland management on crop-specific AVRs. The aim of this study was to identify key influencing factors on AVRs and provide options for NH3 mitigation in China and which could be applied in other countries.

**2 Materials and Methods**

**2.1 Data collection**

We used ‘NH3 volatilization’ or ‘NH3 emission’ as key words to search relevant peer-reviewed articles and academic dissertations, published between January 1, 1980 and August 31, 2021, from the Web of Science and China Knowledge Resource Integrated databases. Publications were carefully reviewed, excluding experiments that were not conducted in China and removing duplicate data by comparing coordinates. In trials that spanned several crop rotations or years, inter-annual and inter-seasonal data were considered as independent observations. Publications were then systematically screened according to the following criteria: 1) only field trials and *in situ* lysimeter studies were included; 2) experiments must include a zero N addition treatment and field management should include consistent methods of N fertilizer application; 3) cumulative NH3 loss and the N application rate must be reported in order to calculate the AVR.

sampling sites

**Fig. 1** Location of experimental sites included in this analysis

A total of 344 studies (including 2002 observations) were included in the analysis (Fig1). Cumulative NH3 loss, crop yield, N use efficiency (calculated by N uptake in the N fertilizer treatment minus that in the zero N addition treatment, divided by the N application rate), meteorological conditions (mean air temperature and rainfall during the growing season), field management and soil properties were collected from each publication. Data were extracted by using digitizer tool in Origin 2019 software.

2.2 Data analysis

2.2.1 NH3 volatilization rate (AVR)

The AVR was calculated using following formula:

Where TNH3 is the cumulative NH3 loss after N fertilizer application, CNH3 is the corresponding control (zero N addition) value, and NAR is the N application rate (kg N ha-1).

The data were separated according to crop type, and included rice, maize, wheat, field-grown vegetables, greenhouse vegetables and others (cotton, grasses and productive forests). The means of crop-specific AVRs and corresponding 95% confidence intervals were calculated by bootstrapping (100 iterations). The differences between crops or field management regimes were assessed using the Kruskal-Wallis non-parameter test followed by a Dunn post-hoc comparison at a 5% level of significance. Statistical analyses were performed in R 4.1.1 using the boot and PMCMRplus packages.

2.2.2 Boosted regression trees model

Influencing factors for each crop-specific AVR were analyzed using the boosted regression trees (BRT) model. BRT is an advanced form of machine learning that can deal with complex nonlinear relationships between the response variable (AVR) and explanatory variables (influencing factors). In this analysis, potential explanatory variables were selected and defined as follows:

(1) Explanatory variables selection and definition:

-Temperature (℃): mean air temperature during crop growth;

-Rainfall (mm): cumulative rainfall during crop growth;

-N types: including urea, ammonium bicarbonate (ABC), organic fertilizer (OF), organic fertilizer plus chemical fertilizer (OF+CF) and others;

-NAR (kg ha-1): N fertilizer application rate;

-NFP: N fertilizer placement including surface (broadcasting to the soil surface) and subsurface (incorporated, deep placement, fertigation and irrigation after broadcasting). For an experiment with multiple application methods, if the subsurface N application exceeded 50% of the total application, the experiment was considered to use subsurface application;

-NAT: N application time, including single applications and split applications;

-Irrigation (mm): total irrigation rate for one crop season (only applicable for greenhouse vegetables).

-S\_pH: soil pH;

-SOM (g kg-1): soil organic matter;

-S\_TN (g kg-1): soil total nitrogen;

-DM: NH3 emission determination method, including dynamic chamber, vented chamber and closed chamber.

(2) Model running and simplification

The BRT model was operated in R software using the gbm and dismo packages. Gaussian error structure was chosen to fit the model because of the nature of the response variable. Instead of fitting a single model, BRT stagewise fitted multiple simple models and then evaluated the feasibility (predictive error) of each single model by adopting 10-fold cross-validation. The boosting method integrated information from simple models to give a collective interpretation. In order to minimize model predictive error and prevent over-fitting, the explanatory variable that made the lowest contribution to the response variable was stepwise removed, and changes of predictive deviance used to evaluate the suitability of simplifying the explanatory variable set, for which decreasing predictive deviance after removing one explanatory variable indicated acceptable simplification. In this way, NAT and DM were removed from the explanatory variable set (Elith et al., 2008).

The optimal settings of the model input parameters such as tree complexity (5, 8 and 10), learning rate (0.1, 0.05 and 0.01) and bag fraction (0.5, 0.6 and 0.75) were obtained by testing the combination systematically and using predictive deviance to assess model performance. The final settings are shown in Table S1. The relative importance of each explanatory variable was calculated and relationships with response variables displayed using partial dependency plots. Additionally, the ggBRT package obtains a 95% confidence interval using bootstrapping (100 iterations), visualized in partial dependency plots (Jouffray et al., 2019).

2.2.3 Linear mixed effect model

A linear mixed effect model was employed to analyze the relationship between AVR and measures of agronomic performance such as N use efficiency (NUE) and yield improved rate (YIR). NUE and YIR were calculated using the following equations:

Where NT and YT are the N uptake and yield from the N fertilizer treatment, respectively; NC and YC are the N uptake and yield from the zero N treatment; NAR is the N application rate (kg N ha-1).

In the linear mixed effect model, NUE or YIR are considered as the response variable, AVR is a fixed effect and each study set is a random effect. The intercept and slope of the models were calculated using the ‘lme4’ package in R 4.1.1. Model performance and differences between two models were analyzed using the ‘car’ package in ANOVA. The R2 of the models was obtained with the ‘MuMIn’ package (Nakagawa et al., 2013).

3 Results

3.1 Crop-specific NH3 volatilization rate

The mean NH3 volatilization rate (AVR) of cropland in China was 9.7% (CI: 9.2-10.1%, N=2002), of which the AVR of the staple crops (rice, maize and wheat) was 10.7% (CI: 10.2-11.2%, N=1611) (Fig. 2). Of the crop-specific AVRs, rice was the largest at 16.2% (CI: 15.0-16.9%, N=700), followed by field-grown vegetables (8.4%, CI: 7.0-9.9%, N=114) and maize (7.8%, CI: 7.0-8.5%, N=445). The lowest AVR was from greenhouse vegetables (1.8%, CI: 1.2-2.2%, N=99).

EF_CROP

**Fig. 2** Crop-specific NH3 volatilization rates (AVRs) for rice, maize, wheat, field-grown vegetables, greenhouse vegetables and others.

3.2 Key influencing factors of crop-specific NH3 volatilization rates (AVRs)

N fertilizer placement (NFP) made the largest contribution to the variation of AVR for maize (17.8%), wheat (40.7%) and field-grown vegetables (17.1%) (Fig. 3). The AVR of subsurface applications was significantly lower than that of surface applications (p<0.05). Soil organic matter (SOM) made the largest contribution to the variation in AVR for rice (22.9%) and greenhouse vegetables (31.9%). High SOM content tended to produce a low AVR. Soil total N content and pH accounted for 5.7-14.6% and 2.9-12.1% of variation in AVR, respectively. Weather conditions during crop growth also made a large contribution to AVR. For example, mean air temperature explained 9.6-16.5% of the variation in AVR, and high temperatures led to high NH3 loss potential. Rainfall accounted for 12.6%, 8.6%, 11.2% and 14.5% of variation in AVR for rice, maize, wheat and field-grown vegetables, respectively; irrigation rate made a 14.0% contribution to AVR variation in greenhouse vegetables. N application rate (NAR) accounted for 7.2-18.7% of the variation in AVR and was positively related to the AVR in rice, wheat and field-grown vegetables. The AVR of ammonium bicarbonate (ABC) applied to rice was higher than that of other fertilizers and using organic fertilizer (OF) alone tended to produce a low AVR (Fig S1).

RICE

MAIZE

WHEAT

OPV

GHV

**Fig. 3** Key influencing factors and their contribution to the variation of the NH3 volatilization rate (AVR)

3.3 Correlation of the NH3 volatilization rate with agronomic performance

Based on the analysis of the linear mixed effect models, the slope model gave the best fit and the fixed effect (AVR) plus random effect (i.e. the different studies) resulted in models with an R2 of 0.6, with a higher NUE generally indicating smaller NH3 losses (Fig 4A, Table S1). A similar relationship was observed between YIR and AVR (Fig 4B, Table S1), with the slope model performing best with an R2 of 0.7.

NUE-slopeYIR-slope

1. (B)

**Fig. 4** Correlation of crop agronomic performance (A. NUE; B. YIR) with the NH3 volatilization rate (AVR)

**4 Discussion**

**4.1 Characteristics of NH3 volatilization rates (AVRs) from major crops**

The data analyzed here produce a lower AVR compared to those from previous studies. For example, Ma et al. (2020) synthesized 614 observations from field experiments in China and found an average AVR of 13.48%; Pan et al. (2016) found an average AVR of 15.9% in East Asia. Based on 495 observations Zhou et al. (2016) reported a mean AVR of 6.5±7.1% for dryland crops and 15.0±11.1% for paddy rice, similar to this study. We found that rice production had the highest potential NH3 loss, which could be due to the high N applications to rice, unsuitable application methods (broadcasting) and flooding water properties (such as high pH and low redox potential) (Wang et al., 2018). The AVRs of maize and wheat were relatively low compared to previous estimates, changes in fertilizer practices may be responsible for the difference. For example, the AVRs of wheat and maize following surface application of N were 17.2% and 11.3%, respectively, which were significant higher compared to those from subsurface application (5.8% and 2.7%, respectively). Zhan et al. (2021) used global data to evaluate the AVRs of various crops and found that cotton had the highest AVR of 23.7%. This is very different from present dataset, where cotton generally had a low AVR (3.1%, CI: 2.1-3.9%, N=43, Fig. S3). This was probably due to the different field management practices in China (dripped irrigation under plastic mulch). Greenhouse vegetable production generally had low AVRs because intensive irrigation prevented the substrate (NH4+-N) from diffusing to surface soil. However, the high N application rates of up to 1000 kg N ha-1yr-1 would lead to large amounts of NH3 loss even at low AVRs. NH3 losses to the environment could be especially large during the intermittent opening of greenhouses for ventilation and cooling (Liao et al., 2019). Field-grown vegetable production also receives large amounts of fertilizer and irrigation water, as for greenhouse vegetable production. However complex meteorological factors (such as wind speed) lower the gas phase resistance to NH3 loss (Vlek and Stumpe, 1978) and so lead to high AVRs.

**4.2 Factors controlling NH3 volatilization from croplands**

Applied N is the main anthropogenic source of NH3. A comprehensive understanding the factors that control the process would aid its mitigation (Zhou et al., 2016). Some studies have used artificial neural networks to analyze the main factors affecting NH3 volatilization after manure application, finding that climate (especially temperature and wind speed during the experimental period) explained 30% of the variation in NH3 losses, with manure composition and management methods also having a significant impact (Lim et al., 2007): meteorological factors explained 25.8%, 25.1%, 20.7% and 29.5% of the AVRs in rice, maize, wheat and field-grown vegetables, respectively. Previous research has suggested that the AVR increases with increasing annual mean precipitation (Ma et al., 2020), but we found no consistent trend. Precipitation was not included in our analysis of greenhouse vegetables, for obvious reasons, but irrigation rate explained 14.0% of the AVR, with the AVR decreasing with increasing irrigation.

Different N fertilizer types had different NH3 loss potentials. As an alkaline fertilizer, ammonium bicarbonate (ABC) is highly volatile: the combination of NH3 and HCO3- causes a rapid increase of soil pH after application, enhancing NH3 loss, and it has been banned for agricultural application in some countries (Bittman et al., 2014). The high NH3 loss potential of urea is also related to increases in soil pH during urea hydrolysis, but soil urease activity and soil buffering capacity are important factors affecting the process (Haden et al., 2011). The average AVR of organic fertilizers is low due to their slow mineralization rate. Volatilization intensity is mainly determined by the properties of the organic fertilizer, such as C/N ratio, NH4+-N content and pH (Xia et al., 2017); the AVR of mixed organic-inorganic fertilizers has been found to be very varied, which may be related to the ratio of organic to inorganic N in the wide range of experiments, with the proportion of organic fertilizer negatively correlated with NH3 emission. The N application rate is an important mediator. A nonlinear response of AVR to application rate has been widely reported in previous studies (Jiang et al., 2017; Zhou et al., 2016). However, the AVRs of crops in this study did not necessarily increase with the N rate, which may be due to the range of management practices such as application method, and which we found to be the main factor controlling AVR. Previous research has shown that each 1 cm increase in the depth of N placement reduced NH3 volatilization by 7% compared with surface application (Rochette et al., 2013). However, deep N application effectively prevented the migration of NH3 and NH4+-N in solution to the soil surface, increasing the adsorption of NH4+-N by the soil and so limiting its availability to crops (Sommer et al., 2004).

Soil properties have a significant impact on the AVR. Soil organic matter content was significantly negatively correlated with NH3 volatilization rate (correlation coefficient: -0.6) (Duan and Xiao, 2000; Sharpe and Harper, 1995), which can be attributed to the high cation exchange capacity in soils with a large SOM content. increasing the storage capacity for NH4+-N and reducing the NH3 volatilization potential. Some research has also found that the OH- buffer capacity is stronger in high SOM soils, which could offset NH3 losses caused by increasing soil pH after fertilizer application, especially for urea (Aitken et al., 1990). Soil pH is another key factor affecting AVR. At high pH, the dissociation of NH4+ will increase and the chemical equilibrium will shift towards NH3, increasing the risk of NH3 loss. In paddy rice production, the pH of the floodwater is the main factor determining NH3 volatilization, but the pH of the floodwater and the soil are not always linearly correlated: the growth of algae in floodwater can consume H+ during photosynthesis which leads to an increase in pH (Thind and Rowell, 2000).

From the perspective of N utilization, excessive N inputs will cause a large N surplus, which is positively correlated with reactive N loss (Chen et al., 2014). The YIR and NUE are important indicators for evaluating the uptake and utilization of N fertilizer by crops, and a higher NUE and/or crop yield are generally associated with low NH3 emissions (Zhang et al., 2015).

**4.3 Mitigation options for NH3 loss from croplands**

Decreasing N fertilizer applications are essential for reducing NH3 emissions from crop production systems. The “Chemical Fertilizer Zero Growth Action” policy was proposed by the Chinese Government in 2015 and has proved very effective in that the input of chemical fertilizers and N fertilizer in particular has decreased by 10.3% and 13.4%, respectively, in 2019 compared with 2015. At the same time, fertilizer utilization efficiency for the three staple crops increased to 40% according to the Chinese Ministry of Agriculture and Rural Affairs (http://www.moa.gov.cn/). In theory, therefore, NH3 emissions from these crops should have been reduced (Liu et al., 2021). To further reduce emissions, even more effective measures are needed. The surface application of N fertilizers is commonly used by smallholder farmers. The use of appropriate machinery for the deep placement of fertilizers is needed to achieve N conservation and control NH3 losses. Using the right type of N fertilizer (another of the ‘4Rs’) is also necessary for achieving a ‘win-win’ for both economic and environmental benefits. Using ammonium or nitrate-based fertilizers in place of fertilizers with high volatilization potential, such as urea or ammonium bicarbonate, or improving the association of livestock and crop production systems, using organic manure to partially or wholly replace chemical fertilizers (Ti et al., 2019; Xia et al., 2017) is therefore an important next step. The very effective NH3 mitigation efficacy of enhanced-efficiency fertilizers such as slow/controlled-release fertilizers and stabilized N fertilizers (with a urease inhibitor, nitrification inhibitor or both) has been demonstrated, e.g. Sha et al. (2021). However, choosing the right mitigation option must focus on the reduction target and take into consideration climate, farming practice and socio-economic conditions.

**5 Conclusion**

Based on data integration from published literature, we analyzed crop-specific NH3 volatilization rates (AVR) of the main crops grown in China and assessed the main influencing factors on AVR using a boosted regression tree model. AVR in the main croplands averaged 9.7%, of which rice had the highest AVR of 16.2%, followed by field-grown vegetables, maize, wheat, with the lowest AVR being for greenhouse vegetables. The N rate, fertilizer placement, meteorological conditions and soil properties are the three key factors affecting AVR. Subsurface placement of N produced significantly lowers AVRs compared to surface application. High yields and N use efficiency were generally associated with low AVRs. The present results will not only enrich the database of regional and specific AVRs in China, but also identify its key influencing factors, providing a strong theoretical basis for the selection and evaluation of NH3 mitigation options.

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