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Global soil-derived ammonia emissions from agricultural nitrogen fertilizer application: A refinement based on regional and crop-specific emission factors

Ruoya Ma[1] | Jianwen Zou[1] | Zhaoqiang Han[1] | Kai Yu[1] | Shuang Wu[1] | Zhaofu Li[1] | Shuwei Liu[1] | Shuli Niu[2] | William R. Horwath[3] | Xia Zhu-Barker[3]

[1]Jiangsu Key Laboratory of Low Carbon Agriculture and GHGs Mitigation, College of Resources and Environmental Sciences, Nanjing Agricultural University, Nanjing, China

[2]Institute of Geographic Sciences and Natural Resources Research, Chinese Academy of Sciences, Beijing, China

[3]Department of Land, Air and Water Resources, University of California, Davis, CA, USA

Correspondence
Shuwei Liu, Jiangsu Key Laboratory of Low Carbon Agriculture and GHGs Mitigation, College of Resources and Environmental Sciences, Nanjing Agricultural University, Nanjing, China.
Email: swliu@njau.edu.cn

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Abstract

Ammonia (NH_3) emissions from fertilized soils to the atmosphere and the subsequent deposition to land surface exert adverse effects on biogeochemical nitrogen (N) cycling. The region- and crop-specific emission factors (EFs) of N fertilizer for NH_3 are poorly developed and therefore the global estimate of soil NH_3 emissions from agricultural N fertilizer application is constrained. Here we quantified the region- and crop-specific NH_3 EFs of N fertilizer by compiling data from 324 worldwide manipulative studies and focused to map the global soil NH_3 emissions from agricultural N fertilizer application. Globally, the NH_3 EFs averaged 12.56% and 14.12% for synthetic N fertilizer and manure, respectively. Regionally, south-eastern Asia had the highest NH_3 EFs of synthetic N fertilizer (19.48%) and Europe had the lowest (6%), which might have been associated with the regional discrepancy in the form and rate of N fertilizer use and management practices in agricultural production. Global agricultural NH_3 emissions from the use of synthetic N fertilizer and manure in 2014 were estimated to be 12.32 and 3.79 Tg N/year, respectively. China (4.20 Tg N/year) followed by India (2.37 Tg N/year) and America (1.05 Tg N/year) together contributed to over 60% of the total global agricultural NH_3 emissions from the use of synthetic N fertilizer. For crop-specific emissions, the NH_3 EFs averaged 11.13%–13.95% for the three main staple crops (i.e., maize, wheat, and rice), together accounting for 72% of synthetic N fertilizer-induced NH_3 emissions from croplands in the world and 70% in China. The region- and crop-specific NH_3 EFs of N fertilizer established in this study offer references to update the default EF in the IPCC Tier 1 guideline. This work also provides an insight into the spatial variation of soil-derived NH_3 emissions from the use of synthetic N fertilizer in agriculture at the global and regional scales.

KEYWORDS

ammonia, emission factor, estimate, fertilizer application, reactive nitrogen

Ruoya Ma and Jianwen Zou contributed equally to this work.

1 | INTRODUCTION

The global excessive release of reactive nitrogen (N) from fertilized soils to the atmosphere along with the subsequent deposition to land surface exert adverse consequences on the environment and human health, such as deteriorating air quality, accelerating soil acidification and constraining ecosystem outcomes (Galloway et al., 2008; Jerrett, 2015). Ammonia (NH_3) volatilization represents a major pathway of soil N loss, showing high variation under different case-specific soil environments (Pan et al., 2016; Singh et al., 2013; Xu et al., 2019). Globally, the use of N fertilizer is expected to increase in the coming decades, particularly in developing countries, where crop N uptake only accounts for 30%–50% of the applied fertilizer N in croplands (Bouwman et al., 2002; Ladha et al., 2005). This low N use efficiency is generally associated with high soil gaseous N loss through volatilization of NH_3 and the processes of nitrification and denitrification, as well as leaching and runoff in croplands (Zhou et al., 2016). Therefore, agricultural practices are highly expected to mitigate NH_3 emissions from N fertilized soils while increasing N use efficiency. A thorough understanding of global patterns of N fertilizer-induced soil NH_3 emissions and their driving factors would provide policy-makers with a reference to improve fertilizer N use efficiency and alleviate negative environmental impacts.

Recently, several studies have addressed N fertilizer-induced NH_3 emissions from agricultural soils by either using meta-analysis or modeling approaches, concluding that generally high NH_3 mitigation potential exists (Pan et al., 2016; Qiao et al., 2015; Ti et al., 2019; Xu et al., 2019; Zhou et al., 2016). These studies have concentrated on the effectiveness of fertilizer options and practices, but limited in scope to establish a worldwide database for a robust bottom-up estimate of global soil NH_3 emissions from agricultural N fertilizer use. Furthermore, these efforts have predominantly focused on application events, but seldom stressed the importance of seasonal or long-term outcomes despite that agricultural practices usually have postevent impacts on soil processes related to NH_3 emissions. Therefore, given that agricultural activities have been reported to dominate the release of global NH_3 to the atmosphere (Bouwman et al., 1997; Paulot et al., 2014; Zhang et al., 2010), a structural insight into global fertilizer-induced NH_3 emissions across N-managed ecosystems is needed.

Emission factor (EF) and process-based model have been frequently adopted to quantify global or regional NH_3 emissions from N fertilizer use. The NH_3 EF represents the percentage of applied N fertilizer that volatilizes as NH_3. The global default value of NH_3 EFs for different fertilizer sources have been recently updated in the Intergovernmental Panel on Climate Change (IPCC) Tier 1 guideline to build emission inventories at the global and regional scales (Bouwman et al., 2002; IPCC, 2019; Pan et al., 2016; Xu et al., 2019). Furthermore, numerous efforts have been made to refine EFs for NH_3 emissions at the regional or national scales (e.g., Europe, United States, and China). Indeed, some studies have proposed an average of EFs ranging from 14% to 21% for synthetic N fertilizer, resulting in an overall amount of 15.7–23.5 Tg N/year of fertilizer N lost as NH_3 globally (Beusen et al., 2008; Bouwman et al., 2002; IFA/FAO, 2003; IPCC, 2019). Bouwman et al. (1997) estimated that the global NH_3 emissions from synthetic N fertilizer use were 9 Tg N in 1990. This estimate was comparable to an earlier evaluation of 8.5 Tg N/year by Schlesinger and Hartley (1992), but lower than a recent estimate of 12 Tg N/year in the year 2000 by Riddick et al. (2016). Using process-based modeling approaches, Xu et al. (2019) showed the dynamics of global NH_3 emissions from N fertilizer use over four historical climate datasets and indicated an increase rate from 1.9 to 16.7 Tg N/year between 1961 and 2010. While these earlier statistics and modeling efforts have provided insights into fertilizer-induced soil NH_3 emissions, they also showed a high degree of uncertainty. To lower the uncertainties of NH_3 estimates, the updated IPCC guideline refined EFs for different fertilizer sources but it did not take other important factors into account, such as region, ecosystem, and cropping type. The incorporation of these factors would help to further constrain predictions of N fertilizer-induced NH_3 emissions at the global scale (Pan et al., 2016). For instance, Europe showed a consistent declined emission trend since the 1980s (Xu et al., 2019). In contrast, southern Asia has undergone an increase in soil NH_3 emissions since the 1990s and the highest emission rate was found in the North Plain of China, which is largely attributed to the expansion of croplands and the increase of N fertilizer consumption (Xu et al., 2019). In addition, soil NH_3 emissions from the use of N fertilizer were crop-type dependent and were dominated by location and specific N fertilizer requirements (Zhang et al., 2010; Zhou et al., 2016). Therefore, using the EFs further refined by these factors with less uncertainties, we would expect to map soil NH_3 emissions from N fertilizer use at the global and regional scales.

This study focused on a global bottom-up estimate of soil NH_3 emissions from the application of synthetic N fertilizer and manure in agricultural systems, including rice paddies and cropping uplands such as wheat, maize, vegetable, and cash croplands. Here 4,590 experimental measurements derived from 324 peer-reviewed publications within an updated and comprehensive dataset were synthesized to quantify global NH_3 emissions from N fertilizer application and to explore potential mitigation strategies. The main objectives of this study were to: (a) outline the spatial variation of soil NH_3 emissions at the global and regional scales using the updated complete regional and crop-based N fertilizer consumption data in 2014; (b) develop the direct NH_3 EFs of N fertilizer by ecosystem type, fertilizer source, region and crop type; and (c) refine agricultural practices to reduce NH_3 emissions from fertilized soils and identify the key factors driving the emission pattern and direct EFs of NH_3.

2 | MATERIALS AND METHODS

2.1 | Data extraction and refinement

We launched a detailed review of the literature published in peer-reviewed journals through the year 2018 (cut-off date on May 22, 2018). All the publications on soil NH_3 emissions were identified from the Web of Science and Google Scholar using different combinations of searching keywords ('ammonia' OR 'NH_3' AND 'emissions' OR 'volatilization' OR 'loss' OR 'release' AND 'fertilizer' OR 'synthetic/chemical N' OR 'manure' OR 'slurry' OR 'FYM' AND 'soil'). We extracted original experimental results from the identified studies providing that soil NH_3 flux data were continuously measured. We also recorded data from these studies on geographic, soil, or other experimental traits in the cases that they were individually or simultaneously available (Tables S1 and S2). Original data in publications were directly extracted from tables and texts. In the cases that data were only reported in figures, GraphClick was used to extract mean values and standard errors. For each literature review, the following original information was collected: soil NH_3 fluxes, direct NH_3 EF of N fertilizer, location (longitude and latitude), general climate attribution (tropical, subtropical and temperate), experimental duration, land-use type, dominated vegetation, mean annual air temperature (MAT) and precipitation (MAP), soil properties [e.g., texture (coarse, medium, and fine), soil pH, mean soil temperature in the depth of 0–20 cm, water-filled pore space (WFPS), soil organic carbon (SOC) content, total N content, soil C/N ratio, and mineral N availability], application rate and source of N fertilizer, as well as method and frequency of NH_3 flux measurement. Other more detailed traits on literature and data sources can be found in the Supplementary Information.

To refine the extracted dataset, we established the following criteria to avoid bias during the screening process; only field measurements were included in this analysis. Data collected from laboratory or pot experiments, as well as soil column-based or modeling simulated results, were excluded. For the data collected from natural habitats, the occasional field NH_3 flux measurements without covering the whole experimental period or non-representative of consecutive measurements were excluded. To test the effectiveness of different management practices on mitigating NH_3 emissions, the controls and treatments within a given study should have the same experimental duration and crop cultivation history. The treatments with no replication or no reported number of replicates or the grouping categories with less than two data pairs were finally discarded from the database to minimize the bias noises in this meta-analysis. The final dataset was comprised of 4,590 field measurements sourced from 324 publications, of which 3,296 measurements were taken from agricultural ecosystems and 1,233 measurements from non-agricultural ecosystems. A subgroup of 263 publications with 3,047 observations reported direct NH_3 EFs of N fertilizer in the database (Figure 3a).

2.2 | Data compilation and analyses

Data were first subjected to a standardization process to allow for comparisons. We calculated the balanced mean values of soil NH_3 fluxes and their EFs with the residual maximum likelihood procedure using GENSTAT release 4.2 to minimize the heterogeneity resulting from missing values and unequal number of observations among the reviewed literature (Payne, 2000). For the studies where only mean soil NH_3 flux data were reported, we calculated the cumulative NH_3 emissions by multiplying the flux data by the length of the measurement period. If only the standard error (*SE*) of a given data mean was available, the standard deviation (*SD*) was calculated as:

$$SD = SE \times \sqrt{n}, \tag{1}$$

where *n* is the number of observations. In the case of no *SD*/*SE* or any variance reported, we assumed 10% of the mean as *SD* (Luo et al., 2006). Fertilizer N-induced NH_3 emissions were calculated by subtracting background emissions from the total NH_3 emissions expressed as a percentage of N applied (Zhang et al., 2019). The direct NH_3 EF of N fertilizer was estimated by the following equation:

$$\text{EF}\ (\%) = \frac{E_\text{t} - E_\text{b}}{\text{N}} \times 100\%, \tag{2}$$

where E_t is the total emissions of NH_3 (kg N/ha) from fertilized treatments, E_b is the background emission of NH_3 (kg N/ha) from the control without N fertilizer application, and N refers to the applied fertilizer N rate (kg N/ha). If data on geographical information and MAT or MAP were missing in the original studies, we extracted relevant data from http://www.google.cn/maps and http://www.worldclim.org/. For studies only reporting data on particle size distribution of soil, we obtained soil texture information based on the International Classification System of Soil Texture. In order to allow for comparisons, the soil pH values measured with $CaCl_2$ were transformed with the formula $\text{pH}_{[H_2O]} = 1.65 + 0.86 \times \text{pH}_{[CaCl_2]}$ (Biederman & Harpole, 2013). In the identified studies, soil volumetric water content was far more frequently reported than WFPS. Thus, we unified them by converting volumetric water content to WFPS using the bulk density of soil. Similarly, if only soil organic matter (SOM) was reported, we converted SOM to SOC using a Bemmelen index value of 0.58.

To further compile data prior to analysis, the selected data were grouped by categories. We divided the soils into five land-use types as rice paddies, tilled cropping upland, no-till cropping upland, grassland, and forest. To address soil NH_3 emissions as affected by different N fertilizer sources, we categorized these data into four groups: synthetic N fertilizer, organic N fertilizer, mixed N fertilizer, and enhanced-efficiency N fertilizer (EENF). Besides fertilizer types, other six key experimental factors including crop residue and biochar amendment, tillage, irrigation, deep placement of fertilizers relative to surface broadcasting, and split relative to single application were also taken into consideration in this analysis. Soil textures were grouped into three general classes as coarse, medium, and fine. Soil pH across all measurements were grouped into two levels as acid/neutral soil (≤7) or alkaline soil (>7). According to the results of normal distribution and threshold analysis of the grouped data, other soil parameters such as soil organic C, total N, C/N ratio, and mineral N were further grouped as ≤1.5% or >1.5%; ≤0.15% or >0.15%; ≤6, 6–12 or >12; and ≤10 mg/kg, 10–30 mg/kg or >30 mg/kg, respectively.

In this meta-analysis, data gathered from studies on soils treated with fertilizer only (102 studies with 1,181 paired measurements) and the soils that were also exposed to other available experimental factors (49 studies with 304 measurements) were analyzed separately to examine the general response of soil NH_3 emissions to fertilization and other typical experimental controls. As a measure of effect size for the meta-analysis, we used the means of soil NH_3 fluxes from control (X_c) and treatment (X_t) groups to calculate the natural log-transformed response ratio (ln*R*). The standard deviations of both control and treatment were included as a measure of variance:

$$\ln R = \ln\left(X_t / X_c\right) = \ln\left(X_t\right) - \ln\left(X_c\right), \quad (3)$$

where X_t and X_c are means of NH_3 fluxes for the treatment and control groups, respectively. Its pooled variance (v) is estimated as follows:

$$v = \frac{s_t^2}{n_t x_t^2} + \frac{s_c^2}{n_c x_c^2}, \quad (4)$$

where n_t and n_c are the sample sizes for the treatment and control groups, while s_t and s_c are the standard deviations for the treatment and control groups, respectively.

A categorical fixed effects model was used to calculate the mean effect size for each category, where it is weighted in inverse proportion to its variance (Adams et al., 1997). Groups with less than two paired observations were removed from this meta-analysis. To account for the intrinsic relevance or non-independence among the responses of variables to fertilizer and experimental factors, the overall mean effect size and the 95% confidential interval (CI) of a given grouping category generated by bootstrapping (9,999 iterations) were estimated with the mixed-effect model in R. Treatment effects were considered significant if 95% CI did not overlap with the line ln$R$ = 0.

TABLE 1 Emission factors (EFs) and EF-derived NH_3 emissions from agricultural N fertilizer application among different regions and crop types at both the global and regional scales

Grouping category	n	EFs (%)	CI (%)	Total emissions (Tg N/year)[a]
Global				
Synthetic N	1,410	12.56	11.91–13.22	13.71 ± 1.08
Manure N	134	14.12	12.04–16.21	3.79 ± 0.23
China				
Synthetic N	614	13.48	12.59–14.36	4.20 ± 0.26
Manure N	33	13.72	11.34–16.10	0.68 ± 0.02
Synthetic N fertilizer use in agriculture				
Regional breakdown				
Eastern Asia	678	13.80	12.90–14.70	4.39 ± 0.28
Southern Asia	139	12.55	10.33–14.78	2.91 ± 0.25
South-eastern Asia	64	19.48	15.34–23.62	1.44 ± 0.06
North America	216	8.75	7.02–10.49	1.27 ± 0.21
South America	181	11.98	10.25–13.71	0.78 ± 0.06
Europe	38	6.00	3.57–8.43	0.87 ± 0.18
Africa	13	12.56	11.91–13.22	0.49 ± 0.04
Oceania	86	9.72	7.94–11.50	0.17 ± 0.01
Crop breakdown				
Global				
Rice	612	13.95	13.01–14.88	2.17 ± 0.07
Wheat	241	12.05	10.29–13.81	2.25 ± 0.17
Maize	302	11.13	9.64–12.61	2.03 ± 0.14
Vegetable	25	14.52	10.04–18.99	1.10 ± 0.16
Other crops	143	8.84	7.04–10.65	1.38 ± 0.14
China				
Rice	358	13.84	12.87–14.81	0.54 ± 0.03
Wheat	109	13.29	10.68–15.90	0.45 ± 0.04
Maize	103	14.33	11.51–17.15	0.67 ± 0.05
Vegetable	21	13.62	8.58–18.67	0.63 ± 0.04
Other crops	23	4.69	3.58–5.79	0.09 ± 0.01

[a]Mean ± 1 *SD* calculated from a given estimation group based on the present dataset.

In addition to meta-analysis procedures, one-way analysis of variance (ANOVA) was performed to test the differences in soil NH_3 emissions and direct NH_3 EFs of N fertilizer among ecosystem types, fertilizer types and species, and other experimental factors. The analysis of covariance (ANCOVA) was adopted to test the discrepancy of NH_3 emissions and direct NH_3 EFs between cropland and non-cropland soils as a relation to soil and environmental controls. We also used the linear mixed regressions to examine the correlations of soil NH_3 emissions and direct NH_3 EFs with the potential driving factors. All statistical analyses were carried out using JMP version 10.0 (SAS Institute, 2007).

2.3 | Scaling-up estimation

Based on both the constant and case-specific NH_3 EFs of N fertilizer estimated in this study, we scaled up the results from this synthesis to global context by distinguishing sources of NH_3 emissions from the application of synthetic N fertilizer versus manure in agricultural systems (Table 1). Fertilizer-induced NH_3 emissions from managed grasslands and forests were excluded from this estimation due to the agricultural focus of this study and the difficulty in quantifying the amount of N excreted during animal grazing and other stray N sources. To further reduce uncertainty, EENF such as the controlled-release N fertilizer and some management options that are not widely employed in agriculture have been also removed from our database for the current estimation. To gain an insight into the overall soil reactive gaseous N losses ($NH_3 + N_2O + NO$) from N fertilizer application at the global scale, we further estimated the fertilizer-induced total of $N_2O + NO$ emissions using the IPCC combined default EF values (Synthetic N: 1.22%, 95% CI 0.94 to 1.51; Manure N: 4.29%, 95% CI −1.15 to 9.70), which are based on the global averages across all ecosystem types as reviewed in the previous literature (IPCC, 2019; Liu et al., 2017). The statistical data on synthetic N fertilizer and manure consumption at the global and regional scales were derived from the updated complete statistical database built in 2014 by the Food and Agricultural Organization (FAO, 2014 Database Collections, Fertilizers by Nutrient Dataset and Livestock Manure Dataset, respectively). The crop-based fertilizer N consumption data were directly taken from the International Fertilizer Industry Association (IFA, 2014 Crop-based Fertilizer Consumption Reports, available at https://www.ifastat.org/plant-nutrition). To increase scientific credibility of this estimate, the incomplete datasets that both FAO and IFA databases shared were used as reference materials to examine their compatibility for a given statistical year. Since regional manure usage data were not fully available, the regional and crop-based NH_3 emissions were only estimated for synthetic N fertilizer using respective case-specific direct NH_3 EFs to reduce uncertainties. Particularly, China as one of the world's major agricultural production countries was highlighted in this study, and we therefore adopted the direct NH_3 EFs established from China-specific database with low uncertainties to give an insight into the role of China in global NH_3 inventory.

3 | RESULTS

3.1 | Soil background NH_3 emissions

Soil background NH_3 emissions are generally defined as those from soils free of anthropogenic N fertilizer input, which have been rarely reviewed in previous studies, especially its contribution to global NH_3 emissions is unclear. To this end, we grouped available data across unfertilized soils to compare the global soil background NH_3 emissions among different ecosystem sources. The mean flux rate of global background NH_3 across unfertilized soils was estimated to be 0.36 mg N m^{-2} hr^{-1}, showing a significant variation among ecosystems (Figure 1a, p = .01).

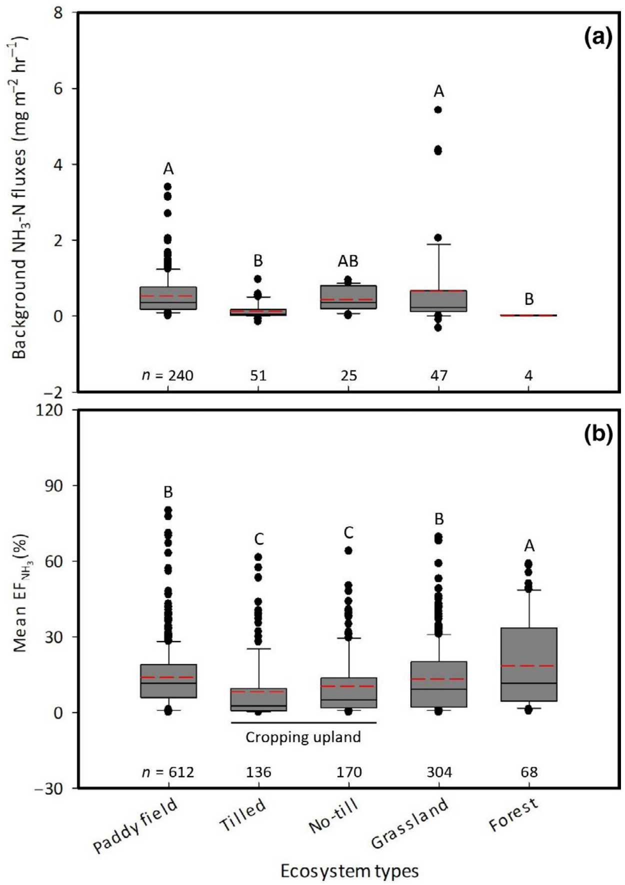

FIGURE 1 Background soil NH_3 fluxes (a) and direct emission factors (EFs) of N fertilizer for NH_3 (b) among ecosystem types. Different letters indicate significant differences in NH_3-N fluxes and EF_{NH_3} among ecosystem types at a statistical probability level of .05. The red dashed and black solid lines, lower and upper edges, and bars and black circles represent the mean and median values, 25th and 75th, 10th and 90th percentiles and outliers of data, respectively. Cropping uplands were further divided into tilled and no-till systems for detailed comparisons. The number of measurements (n) for each ecosystem type is shown next to the x-axis [Colour figure can be viewed at wileyonlinelibrary.com]

Generally, soil background NH_3 fluxes were the highest in grasslands, with a balanced mean of 0.66 mg N m^{-2} hr^{-1}, followed by rice paddies (0.53 mg N m^{-2} hr^{-1}), no-till cropping uplands (0.44 mg N m^{-2} hr^{-1}), tilled cropping uplands (0.13 mg N m^{-2} hr^{-1}), and forests (0.02 mg N m^{-2} hr^{-1}).

3.2 | Direct NH_3 EFs of N fertilizers

Globally, the direct constant NH_3 EFs of synthetic N fertilizer and manure were estimated to be 12.56% and 14.12%, respectively (Table 1). While in China, 13.48% and 13.72% of synthetic N fertilizer and manure, respectively, were found to be emitted as NH_3. When pooling data across different synthetic N fertilizers, the direct NH_3 EFs significantly differed among ecosystem and crop types (Figures 1b and 2b). In general, the direct EF was significantly higher in forests (18.54%) than in grasslands (13.30%) or croplands (12.09%). Among different croplands, rice paddies had the highest EF, while the lowest EF was found in tilled cropping uplands (Figure 1b). Generally, non-cropland soils showed a greater mean NH_3 EF of N fertilizer than in cropland soils. The same amount of N fertilizer applied into no-till cropping uplands was expected to result in higher NH_3 emissions than that applied into tilled cropping uplands (Figure 1b).

From a crop-type perspective, we grouped available data into four major broad crop categories (i.e., cereal, vegetable, cash crop, and forage) and compared the direct NH_3 EFs of synthetic N fertilizer among them (Figure 2b). Vegetable crops averaged to show the largest EF (14.52%), while cash crops had the lowest EF (8.84%). Among cereal crops, rice relative to upland cultivation (i.e. wheat and maize) had the largest EF (13.95%). In China, crop-specific EFs averaged 13.29%–14.33% for the four leading crops (Table 1). For region-specific EFs, the NH_3 EFs of synthetic N fertilizer were greater in Asia than in other regions (Table 1), with an average of 19.48%, 13.80%, and 12.55% in south-eastern Asia, eastern Asia, and southern Asia, respectively. North America, Europe, and Oceania showed relatively lower EFs, of which Europe had the lowest one (6.00%).

3.3 | Soil NH_3 emissions induced by different N fertilizer types and species

When averaged the data across all ecosystem types, the use of N fertilizer significantly increased soil NH_3 emissions by 162%, with a 95% confidence interval (CI) of 153%–171% compared to no fertilizer input. This positive response of NH_3 emissions to N fertilizer application varied with N fertilizer types (Table S3; Figure 3b). Among different fertilizer types, organic N fertilizer induced the greatest increase in soil NH_3 emissions (+304%), of which livestock manure produced greater emissions than green manure, followed by mixed N fertilizer (+192%), synthetic N fertilizer (+157%), and EENF (+128%). When pooling the data from different synthetic N fertilizer species, soil NH_3 emissions had the largest response to the combined use of urea with ammonium nitrate (UAN, +200%), followed closely by urea alone (U, +170%), and compound fertilizer (CF, +166%). Compared to conventional N fertilizer forms, the use of EENF significantly reduced soil NH_3 emissions, to the largest extent (42%) for use of slow-release N fertilizer (Figure 3b). Nitrification inhibitors had the least mitigation effect on soil NH_3 emissions after the application of N fertilizers (Figure 3b).

3.4 | Global and regional soil NH_3 emissions from agricultural N fertilizer use

We estimated global soil NH_3 emissions from synthetic and manure N application in agriculture using the region-specific and global constant NH_3 EFs of N fertilizer synthesized in this study (Table 1; Figure 4a). Overall, global NH_3 emissions from the application of synthetic N fertilizer using region-specific EFs were estimated to be 12.32 Tg N/year in 2014, lower than the estimate of 13.71 Tg N/year

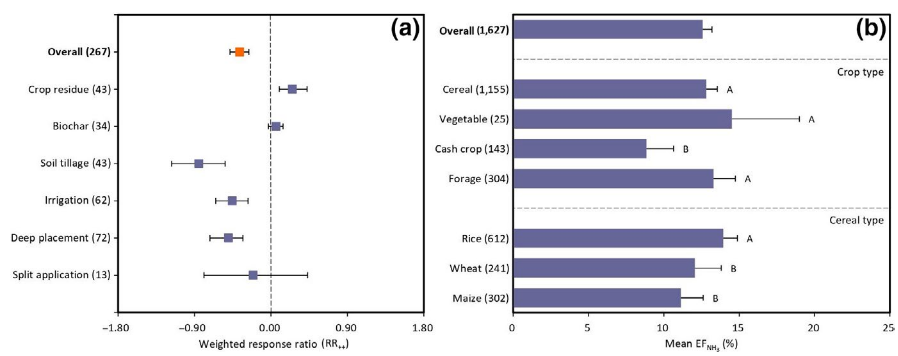

FIGURE 2 Soil NH_3 fluxes in response to experimental factors across ecosystem types (a) and direct emission factors of NH_3 (EFs) partitioned by crop types (b). Symbols in (a) refer to the weighted responses to experimental factors with an interval of 95% confidence. Numerals in the brackets indicate number of measurements. "Overall" refers to the integrated effects of experimental factors as compared with controls without undergoing relevant experimental manipulations. "Deep placement" of N fertilizer was investigated as compared with the control of surface application. "Split application" of N fertilizer was investigated as compared with the control of single application. Different letters in (b) indicate significant pairwise differences in NH_3 emission factors among subgroups within a given category [Colour figure can be viewed at wileyonlinelibrary.com]

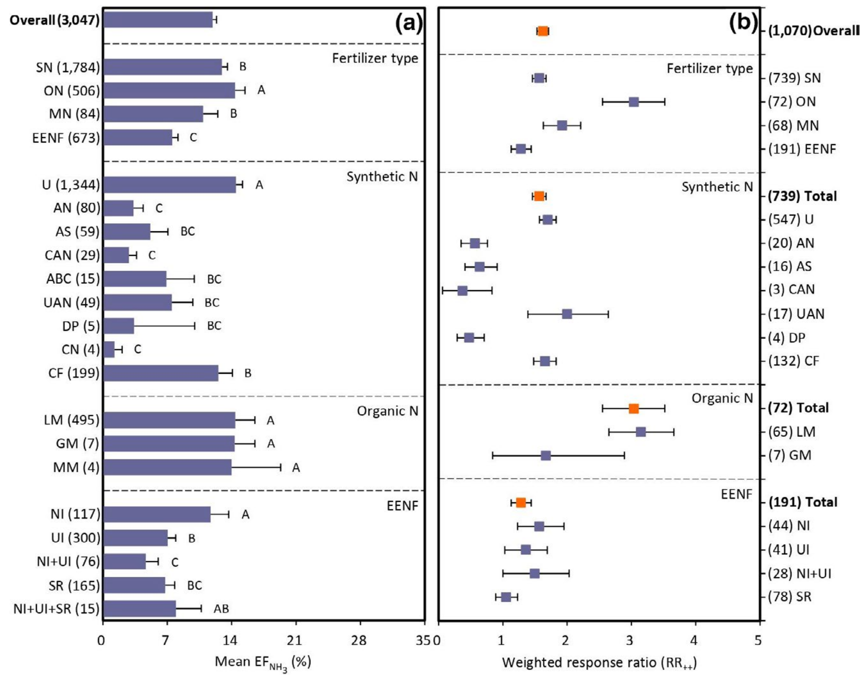

FIGURE 3 Direct NH_3 emission factors of N fertilizer (EFs) partitioned by fertilizer sources (a) and the effectiveness of N fertilizer in driving soil NH_3 emissions (b). Different letters in (a) indicate significant differences at a statistical probability level of .05. Symbols in (b) refer to the mean effect size with an interval of 95% confidence. "Overall" indicates the integrated response across fertilizer sources as compared to controls without fertilizer application. Numerals in the brackets indicate number of measurements. ABC, ammonium bicarbonate; AN, ammonium nitrate; AS, ammonium sulfate; CAN, calcium ammonium nitrate; CF, compound fertilizer; CN, calcium nitrate; DP, diammonium phosphate; EENF, enhanced-efficiency N fertilizer; GM, green manure; LM, livestock manure; MM, mixed manure means mixed livestock manure and green manure; MN, mixed N fertilizer; NI indicates synthetic or organic N fertilizer combined with nitrification inhibitor application; ON, organic N fertilizer; SN, synthetic N fertilizer; SR, slow release fertilizer; U, urea; UAN, urea ammonium nitrate; UI indicates synthetic or organic N fertilizer combined with urease inhibitor application [Colour figure can be viewed at wileyonlinelibrary.com]

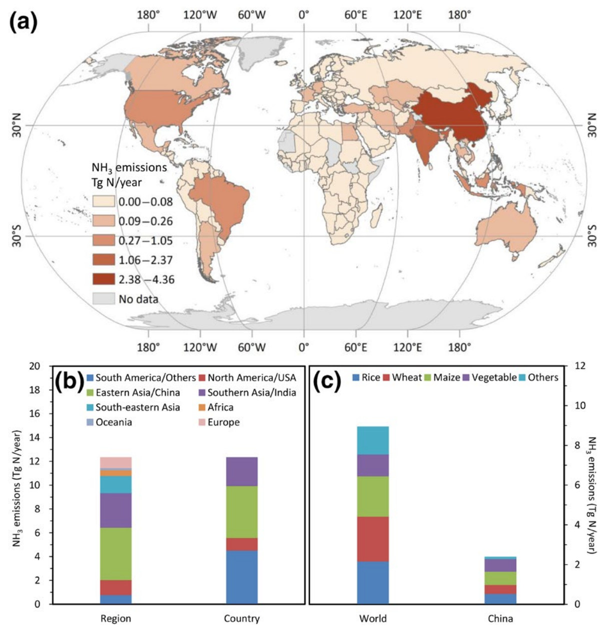

FIGURE 4 Global and regional patterns of soil NH_3 emissions (a, b) and crop-specific NH_3 emissions (c) from agricultural synthetic N fertilizer application estimated by respective case-specific emission factors (EFs) established in this study. All the estimated results are from cropland soils, without including those from grassland and forest with N fertilizer application [Colour figure can be viewed at wileyonlinelibrary.com]

derived from the constant EF established in this study, but substantially higher than the total emissions (3.79 Tg N/year) induced by manure N application globally (Table 1).

Soil NH_3 emissions from synthetic N fertilizer were further examined among regions and countries (Table 1; Figure 4a,b). The Northern Hemisphere dominated global NH_3 emissions from the application of synthetic N fertilizer, especially in Asia, where the estimated rates were as high as 4.39, 2.91, and 1.44 Tg N/year in eastern, southern, and south-eastern Asia, with a contribution of 36%, 24%, and 12% to the global total emissions, respectively. North America and Europe, second only to Asia, contributed to 10.3% and 7.1% of the global total NH_3 emissions, respectively. At the country scale, China (4.20 Tg N/year) among all the countries contributed most to global NH_3 emissions from synthetic N fertilizer input, followed by India (2.37 Tg N/year) and America (1.05 Tg N/year). Soil NH_3 emissions from these three leading countries accounted for over 50% of the global total emissions (Figure 4b).

3.5 | Cropland soil NH_3 emissions from synthetic N fertilizer use

By using crop-specific EFs, we estimated soil NH_3 emissions from the use of synthetic N fertilizer in cereal and vegetable crops in

2014 (Figure 4c). Globally, the four leading crops (i.e., rice, wheat, maize, and vegetable) together emitted 7.55 Tg N/year NH_3, accounting for 85% of the global cropland NH_3 emissions (8.93 Tg N/year) induced by the use of synthetic N fertilizer. In China, upland maize and vegetable production together contributed to more than 50% of the total national cropland NH_3 emissions (2.38 Tg N/year), followed by rice (0.54 Tg N/year) and wheat (0.45 Tg N/year). Compared to maize and wheat cropping systems that were hotspots of NH_3 emissions at the global scale, vegetable together with maize cultivation dominated the total national NH_3 emissions in China.

4 | DISCUSSION

4.1 | Soil background NH_3 emissions depending on ecosystem type

As highlighted in this synthesis, grasslands had the largest background emission of NH_3, which was largely attributed to the stray inputs of livestock manure and urine by herbivores that increased soil ammonium concentration and thereby stimulated NH_3 emissions (Liu et al., 2017). In cropland systems, rice cultivation served as the leading background source of NH_3 emissions, where high temperature and waterlogging could be the main controls that drive NH_3 emissions from rice paddies (Bouwman et al., 2002; Xu et al., 2019). Moreover, the data on NH_3 emissions from grasslands and rice paddies as included in this study were overwhelmingly measured in subtropical and tropical regions that have been documented to be most impacted by N deposition (Song et al., 2020), which also accounted for their role as hotspots of NH_3 emissions due to increased soil deposited N input. Interestingly, compared with tilled cropping soils, no-till cropping soils had higher background NH_3 emissions, as evidenced by the decreased NH_3 emission rates that were found after soil tillage in this analysis (Figure 2a; Table S4). Compared with no-till soils, tillage events may increase soil contact with the surface of clays that adsorbs NH_3, leading to the reduced NH_4^+-N availability in soils, and thereby decreases NH_3 emissions (Bacon & Freney, 1989; Fox & Piekielek, 1993; Grandy et al., 2006). In addition, the greater soil NH_3 emissions in no-till cropping uplands might also be associated with the slightly higher pH environments created by the decomposition of crop residues at the surface of soils (Vanzolini et al., 2017).

4.2 | Soil NH_3 emissions depending on N fertilizer type and species

Both linear and nonlinear positive dependences of soil NH_3 emissions on N fertilizer input have been routinely documented in previous studies (Cui et al., 2014; Jiang et al., 2017; Yan et al., 2003). Compared to mixed or synthetic N formulations, organic N fertilizers were most effective at stimulating soil NH_3 emissions, generally in agreement with other field observations (Hou et al., 2007). This suggests that, relative to the use of synthetic N fertilizer, organic N fertilizer input tends to increase the risk of N loss through NH_3 volatilization. When grouping data by different synthetic N fertilizer types, urea-based fertilizers increased soil NH_3 emissions to the largest extent among all the synthetic N fertilizers. This was partially attributed to the rapid hydrolysis of urea, leading to a localized increase in pH that stimulates soil NH_3 emissions (Bittman et al., 2013). Compared with the conventional N fertilizers, the use of EENFs showed a promise to decrease soil NH_3 emissions, with the slow-release N fertilizers decreasing the emissions to the largest extent. Several reasons might account for the mitigation benefits of EENFs. For example, the use of slow-release N fertilizers delays the availability of N in soils for microbial and enzymatic uptake and utilization, resulting in an extended period that prolongs N availability to crops (Sommer et al., 2004). As proposed by Halvorson et al. (2014), the combined use of urease inhibitor with fertilizer N application is a viable option to reduce soil N losses through gaseous emissions. However, nitrification inhibitors together with N fertilizer use have no evident mitigation effect on NH_3 emissions (Chen et al., 2008; Xia, Lam, Chen, 2017; Xia, Lam, Wolf, 2017). Nitrification inhibitors are designed to block the process of NH_3 oxidation and therefore reduce N_2O emissions and nitrate leaching, but may indirectly promote soil NH_3 volatilization, and subsequently contribute to an increase of indirect N_2O emissions from land surface deposited NH_3 (Lam et al., 2017; Wang et al., 2020).

4.3 | Direct NH_3 EFs varying with ecosystem type and N fertilizer source

The documented direct EFs of N fertilizer for NH_3 have large uncertainties, and remain highly empirical, depending on climate, soil conditions, and site-specific management practices (Pan et al., 2016; Sommer et al., 2004). The direct NH_3 EFs of synthetic N fertilizer significantly differed among ecosystem types and N fertilizer forms (Figures 1b and 3a). On average, the direct EFs of synthetic N fertilizer were significantly greater in forests than in grasslands or croplands. Rice relative to upland cultivation (i.e., wheat and maize) had the largest EF (Figure 1b). Non-cropland soils had an overall greater NH_3 EF of synthetic N fertilizer than in cropland soils (Figure 1b). The greater EFs in non-cropland soils relative to cropland soils in this analysis suggest that fertilizer-introduced soil N has a higher risk of being lost as NH_3 from soils with less human impact, especially in temperate regions (Raymond, 2016). Presumably, N fertilizer applied into non-cropland soils is often subject to less management practices, leading to lower nitrification due to reduced soil aeration for limited oxygen availability, less soil adsorption through contacting, and thereby more soil NH_4^+ availability for NH_3 emissions (Pan et al., 2016; Singh et al., 2013). The earlier IPCC default value of 10% is lower than 12.56% derived from the present analysis (De Klein et al., 2006). The higher constant EF at the global scale in this study partially benefited from the larger sample size of the updated database by including N-managed grasslands and forests with greater fertilizer-induced NH_3 emissions that have been rarely examined in previous studies.

Direct NH_3 EFs also significantly varied with N fertilizer types (Figure 3a). Compared to synthetic N fertilizer (12.95%), manure N sources had significant greater EFs (14.38%), with comparable EFs between livestock (14.39%) and green manure (14.32%). These estimated EFs are similar to those for synthetic N fertilizer (14.00%) but were lower than animal manure (23.00%) estimated by Bowman et al. (2002). In addition to field manure application, however, the stray manure N input due to grazing in grassland and forest was also taken into account in the global estimation by Bowman et al. (2002), which is actually beyond the focus of this study. However, the EF (10.90%) for manure N was relatively lower when it was applied in combination with synthetic N fertilizers. The combined use of manure and synthetic N fertilizers likely stimulated microbial immobilization of N, promotes crop N uptake, and delays hydrolysis of urea, all of which may result in high N use efficiency and less N losses as NH_3 (Sommer et al., 2004; Ti et al., 2019). Among synthetic N fertilizers, urea had the highest EF (14.47%), and calcium nitrate had the lowest (1.25%), indicating that NH_3 losses were most responsive to the use of urea due to its formation of localized higher pH hotspots (Watson & Kilpatrick, 1991). The use of EENFs significantly decreased the EF as compared with conventional N fertilizer forms, to the largest extent for the combined use of nitrification and urease inhibitors.

4.4 | Global, regional and cropland soil NH_3 emissions from synthetic N fertilizer

Globally, the estimated soil NH_3 emissions from synthetic N fertilizers were significantly greater than those derived from manure N input. The use of manure N fertilizers may increase in the future as a conservation practice to promote soil health (Liu et al., 2017), suggesting a potentially intensified global impact of NH_3 emissions from manure N input due to its high direct EF as observed in this analysis. We compared our estimates with previous studies using constant EFs and modeling approaches at the global and regional scales (Table S6). The results showed that our global estimate of NH_3 emissions (12.32 Tg N/year) from synthetic N fertilizer based on region-specific EFs was generally comparable to the estimates of 12.00 and 13.60 Tg N/year that were generated using process-based models by Riddick et al. (2016) and Xu et al. (2019), respectively. However, our estimates based on region-specific EFs were greater than the EF-derived estimates reported in other studies (Table S6). This could be mainly attributed to the differences in the development of datasets for synthetic N fertilizer usage, rather than the estimate methodology used in these studies. The previous estimates using the IPCC default or constant EFs without considering regional discrepancy might have underestimated global NH_3 emissions (Xu et al., 2019).

By adopting the direct EFs of N fertilizer for nitrous oxide (N_2O) and nitric oxide (NO) from a recent worldwide assessment (IPCC, 2019; Liu et al., 2017), we obtained a full accounting of the overall global reactive gaseous N losses ($NH_3 + N_2O + NO$) from the use of synthetic and manure N fertilizers, which were estimated to be 13.86 and 4.87 Tg N/year in 2014, respectively. Using the IPCC default EF of 1% for indirect N_2O emissions from NH_3 volatilization and deposition (De Klein et al., 2006), the losses of NH_3 estimated in this study for synthetic and manure N fertilizers were equivalent to 0.11 and 0.04 Tg N/year indirect N_2O emissions, or 51.81 and 18.76 Tg/year carbon dioxide equivalent (CO_2-e), respectively.

At the country scale, China acted as the leading contributor of global NH_3 emissions and its emission role has been further constrained using the updated China-specific dataset in this study. Our estimate of NH_3 emissions from the use of synthetic N fertilizer in China (4.20 Tg N/year) fell within the range of 2.1–4.7 Tg N/year that were estimated for different reference years by other studies using bottom-up approaches. Our estimate was also close to some recent estimates (3.7–4.2 Tg N/year) based on refined EFs and statistical models (Huang et al., 2012; Kang et al., 2016; Xu et al., 2015, 2016; Yan et al., 2003; Zhang et al., 2011, 2017; Zhou et al., 2016). Among these inventories, our estimates of EFs and synthetic N fertilizer-induced NH_3 emissions from upland and rice croplands in China showed better agreement with the results obtained from different statistical models by Zhou et al. (2016). Nevertheless, the slight difference in estimates between our study and other inventories stems primarily from the current constrained EFs derived from robust China-specific database with low uncertainties and the variation of N fertilizer consumption among different reference years.

Based on the crop-specific EFs established in this study, our estimates showed that the four leading crops of rice, wheat, maize, and vegetable together dominated or represented 85% of the total synthetic N fertilizer-induced NH_3 emissions from croplands. Consistent with a recent process-based modeling results reported by Xu et al. (2019), the cultivation of rice, maize, and wheat together consumed more than 50% of the world's synthetic N fertilizer (Zhang et al., 2017), acting as three major NH_3 emission sources that account for nearly 72% of the global agricultural total emissions in this study (Figure 4c). In China, however, vegetable and maize cultivation dominated the agricultural N fertilizer consumption and therefore contributed to the majority of total synthetic N fertilizer-induced NH_3 emissions. Presumably, the intensified vegetable production characterized by excessive N fertilizer input and frequent irrigation in China promotes soil gaseous N losses such as via NH_3 volatilization and denitrification.

4.5 | Implications for mitigating agricultural NH_3 emissions

The role of optimized fertilizer management regime in mitigating NH_3 emissions has been well synthesized in a recent global meta-analysis by Pan et al. (2016). In this meta-analysis (Figure 2a), soil NH_3 emissions were significantly decreased by soil tillage (−85%), irrigation (−46%), and deep placement of fertilizers (−50%), suggesting that these agricultural practices have potential to mitigate NH_3 emissions from croplands. The negative response of soil NH_3 emissions to tillage events was generally associated with improved soil aeration that promoted the nitrification process via increasing oxygen availability, enhanced soil adsorption via contacting, and thereby decreased soil NH_4^+ availability for NH_3 emissions (Liu et al., 2017). Irrigation was found to decrease soil NH_3 emissions, through transporting N fertilizer from soil surface to plant root zone and diluting localized NH_4^+ concentration (Dawar et al., 2011). The deep placement of fertilizers could lower urease activity, reduce NH_4^+ concentration, increase plant N utilization, and consequently decrease soil NH_3 emissions (Liu et al., 2015; Mohanty et al., 1998). In contrast, soil NH_3 emissions were stimulated by crop residue and biochar amendment (Figure 2a). Our results showed a better agreement with a previous meta-analysis study (Xia et al., 2018), which suggested that the stimulation of NH_3 emissions following crop residue return was mainly attributed to the increased soil urease activity and high NH_4^+ availability from crop residue mineralization. For biochar amendment, in addition to its enhanced soil pH, the oxygen-containing functional groups on the biochar surface may adsorb NH_4^+ and decrease its availability (Kastner et al., 2009). Thus, the role of biochar in driving soil NH_3 emissions depends on the balance between its adsorption and liming effects (Xia et al., 2018).

Soil NH_3 emissions depended largely on soil physiochemical properties. Soil NH_3 emissions decreased as the soil texture changes from coarse and medium soils to fine soils (Figure S1). When experimental soils were grouped into acid/neutral (pH ≤ 7) and alkaline soils (pH > 7), soil NH_3 emissions were significantly greater in the alkaline soils than in the acid or neutral soils and linearly increased with soil pH (Figure 4d; Figure S1). Based on the results of threshold analysis for soil nutrients, soil NH_3 emissions showed a trend to be facilitated in soils with high C and N availabilities, had a pronounced positive correlation with soil mineral N content, and were most enhanced in soils with C/N ratios of 6–12 (Figure 5c; Figure S1). Within this C/N ratio range, less NH_4^+ was immobilized by soil microbes compared to that in the soils with high C/N ratios, which therefore

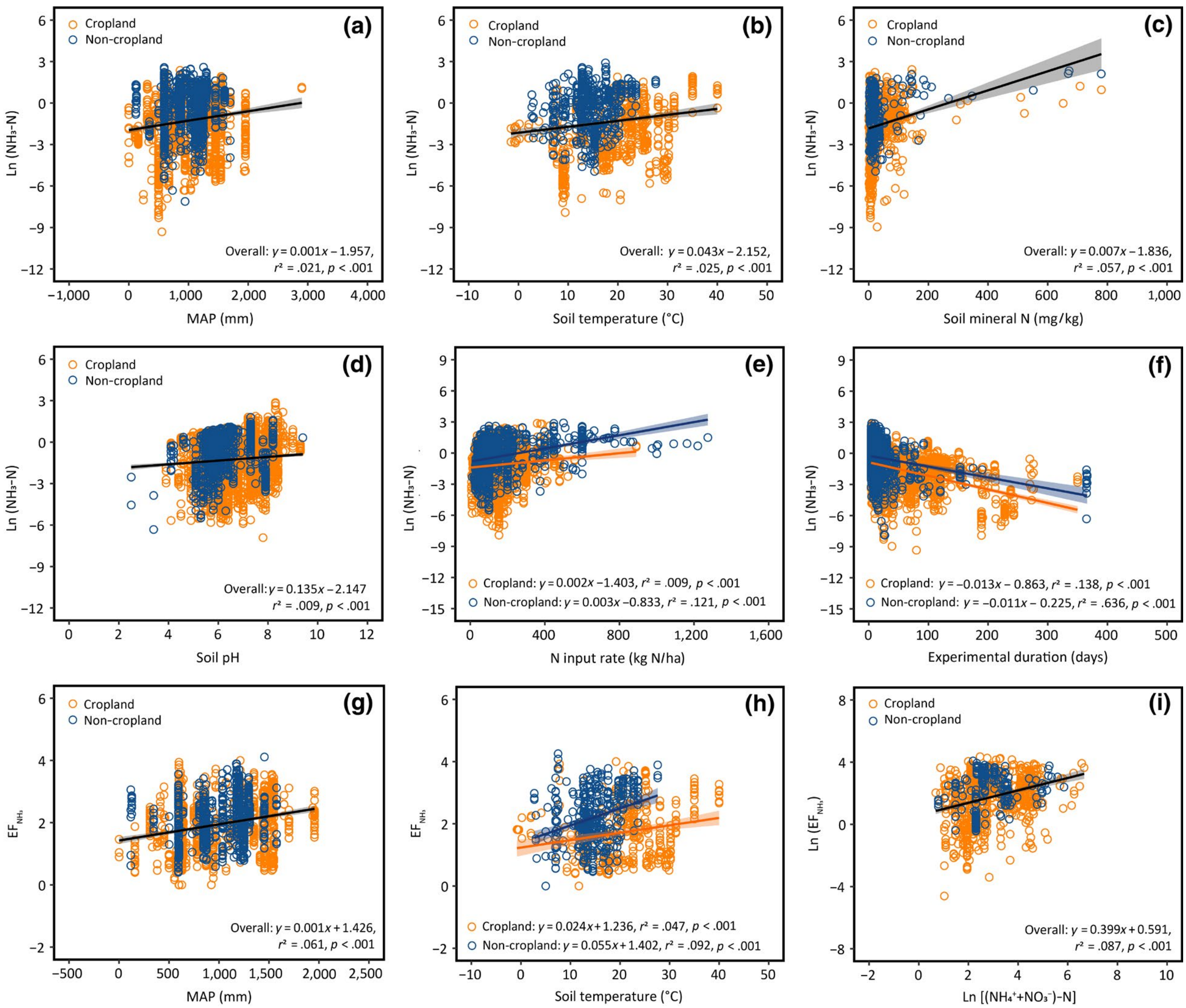

FIGURE 5 Dependence of soil NH_3 fluxes and direct emission factors of N fertilizer for NH_3 (EFs) on soil, environmental, and experimental controls partitioned by agricultural and non-agricultural soils. Mean annual precipitation (a and g), soil temperature (b and h), soil mineral nitrogen (c and i) of the 0–20 cm soil depth, soil pH (d), N input rate (e), and experimental duration (f). Direct emission factors of NH_3 in Figure 4 (g and h) were root-transformed in this analysis [Colour figure can be viewed at wileyonlinelibrary.com]

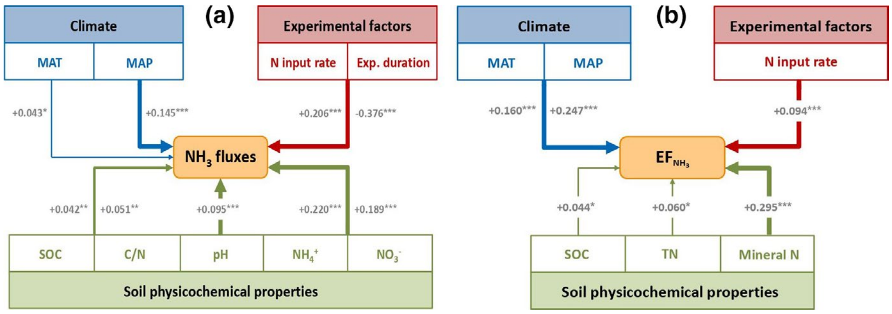

FIGURE 6 A conceptual diagram illustrating soil NH_3 emissions (a) and fertilizer-induced direct emission factors of NH_3 (b) as affected by controlling factors involved in this analysis. Only significant effects were provided in the figure. The arrow width is proportional to the strength of the relationship. The symbols of "*," "**," and "***" following correlation coefficients indicate statistical significance at $p < .05$, .01, and .001, respectively. MAP, mean annual precipitation; MAT, mean annual temperature; SOC, soil organic carbon; TN, total nitrogen [Colour figure can be viewed at wileyonlinelibrary.com]

enhanced the biological pathway that leads to NH_3 production. For a systematic review, an overall schematic response of soil NH_3 and its direct EFs to driving factors is presented in Figure 6.

4.6 | Uncertainties and future research directions

While the efforts have been made to reduce uncertainties in this study, they still exist due to inherent limitations for using the EF-based approach. For example, it was difficult to use EF-based approaches to fully account for the impact of environmental factors and management practices on NH_3 emissions from N fertilizer input. Soil NH_3 emissions were positively correlated with the application rates of N fertilizer, with a more sensitive response occurring in non-cropland soils compared to cropland soils (Figure 5e). This suggests that future studies should give a priority to land use change when examining the factors that affect NH_3 emissions. In addition, soil NH_3 emission rates showed a declining trend with experimental duration, to a larger extent in cropland soils than in non-cropland soils (Figure 5f), suggesting that the annual soil NH_3 emissions might have been overestimated for the field studies where measurements were only conducted in growing seasons. Soil NH_3 fluxes measured using continuous monitoring methods were significantly greater than those using chamber-based methods (static open or closed), suggesting that soil NH_3 emissions were frequency-dependent and largely influenced by the extent to how often the flux peaks were captured. In addition, relative to the use of continuous monitoring methods with low uncertainty, chamber-based methods subject to the influence of multiple environmental parameters and low system stability may have a risk to underestimate N fertilizer-induced soil NH_3 emissions (Yan et al., 2003).

Climate also played a vital role in regulating soil NH_3 emissions (Figure 6; Table S5; Figure S1). Soil NH_3 emissions were positively correlated with soil temperature and MAP (Table S5; Figure 5a,b), consistent with the recent modeling results reported by Xu et al. (2019). Similarly, the EFs of NH_3 linearly increased with MAP and soil temperature (Figure 5g–i). When grouping data by different climate zones, soil NH_3 emissions were the greatest in temperate regions and the lowest in subtropical regions. Tropical soils are generally characterized by low cation exchange capacity (CEC), low organic C content, and low pH, and thereby low NH_3 emissions than in temperate soils. In addition, this study provides the evidence that climate change (e.g., increased temperature) could significantly stimulate NH_3 emissions from managed ecosystems with N fertilizer input. However, few studies have focused on NH_3 emissions under different climate scenarios. Based on the complexity of climate change interactions, multi-factor experimental networks are therefore required to relatively and more accurately predict NH_3 emissions in the future global change contexts.

In conclusion, this study has concentrated on the estimation of N fertilizer-induced soil NH_3 emissions at the regional and global scales. We quantified the global direct NH_3 EFs from the use of both synthetic N fertilizer and manure. The regional synthetic N fertilizer-induced NH_3 EFs showed a large spatial variation, with the highest EF observed in south-eastern Asia and the lowest EF occurring in Europe, depending on the regional discrepancy in the form and rate of N fertilizer use and management practices in agricultural systems. China followed by India and America together contributed to over half of the total global agricultural NH_3 emissions from the use of synthetic N fertilizer. The three main staple crops (i.e., maize, wheat, and rice) together dominated the global total of synthetic N fertilizer-induced NH_3 emissions from croplands. Our results would offer a reference to update the default EF in the IPCC Tier 1 guideline. This study also provides an insight into the spatial variation of soil-derived NH_3 emissions from the use of synthetic N fertilizer in agriculture at the global and regional scales.

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CONFLICT OF INTEREST

The authors declare no competing interests.

AUTHOR CONTRIBUTION

S.L., R.M., and J.Z. initiated and led the research. S.L. and J.Z. designed the investigation. R.M. and K.Y. extracted the data from literature and constructed the database. S.L., R.M., and S.W. performed the statistical analyses. S.L., R.M., and Z.L. created the figures and wrote the paper. All authors provided feedback and contributed to improving and finalizing the manuscript.

DATA AVAILABILITY STATEMENT

All data used for this study are freely available from the corresponding author upon reasonable request.

ORCID

Jianwen Zou https://orcid.org/0000-0003-4689-9237
Shuwei Liu https://orcid.org/0000-0001-8000-0753
Shuli Niu https://orcid.org/0000-0002-2394-2864

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SUPPORTING INFORMATION

Additional supporting information may be found online in the Supporting Information section.

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