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Improved ammonia emission inventory of fertilizer application for three major crops in China based on phenological data

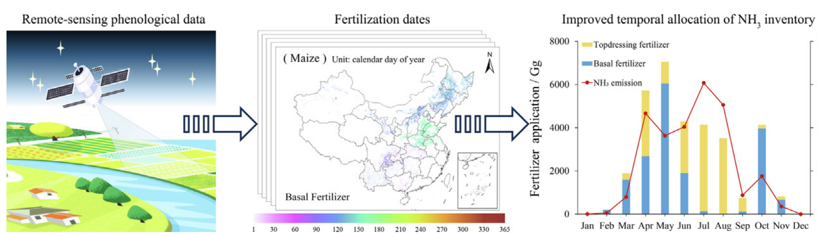
Yongqi Zhao, Baojie Li *, Jinyan Dong, Yan Li, Xueqing Wang, Cong Gan, Yingzhen Lin, Hong Liao

Collaborative Innovation Center of Atmospheric Environment and Equipment Technology, Jiangsu Key Laboratory of Atmospheric Environment Monitoring and Pollution Control, School of Environmental Science and Engineering, Nanjing University of Information Science & Technology, Nanjing 210044, China

HIGHLIGHTS

- A phenology-based method was introduced to establish the NH_3 emission inventory.
- Significant temporal variation in fertilizer application dates was found in China.
- Fertilization dates cover almost the whole year (11–360 calendar day of year).
- 85.91 % of NH_3 emissions occurred between April and August.
- High fertilizer application generally coincided with high NH_3 emission.

GRAPHICAL ABSTRACT



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ABSTRACT

NH_3 has an important impact on atmospheric chemistry, and its reduction has become a potential pathway to alleviate haze pollution. The existing NH_3 emission inventories still have significant uncertainties in terms of their temporal distributions. In this study, we combined satellite remote-sensing phenological data with ground-station phenological data to develop a method for the temporal allocation of NH_3 emissions from fertilizer application. A high-resolution dataset for fertilizer application in China was established. We developed NH_3 emission inventories for the fertilization of three major crops in China, with a resolution of $1/12^{\circ} \times 1/12^{\circ}$. The results showed that there was a significant temporal variation in fertilizer application dates across the country, mainly concentrated in June (17.16 %), July (19.08 %), and August (18.77 %). The majority of fertilizer application for the three major crops occurred during the spring and summer months, with a particular emphasis on April (5.72 Tg), May (7.05 Tg), and June (4.29 Tg). The total NH_3 emission from the three major crops in China in 2019 was 2.73 Tg. The North China Plain (762.23 Gg) and Middle and Lower Yangtze River Plain (606.85 Gg) were identified as the primary regions for high NH_3 emissions from fertilizer application. The results also showed that NH_3 emissions from the three major crops were predominantly observed during summer, with a peak value in July (606.99 Gg), mainly because of the high proportion of topdressing fertilizers. Areas with high fertilizer application generally coincided with areas of high NH_3 emissions. This study may be the first to utilize remote-sensing phenological data to establish the NH_3 emission inventory, which is of great significance for further improving the accuracy of the NH_3 emission inventory.

1. Introduction

Ammonia (NH_3) is a highly reactive gas in the atmosphere and the major alkali gas that plays an important role in atmospheric chemistry (Fowler et al., 2013). NH_3 reacts with acidic pollutants such as SO_2 and NO_x to form fine particulate matter ($PM_{2.5}$), which contributes to haze pollution (Backes et al., 2016; Fu et al., 2017). Deposition of NH_3 and NH_4^+ can

Abbreviations: NH_3, ammonia; $PM_{2.5}$, fine particulate matter; ABC, ammonium bicarbonate; DAP, diammonium phosphate; NPK, compound fertilizers; CEC, soil cation exchange capacity.

* Corresponding author.
E-mail address: baojieli@nuist.edu.cn (B. Li).

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also lead to many other environmental problems, such as eutrophication of water bodies, loss of biodiversity, and soil acidification (Liu et al., 2022; Paerl et al., 2014). A significant increase in NH_3 leads to further loss of biodiversity and accelerated production of sulfate particles (Kirkby et al., 2011; Meng et al., 2018). The relationship between ammonia and climate change is intricate and multifaceted. The amount of NH_3 emissions is greatly affected by temperature and precipitation (Sutton et al., 2013; Ren et al., 2021), while NH_3 also plays an important role in climate change by aerosol generation, nitrogen cycling, and nitrogen deposition (Bellouin et al., 2011; Kanter, 2018; Ornes, 2021). It is clear that the increase in NH_3 emissions will not only exacerbate China's environmental problems but will also have far-reaching impacts on climate change. Therefore, it is important to conduct an in-depth study on NH_3 emissions.

China has launched a series of emission reduction policies aimed at pollutants such as particulate matter, SO_2, NO_x, and VOCs since the issuance of the Air Pollution Prevention and Control Action Plan in 2013; however, the control of NH_3 emissions has not received sufficient attention (Chen et al., 2020). Currently, the NH_3 emission reduction has become more important to alleviate haze pollution (Liu et al., 2019; Zhang et al., 2022a; Zheng et al., 2022). Mitigating $PM_{2.5}$ pollution through NH_3 reduction has been proved to be cost-effective. The global marginal abatement cost of ammonia emissions is only 10 % of that for nitrogen oxide emissions. (Gu et al., 2021, 2023). About 90 % of NH_3 emissions in China come from agricultural sources, of which 30–40 % come from nitrogen fertilizer application (Huang et al., 2012; Zhang et al., 2018). According to recent studies by Li et al. (2021) and Wang et al. (2021), a large amount of NH_3 emissions from agricultural activities in China are highly correlated with severe air pollution, contributing to haze pollution. Additionally, the emissions of NH_3 from agricultural sources are important contributors to $PM_{2.5}$ in the atmosphere. Agricultural NH_3 contributed 29 % of secondary inorganic aerosols and 16 % of $PM_{2.5}$ in 2015 (Han et al., 2020). Reducing agricultural NH_3 emissions is not only important for reducing air pollution and improving the environment and human health but also has a positive effect on improving crop yields and reducing farming costs (Zhang et al., 2020). The heavy use of chemical fertilizers in agricultural production has already caused environmental damage in China (Kanter et al., 2019; Li et al., 2022; Li et al., 2017a). However, the spatial and temporal distribution of NH_3 emissions from fertilizer application is difficult to determine, which poses difficulties for NH_3 reduction and urgently needs to be addressed. Improving the accuracy of the NH_3 emission inventory for fertilizer application can aid in identifying the high-resolution spatial and temporal characteristics of NH_3 emissions, which is vital for developing effective NH_3 emission reduction measures and promoting air pollution control (Zhong et al., 2021).

Many scholars have improved the NH_3 emission inventory for fertilizer application in China. The Emission Database for Global Atmospheric Research (EDGAR) (Olivier and Berdowski, 2001) showed significant spatial variation in NH_3 emissions in China. However, it did not consider the monthly variations in NH_3 emissions. Zhang et al. (2011) collected county-level fertilizer application amounts in China and constructed a county-level NH_3 emission inventory for fertilizer application, which improved the spatial accuracy of the NH_3 emission inventory for fertilizer application in China. The MASAGE_NH_3 inventory (Paulot et al., 2014) utilized a top-down approach to optimize the spatiotemporal allocation of China's NH_3 emission inventory, based on which a new temporal allocation method for fertilizer application was established using global survey data. Some scholars have also introduced the Community Multi-scale Air Quality model (CMAQ model) and the Environmental Policy Integrated Climate model (EPIC agroecosystem model) to estimate NH_3 emissions from fertilizer application, which improved the spatial and temporal resolution of the NH_3 emission inventory in China (Fu et al., 2015). Huang et al. (2012) and Kang et al. (2016) considered the effects of wind speed, temperature, soil acidity, fertilization method, and fertilizer application rates to optimize NH_3 emission factors for fertilizer application. Li et al. (2021) improved fertilizer application dates in China by integrating crop reports and technical guidelines for crop management in each province, which optimized the temporal accuracy of the fertilizer application NH_3 emission inventory. Based on previous studies, substantial progress has been made in establishing an NH_3 emission inventory for fertilizer applications.

Uncertainties remain in estimates of NH_3 emissions according to the results of previous studies (Li et al., 2021; Wang et al., 2021; Zhang et al., 2017). There are also large differences in the temporal allocation methods of previous NH_3 emission inventories for fertilizer application, which is an important source of uncertainty. Temporal allocation methods remained at the provincial scale. However, most existing studies have focused on improving the emission factors rather than on improving temporal allocation methods. In most previous studies, fertilizer application dates were assigned to the same months in each province, neglecting the variability between individual grids, which does not reflect reality and may not provide accurate results. This is not conducive to improving the accuracy of the temporal allocation of the NH_3 emission inventory for fertilizer application, thereby hindering the development of NH_3 emission inventories and limiting the simulation accuracy of atmospheric transport models.

Accurate information on the phenological periods of major crops determines the agronomic management options (Chen et al., 2018; Sakamoto et al., 2013). Crop-specific phenology is a crucial factor in determining the appropriate dates of fertilizer application (Li et al., 2019; Rurinda et al., 2020; Shi et al., 2021). Therefore, it is feasible to utilize key phenological information of crops to improve the temporal accuracy of NH_3 emission inventory. For example, there is a high demand for nutrients during the seeding, tillering, and spike differentiation periods of rice growth. As a result, these three periods are typically targeted for fertilizer application during planting (He et al., 2022; Liu et al., 2020; Zhang et al., 2022b). By establishing a relationship between key phenological periods and fertilizer application dates, it is possible to improve the temporal allocation of NH_3 emissions from fertilizer application and establish high-resolution NH_3 emission inventories.

In this study, we utilized crop phenological data to generate daily-scale fertilizer application dates for the three major crops in China (i.e. maize, rice, and wheat) to optimize the temporal allocation of fertilizer-source NH_3 emission inventories. The main objectives of this study were: (1) to determine the daily-scale fertilizer application dates in China at a raster resolution of 1/12° using phenological data; (2) establish a database of fertilizer application amounts for the three major crops in China at a spatial resolution of 1/12°; (3) develop a high-resolution NH_3 emission inventory for the three major crops in China. This study aimed to improve the accuracy of NH_3 emission inventories to improve the simulation accuracy of atmospheric chemistry models.

2. Data and methods

This study used a 'bottom-up' approach to estimate NH_3 emissions from fertilizer application for three major crops in mainland China: maize, wheat, and rice. Gridded NH_3 emissions (E_{NH3}) were calculated using Eq. (1).

$$E_{NH_3} = \Sigma_i \Sigma_j \Sigma_k A_{i,j,k} \times EF_{i,j,k} \tag{1}$$

In Eq. (1), *i, j,* and *k* denote the grid, crop, and month, respectively. *A* denotes the activity level, that is, the amount of fertilizer applied to each crop (see Section 2.1). *EF* denotes the corresponding emission factors, as illustrated in Section 2.2.

2.1. Fertilizer application amounts

In the establishment of emission inventories, the accuracy of the activity level plays a crucial role (Li et al., 2017b). In 2019, the three major crops accounted for 60.51 % of the total fertilizer application amounts in China (NBS – National Bureau of Statistics, 2020a). The variability in monthly emissions is mainly due to differences in fertilizer application (Huang et al., 2012; Wang et al., 2021). In this study, three major crops that are widely grown in China were considered: maize, rice (early, medium, late, and single-season rice), and wheat (spring and winter wheat). Five types of fertilizers were considered, including urea, ammonium bicarbonate (ABC), diammonium phosphate (DAP), compound fertilizers (NPK), and other fertilizers (Other). In addition, every crop was set to be fertilized three times (i.e. basal fertilizer, 1st topdressing fertilizer, and 2nd topdressing fertilizer). The provincial-level fertilizer application amount database for 2019 for the three major crops in mainland China at the provincial level was obtained using planted area data (NBS – National Bureau of Statistics, 2020a) and fertilizer application per unit of planted area (NDRC – National Development and Reform Commission of China, 2020).

2.2. Emission factors

The selection method of emission factors in this study refers to Li et al. (2021) and Zhang et al. (2018). The effects of soil characteristics, agricultural activities, and meteorological conditions were considered (Bouwman et al., 2002; Zhang et al., 2018). Monthly emission factors corresponding to fertilizer application were calculated as follows:

$$EF = EF_0 \times e^{f_{pH}+f_{CEC}+f_{crop}+f_{method}} \times \alpha \tag{2}$$

In Eq. (2), EF_0 represents the baseline emission factor (Cai et al., 2002; Dong et al., 2009; Zhou et al., 2017), and f represents the effect of soil pH, soil cation exchange capacity (CEC), fertilization method (basal or topdressing fertilizer), and crop type (dryland crops or flooded crops) (Bouwman et al., 2002; Zhang et al., 2018). Soil pH and CEC data were obtained from Harmonized World Soil Database v 1.2 (http://www.fao.org/land-water/databases-and-software/hwsd/en/). Detailed f values are provided in Table S1. In addition, α indicates the monthly scalar used to describe the effect of meteorological factors on NH_3 emissions (Gyldenkærne, 2005; Søgaard et al., 2002; Sommer and Hutchings, 2001; Zhang et al., 2018).

$$\alpha = \left(e^{0.0223T_i+0.0419W_i}\right) / \left(\frac{1}{12}\Sigma_{i=1}^{12} e^{0.0223T_i+0.0419W_i}\right) \tag{3}$$

In Eq. (3), T_i and W_i represent the 2 m air temperature (°C) and 10 m wind speed (m/s), respectively for a given month i. T and W were processed using the ECMWF ERA5 Reanalysis data (https://www.ecmwf.int/en/forecasts/datasets/reanalysis-datasets/era5).

2.3. Improvement of fertilizer application distribution

A 1 km × 1 km resolution harvesting area dataset of the three major crops (Luo et al., 2020b) was utilized to determine the spatial distribution of fertilizer application in China. We calculated the fertilizer application amount for each grid in each province by considering the percentage of harvesting area on each 1/12° × 1/12° grid and the total fertilizer application amount of the three major crops in that province, as illustrated in Eq. (4). This allowed us to obtain the gridded fertilizer application amounts for each province.

$$A_{i,j} = A_{sum-i,j} \times \frac{N_{i,j}}{N_{sum-i,j}} \tag{4}$$

In Eq. (4), i and j denote the 1/12° (about 9.27 km) grid and crop, respectively. $A_{i,j}$ denotes the amount of fertilizer applied to crop j in grid i. $A_{sum\text{-}i,j}$ denotes the total amount of fertilizer applied to crop j in the province where grid i is located. $N_{i,j}$ denotes the area of crop j in grid i, which was obtained from the 1 km resolution harvesting area dataset (Luo et al., 2020b). $N_{sum\text{-}i,j}$ denotes the total area of crop j in the province where grid i is located.

Many previous studies have assigned fertilizer application dates to specific months without considering the variations between grids. This approach does not reflect the actual situation and may not provide accurate results. In this study, we utilized a high-resolution crop phenological dataset (ver. 7, Luo et al., 2020a) for the three major crops in China. We collected crop phenology reports and technical guidelines for crop management from each province and obtained ground observation phenological data (Dataset of Growth and Development of Major Crops obtained from the China Meteorological Data Service Centre, https://data.cma.cn/) to comprehensively evaluate the dates of fertilizer application for maize, rice, and wheat in different regions. We also considered the differences in the ratio of basal and topdressing fertilizers between different regions based on the farmer survey data (Wang et al., 2008). The specific proportions of basal and topdressing fertilizers are shown in Table S2.

The remote-sensing phenological dataset developed by Luo et al. (2020a) was constructed based on the leaf area index and identified three key phenological periods for each of the three major crops in China. The dataset identified the V3, tassel, and maturity stages for maize; the transplanting, tassel, and maturity stages for rice; the greening, tassel, and maturity stages for wheat. We further corrected the dates of tassel stage in the remote-sensing dataset based on the Dataset of Growth and Development of Major Crops using Eq. (5).

$$H_{i,j}' = T_{i,j} + \frac{M_{i,j} - T_{i,j}}{M_{i,j}^* - T_{i,j}^*} \times \left(H_{i,j}^* - T_{i,j}^*\right) \tag{5}$$

where $H_{i,j}'$ represents the corrected date of the tassel stage obtained from remote-sensing phenological data of crop j in grid i; $T_{i,j}$ is the date of the first phenological stage obtained from remote-sensing phenological data of crop j in grid i, i.e., the V3 stage for maize, transplanting stage for rice, and greening stage for wheat; $T_{i,j}$ represents the date of the first phenological stage of crop j obtained from the nearest ground station of grid i; $M_{i,j}$ and $M_{i,j}$ are the dates of maturity stages of crop j obtained from remote-sensing data in grid i and the nearest ground station of grid i, respectively; $H_{i,j}$ is the date of the tasseling stage of crop j obtained from the nearest ground station of grid i. All variables in the formula are in the unit of "calendar day in 2019."

We studied the crop phenology reports of each province published by the Ministry of Agriculture and Rural Affairs of the People's Republic of China (www.moa.gov.cn) and the technical guidelines for crop management published on the Chinese government website (www.gov.cn). We calculated the mean and standard deviation of the time intervals between key phenological periods and fertilizer application dates (Table S3) to improve the method of the temporal allocation of NH_3 emissions from fertilizer application. As many 1 km grids with different fertilization dates coexist in each 1/12° grid, we allocated the annual fertilizer application amounts to months according to the fertilized area proportion of each month using Eq. (6).

$$A_{i,j,m} = A_{i,j} \times x_{i,j,m} \tag{6}$$

In Eq. (6), i, j, and m denote the 1/12° grid, crop, and month respectively. $A_{i,j,m}$ denotes the amount of fertilizer applied to crop j in month m in grid i. $A_{i,j}$ denotes the annual amount of fertilizer applied to crop j in grid i, which was calculated by Eq. (4). $x_{i,j,m}$ denotes the percentage of area fertilized in month m for crop j in grid i.

3. Results and discussion

3.1. Fertilizer application dates for the three major crops in China

In this study, satellite remote-sensing phenological data and ground observation phenological data, as well as crop management opinions, technical guidelines, and farmer survey data from each province in China, were used to identify the optimal fertilizer application dates for the three major crops. The fertilizer application dates were presented in Fig. 1. The study reveals a substantial temporal disparity in the fertilizer application dates throughout the country, with the majority being concentrated in the months of June (17.16 %), July (19.08 %), and August (18.77 %). The fertilizer application dates in China showed a large spatial variation. Fertilizer application generally occurred 1–2 months earlier in the southern region than in the northern region for the same crop. This difference can be attributed to the significant variations in plant phenological characteristics in different climatic conditions (Wolkovich et al., 2012).

The fertilizer application dates of maize are more variable at high latitudes and shows a gradual advance from the coast to the interior. The basal fertilizer of maize is mainly concentrated in May (54.08 %) and June (28.09 %). The first topdressing fertilizer of maize is mainly concentrated in June (16.05 %), July (62.25 %), and August (19.36 %), while the second topdressing fertilizer is mainly concentrated in July (33.34 %) and August (61.04 %). It is also noteworthy that wheat is highly aggregated. Specifically, the basal fertilizer for wheat is mainly concentrated in October (74.85 %) and November (13.16 %), the first topdressing fertilizer and the second topdressing fertilizer are both concentrated in April, 73.41 % and 80.25 % respectively. The basal fertilizer of rice is mainly concentrated in April (30.41 %), May (20.91 %), and June (33.75 %), the first topdressing fertilizer is mainly concentrated in June (38.94 %), July (24.69 %), and August (26.30 %), and the second topdressing fertilizer is mainly concentrated in August (35.48 %), September (23.70 %), and October (16.63 %). It should be noted that rice cultivation in the southern coastal regions of China is practiced in biennial triplication or even annual triplication, and wheat in China includes both winter and spring wheat, so their fertilizer application dates are very complex and cover almost the whole year (11–360 calendar day of year). The specific fertilizer application dates for the three major crops in each province of China are shown in Table S4, and the daily-scale fertilizer application dates map is shown in Fig. S1.

Previous studies have utilized fixed empirical coefficients to determine the temporal allocation. However, these coefficients are often uniform within a province or among provinces and fail to capture the intricate and dynamic nature of agricultural NH_3 emissions. Hence, a more sophisticated approach is required to accurately describe the high-resolution dynamics of agricultural NH_3 emissions. The method used in this study to allocate fertilizer application dates is a localized "phenology-fertilizer" approach, which makes calculations for each grid individually. This approach is more detailed and accurate than previously used methods.

3.2. Spatial and temporal distribution characteristics of fertilizer application amounts

In this study, we found that there were significant differences in the spatial and temporal distribution of fertilizer application amount in China (Fig. 2). The total amount of fertilizer application from three major crops in China in 2019 was 32.51 Tg. Overall, the fertilizer application in China was mainly concentrated in the North China Plain (8.69 Tg) and the Middle and Lower Yangtze River Plain (7.28 Tg). The fertilizer application amounts in China were mainly concentrated in April (5.72 Tg), May (7.05 Tg), and June (4.29 Tg).

Spatially, Henan (10.60 %) and Shandong (9.19 %), located in the North China Plain, were the two provinces with the largest share of fertilizer application amounts for the three major crops in China. Henan and Shandong are the major provinces in China's crop production, and although they account for only 3.40 % of the total land area of mainland China, they accounted for 47.12 % and 18.34 % of the total national production of wheat and maize, respectively, in 2019 (NBS – National Bureau of Statistics, 2020b). Fertilizer application for wheat was identified as the main source in Henan and Shandong, accounting for 61.53 % and 55.14 % of the total fertilizer application amount in the two provinces, respectively. Fertilizer application was also higher in Heilongjiang, the province with the highest fertilizer application amount for maize in China, accounting for 8.00 % of the national application amount. The northeastern black soil region is an

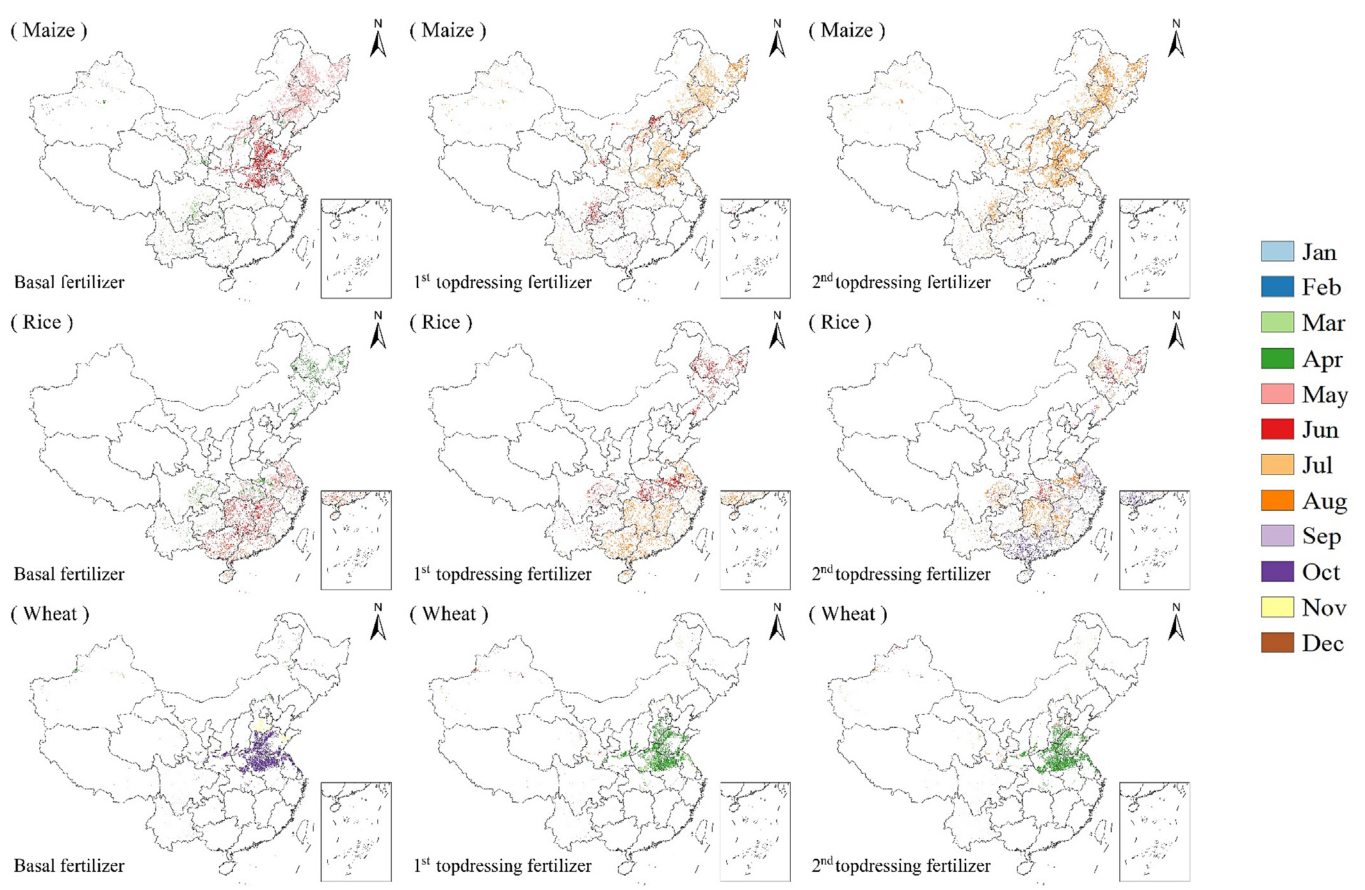

Fig. 1. Months of fertilizer application for maize, rice, and wheat. The three fertilizer applications were basal fertilizer (column 1), first topdressing fertilizer (column 2), and second topdressing fertilizer (column 3).

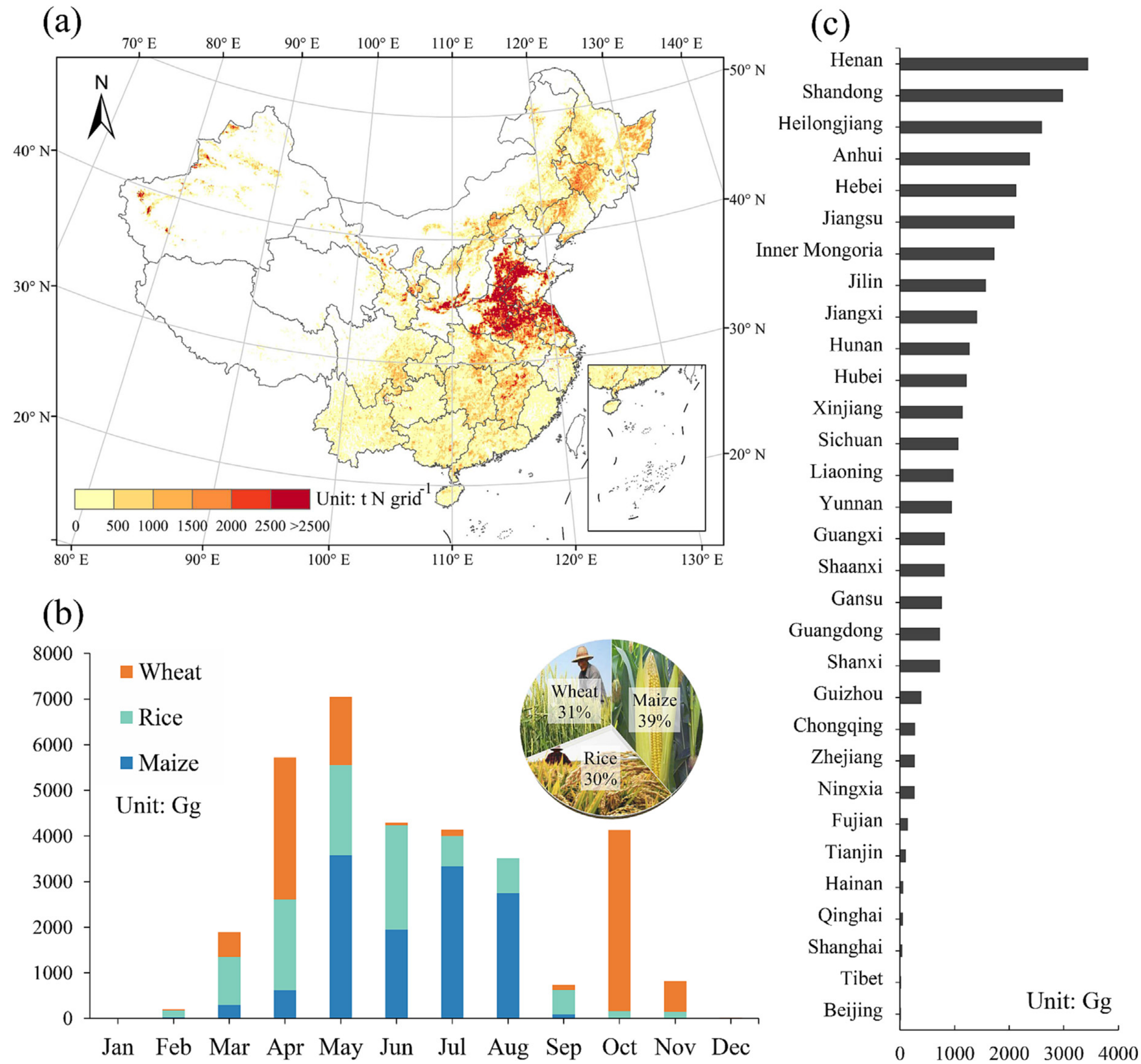

Fig. 2. (a) Total amount of fertilizer used for the three main crops in 2019. (b) Monthly fertilizer amounts applied to three major crops and the fertilizer application proportion of the three crops. (c) Total amount of fertilizer used in each province.

important grain production base in China (Gao et al., 2023), and Heilongjiang is an important province.

Temporally, the distribution of fertilizer application amounts showed significantly lower values in winter and significantly higher values in the months of April to August, with significant peaks in May and October (Fig. 2b). The month with the highest fertilizer application amount for the three major crops in China in 2019 was May (7.05 Tg), which was approximately 2.60 times the amount of the average of 12 months (2.71 Tg), and the month with the lowest fertilizer application amount was January with only 9.12 Gg. This result is in line with China's farming habits, as April and May are the sowing periods for spring maize and spring wheat, and the most important topdressing fertilizer application period for winter wheat, while there is almost no agricultural activity on farmland in winter. The fertilizer application amount proportions of different crops show that maize was fertilized slightly more than the other two crops, accounting for about 39 % of the total amount applied (Fig. 2b).

3.3. Spatial and temporal distribution characteristics of NH_3 emissions

3.3.1. Comparison with previous studies

This study established an NH_3 emission inventory for three major crop fertilizer applications in China in 2019 at a resolution of $1/12^\circ \times 1/12^\circ$. The total NH_3 emission was found to be 2.73 Tg, which is similar to the result compiled by Li et al. (2021), 2.72 Tg. According to Fig. 3, the total NH_3 emissions from fertilizer sources in China range between 3.0 and 7.2 Tg. This study only considered the three major crops, resulting in smaller emission. We used the method proposed by Li et al. (2021) to accurately calculate NH_3 emissions from all crops in 2019 in China and obtained a total amount of 4.5 Tg of NH_3 emissions. This demonstrates the precision of our NH_3 emission inventory. Moreover, our study also boasts a more precise temporal distribution compared to previous studies.

3.3.2. Characteristics of the spatial distribution of NH_3 emissions

The study found that the spatial distribution of NH_3 emissions in China was closely related to the spatial distribution of fertilizer application amount. The areas with high NH_3 emissions were concentrated in the North China Plain (762.23 Gg) and the Middle and Lower Yangtze River Plain (606.85 Gg); this result was consistent with the high-value areas of fertilizer application (Fig. 4a). The specific NH_3 emission results of the three major crops in each province are statistically presented in Table S5. The two provinces with the largest NH_3 emissions in 2019 are Henan Province and Shandong Province, with emissions reaching 308.85 Gg and 263.78 Gg, respectively; meanwhile, the province with the smallest NH_3 emissions is Tibet, with emissions of only 786.60 tons. Henan has the

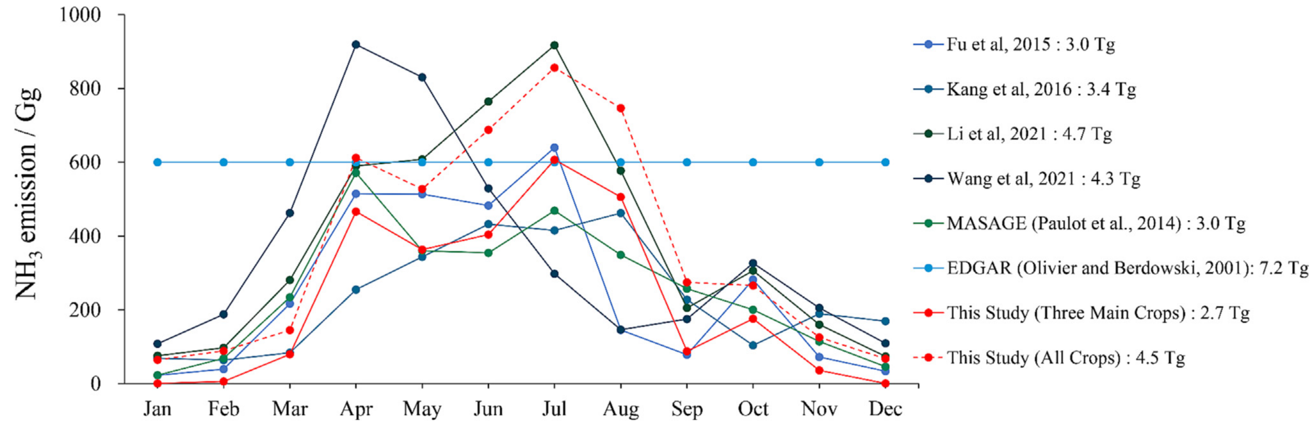

Fig. 3. Comparison of the NH_3 emissions from fertilizers with previous studies.

highest NH_3 emission density at 1.85 t/km^2. It is worth mentioning that Jiangsu's NH_3 emission is only 63.52 % of Henan's, but its emission density is high, reaching 1.83 t/km^2, which is 3.6 times the amount of national average (0.6 t/km^2) and the second highest in the country. The total amount of NH_3 emissions in Jiangsu ranks third in China, reaching 196.18 Gg. Wheat and rice are the largest emission sources in Jiangsu,

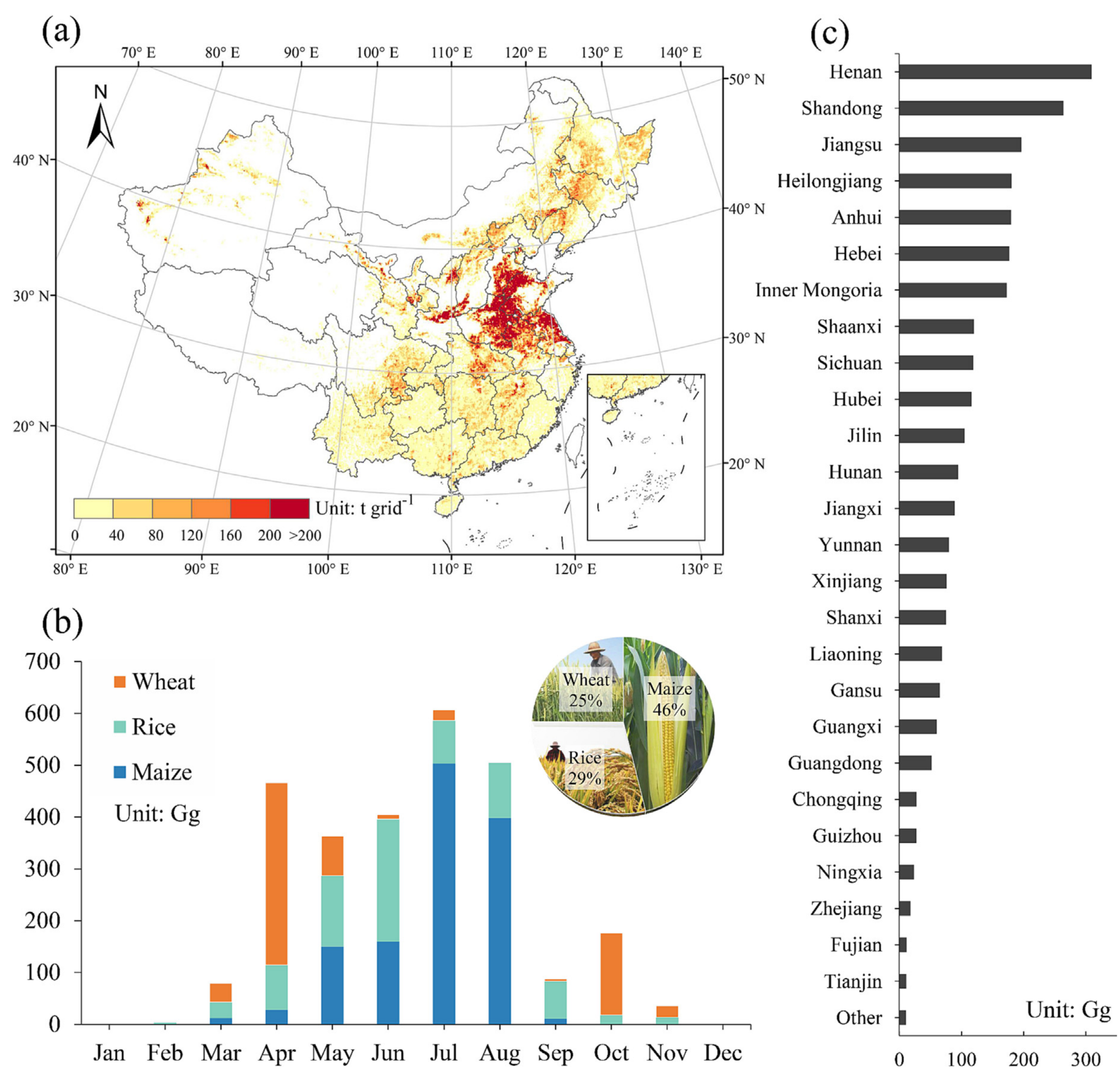

Fig. 4. (a) Total NH_3 emissions from three major crops. (b) Temporal distribution of NH_3 emissions from three major crops and the NH_3 emission proportion of the three crops. (c) NH_3 emission in each province.

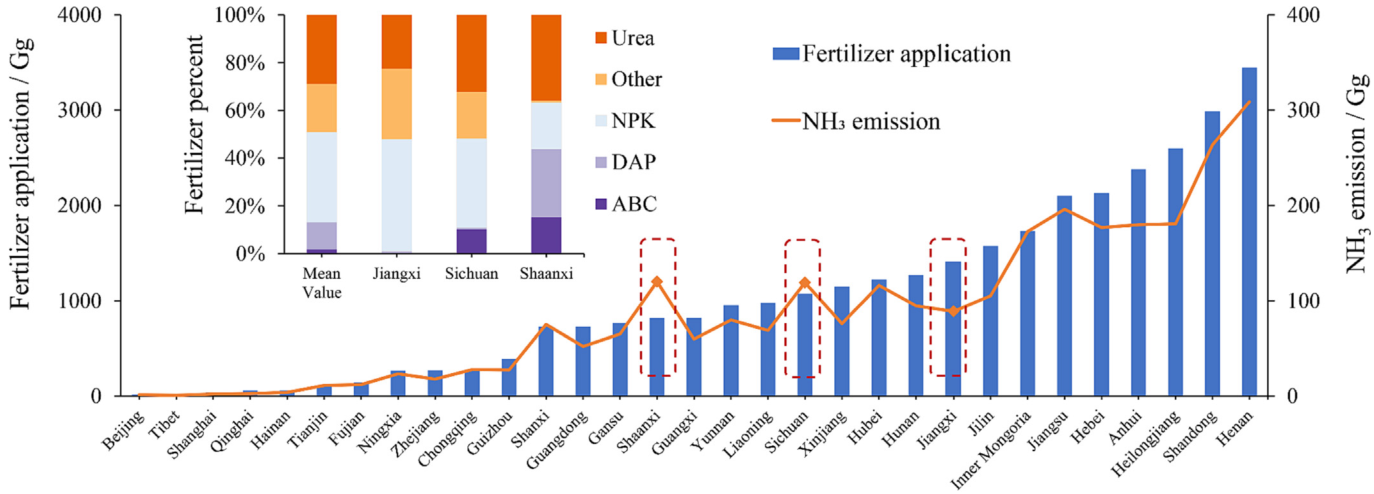

Fig. 5. Relationship between fertilizer application and NH_3 emissions for the three main crops in China, categorized by province. The stacked graph represents the proportion of fertilizer application amount for various types of fertilizers in Jiangxi, Sichuan, Shaanxi, and the national average.

reaching 95.69 Gg and 74.25 Gg, respectively. The distribution of NH_3 emissions from the three major crops is shown in Fig. S2.

A comparison of Fig. 2a and Fig. 4a shows that there are zones that had small fertilizer application amounts with high NH_3 emissions and zones that had large fertilizer application amounts with low NH_3 emissions. The relationship between the amount of fertilizer applied and NH_3 emissions can be clearly demonstrated by organizing the amount of fertilizer applied in each province from smallest to largest (Fig. 5). Anomalously high NH_3 emissions were observed in Shaanxi and Sichuan, whereas anomalously low NH_3 emissions were observed in Jiangxi. The anomalous NH_3 emissions in the three provinces were primarily caused by the varying structures of fertilizer types. Urea and ABC, are significantly more volatile than other commonly used N fertilizers in China (Kang et al., 2016; Klimczyk et al., 2021). The proportions of urea and ABC fertilizers applied in Jiangxi (22.77 %) were significantly lower than the national average (30.69 %), whereas the proportions in Sichuan (42.51 %) and Shaanxi (51.24 %) were significantly higher.

3.3.3. Characteristics of the temporal distribution of NH_3 emissions

The temporal variation of NH_3 emissions is shown in Fig. 4b. NH_3 emissions were distinctly seasonal, with a notable increase during summer and a decrease during winter. Emissions during December, January, and February were almost negligible compared to the other months. The highest values of NH_3 emissions from the three major crops in 2019 occurred in July with 606.99 Gg, followed by August with 505.61 Gg; the lowest NH_3 emission occurred in December with 319.80 t. The peaks in July and August were primarily caused by NH_3 emissions from maize, contributing 82.86 % and 78.76 %, respectively, whereas the high values in April were primarily caused by emissions from wheat, contributing 75.38 %. It is worth noting that the high value of NH_3 emissions from fertilizer application in October was mainly due to wheat, which accounted for 89.74 % of the NH_3 emissions in the month.

A temporal mismatch was observed between fertilizer application amount and corresponding NH_3 emissions for the three major crops in China (Fig. 6). The fertilizer application amounts exhibited a peak in May (7.05 Tg), while lower values were observed in July (4.14 Tg) and August (3.52 Tg). Interestingly, the month of May (363.05 Gg) had minimal NH_3 emission, whereas July (606.99 Gg) and August (505.61 Gg) showed high levels of NH_3 emission. Additionally, low NH_3 emission was observed in October (175.90 Gg), which did not correspond to its high fertilizer application amount (4.13 Tg). The study found high proportions of basal fertilizers in May (85.77 %) and October (96.05 %), when NH_3 emissions were unusually low. Conversely, topdressing fertilizers were predominantly used in July (96.83 %) and August (99.99 %), when NH_3 emissions were unusually high. This mismatch can be attributed to the significantly higher volatilization rate of the topdressing fertilizer than that of the

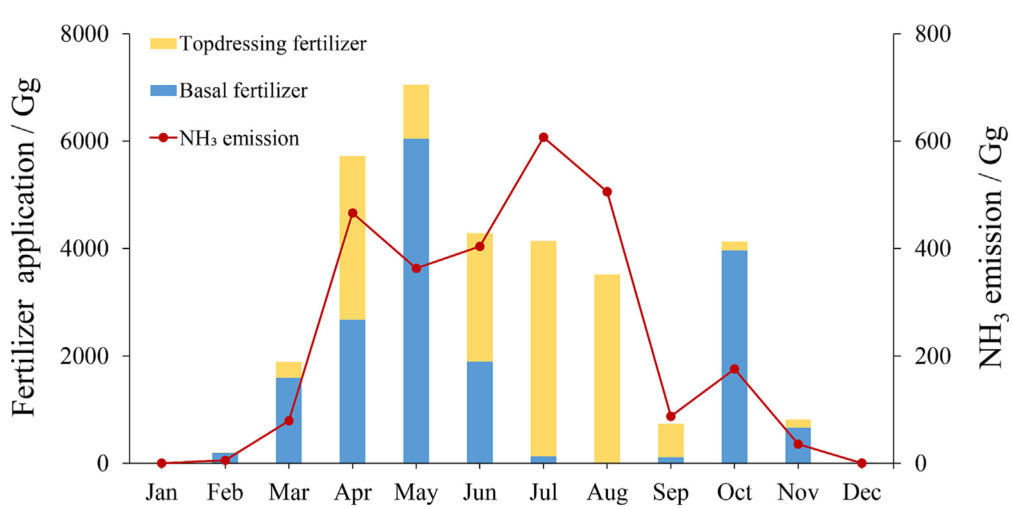

Fig. 6. Temporal distribution of fertilizer application and NH_3 emissions for three major crops.

basal fertilizer (Huang et al., 2012). In addition, high temperatures in summer also contribute to increased NH_3 emissions (Ahmed et al., 2018; Sutton et al., 2013; Yang et al., 2015).

The distribution of NH_3 emissions from fertilizer application in China showed significant temporal differences throughout the year (Fig. 7). During winter in China, NH_3 emissions are minimal in most regions except for some low latitude areas, which aligns with China's farming practices. In other seasons, it can be clearly found that the North China Plain area and the Middle and Lower Yangtze River Plain are the main high value areas. They contribute 50.13 % to total NH_3 emissions, and their emissions are especially high in April (348.08 Gg), July (259.02 Gg) and August (227.74 Gg). Temporally, the increasing trend of NH_3 emissions in these two areas is consistent with the temporal trend of total NH_3 emissions from fertilizer application in China. Therefore, it can be presumed that the peaks of NH_3 emissions from fertilizer sources in China in April, July, and August are mainly attributed to the North China Plain area and the Lower Yangtze River Plain. These two regions are the major cultivation areas of the three major crops (47.56 % of total sown area) in China. The specific NH_3 emissions by month for each province are shown in Table S6.

4. Conclusion

NH_3 is an important part of the nitrogen cycle. There are still many uncertainties in the NH_3 emissions inventory for fertilizer applications. This study aimed to improve the accuracy of the NH_3 emission inventory for fertilizer application by improving the temporal distribution method. In this study, we successfully established daily-scale fertilizer application dates at a spatial resolution of 1/12° by combining satellite remote-sensing phenological datasets and ground-station phenological data, and by exploring the relationship between fertilizer application dates and phenological periods with crop management advice and technical guidelines. Finally, we optimized the temporal allocation method of NH_3 emission inventory for fertilizer application, constructed a fertilizer application amount database for the three major crops in China, and established an improved NH_3

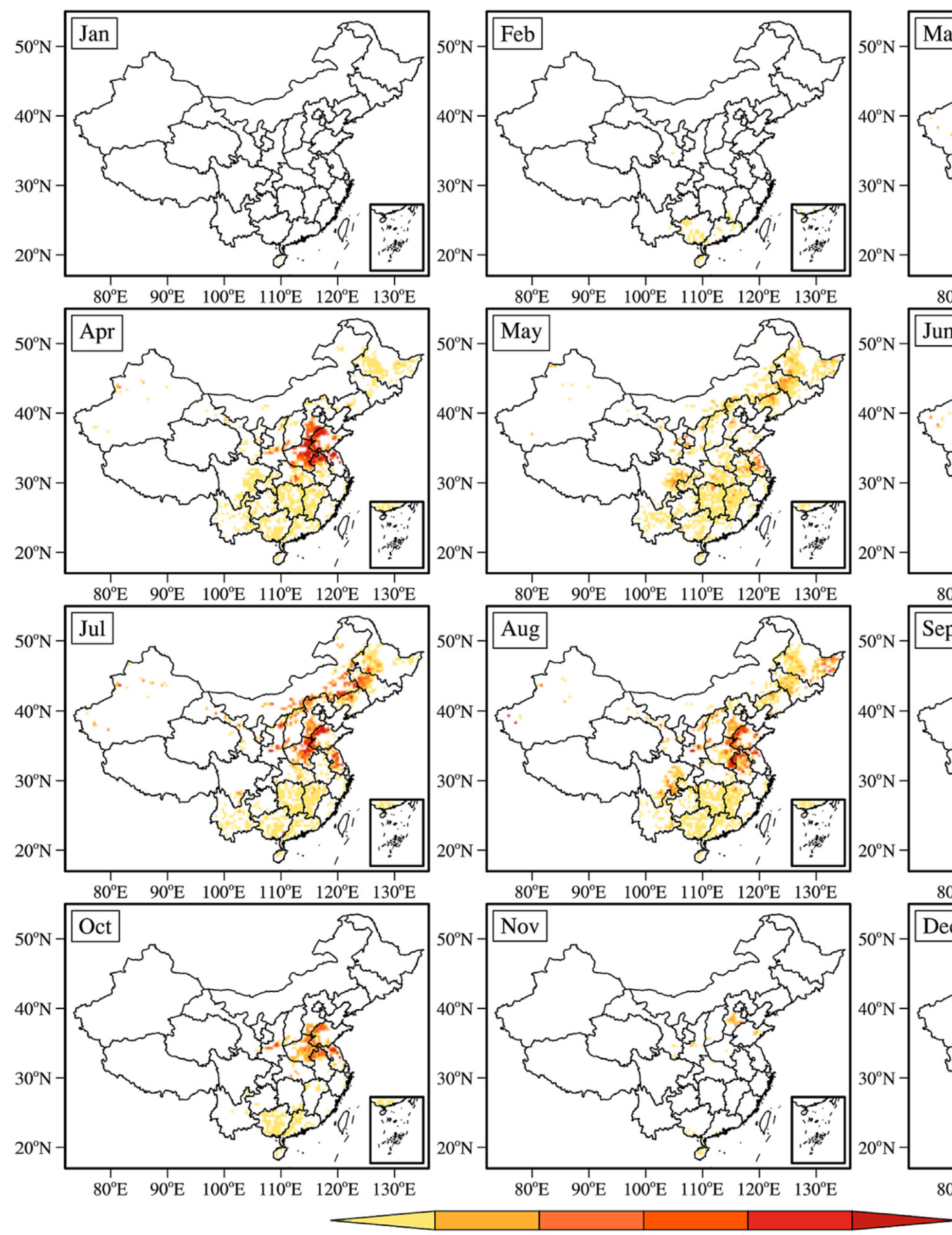

Fig. 7. Distribution of NH_3 emissions from fertilizer application throughout the year.

emission inventory of the three major crops in China in 2019 at a spatial resolution of 1/12°.

In this study, we analyzed the spatial and temporal allocation of fertilizer application dates, fertilizer application amounts, and NH_3 emissions for three major crops in China. The results show that the fertilizer application dates vary greatly across the country, mainly concentrated in June (17.16 %), July (19.08 %) and August (18.77 %). The month with highest fertilizer application amount was May, 7.05 Tg. The total NH_3 emission from fertilizer application for the three major crops in China in 2019 was 2.73 Tg. Significant spatial and temporal differences were found in NH_3 emissions from the three major crops in China. The North China Plain (762.23 Gg) and the Middle and Lower Yangtze River Plain (606.85 Gg) were the main high-value areas of NH_3 emissions, where the fertilizer application amounts were also large. NH_3 emissions from the three major crops in China in 2019 were mainly distributed in summer, with the highest value occurring in July, 606.99 Gg. In most cases, the high fertilizer application and high values of NH_3 emissions in China overlap spatially and temporally. The mismatch between them is mainly due to the proportion of basal and topdressing fertilizers and the different types of fertilizers. The results indicate that the use of urea and ABC should be replaced or properly managed because of their significant volatility, especially in Shaanxi, Sichuan, and Jiangsu. Improved managements such as utilizing urease inhibitors and slow-release fertilizer, deep placement of fertilizer, and use of drip irrigation are suggested to mitigate cropland NH_3 (Yang et al., 2022).

This study had some limitations that should be addressed. First, the optimization of fertilizer application dates was restricted to three major crops (maize, rice, and wheat) in China owing to a lack of remote-sensing phenological data. In future studies, we aim to extend this method to more crops and utilize it to generate NH_3 emission inventories on a decadal or daily scale. Secondly, atmospheric chemistry models were not used to validate the inventory established in this study because it only included NH_3 emissions from the three major crops. Therefore, in future studies, we plan to construct NH_3 emission inventories for all crops and other sources and validate them using an atmospheric chemistry model. Thirdly, fertilization in this study was under the assumption that each crop was fertilized three times according to a fertilization guide book for China (Zhang et al., 2009). Then the fertilization timings for each crop were set to periods that are typically targeted for fertilizer application (Table S3). It is a challenge to obtain accurate statistics on fertilizer application methods for farmers nationwide. In future studies, we will conduct field surveys to confirm the exact frequency of fertilizer applications in China.

CRediT authorship contribution statement

Yongqi Zhao: Conceptualization, Methodology, Software, Writing – original draft, Data curation. **Baojie Li:** Conceptualization, Writing – review & editing, Resources, Supervision. **Jinyan Dong:** Investigation, Resources. **Yan Li:** Resources. **Xueqing Wang:** Resources. **Cong Gan:** Resources. **Yingzhen Lin:** Investigation. **Hong Liao:** Investigation.

Data availability

Data will be made available on request.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.165225.

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