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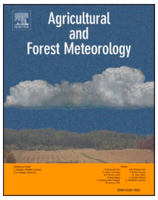

Differentiated strategies for synergistic mitigation of ammonia and methane emissions from agricultural cropping systems in China

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ABSTRACT

Ammonia (NH_3) and methane (CH_4) emissions from agricultural cropping systems in China have substantially impacted environmental quality and climate change, which become the key sector for synergizing pollution control and carbon reduction. However, the experiences of synergistic NH_3 and CH_4 emission governance at fine scales in this sector are still lacking. Here, we estimated NH_3 and CH_4 emissions from agricultural cropping systems in China in 2019 and identified regional characteristics of these emissions using advanced spatial analysis techniques. Subsequently, we developed an innovative synergistic mitigation zoning identification method to establish targeted and spatially differentiated reduction measures, addressing a critical gap in mitigation strategies for agricultural cropping systems in China. The results showed that NH_3 and CH_4 emissions from the three most important crops were 3.04 Tg N yr^{-1} and 8.13 Tg C yr^{-1}, respectively. NH_3 and CH_4 emissions exhibited the highest values in Eastern China, but the highest emission densities of NH_3 (49.8 kg N ha^{-1} yr^{-1}) and CH_4 (394 kg C ha^{-1} yr^{-1}) were observed in Northwest and South China, respectively. Further, bivariate local indicators of spatial association (LISA) maps revealed that the middle and lower reaches of the Yangtze River were hotspots for simultaneous high emissions of NH_3 and CH_4. Finally, based on the identified eight categories of zones, region-specific mitigation measures were proposed, including improved fertilization practices, irrigation methods, and straw management. These targeted, spatially differentiated mitigation strategies provide a reference framework and feasible approach for synergistic pollution control and carbon reduction in China's agricultural sector.

1. Introduction

Ammonia (NH_3) and methane (CH_4) emissions from agricultural cropping systems have profound impacts on environment and climate. Spatially precise regionalization of NH_3 and CH_4 from agricultural cropping systems and proposing targeted and differentiated control strategies are effective approaches to achieve pollution reduction and carbon mitigation. As a major alkaline gas in the atmosphere, NH_3 reacts with acidic gases (e.g., SO_2 and NO_x) to generate secondary aerosols, which can not only contribute to haze pollution (Fu et al., 2017), but also exert a cooling effect on the climate by influencing the global radiation budgets (Behera et al., 2013; Henze et al., 2012; IPCC, 2021). Additionally, it can affect biodiversity and water eutrophication (Clarisse et al., 2009; Paerl et al., 2014). CH_4 is the second largest greenhouse gas responsible for climate change (IPCC, 2021), while it also contributes to pollution episodes and adverse health effects by promoting the formation of tropospheric ozone (O_3) (Fiore et al., 2002; Kirschke et al., 2013; Turner et al., 2016). Over 80 % of NH_3 and 40 % of CH_4 in China are emitted from agriculture, of which up to 46 % of NH_3 and 40 % of CH_4 are caused by the agricultural cropping systems (Chen et al., 2022b; Li et al., 2021a). The urgency of the climate and environmental crises requires a shift to net-zero emissions in agriculture cropping systems, targeting not only the reduction of greenhouse gas emissions but also the synergistic optimization of the carbon and

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nitrogen nexus (Rosa and Gabrielli, 2023; Pan et al., 2022). Ongoing enhancements in cropland management practices will contribute to improved air quality, climate change mitigation and sustainable agriculture (Nayak et al., 2015; Ti et al., 2019).

Estimating NH_3 and CH_4 emissions accurately from agricultural cropping systems is crucial for developing effective emission reduction strategies. Previous estimates were mainly based on emission factors (EFs) (Ma, 2020; Wang et al., 2022), empirical regression model simulations (Sun et al., 2020; Wang et al., 2021a), and process-based model simulations (Chu et al., 2023; Zhang and Chen, 2014). The Intergovernmental Panel on Climate Change provides standardized EFs for NH_3 and CH_4 emissions. However, this method has high uncertainties as it cannot reflect the high variety of climate, soil and agricultural management practices factors (Xu et al., 2019). Further, empirical regression models have been developed to incorporate these factors into emission estimations. Wu et al. (2021) and Sun et al. (2020) employed stepwise regression models to estimate NH_3 volatilization from farmlands and CH_4 emissions from rice paddies in 2015 in China, respectively. Nevertheless, the accuracy of empirical regression models heavily relies on the quality and representativeness of the input data (Sun et al., 2020), and selecting appropriate independent variables to explain variations in the dependent variable poses a challenge in establishing accurate models.

Instead, process-based models have the superiority to simulate C and N biogeochemistry, considering various driving factors in agricultural cropping systems, such as biogeochemical processes, meteorological fields, soil properties and management practices, to compensate for the limitations of the mentioned methods. These process-based models include DeNitrification-DeComposition Model (DNDC) (Li et al., 2002), Water Heat Carbon Nitrogen Simulatorl Model (WHCNS) (Shi et al., 2022), Dynamic Land Ecosystem Model (DLEM) (Zhang et al., 2020a) and DayCent (Parton et al., 1998). For example, DNDC has been widely applied to estimate NH_3, CH_4, N_2O, and other gas emissions from various crops by integrating biogeochemical processes (including nitrification, denitrification, and fermentation), plant growth and agricultural management (Wang et al., 2021b; Yang et al., 2022, 2021). Moreover, to reduce the uncertainties of fine-scale zoning results, it is important to utilize simulation outputs from a single model to ensure data consistency. Therefore, we use the DNDC to comprehensively evaluate NH_3 and CH_4 emissions from the three most important crops in China.

Facing the escalating global challenge of environmental pollution and climate change, scholars have conducted extensive research on NH_3 and CH_4 to devise mitigation strategies (Ocko et al., 2021; Xu et al., 2024b). The *Implementation Plan of Agricultural Non-point Source Pollution Control and Supervision Guidance* issued in 2021 emphasized the importance of precise agricultural zoning management (MEE – Ministry of Ecology and Environment of the People's Republic of China, 2021). However, there is currently a lack of targeted and spatially differentiated synergistic mitigation strategies for NH_3 and CH_4 emissions in agricultural cropping systems. Existing studies have only focused on individual gas emission characteristics (Ma, 2020; Pan et al., 2018; Zhang and Chen, 2014; Zhang et al., 2022), with little research integrating NH_3 and CH_4 emissions. Research on zoning control based on the NH_3 and CH_4 emission characteristics in agriculture is even more limited. Various policy strategies have been proposed to mitigate NH_3 and CH_4 emissions from the agricultural sector in China (Li et al., 2023; MARA – Ministry of Agriculture and Rural Affairs of the People's Republic of China, 2022). These include specific measures for controlling NH_3 and CH_4 emissions from farmlands. However, due to differences in climatic conditions, agricultural production, and other factors in different regions, the effectiveness of these emission reduction measures may be limited, making their implementation on a broader scale challenging. The effectiveness of NH_3 and CH_4 emission reduction measures in various regions needs to be evaluated. Considering the complexity of the spatial interaction relationship between NH_3 and CH_4 emissions (such as double-high emissions, high NH_3 emissions, and high CH_4 emissions) and the variations in environmental pollution control efforts or agricultural production among different regions, there is an agent need to carry out research on the differentiated strategies for synergistic mitigation of NH_3 and CH_4 emissions at the grid scale.

Given the aforementioned knowledge gaps, existing research has been insufficient to provide specific and precise policy recommendations for synergistic mitigation in China's agricultural cropping systems. To address these gaps, the main objectives of this study are: (1) to evaluate NH_3 and CH_4 emissions from the agricultural cropping systems in China in 2019 using the DNDC model; (2) to compare distinctive regional characteristics of NH_3 and CH_4 emissions using geospatial statistical analysis, identifying both common and individual regional hotspots; and (3) to propose an innovative synergistic mitigation zoning identification method combining NH_3 and CH_4 emission densities, the bivariate LISA mapping and environmental pollution control investment levels to provide specific agricultural management practice guidance. The spatially differentiated synergistic mitigation strategies proposed in this study provide scientific evidence and policy insights for promoting the synergy of pollution control and carbon reduction in the agricultural sector, specifically crop production, and for achieving net-zero emissions.

2. Data and methods

2.1. DNDC model

The DeNitrification-DeComposition (DNDC) model is a biogeochemical model used to trace the cycle of C and N in agricultural systems (Li, 2007; Li et al., 1992). The DNDC model incorporates a comprehensive suite of biophysical and biogeochemical processes that govern C and N transport and transformation in plant-soil-climate systems (Li et al., 2011). It has been widely applied to simulate NH_3 emissions, greenhouse gas emissions, and yields of crops such as rice, wheat, and maize (Abdalla et al., 2020; Guo et al., 2023; Zhou et al., 2023). The DNDC model consists of six interacting sub-models: soil climate, plant growth, decomposition, nitrification, denitrification, and fermentation (Li et al., 1997). The "soil climate" sub-model simulates soil temperature, moisture, and Eh profiles using soil and meteorological data. The "plant growth" sub-module simulates the crop growth dynamics based on crop characters, climate, soil properties, and management practices. The "decomposition" sub-model mainly describes the decomposition process of soil organic matter. The "nitrification" sub-model tracks growth of nitrifiers and oxidation of ammonium to nitrate. The "denitrification" sub-model simulates the reduction of NO_3^- to NO_2^-, NO, N_2O, and N_2 under the action of denitrifying bacteria. The "fermentation" sub-model simulates methane production, oxidation, and transport under submerged conditions (Li, 2000; Li et al., 2011).

The DNDC model calculates liquid-phase NH_3 concentration based on NH_4^+ and OH^- concentrations. Soil gas-phase NH_3 concentration is directly controlled by liquid-phase NH_3 concentration and soil temperature. Volatilization of NH_3 to the atmosphere are a function of soil air-filled porosity and clay content (Balasubramanian et al., 2017; Li, 2000). Methane fluxes are predicted by modeling CH_4 production, oxidation, and emission processes. CH_4 production rate is a function of soil organic carbon (SOC) or dissolved organic carbon (DOC), soil redox potential (Eh), and temperature. CH_4 oxidation rate depends on soil CH_4 concentration and redox potential (Eh). CH_4 is emitted from soil to atmosphere primarily via plant-mediated transport or ebullition (Guo et al., 2023; Tang et al., 2024).

The 2019 NH_3 and CH_4 emissions from China's three principal grain crops were simulated using the latest DNDC95 version (http://www.dndc.sr.unh.edu/) at a 0.25° × 0.25° spatial resolution, excluding emissions from natural soils and livestock. Meteorological, soil, crop and management parameters were assigned to each grid, assuming that the attributes within each grid were uniform. Daily NH_3 and CH_4 emission fluxes were estimated for each grid, with cumulative emissions calculated over each crop's growing period. Here, we focused on major cropping systems comprising the three most important crops: rice (early, late, and single-season), maize, and wheat (spring and winter).

2.1.1. *Input data for the DNDC model*

The input data for the DNDC model includes geographic data (site name, latitude, etc.), meteorological data (daily maximum and minimum temperatures, daily precipitation), soil data (land use types, texture types, bulk density, clay content, etc.), crop data (crop types, planting and harvesting dates, etc.), and fertilizer application data (number of applications, application dates, amounts of fertilizer applied, etc.). In this study, some parameters were set to default values within the model (Table S1).

Meteorological data were obtained from climatic stations of the China Meteorological Administration (https://data.cma.cn/, daily value dataset of Chinese ground climate data (V3.0)). According to the location of the meteorological stations, China was divided into more than 800 polygons using the Tyson polygon method. Each polygon corresponds to a meteorological station, and the meteorological data of all grids within the polygon are equal to the data of the meteorological station. Soil data were obtained from Harmonized World Soil Database v 1.2 (http://www.fao.org/land-water/databases-and-software/hwsd/en/) with a spatial resolution of 1 km × 1 km.

The spatial distribution maps of each crop were derived from Li et al. (2021a) and Monfreda et al. (2008). Provincial-level data on the planting area and fertilizer application rates for these three most important crops were obtained from the *China Agricultural Yearbook 2020* (NBS – National Bureau of Statistics, 2020a) and the *National Data on the Cost and Profit of Agricultural Products 2020* (NDRC – National Development and Reform Commission of China, 2020). Referring to the *Fertilizer Pure Amount Conversion Table* (www.moa.gov.cn), the obtained fertilizer application rates per acre were converted into pure nitrogen. Every crop was set to be fertilized three times, with application dates determined based on critical phenological periods (Li et al., 2021a; Zhao et al., 2023). The application of farmyard manure or green manure was not considered due to a lack of detailed statistical data (Liu et al., 2022b; Wang et al., 2021b). The planting (transplanting) and harvesting dates for the three most important crops were sourced from Zheng (2015).

2.1.2. *Evaluation of simulated ammonia and methane emissions*

We used the ground-based measurements of NH_3 and CH_4 from public papers to evaluate the accuracy of DNDC simulation. Field measurements on NH_3 and CH_4 emissions were collected from websites such as CNKI and Web of Science, using keywords such as 'Ammonia volatilization', 'NH_3', 'methane', 'CH_4', and 'experiment', screened according to certain criteria (Supplementary Sections S1). We finally obtained 32 data pairs of NH_3 fluxes from 29 studies (Table S2) and 30 data pairs of CH_4 fluxes from 26 studies (Table S3) over a time span of 1992–2020. These NH_3 and CH_4 observation data mainly covered the typical intensive cultivated areas of the three most important crops, representing the overall simulated NH_3 and CH_4 emissions from DNDC across China. Observed values were compared with simulated values in the corresponding grid based on the observation point's geographic location.

2.2. *Bivariate spatial correlation analysis of NH_3 and CH_4*

Spatial autocorrelation effectively reveals distribution characteristics and relationships of spatial data, for example by using Moran's I index. Local Moran's I is the most commonly used local indicators of spatial association (LISA) statistic (Anselin, 1995). Bivariate LISA, based on Local Moran's I, identifies the degree of spatial autocorrelation at specific locations for different variables and generates cluster maps to reveal spatial correlations between two variables (Anselin et al., 2002; Lee, 2001; Ogneva-Himmelberger and Huang, 2015). This method has been extensively applied in air and soil pollution studies, such as examining spatial relationships between $PM_{2.5}$ concentrations and CO_2 emissions (Li et al., 2020), and between heavy metals and polycyclic aromatic hydrocarbons (PAHs) concentrations (Wu et al., 2019a). In this study, we employed bivariate local Moran's I to characterize the spatial interaction of NH_3 and CH_4 emissions. The analysis used gridded emission data (0.25° × 0.25° resolution) for NH_3 and CH_4 simulated by the DNDC model, encompassing over 16,000 grids. The bivariate local Moran's I (I_{kl}) is calculated as follows:

$$I_{kl} = Z_k^i \sum_{j=1}^{n} w_{ij} Z_l^j$$

where $Z_k^i = \left(X_k^i - \overline{X}_k\right)/s_k^2$, $Z_l^j = \left(X_l^j - \overline{X}_l\right)/s_l^2$. X_k^i is the value of NH_3 emissions at grid i; X_l^j is the value of CH_4 emissions at neighboring grid j. Their units are t N and t C, respectively. $\overline{X}_k$ and $\overline{X}_l$ are the average values of NH_3 and CH_4 emissions, respectively; s_k^2 and s_l^2 means the variances of NH_3 and CH_4 emissions, respectively; n represents the total number of samples in the study area; and w_{ij} is the spatial weight matrix, which can be represented based on a distance weighting between locations i and j. I_{kl} can quantitatively measure the degree of association between NH_3 emissions in grid i and CH_4 emissions in neighboring grid j. A high positive I_{kl} represents that the location under study has values similar to its neighbors, either both high or both low. A high negative I_{kl} represents that the location under study is obviously different from the values of its surrounding locations. $I_{kl}=0$ represents no spatial correlation between NH_3 and CH_4 emissions.

We used Geoda (http://geodacenter.github.io/) to calculate the bivariate Local Moran's I. The statistical significance of bivariate Local Moran's I was tested using 999 permutations, and the significance level was $p < 0.05$, which can ensure robustness in identifying spatial patterns and relationships (Anselin, 1995; Biswas et al., 2024).

2.3. *Synergistic mitigation zones of NH_3 and CH_4 in agricultural cropping systems*

Based on bivariate LISA analysis, although we have identified the spatial interaction of NH_3 and CH_4, considering the variations in environmental governance intensity across different regions, further zoning based on bivariate LISA maps was necessary. We proposed an innovative synergistic mitigation zoning identification method and divided the zones using three indicators: (1) whether the NH_3 and CH_4 emission densities exceeded the standards. This study defined the total NH_3 emissions from the three most important crops/total planting area of the three most important crops (32 kg N ha^{-1} yr^{-1}) and the total CH_4 emissions from paddy fields/total planting area of rice (274 kg C ha^{-1} yr^{-1}) in China in 2019 as the standard values to measure the average NH_3 and CH_4 emission densities in each grid, respectively. Due to the lack of corresponding standard values in the relevant literature, we chose to use the average emission densities for our assessment; (2) the bivariate LISA map of NH_3 and CH_4, which reflected different combinations of their spatial distributions; (3) the proportion of environmental pollution control investment, we judged the level of environmental pollution control investment by whether it exceeded the national average (1.2 % yr^{-1}). If the proportion of environmental pollution control investment in a region was above the national average, it was considered high; conversely, it was considered low. The proportion of environmental pollution investment to GDP is a crucial indicator of how seriously an area prioritizes environmental protection. A reasonable proportion of environmental pollution investment can effectively reduce NH_3 and CH_4 emissions from agriculture, and differential management should be implemented to avoid high environmental pollution control costs in areas with lower agricultural gas emissions and insufficient control in areas with higher agricultural gas emissions. Data on the proportion of environmental pollution control investment which covered all anthropogenic sectors at the city level were sourced from the China Urban

Statistical Yearbook (NBS – National Bureau of Statistics, 2020b) and various provincial and municipal statistical yearbooks, environmental statistics bulletins, etc.

According to the combination of the three indicators, eight types of collaborative control zones can be classified (Table 1). For example, if the NH_3 and CH_4 emission densities in a region exceeded the standards, the bivariate LISA map showed a high-high area, and the proportion of environmental pollution control investment in GDP was low, this area was a Double-high emissions investment insufficient zone.

3. Results

3.1. *Total amount and source of NH_3 and CH_4 emissions from agricultural cropping systems*

The total NH_3 and CH_4 emissions from the three most important crops in China in 2019 simulated by the DNDC model were 3.04 Tg N yr^{-1} and 8.13 Tg C yr^{-1}, respectively. Table 2 presents a quantitative comparison of this study with other research results, our findings were close to previous research (NH_3: 2.23–3.49 Tg N; CH_4: 7.33–8.97 Tg C). Moreover, a comparison between the model simulations and field observation data (Fig. 1) revealed high accuracy in our results (the R^2 of NH_3 and CH_4 were 0.80 and 0.81, respectively).

Obvious contribution variances from different crops in NH_3 and CH_4 emissions were identified. For NH_3 emissions, values from rice, maize, and wheat were 1.12, 1.41, and 0.50 Tg N, respectively. The average NH_3 emission density from the three most important crops was 32.1 kg N ha^{-1} yr^{-1}, with rice having a higher emission density (37.7 kg N ha^{-1} yr^{-1}) than maize (34.2 kg N ha^{-1} yr^{-1}) and wheat (21.1 kg N ha^{-1} yr^{-1}). Studies indicate that paddy fields generally have higher ammonia volatilization losses than upland fields (Liu et al., 2022a; Xu et al., 2024a), due to accelerated urea hydrolysis under flooding conditions (Hayashi et al., 2006). Additionally, the higher emission density of maize compared to wheat is mainly attributed to its fertilization dates being concentrated in spring and summer, and the high temperature can enhance NH_3 volatilization (Yang et al., 2015). In contrast, wheat is generally fertilized in spring and autumn during lower temperature. For CH_4 emissions, values from early, late, and single-season rice were 0.94, 2.30, and 4.90 Tg C, respectively. The average CH_4 emission density from rice paddies was 274 kg C ha^{-1} yr^{-1}, with late rice having an emission density of 462 kg C ha^{-1} yr^{-1}, which was 2.19 times and 1.91 times that of early rice (211 kg C ha^{-1} yr^{-1}) and single-season rice (242 kg C ha^{-1} yr^{-1}), respectively, similar to the findings of Wang et al. (2021b) (1.85 and 1.76 times). It is reported that high temperature stimulates CH_4 emission (Yue et al., 2005), while excessive nitrogen application can inhibit CH_4 emission (Yao et al., 2012). This may have contributed to the differences in emission densities among different rice. Single-season rice was the largest contributor to CH_4 emissions (60.3 %), a result of its large cultivation area.

Table 1
Zone division conditions for NH_3 and CH_4 synergistic mitigation.

NH_3 and CH_4 emission densities: Exceed standards	Bivariate LISA map	Proportion of environmental pollution control investment: High or Low	Classification Results
NH_3 and CH_4 yes	high-high	Low	Double-high emissions investment insufficient zone
NH_3 and CH_4 yes	high-high	High	Double-high emissions poor governance zone
NH_3 yes	high NH_3-low CH_4 or high-high	Low	High NH_3 emissions investment insufficient zone
NH_3 and CH_4 yes	high NH_3-low CH_4	Low	
NH_3 yes	high NH_3-low CH_4 or high-high	High	High NH_3 emissions poor governance zone
NH_3 and CH_4 yes	high NH_3-low CH_4	High	
CH_4 yes	low NH_3-high CH_4 or high-high	Low	High CH_4 emissions investment insufficient zone
NH_3 and CH_4 yes	low NH_3-high CH_4	Low	
CH_4 yes	low NH_3-high CH_4 or high-high	High	High CH_4 emissions poor governance zone
NH_3 and CH_4 yes	low NH_3-high CH_4	High	
No	low-low	High or Low	Clean zone
Others			Potential risk zone

Table 2
Comparison of estimated NH_3 and CH_4 emissions from the three most important crops with other studies (The unit of NH_3 emissions is Tg N yr^{-1}, the unit of CH_4 emissions is Tg C yr^{-1}).

Gas type	Base year	Total	References
NH_3	2010	3.33	Xu et al. (2024a)
	2015	3.49	Wu et al. (2021)
	2016	2.23	Li et al. (2021a)
	2018	3.27	Xu et al. (2021)
	2018	3.46	Ma et al. (2022)
	2019	3.04	This study
CH_4	1981–2010	7.89	Tian et al. (2018)
	2015	7.33	Huang et al. (2019)
	2015	8.59	Gong and Shi (2021)
	2010–2020	8.97	Yang and Shi (2022)
	2020	7.34	Duan et al. (2023)
	2019	8.13	This study

3.2. *Spatial distribution pattern of NH_3 and CH_4 emissions*

The total NH_3 and CH_4 emissions from the three most important crops in China in 2019 exhibited significant spatial variability (Fig. 2). High NH_3 emissions were predominantly concentrated in the Northeast Plain, North China Plain, and the middle and lower reaches of the Yangtze River Plain. High CH_4 emissions were predominantly concentrated in the middle and lower reaches of the Yangtze River Plain and South China. The region with the highest NH_3 and CH_4 emissions was East China, calculated to be 815 Gg N and 3090 Gg C, respectively, accounting for 27.0 % and 38.5 % of total national emissions. The three provinces with the highest NH_3 emissions were Heilongjiang (255 Gg N), Jiangsu (246 Gg N), and Henan (215 Gg N). The three provinces with the highest CH_4 emissions were Hunan (967 Gg C), Anhui (960 Gg C), and Hubei (920 Gg C).

The emphasis of our study was on the NH_3 and CH_4 emission densities in each region (Fig. 3). Generally, the southern region exhibited higher NH_3 and CH_4 emission densities compared to the northern region, with NH_3 and CH_4 emission densities in the south 1.25 times and 12.3 times those in the north, respectively. Studies show that NH_3 and CH_4 emissions are significantly positively correlated with air temperature (Wu et al., 2021; Zhang et al., 2011). In terms of region, South China had the highest CH_4 emission density in China, reaching 394 kg C ha^{-1} yr^{-1}. Despite the relatively lower total NH_3 emissions in Northwest China, accounting for only 11.06 % of the national total NH_3 emissions, its emission density was the highest as high as 49.8 kg N ha^{-1} yr^{-1}, ranking first nationwide. This phenomenon can be attributed to the excessive use of nitrogen fertilizers in the region, with an average nitrogen application rate of 273 kg N ha^{-1} yr^{-1} for the three most important crops, surpassing other regions in China. A significant portion of nitrogen that is not absorbed by crops is directly converted into NH_3 emissions into the atmosphere. In terms of province, Shanghai had the

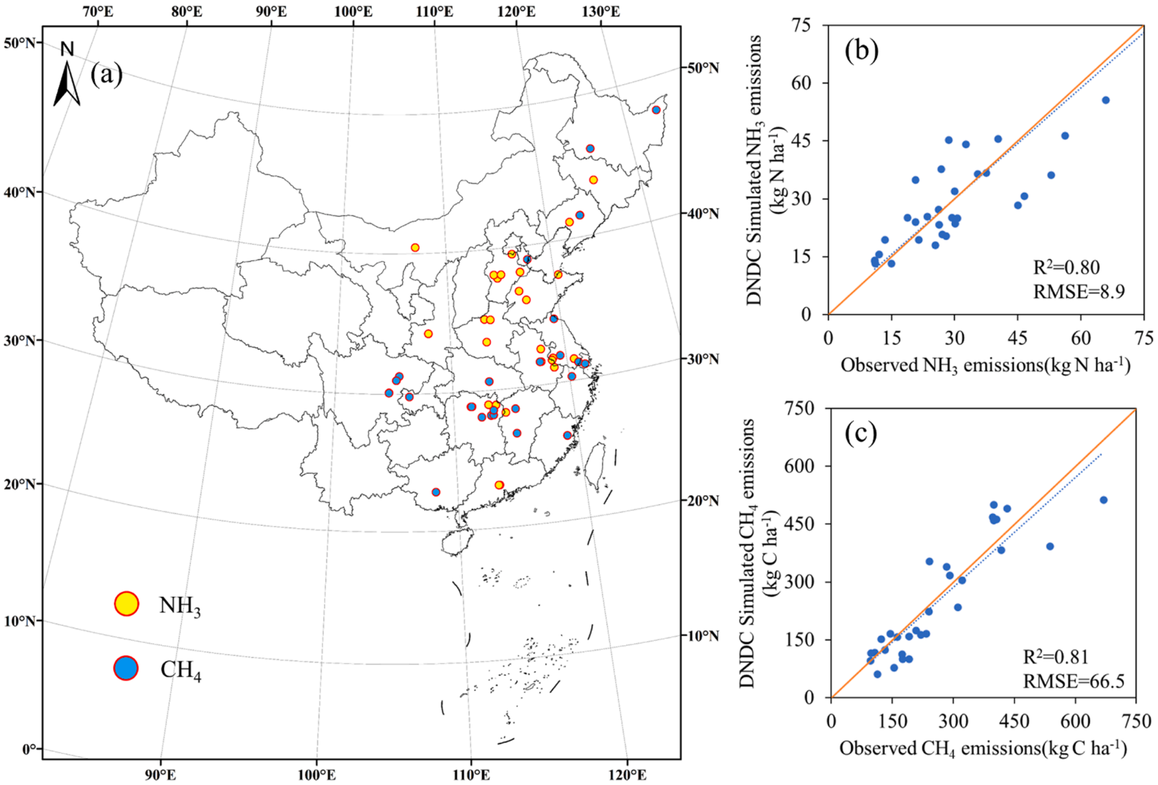


Fig. 1. Comparison of field observations with DNDC simulations. (a) Spatial distribution of NH_3 and CH_4 observation sites in China; (b) Scatterplots between the measured and simulated cropland NH_3 emissions; (c) Scatterplots between the measured and simulated cropland CH_4 emissions.

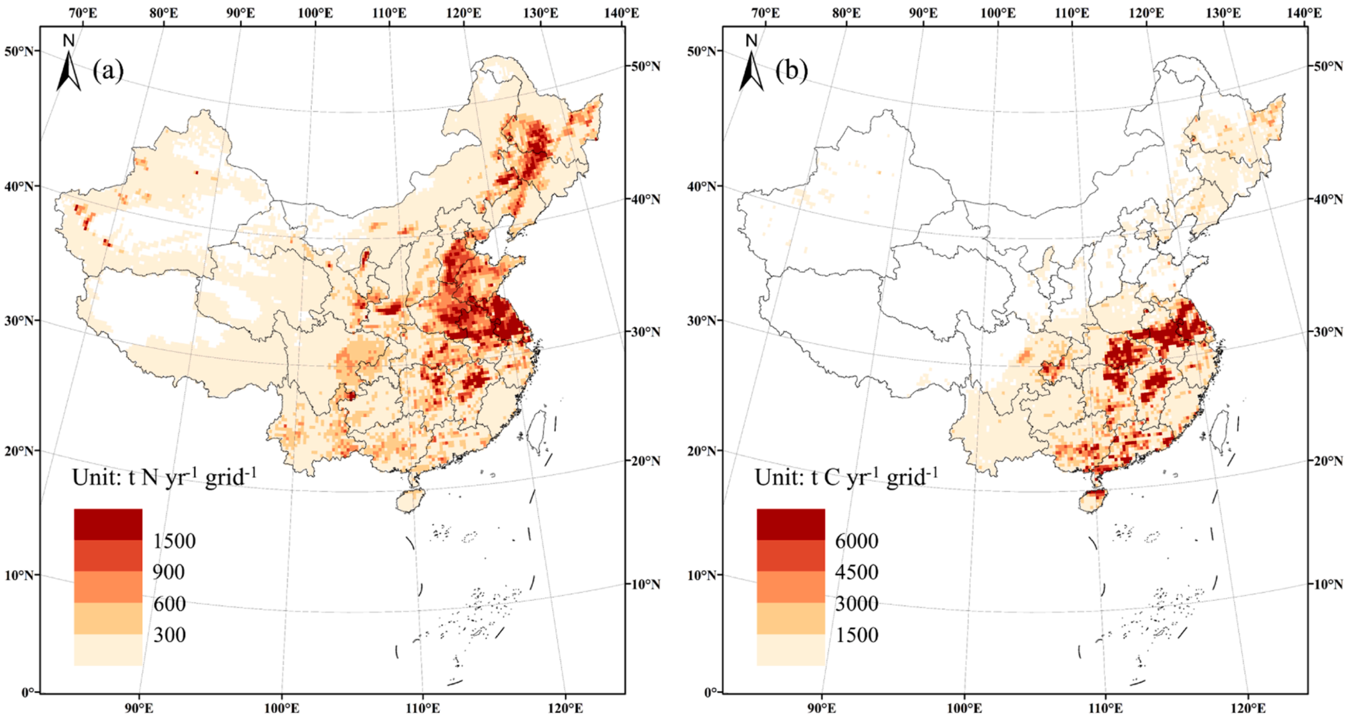


Fig. 2. Spatial distribution of 2019 (a) total NH_3 emissions from three most important crops and (b) total CH_4 emissions from paddy fields.

highest NH_3 emission density. Despite its small cultivated area (115×10^3 ha), which accounted for only 0.12 % of the national total cultivated area, the NH_3 emission density was as high as 74.1 kg N ha^{-1} yr^{-1}, 2.3 times the national average, mainly due to its nitrogen application rate being 1.4 times higher than the national average. Hainan had the highest CH_4 emission density at 599 kg C ha^{-1} yr^{-1}, 2.19 times the national average, attributed to its humid and warm climate. Specifically, although Heilongjiang cultivated a large area of rice (3.81×10^6 ha), the CH_4 emissions were not significantly high due to its lower emission density (93.1 kg C ha^{-1} yr^{-1}) compared to other provinces, consistent

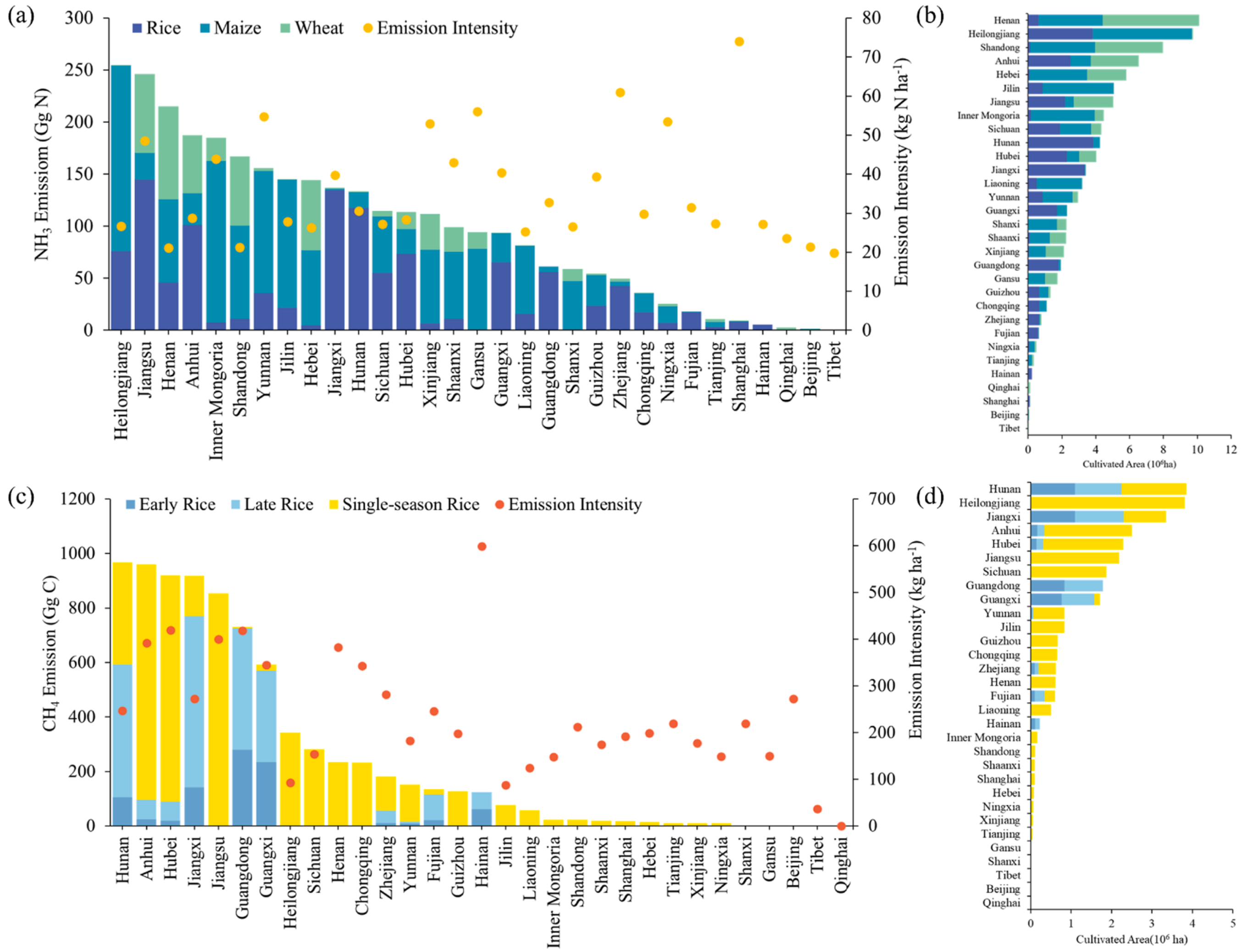


Fig. 3. Provincial (a) Total NH_3 emissions and emission density; (b) Planting area of three most important crops; (c) Total CH_4 emissions and emission density; (d) Planting area of paddy field.

with other studies (Gong and Shi, 2021; Wang et al., 2021b). The air temperature of Heilongjiang was far below the average temperature, which may be the main reason for the low CH_4 emission density.

3.3. Spatial interactions between NH_3 and CH_4

Bivariate LISA maps can clearly show the spatial interaction relationship of NH_3 and CH_4. Due to the influence of planting structure and management practices, the clustering patterns of combined NH_3 and CH_4 can be presented as five categories on a bivariate LISA map: high-high, low-low, high-low, low-high, and not-significant (Fig. 4). The high-high cluster patterns were aggregated mainly in the middle and lower reaches of the Yangtze River, the Sichuan Basin, East China, and some parts of eastern Heilongjiang. Among these regions, the middle and lower reaches of the Yangtze River (including Jiangsu, Zhejiang, Anhui, Shanghai, Jiangxi, Hubei, and Hunan) exhibited a more concentrated high-high clustering, accounting for approximately 29.1 % and 60.0 % of the national NH_3 and CH_4 emissions, respectively. This occurrence was largely attributed to the intensive cultivation of rice in this region, which accounted for over half of rice cultivation in China in 2019 (NBS – National Bureau of Statistics, 2020a). The high NH_3-low CH_4 region was mainly distributed in northern China and parts of Yunnan, where the high NH_3 emissions were dominated by maize and wheat. The high N fertilizer application rates and minimal CH_4 emissions during the growth of dryland crops led to these cluster patterns. The low NH_3-high CH_4 region was mainly distributed in southern China, where the high CH_4 emissions were dominated by double cropping rice, potentially influenced by water and fertilizer management practices in paddy fields (Gu et al., 2022; Yang et al., 2020). The low-low clusters were aggregated in northwestern and southwestern China. Moreover, a majority of grids fall into the "not significant" category, where the local Moran' I cannot effectively compare the emissions in sub-regions (Anselin, 1995; Li et al., 2021b). In other words, there was no significant spatial correlation between local NH_3 emissions and nearby CH_4 emissions.

In summary, the distinct spatial clustering patterns identified through bivariate LISA analysis offer valuable and objective insights into the spatial interactions and distribution of NH_3 and CH_4 emissions across different regions of China. It could provide basic information for synergistic mitigation zoning.

3.4. Synergistic mitigation zoning and policy recommendations

Refined zoning is crucial for achieving precise control of NH_3 and CH_4 emissions and optimizing the allocation of environmental pollution control investment. We first sorted out the current major NH_3 and CH_4 emission reduction measures and reduction potentials in agricultural cropping systems (Table 3). For the mitigation efficiency of specific reduction strategies, we first selected results from meta-analyses. Next, we prioritized results from field experiments with multiple control

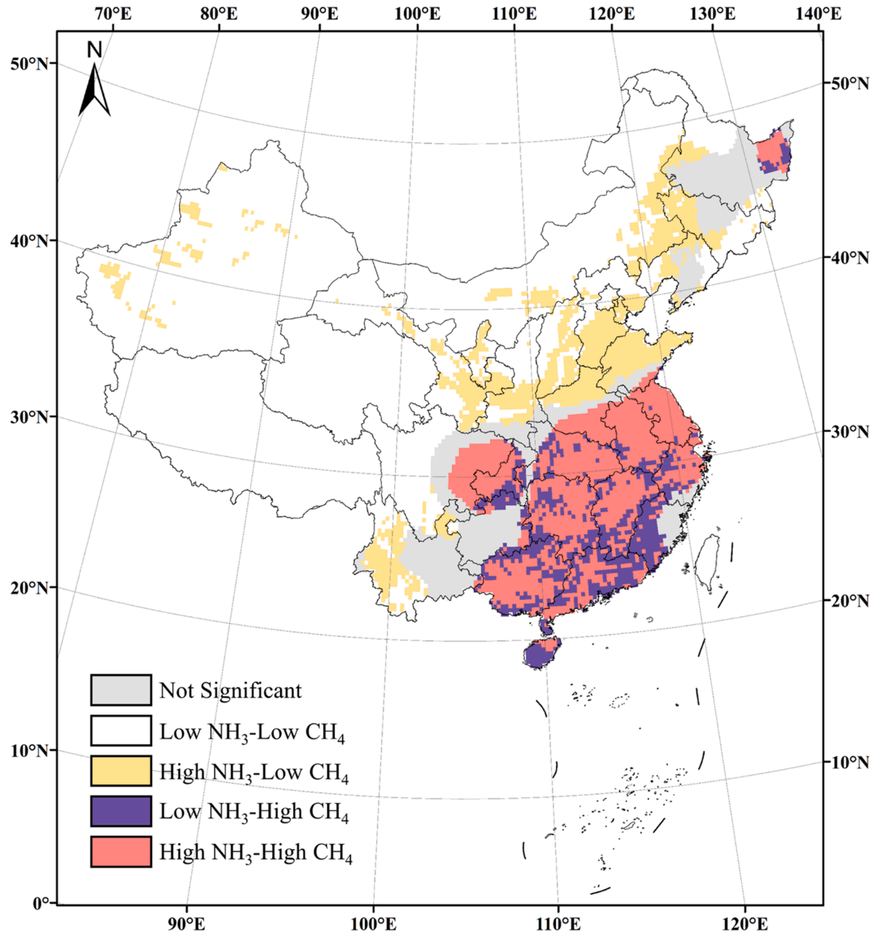


Fig. 4. Bivariate LISA mapping of NH_3 and CH_4.

groups. Based on the zoning results, agricultural systems, climate conditions, and agricultural economics of each region, we have developed differentiated and locally applicable control strategies for emission reduction (Fig. 5).

The characteristics and emission reduction strategies of each zone were summarized as follows:

(1) Double-high emissions investment insufficient zone and Double-high emissions poor governance zone: The Double-high emissions investment insufficient zone accounted for 19.6 % of the total area of the Double-high emissions zone, mainly concentrated in Xinyang City in Henan Province, Liuan City and Hefei City in Anhui Province, and so on. Taking Xinyang City as an example, the proportion of environmental pollution control investment in Xinyang City was the lowest among cities with insufficient investment, standing at only 57.9 % of the national average level. The lack of funds may result in insufficient capacity to address pollution from agricultural cropping systems. Double-high emissions poor governance zone accounted for a significantly higher proportion (80.4 %) of the Double-high emissions zone compared to Double-high emissions investment insufficient zone, mainly concentrated in Jiangsu, Guangdong, Guangxi, and eastern Anhui. In Jiangsu Province, for instance, the proportion of environmental pollution control investment was 1.39 times the national average level; however, the high emission trends of NH_3 and CH_4 have not been alleviated, with NH_3 and CH_4 emissions accounting for 8.16 % and 10.6 % of the national emissions, respectively. Higher investments were likely to be allocated to pollution control in sectors such as industry. Therefore, these regions need to strengthen the management and supervision of investment projects in agricultural governance, seek optimal reduction measures, and ensure the maximization of investment utilization.

Priority should be given to NH_3 and CH_4 mitigation measures with lower cost or labor inputs. For example, measures such as controlled irrigation and optimized N and water management have been shown to achieve synergistic reduction of NH_3 and CH_4 emissions from paddy fields (Liang et al., 2017; Xu et al., 2012; Yang et al., 2012). Furthermore, higher-cost slow/controlled-release urea and biochar can be applied in poor governance zone to achieve synergistic reduction of NH_3 and CH_4 emissions (Guo et al., 2019; Sun et al., 2019; Wang et al., 2019). It is necessary to increase subsidies or investments to purchase such fertilizers or soil amendments. In the plains of the middle and lower reaches of the Yangtze River, where rice cultivation is concentrated and facility agriculture is well developed, nitrogen fertilizer deep placement and the promotion of rice-fish or rice-shrimp co-culture can be employed to alleviate the local situation of double-high emissions (Fang et al., 2023; Liu et al., 2022c, 2015, 2020b). Combined strategies for NH_3 and CH_4 reduction can also be used to mitigate double-high emissions in specific regions. Taking Jiangsu as an example, the application of urease inhibitors has been shown to reduce NH_3 emissions (Sha et al., 2022), while water-saving irrigation techniques (alternating wetting and drying, controlled irrigation) and the cultivation of high yields and low methane emissions rice varieties can reduce CH_4 emissions (Qin et al., 2015; Wang et al., 2020a).

(2) High NH_3 emissions investment insufficient zone and High NH_3 emissions poor governance zone: High NH_3 emissions investment insufficient zone was mainly distributed in Inner Mongolia, Xinjiang, Hunan, Yunnan, and other places. High NH_3 emissions poor

Table 3
Emission reduction measures of NH_3 and CH_4 in agricultural cropping systems.

Crop type	No.	Optimization direction	Concrete measure	NH_3 mitigation efficiency (%)	CH_4 mitigation efficiency (%)	Reference
Rice paddy	(1)	Fertilizer type	Slow/Controlled-release urea [a]	65	20	Guo et al. (2019)
	(2)	Fertilizer type	Composted livestock manure [a]	NA	56	Chen et al. (2011)
	(3)	Fertilizer type	Substituting manure for mineral N [a]	36	–	Xia et al. (2021)
	(4)	Tillage method	No-tillage [b]	–	23	Maucieri et al. (2021)
	(5)	Irrigation method	Controlled irrigation [a]	14	79	Xu et al. (2012); Yang et al. (2012)
	(6)	Irrigation method	Alternating wetting and drying [a]	NA	38	Wang et al. (2020a)
	(7)	Straw management	Straw strip-mulched onto the field surface [a]	NA	32	Ma et al. (2009)
	(8)	Straw management	Application during non-rice growing season [b]	NA	24–43	Xie et al. (2010)
	(9)	Straw management	Rotten straw return /Cow-digested straw return [a]	NA	34–72	Wang et al. (2023)
	(10)	Fertilization practice	Nitrogen fertilizer deep placement [a]	15–45	36–39	Liu et al. (2015); Liu et al. (2020b)
	(11)	Fertilization practice	Nitrogen reduction [a]	39–55	–	Zhu et al. (2021)
	(12)	Rice cultivar	Rice varieties with high yields and low methane emissions [a]	NA	27	Qin et al. (2015)
	(13)	Integrated water and fertilizer management	Optimized N and water management [a]	28	21	Liang et al. (2017)
	(14)	Soil additives	Biochar [a]	20	20–51	Sun et al. (2019) Wang et al. (2019)
	(15)	Soil additives	Urease inhibitor [b]	37	NA	Sha et al. (2022)
	(16)	Planting pattern	Rice-aquaculture co-culture systems [a]	16–20	14	Liu et al. (2022c) Fang et al. (2023)
	(17)	Planting pattern	Drained in the winter or paddy-upland rotation [b]	NA	42–56	Shi et al. (2010)
	(18)	Planting method	Direct seeding crop establishment [a]	NA	16–54	Corton et al. (2000)
	(19)	Sowing Date	Optimizing delayed sowing [a]	NA	30	He et al. (2023)
Upland crop	(20)	Fertilizer type	Substituting manure for mineral N fertilizer [b]	49–50	NA	Ren et al. (2022)
	(21)	Fertilizer type	Slow-release fertilizer [a]	66–79	NA	Hu et al. (2020b)
	(22)	Irrigation method	Drip irrigation [a]	34	NA	Wang et al. (2020b)
	(23)	Straw management	Straw returning [a]	37–39	NA	Lyu et al. (2020)
	(24)	Fertilization practice	Nitrogen fertilizer deep placement [a]	64	NA	Hu et al. (2020b)
	(25)	Fertilization practice	Irrigation after fertilization [a]	83	NA	Viero et al. (2017)
	(26)	Fertilization practice	Nitrogen reduction [a]	24	NA	Liu et al. (2020a)
	(27)	Soil additives	Urease inhibitor [a]	41–96	NA	Sha et al. (2020)
	(28)	Soil additives	Biochar [a]	35	NA	Wu et al. (2019b)
	(29)	Planting method	Ridge furrows with plastic film mulching [a]	30–64	NA	Shangguan et al. (2012)

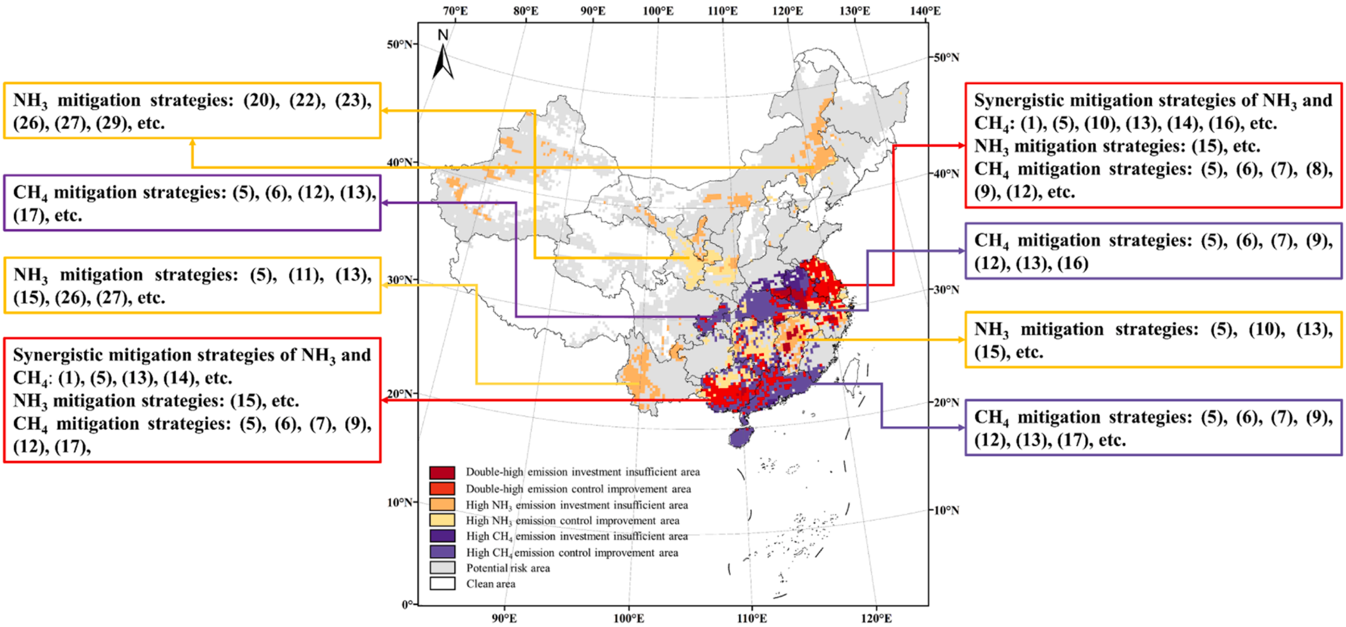
Note: “a” indicates that the data were from the results of the field experiment, and “b” indicates that the data were from the results of a meta-analysis; “-” denotes that implementing a reduction strategy for NH_3 or CH_4 may potentially increase emissions of another gas.

governance zone was mainly distributed in Gansu, Shaanxi, Hunan, Guangxi, and other places.

For Northwest China and Inner Mongolia, with relatively low NH_3 emissions, and the planting area is small, accounting for only 11.8 % of the country, it is relatively easy to improve the current situation of high NH_3 emissions. Some simple and economic measures like reducing nitrogen fertilizer use, combining organic and inorganic fertilizers, using urease inhibitors, and returning straw to the field can be implemented (Liu et al., 2020a; Lyu et al., 2020; Ren et al., 2022; Sha et al., 2020). In addition, given the shortage of water resources in the north, ridge furrows with plastic film mulching and drip irrigation technologies can be promoted (Shangguan et al., 2012; Wang et al., 2020b). Nitrogen fertilizer deep placement and optimized N and water management can reduce the high levels of NH_3 emissions in southern China from rice fields.

(3) High CH_4 emissions investment insufficient zone and High CH_4 emissions poor governance zone: Overall, the level of environmental pollution control investment in the high CH_4 emissions zone was relatively high, with the poor governance zone being 6.33 times that of the investment insufficient zone. Therefore, High CH_4 emissions poor governance zone should be the focus of attention. For instance, CH_4 emissions from rice fields in Hubei reached as high as 920 Gg C, ranking third in China. The proportion of environmental pollution control investment across the entire province exceeded the national average by 38.0 %. In such areas, it is essential to ensure sufficient investments for implementing scientific and reasonable emission reduction measures to reduce CH_4 emissions from rice paddies.

Considering the universal irrigation methods in China such as seasonal continuous flooding (F), flooding-drainage-reflooding (F-D-F), and flooding-drainage-reflooding-moist (F-D-F-M), as well as the critical impact of water management on CH_4 emissions from rice (Qian et al., 2023; Zou et al., 2007), promoting water-saving irrigation techniques, adopting optimized N and water management, and planting rice varieties with high yields and low methane emissions are universally applicable mitigation measures (Liang et al., 2017; Qin et al., 2015; Wang et al., 2020a; Xu et al., 2012). Because of the characteristics of the hot and humid climate in South China, it is advisable to adjust the planting pattern to rice-upland rotation or multi-cropping. In the Sichuan-Chongqing area, the winter paddy field accounts for a large proportion, so the planting pattern of draining water in the leisure season or rice-upland rotation can be implemented (Shi et al., 2010). Additionally, effective management of straw residues can play a crucial role in improving soil chemical properties and reducing CH_4 emissions from rice fields (Lv et al., 2023; Sanchis et al., 2012). Practices such as straw strip-mulched onto the field surface, rotten straw return or cow-digested straw



NH_3 mitigation strategies: (20), (22), (23), (26), (27), (29), etc.

CH_4 mitigation strategies: (5), (6), (12), (13), (17), etc.

NH_3 mitigation strategies: (5), (11), (13), (15), (26), (27), etc.

Synergistic mitigation strategies of NH_3 and CH_4: (1), (5), (13), (14), etc.
NH_3 mitigation strategies: (15), etc.
CH_4 mitigation strategies: (5), (6), (7), (9), (12), (17),

Synergistic mitigation strategies of NH_3 and CH_4: (1), (5), (10), (13), (14), (16), etc.
NH_3 mitigation strategies: (15), etc.
CH_4 mitigation strategies: (5), (6), (7), (8), (9), (12), etc.

CH_4 mitigation strategies: (5), (6), (7), (9), (12), (13), (16)

NH_3 mitigation strategies: (5), (10), (13), (15), etc.

CH_4 mitigation strategies: (5), (6), (7), (9), (12), (13), (17), etc.

Fig. 5. Synergistic mitigation zones of NH_3 and CH_4 in agricultural cropping systems in China. The numbers in the figure represent the various mitigation strategies in Table 3.

return can effectively reduce CH_4 production (Ma et al., 2009; Wang et al., 2020a). In paddy-wheat rotation areas like Hubei, considering the application of straw during non-rice growing season is also a viable option (Xie et al., 2010).

(4) Potential risk zone: Mainly distributed around regions with high NH_3 emissions, high CH_4 emissions, or Double-high emissions. The potential risk zone was large, occupying over 45 % of China's land area. To prevent becoming new high NH_3 or CH_4 emissions hotspots, these areas should be prioritized for prevention and control by strengthening the monitoring of NH_3 and CH_4 emissions from cultivation, as well as improving the management of agricultural production.

(5) Clean zone: Mainly distributed in Tibet, Qinghai, and other areas where crop cultivation is small or unsuitable, accounting for 37 % of the national land area. These areas should maintain their current status to prevent the generation of NH_3 or CH_4 emissions.

4. Discussion

4.1. Uncertainties in NH_3 and CH_4 emissions

NH_3 and CH_4 emissions from the three most important crops in China were simulated by the DNDC model. The total NH_3 and CH_4 emissions in 2019 were 3.04 Tg N yr^{-1} and 8.13 Tg C yr^{-1}, respectively, which fall within the range of previous estimates (Table 2). We also evaluated the

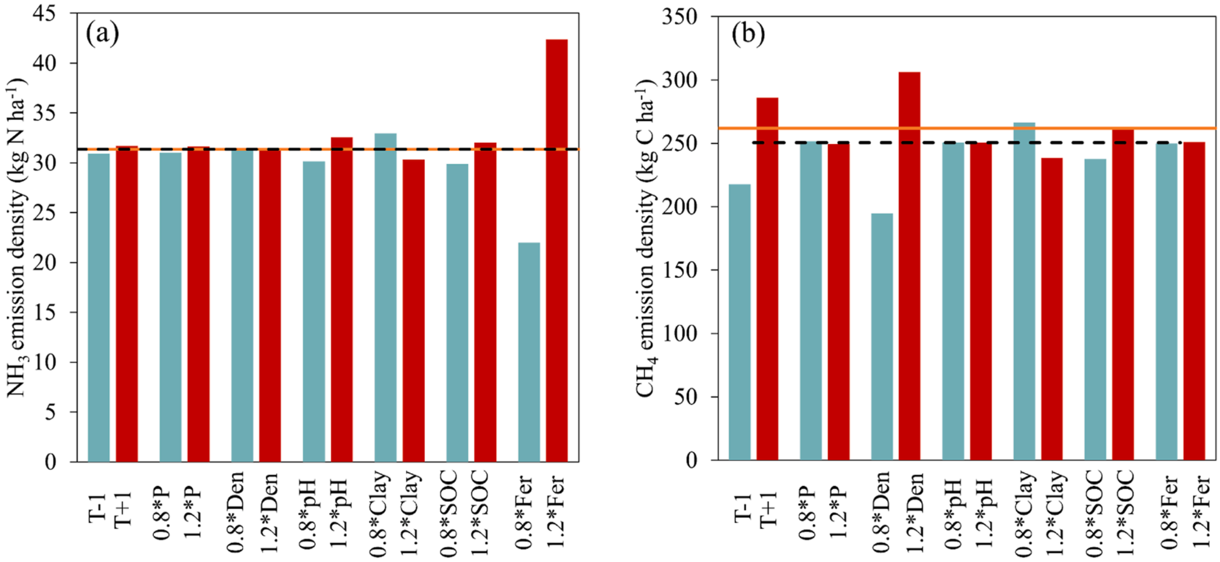


Fig. 6. Sensitivities of different variables for estimating (a) NH_3 emission densities and (b) CH_4 emission densities. T: air temperature (°C) (from "baseline-1" to "baseline+1"). P: precipitation (mm) (from 0.8 to 1.2 times the baseline). Den: soil bulk density (g cm^{-3}) (from 0.8 to 1.2 times the baseline). pH: soil pH (from 0.8 to 1.2 times the baseline). Clay: soil clay content (%) (from 0.8 to 1.2 times the baseline). SOC: soil organic carbon content (kg C kg^{-1}) (from 0.8 to 1.2 times the baseline). Fer: nitrogen fertilizer application (kg N ha^{-1}) (from 0.8 to 1.2 times the baseline). The orange line represented average observations value. The black line represented average simulated value in the baseline scenario.

model performance based on the ground-based measurements over the period 1992–2020. The simulated NH_3 and CH_4 emissions are generally consistent with the measurement (NH_3: R^2=0.80, RMSE =8.90 kg N ha^{-1}, n = 32; CH_4: R^2=0.81, RMSE =66.2 kg C ha^{-1} n = 30) (Fig. 1).

Sensitivity analysis was employed to assess the response of the DNDC simulated results to variation in input parameters (Qin et al., 2013). We conducted one baseline simulation and multiple scenario simulations with altered input parameters. Seven major input parameters were selected for the analysis: air temperature, precipitation, soil bulk density, soil clay content, soil pH, soil organic carbon (SOC) content, and nitrogen fertilizer application (Balasubramanian et al., 2017; Chu et al., 2023; Xu et al., 2023) (Fig. 6). These scenario simulations were performed across all observation sites to ensure representation. In each scenario, we adopted a single-factor variation method, wherein only one parameter was adjusted while maintaining others constant.

In general, annual NH_3 and CH_4 emission densities were within ±35 % and ±25 %, respectively (Fig. 6). Changes in temperature and the fertilizer applied to the three grain crops in China in the last decade were within ±0.5 °C and ±15 %, respectively, with relatively small changes. Therefore, the simulation results from a single year can be considered representative of the emission patterns in recent years. In future research, we will further use a climatology for the DNDC model simulation to study interannual variations in emissions.

Fertilizer application greatly affects NH_3 emissions, a 20 % increase in nitrogen fertilizer application could cause a 35 % increase in NH_3 emission density. Excessive use of nitrogen fertilizer is mostly lost through volatilization (Ahmed et al., 2017). In addition, soil pH and SOC had a positive impact on NH_3 emissions, while clay content inhibited NH_3 emissions, resulting in changes in NH_3 emissions within ±10 %. An increase in air temperature and bulk density significantly increases CH_4 emissions, with a 1 °C increase in air temperature and a 20 % increase in soil bulk density causing 14 % and 22 % increases in CH_4 emissions, respectively. High temperature significantly promotes the activity of organic matter fermentation (Yvon-Durocher et al., 2014), while high bulk density impedes soil permeability leading to oxidation of produced CH_4 (Hu et al., 2020a; Zhang et al., 2012), which ultimately promotes CH_4 emissions. SOC and clay content also play an important role in CH_4 emissions, causing CH_4 emission density variations in the range of ±7 %.

4.2. Limitations and implications of NH_3 and CH_4 mitigation strategies in agricultural cropping systems

This study established spatially differentiated strategies for synergistic pollution control and carbon reduction in China's agricultural cropping systems. The proposed mitigation strategies aimed to address the negative impacts of agricultural cropping systems on the environment and climate, providing integrated guidance for policymakers. Here, we focused on NH_3 and CH_4 emissions from major cropping systems comprising the three most important crops. Although only rice emits CH_4, however, maize (25.9 %) and wheat (14.8 %) are the significant sources of NH_3 emissions in Chinese agricultural cropping systems (Li et al., 2021a), which cannot be ignored. We recognized that livestock is also a major source of agricultural NH_3 and CH_4 emissions (Duan et al., 2023; Li et al., 2021a), and manure application indirectly affects crop emissions. However, given that the version of the DNDC model used in this study is unable to quantify NH_3 and CH_4 emissions from livestock, and due to the lack of detailed statistical data on the application of farmyard manure or green manure (Liu et al., 2022b; Wang et al., 2021b), emissions related to livestock were not considered in this study. In the future, livestock-related NH_3 and CH_4 emissions should be incorporated for a comprehensive assessment to provide guidance for pollution reduction and carbon mitigation across the entire agricultural sector.

Although cropping systems also emit nitrogen oxides (NO_x) and nitrous oxide (N_2O), again with negative impacts on climate and the environment (Chen et al., 2022a; IPCC, 2021; Jacob et al., 1996). However, NO_x emissions from the agricultural sector account for only 0.06 % of total emissions (Kim et al., 2023), which has a minimal impact. N_2O, on the other hand, is a long-lived greenhouse gas (> 100 yr) compared to CH_4 (12 yr), and the benefits of reducing its emissions are not significant in the short term. Therefore, we focused on exploring mitigation strategies for NH_3 and CH_4, which are prominent in agricultural cropping systems. This would lead to more immediate and significant effects in mitigating climate change and reducing environmental pollution (Ocko et al., 2021; Saunois et al., 2016).

We sorted out the current major NH_3 and CH_4 mitigation measures, most of which effectively reduce emissions (Table 3). The DNDC model was not used to test these strategies due to challenges in obtaining detailed agricultural management information for various regions. For instance, determining precise nitrogen fertilizer reduction amounts or optimal nitrogen-to-organic fertilizer ratios for each region is difficult. Future studies should employ the DNDC model to quantify the specific emission reduction potentials of these strategies. To ensure practical applicability, the mitigation strategies we selected were relatively low-cost and easy to implement at the spatial scale. For example, the average market price of organic and controlled-release fertilizers are only USD 157/t and USD 357/t (Zhang et al., 2020b), respectively. Measures with higher costs or limited applicability were excluded. In the future, the economic feasibility of implementing these mitigation measures needs to be further investigated through a cost-benefit analysis. In addition, attention should be paid to potential trade-offs and synergies between the various gases emitted from cropland when implementing NH_3 and CH_4 mitigation strategies. Although few strategies may lead to more other air pollutants or greenhouse gases (e.g., controlled irrigation significantly reduced CH_4 emissions by 79 % but slightly raised N_2O emissions by 10 %) (Yang et al., 2012), it is important to note that most of the suggested strategies have co-benefits. Specifically, our recommendation aimed at curbing NH_3 emissions, such as reducing N nitrogen fertilizer application, would decrease N_2O and NO_x emissions as well (Cui et al., 2024; Gu et al., 2023; Pan et al., 2022). This synergistic effect underscores the potential for our proposed strategies to contribute to broader environmental benefits beyond just NH_3 and CH_4 mitigation.

This study primarily proposes spatially oriented mitigation strategies for NH_3 and CH_4 emissions from agricultural cropping systems, with an emphasis on annual-scale approaches. Although intra-annual variations influence strategy implementation, developing specific, temporally refined emission reduction strategies is complex. Future research should address the impact of intra-annual temporal changes on mitigation strategies.

5. Conclusions

Differentiated and region-specific synergistic mitigation strategies are essential for addressing climate change and reducing environmental pollution However, there remains a significant research gap in the field of agricultural cropping systems regarding such strategies. Therefore, our study employed an integrated approach combining agricultural process modeling (DNDC model), spatial statistical analysis (Bivariate LISA method), and zoning mitigation strategies. This multifaceted methodology enables the establishment of targeted and spatially differentiated farmland management practices, thus contributing to the development of more effective and localized mitigation strategies in agricultural settings.

The total amount of NH_3 and CH_4 emissions from the three most important crops were estimated to be 3.04 Tg N yr^{-1} and 8.13 Tg C yr^{-1} in China in 2019, respectively. Bivariate LISA mapping revealed distinct regional hotspots for both combined and individual NH_3 and CH_4 emissions. Notably, the middle and lower reaches of the Yangtze River emerged as common hotspots of NH_3 and CH_4 emissions. Based on the identification of eight zone categories, region-specific emission mitigation strategies were developed. These strategies emphasize the implementation of effective management practices in farmland, particularly optimized fertilization, irrigation, and straw management.

Our findings indicate significant potential for mitigating NH_3 and CH_4 emissions from agricultural cropping systems. Future research should focus on comprehensively assessing and quantifying the potential impacts and net benefits of implementing these NH_3 and CH_4 mitigation strategies. Additionally, we recommend conducting cost-benefit analyses to evaluate the economic feasibility of various farmland management practices. This holistic approach will ensure the development of sustainable and economically viable mitigation strategies for agricultural emissions.

CRediT authorship contribution statement

Baojie Li: Writing – original draft, Software, Methodology, Data curation, Conceptualization. **Wanglijin Gu:** Writing – review & editing, Resources, Conceptualization. **Yongqi Zhao:** Resources, Investigation. **Zhifei Zhang:** Resources. **Xiaorui Wang:** Resources. **Yunkai Yang:** Data curation. **Zhihui Shen:** Investigation. **Hong Liao:** Supervision. **Qing Zhu:** Supervision.

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.

Data availability

Data will be made available on request.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.agrformet.2024.110250.

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