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Roles of historical land use/cover and nitrogen fertilizer application changes on ammonia emissions in farmland ecosystem from 1990 to 2020 in China

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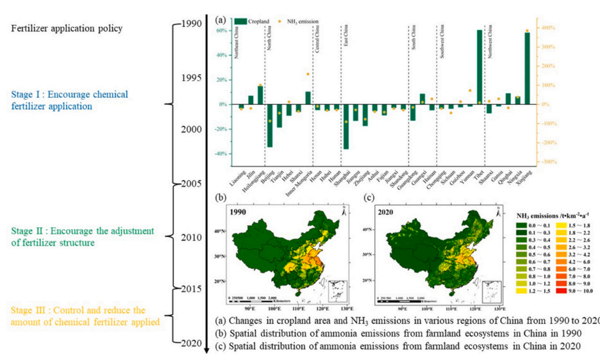
HIGHLIGHTS

- Over the past 30 years, NH_3 emissions in China's farmland ecosystem ranged between 3294.75 Gg and 4064.20 Gg.
- LULC and N-fertilizer application are important factors affecting NH_3 emissions from farmland ecosystem.
- The direction and the degree of strictness in policy directly affects the effect timeliness of NH_3 emission change.

GRAPHICAL ABSTRACT



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ABSTRACT

In the past decades, China has witnessed significant changes in its land use/land cover (LULC) pattern. These changes have led to a direct impact on ammonia (NH_3) emissions in soil background, and indirectly affected the total nitrogen fertilizer (N-fertilizer) application, crop planting amount and the resulting straw mass through the changes of cropland area. Great changes have also taken place in the amount and structure of fertilizer application in China, which affects the NH_3 emissions from farmland ecosystems caused by N-fertilizer application. The aforementioned changes have led to significant alterations in NH_3 emissions from China's farmland ecosystems over the past 30 years. The process of these changes remains to be analyzed, and the contributions of LULC changes and N-fertilizer application in this process are yet to be assessed. This study aims to investigate the NH_3 emission changes and spatiotemporal variation characteristics from farmland ecosystems during 1990 and 2020 due to the LULC changes. Additionally, the study employs scenario analysis method to discuss the effects of LULC changes and N-fertilizer application changes on NH_3 emissions in farmland ecosystems. Results indicate that there is evident spatiotemporal heterogeneity in China's LULC pattern, particularly in eastern China. The southeast region is predominantly characterized by the conversion of cropland into construction land. Moreover,

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some regions such as Northwest China and Northeast China have experienced the conversion of other land types into cropland, significantly influenced by national development policies. From 1990 to 2020, the national NH_3 emissions from farmland ecosystem range from 3294.75 Gg to 4064.20 Gg. NH_3 emissions and their interannual variation in farmland ecosystems exhibit significant differences across various regions. The regions with higher contributions to NH_3 emissions in farmland ecosystems are East China, Central China, and North China, accounting for 25.32 %–37.26 %, 18.85 %–22.46 % and 11.24 %–18.50 % of the total emissions, respectively. NH_3 emissions in each region are influenced by cropland area, N-fertilizer application, and regional development characteristics. Compared to LULC changes, changes in N-fertilizer application have a more pronounced impact on NH_3 emission changes in farmland ecosystems. From 1990 to 2020, the contribution (increase or decrease) of N-fertilizer application changes to NH_3 emission changes in farmland ecosystems in China ranges from 0.11 % to 16.61 %, while the contribution (increase or decrease) of LULC changes ranges from 0.47 % to 2.38 %. South China demonstrates a unique situation regarding the influence of LULC changes. This region has a relatively small cropland area, and fluctuations in cropland area significantly affect NH_3 emissions in farmland ecosystems. The influence of policies is evident. From the changes in cropland area in Northwest China and Northeast China to changes in N-fertilizer application, policy changes have consistently impacted the changes in NH_3 emissions in China's farmland ecosystems. From "soft policies" involving encouragement and guidance to "hard policies" encompassing the establishment of necessary targets, the degree of strictness in policy directly affects the timeliness of policies effectiveness. The results of this study indicate that reducing the application of N-fertilizers is the primary approach to reducing NH_3 emissions in China's farmland ecosystems. In terms of policy guidance, compared to implementing structural and pathway adjustments, implementing clear total control of fertilizer usage is a timely and effective choice for reducing NH_3 emissions.

1. Introduction

Ammonia (NH_3) plays a crucial role in the global nitrogen cycle and is the primary alkaline trace gas in the atmosphere (Galloway et al., 1996). It acts as a neutralizer for the oxidation products of sulfur dioxide and nitrogen oxide, participating in the formation of secondary inorganic salts and contributes to the increase in atmospheric fine particulate matter ($PM_{2.5}$) concentration, which has implications for urban visibility (Li et al., 2018). Secondary inorganic salts account for approximately 60 % of the total $PM_{2.5}$ mass (Huang et al., 2014; Ianniello et al., 2011; Niu et al., 2006; Wu et al., 2015), which is a critical factor in haze pollution formation in China (Huang et al., 2020). Reducing NH_3 emissions can effectively decrease the concentrations of secondary inorganic ions in the atmosphere and consequently reduce $PM_{2.5}$ concentrations (Ye et al., 2019). It has also been reported that NH_3 emissions have a greater impact on $PM_{2.5}$ concentrations than other $PM_{2.5}$ precursors, especially during severe haze conditions (Wu et al., 2016), which pose significant risks to human health (Behera et al., 2013; Pozzer et al., 2017). Mitigating NH_3 emissions by 38–67 % has the potential to reduce $PM_{2.5}$ concentrations by 8–20 % and prevent premature deaths of 90–240 thousand people (Zhang et al., 2020). Furthermore, NH_3 emissions can indirectly influence surface ozone concentrations. Liu et al. (2021) found that increasing agricultural NH_3 emissions would result in a 20 % rise in global nitrogen deposition and a 2–3 ppbv increase in surface ozone levels by 2050.

The sources of NH_3 emissions encompass various activities such as agriculture, biomass burning, human excrement, chemical production, waste disposal, and vehicle exhaust (Clarisse et al., 2009; Huang et al., 2012; Zeng et al., 2018). Agriculture serves as the primary source of NH_3 emissions. It is estimated that N-fertilizer application and livestock and poultry farming account for 57 % of global NH_3 emissions (Bouwman et al., 1997). The prevalence of intensive agricultural practices has led to a rapid increase in NH_3 emissions during the 20th century (Erisman et al., 2008; Galloway et al., 2004; Galloway et al., 2008). Global agricultural NH_3 emissions increased by 78 % from 1980 to 2018, with a 128 % increase in farmland emissions and a 45 % increase in livestock emissions (Liu et al., 2022). For Asian countries, the contribution reached as high as 80 % (Streets et al., 2003; Kurokawa et al., 2013). In China, NH_3 from N-fertilizer application and livestock and poultry farming accounts for 88 % of total anthropogenic NH_3 emissions (Zhang et al., 2018).

Small farm size has caused the insufficient driving force for the input of new agricultural technologies (e.g. soil testing and deep placement of fertilizers by machinery), which is one of the main drivers of high farmland NH_3 emissions in China (Gu et al., 2020). Xu et al. (2022) discovered that replacing 40–60 % of synthetic fertilizer with livestock manure can achieve the greatest synergy by improving crop yield and reducing NH_3 emissions. Over the past decades, China's total NH_3 emissions have increased by 2.4 times, from 4.7 $Mt \cdot a^{-1}$ (in terms of nitrogen) in 1980 to 11 $Mt \cdot a^{-1}$ in 2016 (Fu et al., 2020).

Previous researches about agricultural NH_3 emissions have focused on analyzing the emission trends and studying their temporal and spatial distribution characteristics. For instance, Xu et al. (2019) employed a process-based Dynamic Land Ecosystem Model (DLEM) coupled with the bidirectional NH_3 exchange module in the Community Multiscale Air-Quality (CMAQ) model (DLEM-Bi-NH_3). This integration could assess the temporal and spatial characteristics of NH_3 emissions at both global and regional scales, including crop-specific NH_3 emissions. In another study, Ma et al. (2021) utilized bibliometric methods to establish a comprehensive dataset of NH_3 emissions resulting from agricultural nitrogen fertilizer application on a global scale. They further examined the variations in NH_3 emissions and emission factors across different regions. Their research provided valuable insights into the spatial changes of NH_3 emissions originating from soil sources due to the use of synthetic nitrogen fertilizer in agriculture, both globally and regionally. Furthermore, Chang et al. (2019) conducted a comprehensive evaluation of the contribution of agricultural and non-agricultural emissions to atmospheric NH_3 in China's megacities. They employed a combination of monitoring experiments, numerical simulations, and isotope-based estimates of environmental NH_3 source allocation. The findings from their study revealed that both non-agricultural and agricultural sources significantly contributed to urban atmospheric NH_3 levels.

With the rapid urbanization and mobility process in China over the past three decades, there have been significant changes in land use/land cover (LULC) in certain regions (Luo and Zhang, 2022). These changes have led to direct impact on NH_3 emissions in soil background, and indirectly affected the total N-fertilizer application, crop planting amount and the resulting straw amount through the change of cropland area. In recent decades, great changes have also taken place in the amount and structure of fertilizer application in China (Guo and Wang, 2021; He et al., 2020), which affects the NH_3 emissions of farmland ecosystems caused by nitrogen fertilizer application. According to a study by Yang et al. (2022b), a reduction of 26 % in NH_3 emissions from farmland can be achieved if the annual N-fertilizer amount is decreased by 0.5 % from 2010 to 2100. However, research on the long-term impact of LULC changes and N-fertilizer application on NH_3 emissions remains unclear due to lack of a dynamic evaluation of NH_3 emissions from farmland ecosystems. Therefore, this study aims to investigate the impact of historical LULC changes and N-fertilizer application as well as policies on NH_3 emissions in China's farmland ecosystems. By utilizing remote sensing inversion results from Landsat satellite data, LULC changes from 1990 to 2020 are investigated. Sensitivity analysis is further conducted to examine the influence of LULC changes and N-fertilizer application changes over the past 30 years on NH_3 emissions in farmland ecosystems. The policy impact on NH_3 emissions during different stages is also discussed. The findings of this research will guide to future polices on NH_3 emission control.

2. Methodology

2.1. LULC change interpretation from satellite remote sensing images

To characterize the land surface changes over China, LULC data were extracted from Landsat-5/7/8 imageries acquired from the official website of the United States Geological Survey (USGS). These data cover the LULC scope of 31 provinces and autonomous regions in mainland China, which are divided into seven regions: Northeast China, North China, Central China, East China, South China, Southwest China, and Northwest China, as what we did in our previous study (Huang et al., 2021). The list of provinces under each region can be found in Table S1. We extracted the data of year 1990, 1995, 2000, 2005, 2010, 2015, and 2020 and applied to a 36 km × 36 km grid using the ArcGIS 10.6 software. The classification system adopted for this data is the land cover I and II classification system, which has six categories: forest, grassland, wetland, cropland, construction land, and other land.

2.2. NH_3 emissions calculation

The main sources of NH_3 emissions in farmland ecosystems encompass N-fertilizer application, soil background, nitrogen-fixing plants, and straw composting. In this study, the NH_3 emissions ($E(NH_3)$) were calculated using the emission factor method, as outlined in Eq. (1):

$$E(NH_3) = \sum_{i,j} A_{i,j} EF(NH_3)_{i,j} \tag{1}$$

where $A_{i,j}$ is the activity data of regions (i) and sources (j); $EF(NH_3)_{i,j}$ is the emission factor data of the different regions and sources. The calculations for $E(NH_3)$ from the different source sectors are described in the supplement material.

2.3. Setup of scenarios

The historical NH_3 emissions in farmland ecosystems during 1990 to 2020 are driven by both changes in LULC and N-fertilizer application. To analyze their effects on agricultural NH_3 emission, thirteen scenarios were established and classified into three main categories (Table S2).

S_1 represents a fundamental category that depicts the actual scenario of NH_3 emissions from farmland ecosystems in China during 1990. By dividing the NH_3 emissions generated by the N-fertilizer application part in 1990 by the cultivated land area in that year, we can get the NH_3 emissions generated by the N-fertilizer application part per unit area under the N-fertilizer application level in 1990, which is called η. The category S_2 is established with η. S_2 selects the cropland data of 1995, 2000, 2005, 2010, 2015 and 2020. Among the four source sectors of NH_3 emission in farmland ecosystem, NH_3 emissions from soil background, nitrogen-fixing plants and straw composting were selected in the corresponding year, while NH_3 emission from N-fertilizer application was calculated by multiplying the corresponding year's cropland area by η. Based on the above content, the category S_2 is established, which is the NH_3 emission level of farmland ecosystem in different years when the N-fertilizer application condition is always the 1990 level. By comparing S_1 and S_2 categories, the effects of LULC changes on NH_3 emission from farmland ecosystem were dissected. After this, the category S_3 is established, which is the actual NH_3 emissions from farmland ecosystems in 1995, 2000, 2005, 2010, 2015 and 2020. By comparing S_2 and S_3 categories, the effects of N-fertilizer application changes on NH_3 emissions from farmland ecosystem were dissected.

3. Results and discussion

3.1. LULC changes in China from 1990 to 2020

The changes in various land areas across China from 1990 to 2020, with a resolution of 36 km × 36 km based on Landsat remote sensing interpretation, are depicted in Figs. S1–S6. From the spatial distribution perspective, there is evident spatiotemporal heterogeneity between the southeastern and northwestern sides of the Hu Huanyong Line (Hu Line) (Kong et al., 2022), the location of which is shown in Fig. 1. Grassland and other land are predominantly found on the northwest side of the Hu Line, while the southeast side consists of a higher proportion of forest, cropland, and construction land. The distribution of wetland is more dispersed.

By comparing the changes in cropland and construction land from 1990 to 2020 (Fig. 1), it can be observed that the spatiotemporal heterogeneity along the Hu Line is also evident in other regions of China, excluding the Northwest China and Northeast China. The southeastern side of the Hu Line is densely populated, and over the past three decades of rapid economic and urbanization development, a significant amount of cropland has been converted into construction land. Consequently, there is a noticeable overlap between the increase in construction land and the decrease in cropland. However, in Northwest China, particularly in the Xinjiang Uygur Autonomous Region, and Northeast China, economic development has been relatively slower, resulting in a smaller increase in construction land. The significant expansion of cropland in these two regions can be attributed to national policies such as "West China Development" and "Great Northern Wilderness Development," which have facilitated the reclamation of a substantial amount of wasteland (Wang et al., 2022).

3.2. Changes of NH_3 emissions in farmland ecosystems from 1990 to 2020

As depicted in Fig. 2a, there are notable variations in NH_3 emissions and their interannual fluctuations within farmland ecosystems across different regions. Surprisingly, the regions exhibiting a high contribution of NH_3 emissions from farmland ecosystems are East China (25.32 %–37.26 %), Central China (18.85 %–22.46 %), and North China (11.24 %–18.50 %), which contradicts the ranking based on cultivated land area in each region (Table S1). Although Northeast China possesses a large expanse of cropland, its regional farmland ecosystem emits relatively less NH_3. This can be attributed to the adoption of "deep ploughing and careful cultivation" practices, a balanced application of chemical and organic fertilizers, and the pursuit of "reducing the amount and increasing the efficiency" of fertilizer usage in this region. Conversely, Central China, despite having a moderate cropland area compared to other regions, experiences significant NH_3 emissions in its farmland ecosystem. This is primarily due to its status as a major grain-producing region in China, where croplands are predominantly used for food crops. To ensure high grain yields, a substantial amount of chemical fertilizers is applied. The application of N-fertilizers, crop types, and straw composting all contribute to the upper limit of ammonia emission from the farmland ecosystem in Central China. In 1990, the NH_3 emissions from the farmland ecosystem in East China reached their highest level in the past 30 years, primarily driven by the early rapid economic development in that region and the unregulated use of chemical fertilizers during the initial stages. Although subsequent years witnessed fluctuations, an overall declining trend was observed. The annual

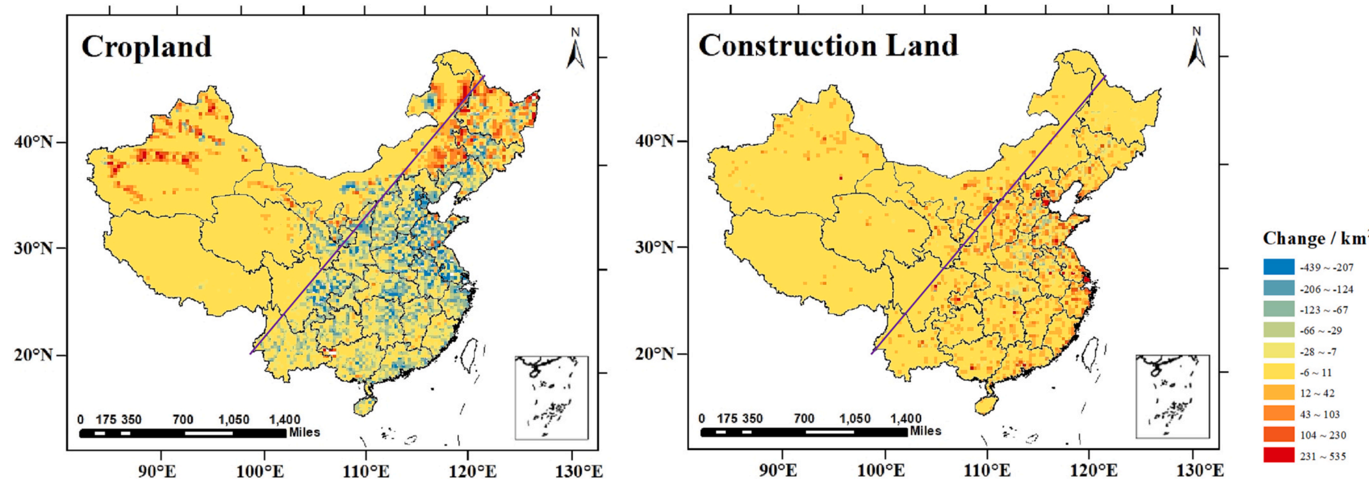

Fig. 1. Changes in cropland area and construction land area in China from 1990 to 2020 (the black line in the figure is Hu Huanyong Line).

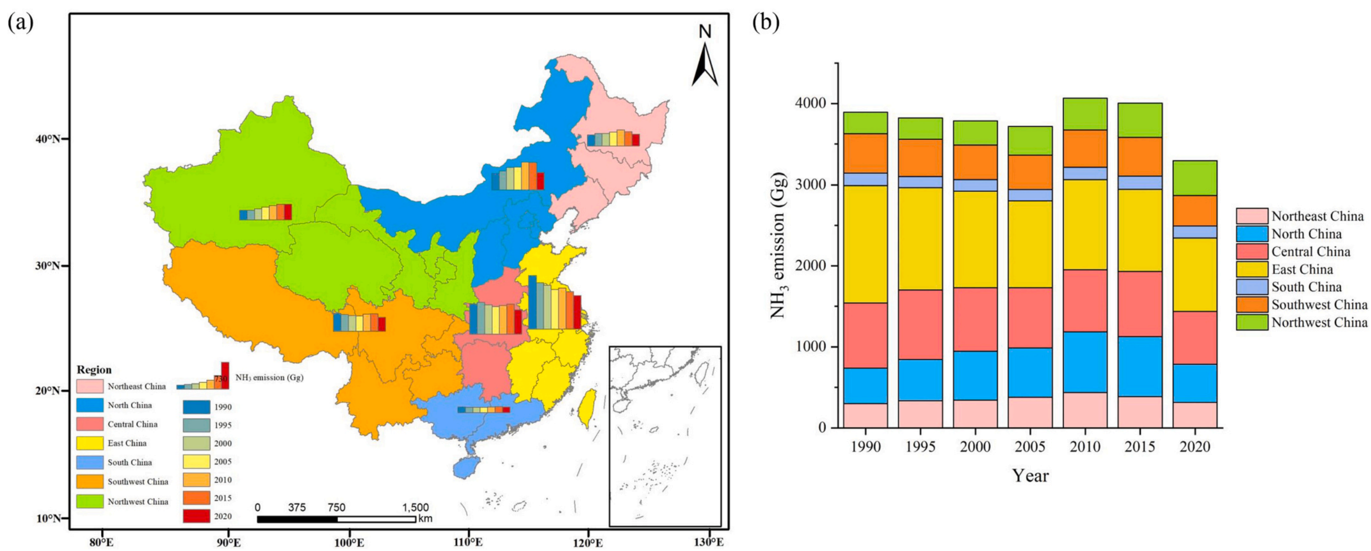

Fig. 2. NH_3 emissions from farmland ecosystems by regions in China from 1990 to 2020.

increase in NH_3 emissions from the farmland ecosystem in Northwest China can be attributed to the expansion of cropland resulting from the "West China development" initiative mentioned earlier. Among the seven regions in China, the farmland ecosystem in South China exhibits the lowest NH_3 emissions. This can be attributed, on one hand, to the relatively small area of cropland in this region and, on the other hand, to the high acidity of the soil in the region. From 1990 to 2020, NH_3 emissions in China's farmland ecosystem ranged from 3294.75 Gg to 4064.20 Gg. Analyzing the temporal variation of NH_3 emissions in China's farmland ecosystem (Fig. 3b), it is evident that over the past 30 years, it underwent a process characterized by a minor initial decline, followed by a significant rise, another minor decline, and ultimately a substantial decrease. This trend may be influenced by changes in cropland area and variations in the application of N-fertilizers, necessitating further investigation in subsequent research.

By analyzing the changes in cropland and NH_3 emissions across China's provinces from 1990 to 2020, it is evident that different provinces exhibit distinct changing patterns. The following five situations can be identified based on the observed trends:

(1) In economically developed and highly urbanized provinces such as Beijing, Tianjin, and Shanghai, the cropland area and NH_3 emissions from farmland ecosystems have significantly reduced, with decreases of 18.58 %–36.20 % and 45.54 %–90.94 %, respectively. This decline can be attributed to urbanization processes.

(2) In provinces like Heilongjiang, Inner Mongolia, and Xinjiang, the cropland area and NH_3 emissions from farmland ecosystems have increased by 10.40 %–58.37 % and 100.71 %–384.95 %, respectively, over the past three decades. This growth can be attributed to the implementation of policies such as the "West China development" and "Great Northern Wilderness development," which have stimulated local agricultural activities. However, the situation in Tibet is slightly different. In the past 30 years, the cropland area has increased significantly by 60.42 %, while NH_3 emissions have increased only slightly by only 9.25 %. This can be explained by two factors: First, highland barley is the main crop in Tibet, accounting for more than 50 % of Tibet's planted crops, and nitrogen-fixing crops are rarely cultivated, resulting in lower N-fertilizer application compared to other provinces. Second, although the rate of change in cropland area in Tibet appears high, the total cropland area is actually quite small (Table S1).

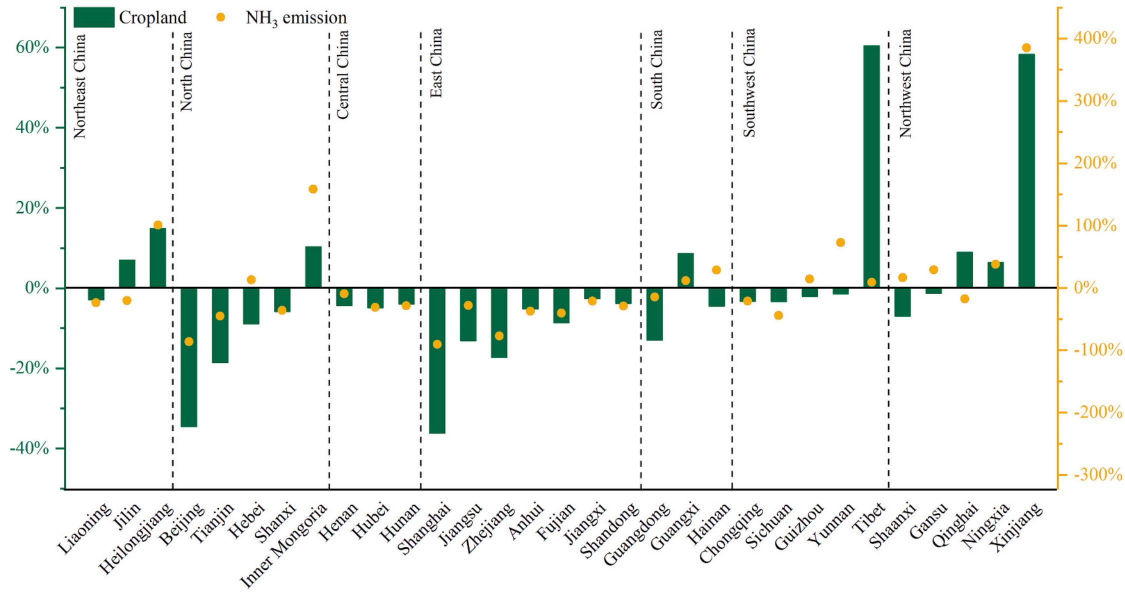

Fig. 3. Changes in cropland area and NH_3 emissions in various regions of China from 1990 to 2020.

(3) In Jilin and Qinghai provinces, NH_3 emissions have decreased while the cropland area has increased. In Jilin and Qinghai provinces, NH_3 emissions decreased with the increase of cultivated land area. Over the past 30 years, the cropland area in the two provinces has increased by 7.03 % and 9.02 % respectively, and NH_3 emissions have decreased by 20.44 % and 17.67 % respectively. This phenomenon could be attributed to changes in local crop planting structure or fertilization structure. For example, in the past 30 years, the area under bean cultivation in Jilin has decreased by 30.76 %. N-fertilizer application in Jilin and Qinghai decreased by 8.39 % and 22.96 %, respectively during 1990 to 2020.

(4) Provinces with limited economic development and small per capita cropland area, such as Hebei and Yunnan, may still experience excessive fertilization due to the pursuit of economic benefits. Over the past 30 years, N-fertilizer application in Hebei and Yunnan increased by 9.24 % and 166.00 %, respectively. Consequently, despite a decrease in cropland area (8.94 % in Hebei and 1.49 % in Yunnan), NH_3 emissions tend to increase in these regions (13.26 % in Hebei and 73.02 % in Yunnan).

(5) Apart from the provinces falling into the aforementioned categories, the cropland area and NH_3 emissions in the remaining provinces have exhibited some fluctuations but overall have remained relatively stable.

3.3. *Cause analysis of NH_3 emission change in farmland ecosystem*

3.3.1. *Roles of LULC and N-fertilizer application changes on NH_3 emissions from farmland ecosystems*

Overall, NH_3 emissions from China's farmland ecosystems exhibited significant fluctuations between 1990 and 2020 (Fig. 4). The impact of N-fertilizer application changes on NH_3 emission changes in farmland ecosystems were found to be more significant compared to LULC changes. From 1990 to 2020, the contribution (increase or decrease) of N-fertilizer application changes to NH_3 emission changes in farmland ecosystems in China ranges from 0.11 % to 16.61 %, while the contribution (increase or decrease) of LULC changes ranges from 0.47 % to 2.38 %. The contribution rates of N-fertilizer application changes and LULC changes to NH_3 emission changes from farmland ecosystems in different regions of China from 1990 to 2020 are shown in Table S3. Analyzing the variation patterns across different regions reveals that LULC change had a more pronounced influence in South China. During 1995 to 2000, LULC change contributed 7.82 % of the increase in NH_3 emissions in South China. This can be attributed to the fact that South China has the smallest cropland area among all regions, and a fluctuation of cropland area will have a more significant impact on NH_3 emission in this region than in other regions. N-fertilizer application changes in East China have effectively reduced NH_3 emissions from farmland ecosystems in most years. East China experienced early economic development, leading to peak N-fertilizer application in the 1980s. Consequently, people became aware of the drawbacks of excessive fertilizer application earlier, prompting adjustments in fertilizer quantity and composition (Li, 2008). Between 2005 and 2010, changes in N-fertilizer application in each region positively contributed to NH_3 emissions in regional farmland ecosystems. However, except for northwest China, N-fertilizer application in other regions made a significant negative contribution to NH_3 emissions between 2015 and 2020. This phenomenon warrants further investigation into its underlying causes.

3.3.2. *Analysis of the influence of related policies implemented*

As mentioned earlier, the alteration in N-fertilizer application has had a noticeable impact on NH_3 emission changes within China's farmland ecosystems. Our study revealed that during the periods from 2005 to 2010 and from 2015 to 2020, changes in N-fertilizer application generally resulted in a significant "one increase and one decrease" effect on NH_3 emissions in farmland ecosystems across different regions of China. To delve into the underlying reasons behind these two changes, we compiled all 31 national-level policies and measures related to fertilization in China from 1990 to 2020 (Table S4). These policies and measures can be broadly categorized into three stages (Fig. 5). The first stage (1990–2005) focused on agricultural output, emphasizing the adequate supply of chemical fertilizers and promoting their application. However, during this stage, limited attention was given to the potential drawbacks of excessive chemical fertilizer use. The second stage (2005–2015) witnessed a shift towards adjusting the application ratio of chemical fertilizers based on local conditions and the development of organic fertilizers. There was an increasing recognition of the need to adopt more environmentally-friendly fertilization practices. The third stage (2015–2020) aimed at controlling or reducing the number of chemical fertilizers applied in accordance with local conditions. It emphasized the importance of applying fertilizers in a scientifically guided manner to reduce their quantity while increasing efficiency. By analyzing these stages, we aim to uncover the underlying factors contributing to the observed changes in NH_3 emissions, providing

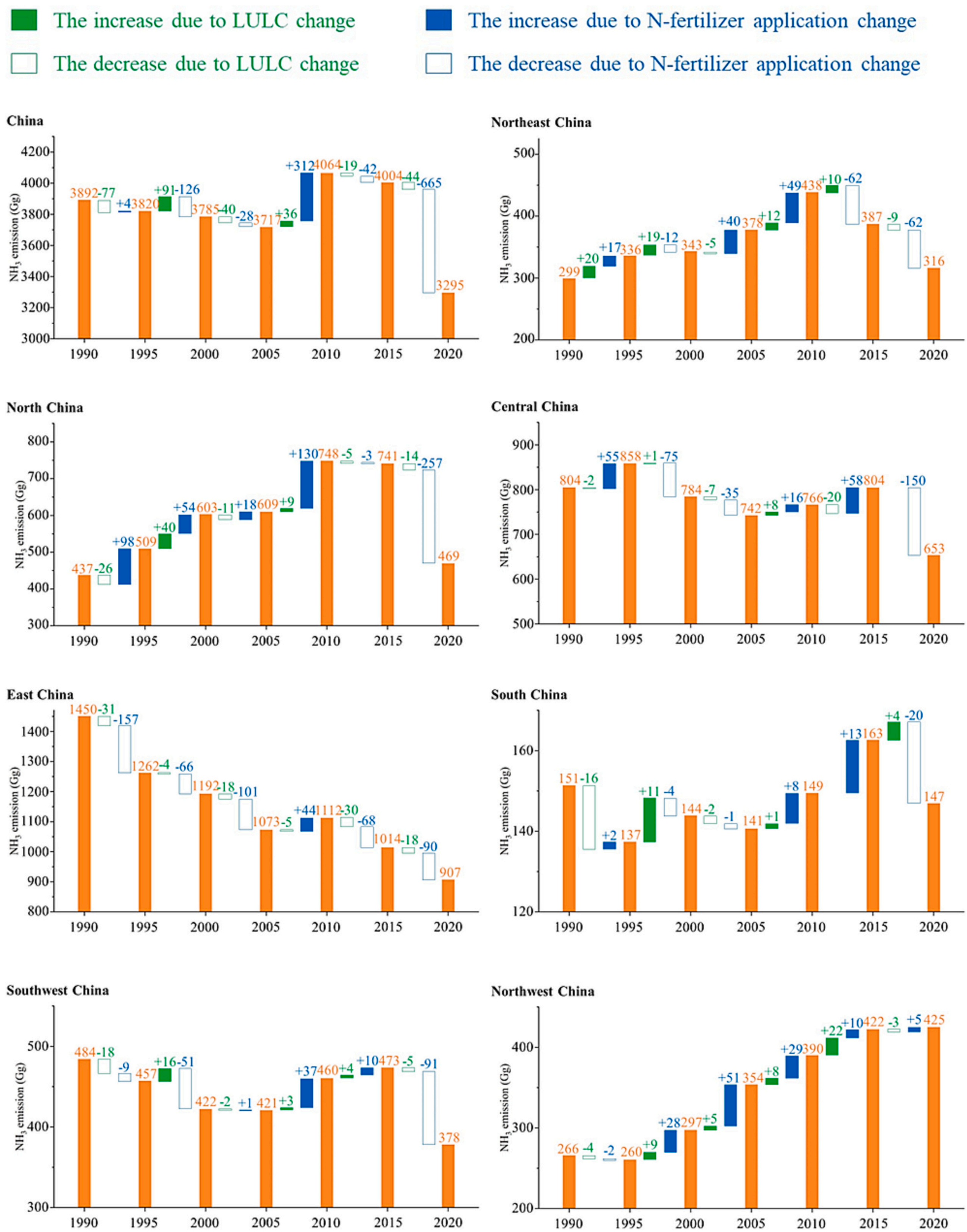

Fig. 4. Change of NH_3 emission caused by land use/land cover and N-fertilizer application/Gg.

valuable insights for sustainable agricultural practices in China.

From the perspective of China's fertilization policy, it appears that the period between 2005 and 2010 marks the onset of the second stage mentioned earlier. It would be expected, based on common sense, that fertilizer application in China should have decreased to some extent during this period. However, the reality was quite the opposite. In fact, the application of N-fertilizers underwent a significant increase, leading to elevated NH_3 emissions from farmland ecosystems in China. This

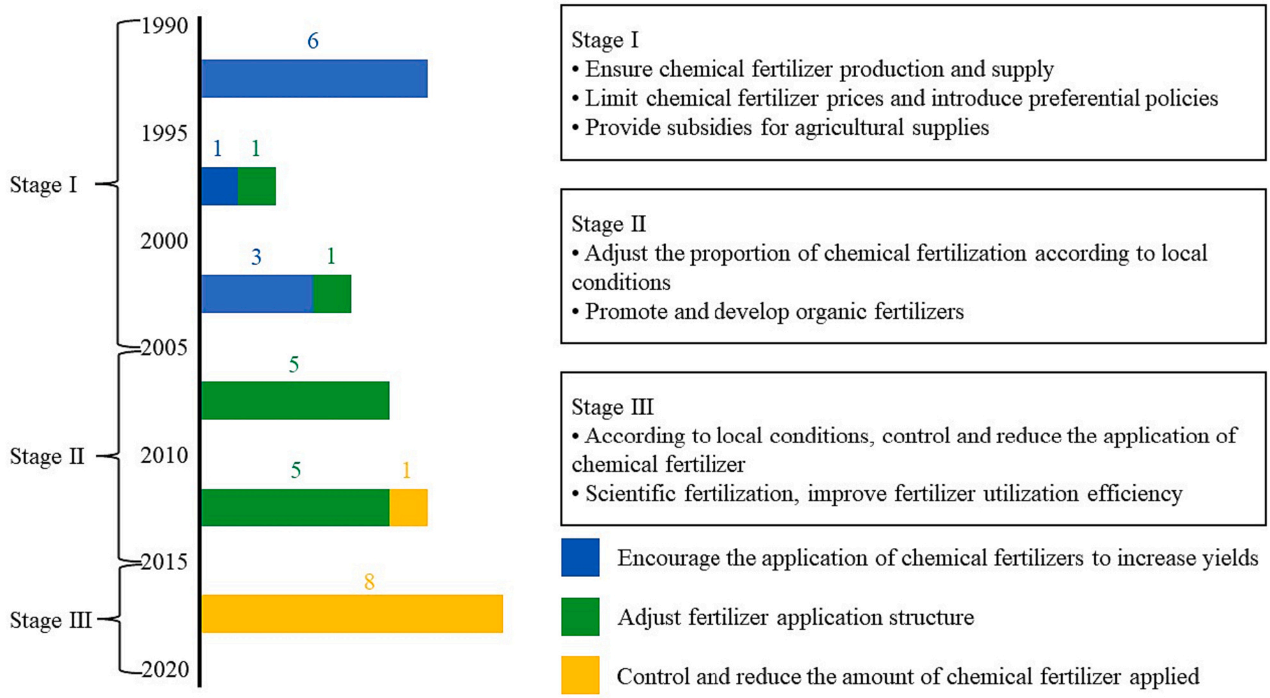

Fig. 5. Changes and development schedules of fertilizer application policies in China.

phenomenon could possibly be attributed to a certain "lag" resulting from the implementation of the first and second stages of the policy, as discussed by Yang et al. (2022a) to some extent. On the other hand, the period between 2015 and 2020 represented another phase of change in the fertilization policy, and it seems that the policy effect was immediate during this time. The impact of changes in nitrogen fertilizer application on NH_3 emissions from farmland ecosystems effectively demonstrated the success of the third stage policy in promoting fertilizer reduction and efficiency improvement. Notably, no "lag" was observed in the implementation of the third policy during this period.

Therefore, we propose the concept of "soft policy" and "hard policy". "Soft policy" resembles the initial and intermediate stages of China's fertilizer application policy in our research. It involves creating favorable objective conditions for achieving policy objectives, advocating the realization of policy goals, and relying to a greater extent on the initiative of those implementing the policy. On the other hand, "hard policy" refers to the type of policies mentioned in the third stage of our study, which stipulate specific implementation measures based on goals and demands, providing implementers with clear targets. The implementation of "soft policies" often exhibits delayed and uncertain effects, whereas the implementation of "hard policies" tends to be effective immediately, with the differences primarily lying in the degree of effectiveness. In the realm of environmental governance, "hard policy" undoubtedly emerges as a more efficient and dependable choice. Nevertheless, it is worth noting that "soft policies", despite occasional delays, can also produce unintended consequences due to their inherent flexibility.

3.4. Comparison with previous studies

Table S5 presents a comparison of NH_3 emissions from farmland ecosystems in our study with previous studies. Overall, our findings align with those of Zhao and Wang (1994), Yan et al. (2003), and Huang et al. (2012). However, Kang et al. (2016) and Liao et al. (2022) employed a different approach to calculate NH_3 emissions, focusing on the application of N-fertilizer using widely grown crops and reference values for various crop fertilizer application rates. In contrast, our study collected fertilizer application data from provincial statistical yearbooks and multiplied it by the corresponding coefficient to estimate NH_3 emissions resulting from N-fertilizer application. Consequently, their results for N-fertilizer application were slightly lower than ours. Considering the fertilization situation in China, the complexity of fertilizer application has increased due to the prevalence of small farms (Gu et al., 2020). Excessive fertilization has been a persistent issue on croplands in China over the past few decades (Liao et al., 2022). Consequently, estimating the fertilization situation solely based on crop planting might lead to underestimation. In Huang et al.'s study, the contribution of compost to emissions was significantly higher compared to our study (Huang et al., 2012). However, both studies utilized the same emission factors. The divergence lies in the calculation method, as we referred to a relatively recent article (Zhou et al., 2017) for our approach. Furthermore, when comparing our study with others, it is worth noting that our estimation of NH_3 emissions from straw compost aligns more closely with the findings of other studies. Zhang et al. (2018) selected a higher emission factor for fertilizer application, suggesting that urea topdressing and surface application could substantially increase NH_3 emissions. Ma (2020) did not consider differences in NH_3 emissions caused by different types of fertilizers in their calculation of fertilizer application. Instead, they employed uniform emission factors, resulting in relatively high NH_3 emissions.

Satellite observations provide an additional means of verifying NH_3 emissions. Although the NH_3 emissions from the farmland ecosystems studied in our research represent only a portion of the total ammonia emissions, which cannot be quantitatively validated through satellite observations, they can still be qualitatively confirmed based on their spatial distribution. Our findings (Fig. S7) exhibit a strong concurrence with the spatial patterns of NH_3 emissions and the vertical column densities (VCDs) of NH_3 measured by the infrared atmospheric sounding interferometer (IASI) satellite for the year 2015 across China, as demonstrated in a previous study (refer to Fig. 1 in Pan et al., 2018). The areas characterized by high NH_3 emissions, such as North China, Sichuan Basin, and Northeast China, are accurately estimated in our study. Additionally, our study effectively validates the previously overlooked high NH_3 emissions in Xinjiang, which were not adequately captured in previous study (Zhang et al., 2017). This confirmation supports the notion that our study provides an accurate depiction of the spatial distribution of NH_3 emissions in China.

3.5. Uncertainty analysis

Generally, there is uncertainty in interpreting infrared aerial remote sensing images, especially when extracting data on LULC types. In the process of extraction and classification, there may exist uncertainties arising from subjective judgment, category ambiguity, and differences in extraction methods. The Landsat dataset typically employs the random forest method as a classifier for LULC classification (Su et al., 2023). Its overall classification accuracy can reach over 80 %, with a recognition accuracy for farmland exceeding 95 % (Zhao et al., 2019). Comparing the interpretation results of farmland in this study with previous research (Table S6), it can be observed that the interpretation results are quite similar, indicating a high level of credibility. Therefore, despite the current uncertainties in land use/land cover results, they are relatively small and do not significantly impact the main conclusions of this study. To further reduce the aforementioned uncertainties in the future, it may primarily depend on further improvements and enhancements in classification methods.

Additionally, there are uncertainties in estimating NH_3 emissions, primarily due to challenges in accurately obtaining and aligning activity data and emission factor data. The EF(NH_3) used in this study for the farmland ecosystem is sourced from the Technical Guidelines for the Preparation of Atmospheric Ammonia Emissions Inventory by the Ministry of Ecology and Environment of the People's Republic of China (2014). However, the emission factor is adjusted based on the specific conditions in this region (eg. temperature, precipitation, pH and amount of fertilizer usage), although complete localization of parameter selection may not be achieved. Consequently, the selected emission factor may have an impact on the estimated NH_3 emission results. Moreover, the data on the amount of N-fertilizer application, as well as the area and yield of nitrogen-fixing plants, were obtained from the municipal statistical yearbook (with partial data from the published bureau of agriculture and rural areas or provincial statistical yearbook), while individual data were derived from calculations or literature. Collectively, these aforementioned factors exert a certain influence on NH_3 emissions from all sources. The emission factor of NH_3 may also vary to some extent with climate factors such as temperature and precipitation, which are not directly considered in the emission inventory technical guidelines at present, leading to some uncertainty in the calculation of NH_3 emissions. In this study, regional variations were considered, indirectly accounting for temperature factors. Soil pH was also taken into account as an influencing factor in the calculation of NH_3 emissions from nitrogen fertilizer application, which indirectly considers precipitation. Of course, in the future development of emission inventories, a more comprehensive consideration of these climate factors may further improve the accuracy of NH_3 emissions in farmland ecosystems.

Additionally, although efforts have been made to separate the impact of LULC in scenario setting, it is not possible to completely eliminate other indirect potential factors. In this study, although an attempt has been made to disentangle the contributions of LULC changes and N-fertilizer application changes to NH_3 emissions in farmland ecosystems through scenario analysis, it should be noted that LULC changes can indirectly affect the N-fertilizer application amounts by altering crop planting areas and crop planting structures. This impact exhibits regional variability and is challenging to precisely quantify.

Overall, the selection of emission factors and activity levels for calculating NH_3 emissions in farmland ecosystems adopted in this study is a common choice for bottom-up assessments of NH_3 emission levels. This study has made efforts to account for regional and species differences in the emission factors and activity level data as much as possible. Section 3.4 provides a comparison with previous studies, which further underscores the credibility of this calculation methodology. Therefore, while there is a certain degree of uncertainty associated with NH_3 emissions in farmland ecosystems, this does not alter the conclusion drawn earlier that changes in N-fertilizer application have a more significant impact on NH_3 emissions in farmland ecosystems than LULC changes.

4. Conclusions

Over the past three decades, China has experienced significant changes in its LULC pattern. These changes have had a direct impact on NH_3 emissions in soil background, and indirectly affected the total N-fertilizer application, crop planting amount and the resulting straw mass through the change of cropland area. Great changes have also taken place in the amount and structure of fertilizer application in China, which affects the NH_3 emissions of farmland ecosystems caused by N-fertilizer application changes as well as policy constraints. This study utilized Landsat series satellite remote sensing images to interpret LULC data, analyzed LULC change, and estimated the long-term changes of NH_3 emissions in farmland ecosystem from 1990 to 2020 using the bottom-up method. Furthermore, the study examined the spatio-temporal variation characteristics of NH_3 emissions in the farmland ecosystem and discussed the effects of LULC change and N-fertilizer application on NH_3 emissions through scenario analysis. The results demonstrated that the spatiotemporal heterogeneity of LULC patterns in China is evident on either side of the Hu Huanyong line. The south-eastern side of the Hu Line, characterized by a large population and a developed economy, primarily exhibits a transformation of croplands into construction lands. Moreover, the Northwest China and Northeast China have experienced the conversion of other land types into croplands, a shift significantly influenced by national development policies. From 1990 to 2020, NH_3 emissions in China's farmland ecosystem ranged from 3294.75 Gg to 4064.20 Gg. There were notable variations in NH_3 emissions and their interannual fluctuations among farmland ecosystems in different regions. The regions of East China, Central China, and North China exhibited a comparatively higher contribution to NH_3 emissions in farmland ecosystems, accounting for 25.32 %–37.26 %, 18.85 %–22.46 % and 11.24 %–18.50 % of the total contribution, respectively. The NH_3 emissions in each region's farmland ecosystems were influenced by factors such as cropland area, N-fertilizer application, and regional development characteristics. Through scenario analysis, we examined the impacts of LULC changes and N-fertilizer application changes on NH_3 emissions in agricultural ecosystems. Our findings indicate that N-fertilizer application changes have a more pronounced effect on NH_3 emission changes compared to LULC changes. From 1990 to 2020, the contribution (increase or decrease) of N-fertilizer application changes to NH_3 emission changes in farmland ecosystems in China ranges from 0.11 % to 16.61 %, while the contribution (increase or decrease) of LULC changes ranges from 0.47 % to 2.38 %. LULC change in South China exhibits unique characteristics. During 1995 to 2000, LULC change contributed 7.82 % of the increase in NH_3 emissions in South China. The relatively small cropland area in this region contributes significantly to the fluctuations in NH_3 emissions within the farmland ecosystem. Moreover, the impact of policies is evident. Policy changes have consistently influenced NH_3 emissions in China's farmland ecosystems, as observed through the alterations in cropland area in northwest China and Northeast China, as well as changes in N-fertilizer application. The effectiveness of the policy varies depending on its level of stringency, ranging from the supportive and advisory nature of 'soft policies' to the firm and goal-oriented approach of 'hard policies.' The degree of toughness directly determines the promptness of the policy's impact.

CRediT authorship contribution statement

N.C. Shen and W. J. Wang conducted the data analysis and calculation. N. C. Shen prepared the manuscript with contributions from all coauthors. Q. Wang and M. Wang contributed to land use/cover data interpretation. J. N. Tan, L. Huang and Y. J. Wang contributed to data interpretation. L. Li provided the initial motivation to this study, designed the research, edited and reviewed the paper.

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.scitotenv.2023.167565.

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