https://doi.org/10.5194/egusphere-2025-3090
Preprint. Discussion started: 10 July 2025

Spatial-temporal variations of atmospheric NH_3 concentration and its dry deposition across China based on one decade of satellite and ground-based observations

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https://doi.org/10.5194/egusphere-2025-3090
Preprint. Discussion started: 10 July 2025

Abstract: Ammonia (NH_3), a key alkaline gas in the atmosphere, significantly influences ecosystem nitrogen cycling and the formation of fine particulate matter ($PM_{2.5}$). However, limited ground-based monitoring hinders understanding of NH_3's spatial and temporal dynamics and its dry deposition across China, which is ranked as one of global largest NH_3 emission hotspots. This study integrated 2013–2023 satellite-derived NH_3 column concentrations from the Cross-track Infrared Sounder (CrIS) with ground in-situ observations. We used the GEOS-Chem transport model and a random forest algorithm to simulate NH_3 dry deposition fluxes and explore the driving forces behind observed trends. Our results show that NH_3 concentrations were the highest in the North China Plain (>10 ppb), with notable annual and seasonal increases. NH_3 concentration in 2023 were 14–31% higher than in 2013. CrIS retrievals aligned well with in-situ data, though were generally about twice as high. Dry deposition fluxes exhibited a clear east-west gradient, with maxima in the North China Plain and Sichuan Basin. Increases in NH_3 concentrations and deposition were most pronounced in urban, cropland, and forest regions, with urban areas experiencing the fastest growth and grasslands the highest total deposition. The national mean NH_3 concentration and dry deposition flux were 4.98 ppb and 0.51 g m^{-2} yr^{-1}, respectively. Anthropogenic emissions explained 77% of the variability in NH_3 concentration trend, while meteorological factors accounted for the remainder. 70%–80% of deposition trend was governed by atmospheric NH_3 concentration changes. This study highlights growing ammonia pollution and informs nitrogen management strategies in China.

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1 Introduction

Ammonia (NH_3), as the most abundant alkaline gas in the atmosphere, readily reacts with acidic species such as nitric acid and sulfuric acid to form secondary inorganic aerosols. These aerosols contribute significantly to fine particulate matter ($PM_{2.5}$), thereby adversely affecting human health, air quality, and atmospheric visibility (Na et al., 2007; Hauglustaine et al., 2014; He et al., 2001). Reducing NH_3 emissions has been identified as a cost-effective strategy for mitigating air pollution (Pinder et al., 2007; Wu et al., 2016). Besides, excessive atmospheric NH_3 can also deposit onto terrestrial and aquatic ecosystems through dry and wet processes, leading to soil acidification, eutrophication, and biodiversity loss (Hernández et al., 2016; Fu et al., 2017; Hu et al., 2021). Therefore, monitoring and quantifying atmospheric NH_3 concentrations and deposition rates within different land use, especially at global emission hotspots, are critical for informing nitrogen management strategies and protecting air, soil, and water resources, as well as human health (Liu et al., 2017a; Griffis et al., 2019).

As the world's largest agricultural country, China is also among the top NH_3 emitters globally, with an estimated annual emission of 8.4 Tg NH_3 yr^{-1}—far exceeding emissions from Europe (3.1 Tg yr^{-1}) and North America (2.8 Tg yr^{-1}) (Paulot et al., 2014). In recent years, the proportion of NH_3 deposition in total nitrogen (N) deposition has been increasing steadily, accounting for approximately 67% in China by 2020 (Liu L et al., 2024). This upward trend is expected to continue, driven by declining NO_x emissions due to pollution control policies and rising NH_3 emissions associated with global agricultural intensification (Erisman et al., 2008; Goldberg et al., 2021; Pinder et al., 2008).

NH_3 deposition in China is nearly double that of the EU (Liu L et al., 2024), mainly due to excessive nitrogen fertilizer application. In 2014, agricultural NH_3 volatilization accounted for 12 Tg N yr^{-1} globally, with China contributing about 34% (Ma et al., 2020). Anthropogenic activities have nearly doubled NH_3 emission over the past few decades, with cropland and livestock sources making up around 80% of the global total emissions. Non-agricultural sources—such as wastewater treatment, human excreta, and transportation—remain relatively minor (Behera et al., 2013; Zhu et al., 2015; Van Damme et al., 2018). Although the growth rate

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of both agricultural and non-agricultural emissions has slowed in recent years, the absolute emissions continue to rise (Chen J et al., 2023).

Atmospheric NH_3 concentration serves as a key indicator of emission intensity; therefore, accurately quantifying its spatiotemporal variations and identifying the underlying drivers is essential for constraining emission estimates, evaluating the ecological and environmental impacts and informing effective mitigation strategies. Due to its high reactivity and predominant agricultural sources, NH_3 exhibits pronounced temporal and spatial variability. To date, China operates two national observation networks dedicated to monitoring NH_3 concentrations and deposition: The National Nitrogen Deposition Monitoring Network (NNDMN, established in 2004) and the Ammonia Monitoring Network of China (AMoN-China, established in 2015). While these networks provide high-quality measurements, their sparse spatial coverage limits their ability to characterize regional patterns, even in the most advanced global networks (Liu et al., 2017a; b). Additionally, few sites offer long-term (>10 years) continuous data records (Wang et al., 2023), posing challenges for trend analysis. The limited availability of NH_3 monitoring data impedes our understanding of its spatial-temporal patterns and impacts on air quality, climate, and ecosystems.

In addition to surface monitoring, the chemical transport models (CTMs, i.e. GEOS-Chem, WRF-Chem) are widely used to simulate NH_3 concentrations and dry deposition, as they incorporate processes such as emission, transport, deposition, and chemical transformation (Hu et al., 2020; 2021; Lu et al., 2020). However, their accuracy is constrained by uncertainties in emission inventories and model parameterizations, where the bias in both NH_3 emissions and other pollution species (e.g. NO_x and SO_2) can lead to considerable uncertainty in simulating NH_3 concentration and corresponding deposition to ground (Van Der Graaf et al., 2022; Liu et al., 2024). NH_3 emission estimates remain highly uncertain due to outdated activity data, poorly constrained emission factors, and underrepresented sources as cities. Compared to other air pollutants, NH_3 exhibits greater variability and uncertainty, particularly because of its diverse agricultural sources (Beusen et al., 2008; Behera et al., 2013).

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Recent advances in satellite remote sensing offer new opportunities to monitor atmospheric NH_3. The first global NH_3 distribution map was derived in 2009 using data from the Infrared Atmospheric Sounding Interferometer (IASI) onboard the MetOp-A satellite. Since then, other hyperspectral infrared instruments, such as the Cross-track Infrared Sounder (CrIS), the Atmospheric Infrared Sounder (AIRS), the Tropospheric Emission Spectrometer (TES) and the Geostationary Interferometric Infrared Sounder (GIIRS) on board China's FengYun-4B satellite, have been used to retrieve NH_3 columns (Shephard et al., 2015; Someya et al., 2020; Chen J et al. 2023; Zeng et al., 2023). Satellite observations provide wide spatial coverage and continuous temporal resolution, helping to fill gaps in ground networks. Satellite-derived NH_3 data represent column-averaged values, and. the lowest atmospheric level from satellite retrievals mainly represents averages within the boundary layer height. Therefore, satellite retrievals cannot directly replace surface-level concentrations which are critical for dry deposition estimation, which heavily depends on near-surface NH_3 levels and deposition velocities (Lei et al., 2021; White et al., 2023; Liu S et al., 2024).

Despite these limitations, satellite observations have been increasingly used to constrain NH_3 emissions, assess deposition flux, and identify trends (Chen et al., 2021; Kharol et al., 2018; Van Damme et al., 2021). For instance, Liu L et al. (2019) estimated global surface NH_3 concentrations from IASI data and identified high concentrations (>6 μg N m^{-3}) in the North China Plain and northern India. Linear trend analysis from 2008 to 2016 revealed strong increases in eastern China (>0.2 μg N m^{-3} yr^{-1}). More recently, satellite data have been used to investigate urban NH_3 concentrations globally, showing a significant rise (1.2% yr^{-1}) in 2008–2019 (Liu S et al., 2024). These studies demonstrate the utility of satellite retrievals in characterizing NH_3 pollution and its spatiotemporal evolution, especially in regions lacking surface monitoring.

With the growing importance of NH_3 in nitrogen deposition—now comprising the majority of dry N deposition (Russell et al., 2003)—accurate estimation of NH_3 dry deposition is becoming increasingly critical. Kharol et al. (2018) reported that NH_3 contributed more than NO_2 to dry N fluxes over much of North America in the warm season. Liu L et al. (2019) used satellite-

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derived data to estimate global NH_3 dry deposition during 2008–2016, with results broadly consistent with ground measurements, highlighting the potential for remote sensing to fill spatial gaps in deposition assessment. In China, satellite observations show that high NH_3 concentrations are mainly concentrated in the North China Plain, Northeast China, and the Sichuan Basin, while low concentrations are found on the Tibetan Plateau (Liu et al., 2017b). However, long-term studies remain scarce, and the drivers of spatiotemporal variation in NH_3 concentrations and dry deposition under rapid urbanization, land-use change, climate change, and shifts in fertilizer use have not been fully quantified, remaining as a large knowledge gap in understanding and constrain global nitrogen cycle at these emission hotspots.

To robustly constrain and quantify the spatiotemporal variations in near-surface NH_3 concentrations and dry deposition over the past decade, we integrated multiple data sources and analytical approaches. These included high-resolution satellite-derived NH_3 retrievals from 2013 to 2023, ground-based observational datasets, simulations from the GEOS-Chem chemical transport model, and dry deposition velocity estimates derived using a random forest algorithm. This study aims to address the following key scientific questions: (1) What are the spatiotemporal patterns of near-surface NH_3 concentrations across different land cover types in China over the past decade? (2) What are the temporal trends in NH_3 dry deposition across China during this period, and what are the primary driving factors? (3) How do NH_3 concentrations and dry deposition in China compare with those in other regions of the world? By addressing these questions, this study seeks to advance understanding of the nitrogen cycle in China and provide a scientific foundation for evaluating ecological impacts and informing targeted strategies for nitrogen management and sustainable agriculture.

2 Materials and Methods

2.1 Satellite-based atmospheric NH_3 concentration

The satellite-based atmospheric NH_3 concentration used in this study is CrIS (version 1.6.4). The CrIS is a hyperspectral infrared sounder onboard the Suomi National Polar-orbiting Partnership (Suomi NPP) and NOAA-20 satellites (Shephard et al., 2020). Operating in a sun-synchronous orbit at an altitude of approximately 824 km, CrIS provides global coverage twice

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daily, with local overpass time around 13:30 (daytime) and 01:30 (nighttime). The instrument has a swath width of up to 2200 km, with a nadir spatial resolution of approximately 14 km (Zavyalov et al., 2013). CrIS onboard Suomi NPP operated from May 2012 to May 2021, while that onboard NOAA-20 has been operational since March 8, 2019.

In this study, the lowest atmospheric layer of CrIS-derived NH_3 concentrations was utilized, representing the column average from the surface to approximately 900 m above ground level. As this study focuses on China, we used NH_3 data over regions of 73°–136°E and 3°–54°N. To ensure data reliability, only high-quality retrievals were included, filtered using a Quality Flag (QF) ≥ 3 and Cloud_Flag = 0. The analysis period spans from 2013 to 2023, covering both satellite missions, and provides an 11-year, near-continuous time series of atmospheric NH_3 observations over China. To assess the consistency between the two satellite missions, a regression analysis was performed using monthly averaged NH_3 concentrations from the overlapping period (2019–2021), revealing strong agreement across China (Figure S1, *SI*). For subsequent analyses, the original satellite retrievals were resampled to a uniform spatial resolution of 0.1° × 0.1°.

2.2 Ground-based observations of atmospheric NH_3 concentration

The dry deposition of NH_3 is the product of ground level (usually calculated by site-based observations of 1~1.5 m height) NH_3 concentration and dry deposition velocity. Our previous observation and modeling study in U.S. Corn Belt has found significant vertical gradient within boundary layer height (~1-2 km) (Griffis et al., 2019; Hu et al., 2020; 2021), therefore, the column-averaged NH_3 concentration should be converted to ground level, which will further used to derive dry deposition flux. To validate and calibrate satellite-derived NH_3 concentrations, we used measurements from the National Nitrogen Deposition Monitoring Network (NNDMN), which was stablished since 2010 and comprises 43 monitoring sites across China, encompassing different land use types especially for urban, rural (cropland), and background (coastal, forest, and grassland) regions. The network provides high-quality observations of atmospheric reactive nitrogen (Nr) species in gas, particulate, and precipitation phases, including measurements of both wet and dry nitrogen deposition (Xu et al., 2015).

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NNDMN employs two monitoring methods: the long-term active denuder for long-term atmospheric sampling (DELTA) and the low-cost, passive Active Leading Passive High Absorption (ALPHA) sampler (Flechard et al., 2011). Monthly surface NH_3 concentrations are primarily monitored using DELTA, with a few sites utilizing ALPHA. Xu et al. (2015) demonstrated that these two methods yield statistically consistent NH_3 measurements. Considering the typically low NH_3 concentrations at background sites, this study selected 24 representative urban and rural stations for calibration to improve the reliability of subsequent NH_3 dry deposition estimates. The locations of monitoring sites and land cover types across China are shown in Figure. 1a.

2.3 Estimation of NH_3 dry deposition

Dry deposition flux of atmospheric NH_3 was estimated by multiplying the surface NH_3 concentration with the dry deposition velocity, following the equation:

$$F = C \times V_d \qquad (1)$$

Here, F denotes the dry deposition flux, C is the surface NH_3 concentration (ppb) obtained from satellite retrievals and subsequently corrected using ground-based measurements, and V_d is the dry deposition velocity (cm s^{-1}), which is highly variable in space and time due to its sensitivity to land surface characteristics and meteorological conditions.

The most widely used approach to derive V_d is by model simulation, here we first used the GEOS-Chem chemical transport model to simulate spatial-temporal varied V_d across China. However, considering the GEOS-Chem model requires substantial computational resources for one decade, and to further improve spatial resolution and computational efficiency, a random forest machine learning algorithm was then applied to simulate dry deposition velocities based on output from GEOS-Chem model (see more details in Section 2.4).

2.4 Simulation of Dry Deposition Velocity

2.4.1 Simulation of V_d by using GEOS-Chem model

We applied a hybrid modeling approach that combines the GEOS-Chem model with a random

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forest regression algorithm to estimate NH_3 dry deposition velocities across China. GEOS-Chem is a global 3-D chemical transport model driven by meteorological inputs from NASA's Goddard Earth Observing System (GEOS), developed for simulating atmospheric composition and chemistry (Eastham et al., 2014). In this study, we used GEOS-Chem v13.3.1 to simulate NH_3 dry deposition velocity over China for the year 2015. The model was driven by assimilated meteorological data from NASA's MERRA-2 reanalysis. Simulations were conducted on a nested horizontal grid of 0.5° × 0.625° covering the domain of 60°E–149.375°E and 11°S–54.5°N (Lu et al., 2024).

2.4.2 Simulation of V_d by using random forest machine learning algorithm

To improve the spatial resolution and model efficiency, we used the GEOS-Chem model based V_d simulations to train a random forest model that can predict dry deposition velocities under various meteorological and land surface conditions and with finer spatial resolution for decade's period. This data-driven approach enables downscaling to a 0.25° resolution and extends predictions to whole study period from 2013 to 2023 by using ERA5 reanalysis data.

The random forest (RF) algorithm is a widely adopted ensemble machine learning method that integrates multiple decision trees using the bagging strategy to capture complex nonlinear relationships between predictors and response variables. This approach is widely used in atmospheric environment assessments, nitrogen management in agriculture, and model validation studies, providing a robust framework for evaluating the ecological impacts of NH_3 deposition (Asadi et al., 2021; Ai et al., 2024; Zhang et al., 2024). As shown in Figure. S2, the RF model was trained on multiple bootstrapped datasets and evaluated by aggregating outputs from multiple trees to obtain stable and accurate predictions. We selected five meteorological variables from ERA5 reanalysis data as predictors: planetary boundary layer height, 10 m wind speed, volumetric soil water of surface layer, surface temperature, and total precipitation. The dataset was randomly split into a training set (60%) and a validation set (40%), the comparisons between using two approaches will be evaluated in Section 3.4.1.

2.5 Geographical division in China and other supporting data

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To investigate spatial heterogeneity in interannual trends, China was divided into nine subregions based on the classification system from the Resource and Environmental Science Data Center (Figure. 1b). These regions include: Northeast China Plain, Yunnan–Guizhou Plateau, Northern Arid and Semi-Arid Region, Southern China, Sichuan Basin and Surrounding Areas, Middle–Lower Yangtze Plain, Qinghai–Tibet Plateau, Loess Plateau, and Huang–Huai–Hai Plain. Table 1 summarizes the dominant land cover types and their proportional areas within each subregion, and the provinces contained in each region are listed in Table S1 (*SI*), the details of main land use categories and corresponding proportions in each region are also displayed in Figure 1b and Text S2 (*SI*).

To clarify the characteristics of atmospheric NH_3 concentrations and dry deposition flux across different land surface types, we utilized the 30-meter resolution China annual Land Cover Dataset (CLCD) to classify surface types. The CLCD is the first annual land cover product for China derived from Landsat imagery, covering the period from 1985 to 2022 (Yang et al., 2021). The dataset categorizes land cover into nine classes: cropland, forest, shrubland, grassland, water bodies, snow/ice, bare land, impervious surfaces, and wetlands. Based on this classification, we conducted a systematic analysis of the spatial variation and temporal trends in NH_3 concentrations and dry deposition fluxes across different land surface types.

In this study, multiple emission inventories of SO_2, NOx, and NH_3 were utilized to investigate the drivers behind changes in atmospheric NH_3 concentrations and to assess potential future trends. The emission inventories for SO_2 and NOx include: (1) the Inversed Emission Inventory for Chinese Air Quality (CAQIEI, https://www.scidb.cn/en/detail?dataSetId=81cc0de9c68b4a4981e2f295ac612fbf); (2) the Multi-resolution Emission Inventory for China (MEIC, http://meicmodel.org.cn/?page_id=560); (3) the Air Benefit and Cost and Attainment Assessment System - Emission Inventory (ABaCAS, https://abacas-dss.com/abacasChinese/Default.aspx); (4) the Community Emissions Data System (CEDS, https://github.com/JGCRI/CEDS/); and (5) the Emissions Database for Global Atmospheric Research (EDGAR, https://edgar.jrc.ec.europa.eu/dataset_ap81#p3). Due to the relatively late

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development of ammonia (NH_3) research and the limited availability of comprehensive emission inventories, this study employed only two datasets—EDGAR v8.1 and MEIC—for NH_3 emission analysis. In addition, the Dynamic Projection model for Emissions in China (DPEC, http://meicmodel.org.cn/?page_id=1917), developed by Tsinghua University, was used to project future emission trends. Further details on all six emission inventories are provided in Text S3 and Table S2 (*SI*).

2.6 Quantification of influencing factors to annual trend of NH_3 concentration and dry deposition

2.6.1 Simulation of ground NH_3 concentration by using random forest model

To assess the contributions of meteorological conditions and emissions to NH_3 concentrations over study period, we constructed another random forest model to simulate atmospheric NH_3 concentration, where the CrIS-retrieved NH_3 concentrations for 2022 were used as the model output considering the most updated emission inventory is available for 2022, and input parameters included five ERA5-derived meteorological variables (boundary layer height, wind speed, soil moisture, temperature, and precipitation) and three emission datasets from the EDGAR inventory (SO_2, NO_x, and NH_3 emissions). To isolate the effects of emissions and meteorology, we conducted a sensitivity experiment using the 2022-trained model as the baseline. By holding emissions constant or regressing meteorological data back to 2013 (and vice versa), we simulated NH_3 concentrations attributable solely to changes in meteorology or emissions. The contributions of each factor were then normalized to calculate the percentage influence on NH_3 concentration changes.

2.6.2 Quantification of influencing factors to annual trends of NH_3 concentration

We further used the logarithmic differentiation method to decompose the relative contributions of NH_3 concentration and dry deposition velocity to the overall change in dry deposition flux. The logarithmic form allows the multiplicative relationship to be transformed into an additive form, making it suitable for quantifying variable impacts, particularly when concentration and velocity change in opposite directions. The decomposition is based on the following:

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$$\Delta \ln F = \Delta \ln C + \Delta \ln V_d \qquad (2)$$

The respective contributions of concentration and deposition velocity are calculated as:

$$\eta_C = \left| \frac{\Delta \ln C}{\Delta \ln F} \right| \qquad (3)$$

$$\eta_{V_d} = \left| \frac{\Delta \ln V_d}{\Delta \ln F} \right| \qquad (4)$$

where Δln denotes the change in the natural logarithm, η_C and η_{V_d} represent contributions from NH_3 concentration and dry deposition velocity. These contributions were normalized to provide intuitive percentage values. This method is particularly effective in quantifying dynamic and opposing changes and does not assume linear relationship, offering a more robust analysis than traditional linear regression. Additionally, the Mann-Kendall (MK) trend test was employed to statistically evaluate the temporal trends in NH_3 concentrations over the study period.

3 Results and Discussions

3.1 Spatial patterns of near-surface NH_3 concentration and its trend analysis

Using CrIS satellite-derived near-surface NH_3 concentrations from 2013 to 2023, we generated a high-resolution (0.1° × 0.1°) monthly averaged dataset of NH_3 distributions across China over an 11-year period. We first displayed the annual averaged spatial patterns and its trend from 2013-2023 at both the national scale and within specific subregions, followed by an analysis of seasonal variations (Figures. 2a–j and Figures. S3–S7, *SI*). The results of annual average from indicate that the North China Plain (also known as the Huang-Huai-Hai Plain) consistently exhibited the highest NH_3 concentrations (>10 ppb) during the study period (Figure 2a). This region is recognized as one of China's most intensive agricultural zones, accounting for approximately 25% of China's total arable land area and grain production (Song et al., 2024), and is thus subject to frequent fertilizer application, contributing significantly to elevated NH_3 emissions.

The secondary NH_3 concentration hotspots were observed in the Guanzhong Plain in Shaanxi

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Province and the southeastern margin of the Tibetan Plateau. The Guanzhong Plain region is another major agricultural production area in western China, with cultivated land accounting for 49% of Shaanxi Province's total arable area. Intensive fertilizer uses and related activities are the main sources of NH_3 emissions in this region. The elevated NH_3 concentrations in southeastern Tibet are likely attributed to emissions from extensive livestock farming, particularly yak and sheep husbandry. In addition to these agricultural and pastoral regions, relatively high NH_3 concentrations were also observed in arid zones such as Xinjiang and Inner Mongolia. However, these apparent NH_3 enhancements are likely artifacts of satellite retrievals influenced by surface radiative properties. High surface thermal contrast in desert regions may amplify the apparent NH_3 spectral signature during the satellite overpass time of midday (13:00), leading to overestimation of NH_3 concentrations due to limitations in retrieval algorithms and thermal contrast biases (Liu et al., 2020b).

To further explore spatial patterns in temporal change, the pixel-wise trend analysis of annual NH_3 concentrations was also conducted (Figure. 2b). Significant positive trends (>0.4 ppb yr^{-1}) were concentrated in the central and eastern parts of China, particularly in major agricultural zones with intensive crop fertilization, where the white areas in the figure indicate trends that were not statistically significant at the 0.05 level. These results are consistent with findings by Warner et al. (2017), who reported a substantial increase in NH_3 concentrations over eastern China using AIRS data from 2002 to 2016. Our study extends this trend through 2023, indicating that NH_3 concentrations in these regions have continued to rise significantly in recent years. In contrast, western China generally showed stable or declining trends. Although northern Xinjiang exhibited moderate NH_3 increases in areas where the trend passed significance testing, other parts of the west demonstrated declining trends. This pattern may be associated with grassland restructuring policies implemented by the Chinese government to reduce overgrazing and restore degraded ecosystems, thereby lowering NH_3 emissions from pastoral sources.

The spatial patterns of NH_3 concentration increases correspond closely to regions of high population density and agricultural land use, such as the North China Plain and Sichuan Basin.

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These areas are also hotspots for reductions in SO_2 and NO_x emissions due to stringent air pollution control measures as displayed in Figures. S8–S9 (*SI*). The decline in acid gases may reduce atmospheric neutralization capacity, thereby enhancing the lifetime and apparent abundance of NH_3 in the atmosphere (Dong et al., 2023), contributing to the pronounced upward trends observed in these regions.

We also displayed the seasonal variations and its trend during 2013-2023, clear seasonal differences in NH_3 spatial distribution were observed during whole study period (Figure 2c–j, Figures S4–S7). In spring, the NH_3 distribution resembled the annual pattern but exhibited concentrations approximately 14% higher. The Huang-Huai-Hai Plain showed especially concentrated and elevated values, likely due to extensive fertilizer use during spring planting. In contrast, the northwest exhibited little seasonal deviation from annual averages, as emissions are more influenced by pastoral activities than by seasonal patterns of fertilization in agricultural regions. In autumn, NH_3 levels declined sharply, despite localized fertilizer application, primarily due to reduced emissions and cooler temperatures. High concentrations remained in Shandong Province and adjacent regions. Winter concentrations were the lowest, reflecting widespread agricultural dormancy and low temperatures, although lower thermal contrast and reduced NH_3 signal strength increase retrieval uncertainties.

In summer, NH_3 concentrations peaked across China, with higher concentration regions expanding westward into semi-arid areas. This peak seasonality contrasts with trends in Europe and the U.S., where springtime peak is also more typical. In China, summer fertilization for maize cultivation—often involving both mineral and organic fertilizers—contributes to the observed summer peak (Paulot et al., 2014). Elevated temperatures further enhance volatilization from manure and urban waste, intensifying atmospheric NH_3 levels. Liu et al. (2024) reported that temperature increases accounted for up to 20% of urban NH_3 increases between 2008 and 2019. Notably, elevated NH_3 levels were also observed along the Yangtze River basin, corresponding to fertilizer use in rice paddies.

The spatial distributions of 11-years' trend analyses for each season are also displayed (Figures

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2d, f, h and j), they show significant increases across eastern China, particularly during summer and autumn. Overall, these results indicate the annual trend of surface NH_3 concentration occurred throughout each season but with obvious seasonal difference. Winter trends were the weakest in magnitude and spatial extent. Consistent with annual patterns, the North China Plain and Sichuan Basin showed the most pronounced increases. Western China showed few significant trends, with slight increases in northern Xinjiang during summer and autumn, and declines in other areas.

3.2 Temporal variation of near-surface NH_3 concentrations for different regions

We also calculated the temporal variation of annual NH_3 concentrations and across different seasons from 2013 to 2023 (Figure 3a). Over this period, the annual mean NH_3 concentration in China increased by 22%, with seasonal increases of 14% in spring, 31% in summer, 26% in autumn, and 18% in winter, respectively. Among these seasons, summer exhibited the highest mean concentration (3.60 ppb), followed by spring (3.28 ppb), with annual, autumn, and winter means recorded at 2.88 ppb, 2.63 ppb, and 2.00 ppb, respectively (Table 2). The Mann-Kendall trend test results (Table 2) indicated statistically significant upward trends for spring, summer, autumn, and annual mean concentrations ($p < 0.05$). Although winter showed a positive trend ($Z > 0$), it did not reach statistical significance. The seasonal rates of increase, in descending order, were: summer (0.065 ppb yr^{-1}), autumn (0.050 ppb yr^{-1}), annual (0.045 ppb yr^{-1}), spring (0.039 ppb yr^{-1}), and winter (0.023 ppb yr^{-1}). The most pronounced increase during summer from 2013 to 2023 also aligns with previous findings by Liu et al. (2018), which only analyze the North China Plain region from 2008 to 2016, but their trend is slightly lower than our results.

The summer increasing trend of atmospheric NH_3 is likely closely related to global warming (Figure S10, *SI*), previous studies shown that over 40% of fertilizer application and approximately 25% of livestock emissions occur during the summer months (Xu et al., 2015; Kang et al., 2016), which enhances NH_3 volatilization from ground to atmosphere. The slower rate of increase in spring may be associated with China's national fertilizer reduction policies, such as the "Action Plan for Fertilizer Reduction by 2025". Fertilizer use increased until peaking in 2015 and subsequently declined for eight consecutive years, resulting in a 15.1%

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reduction from 2013 to 2023, with the national application totaling 50.22 million tons in 2023 (Figure. S11).

The decrease in chemical fertilizer use, combined with the adoption of organic fertilizers, has contributed to a gradual slowdown in the rise of NH_3 concentrations. By 2024, the nitrogen uses efficiency (NUE) for rice, maize, and wheat reached 42.6%, helping to reduce fertilizer input without compromising yields and mitigating NH_3 emissions and nutrient pollution. Zhan et al. (2020) identified improving NUE as the most effective and cost-efficient strategy for NH_3 mitigation in agriculture, a finding supported by cost-benefit assessments (Zhang et al., 2020). Autumn also showed a substantial increase in NH_3 concentrations, second only to summer. Current emission reduction efforts have primarily focused on spring and summer, reflecting crop planting cycles, while autumn has often been overlooked, contributing to this seasonal gap in mitigation. These findings highlight the need for seasonally and crop-specific emission control strategies in future NH_3 management efforts.

Significant spatial heterogeneity was observed in the interannual variation of NH_3 concentrations across different regions. Figure 3b illustrates long-term trends in NH_3 concentrations for nine subregions. Most regions exhibited increasing trends, with the Huang-Huai-Hai Plain standing out for its consistently elevated concentrations—approximately twice as high as the national average (Table 3). This region is China's primary agricultural zone, characterized by high population density and intensive agricultural activity, both of which contribute to substantial NH_3 emissions. Additionally, it has been a focal area for SO_2 and NO_x emission reductions, and the combined effects of high emissions and reduced atmospheric neutralization capacity have led to persistent NH_3 accumulation.

MK trend analysis further revealed statistically significant upward trends in the Huang-Huai-Hai Plain, the Northern Arid and Semi-Arid Region, the Loess Plateau, the Middle-Lower Yangtze Plain, South China, the Northeast China Plain, and the Sichuan Basin and its surrounding regions. The Huang-Huai-Hai Plain showed the steepest increase, with an average annual rise of 0.24 ppb, corresponding to a 7% per year growth rate—3.3 times the national

https://doi.org/10.5194/egusphere-2025-3090
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average. The primary driver of this sharp increase is the marked reduction in atmospheric SO_2, which has disrupted the NH_3–acid gas neutralization balance (Xu et al., 2019a). The Loess Plateau ranked second, with an average increase of 0.14 ppb per year. In contrast, the Yunnan–Guizhou Plateau exhibited a mild, non-significant increase, with relatively stable concentrations. The Tibetan Plateau showed a slight downward trend, which also lacked statistical significance ($p > 0.05$), indicating a relatively stable NH_3 regime in this high-altitude, low-emission region.

3.3 Comparison between satellite and ground-based NH_3 observations and satellite data correction

As stated in Section 2.1, although satellite-based observations provide high spatial resolution and long-term data for atmospheric NH_3 studies, they represent near-surface column average between ground to around 1 km, where large vertical gradient has been reported because of its reactive nature of ammonia and its role in chemical transformation processes (Hu et al., 2020; 2021; Griffis et al., 2019). Besides, the dry deposition of NH_3 is the product of ground level (usually calculated by site-based observations of 1~1.5 m height) NH_3 concentration and dry deposition velocity. Therefore, to enable accurate estimation of NH_3 dry deposition, we conducted a comparative analysis between satellite-derived and multiple years observations at 24 ground-based NH_3 sites, and their relationship will be used to correct column averages to ground level height.

As shown in Figure 4a, satellite and ground-based NH_3 concentrations exhibit a strong correlation ($R = 0.79$), with a coefficient of determination (R^2) of 0.62 and a root mean square error (RMSE) of 3.56 ppb. However, the ground-based measurements are, on average, approximately twice as high as those retrieved by satellite. This discrepancy can be attributed to the vertical gradient of NH_3 in the atmosphere: ground-based sensors typically operate at heights of 1–1.5 m, while satellite instruments, even in their lowest vertical layer, observe at altitudes hundreds of meters above ground. Many pioneer studies have demonstrated that when the land surface acts as an NH_3 source, its vertical distribution decreases logarithmically with height. For example, our previous studies of tall tower observations in the United States

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reported an NH_3 mixing ratio gradient of −0.27 ppb per 100 m, with modeled gradients ranging from −0.21 to −0.84 ppb per 100 m (Hu et al., 2020; 2021; Griffis et al., 2019), showing good agreement between observations and simulations. When using the gradient of above reported values, the average of 0-1000 m column NH_3 concentration should be around 1~ 4 ppb lower than ground level, this pronounced vertical gradient is a major reason for the systematic underestimation of NH_3 by satellites.

To address this bias, we used the regression relationship derived from Figure 4a to calibrate the satellite retrievals. After correction, a new regression (Figure 4b) shows a nearly 1:1 agreement between satellite and ground-based measurements, with the RMSE reduced to 1.69 ppb. To further assess the correction effectiveness, we selected the year 2015—when both satellite and ground data are available—for analysis. As shown in Figure 4c, the corrected satellite-derived NH_3 concentrations closely match ground observations across almost all sites, confirming the reliability of the correction approach. This calibration function was then applied to the full 2013–2023 satellite dataset to improve the reliability of NH_3 dry deposition estimates. Table 3 illustrated the corrected average NH_3 concentrations across different regions, with the Huang-Huai-Hai Plain exhibiting the highest value of 11.36 ppb. This was followed by the Northern Arid and Semi-Arid Region (6.93 ppb), the Qinghai–Tibet Plateau (6.48 ppb), and the Loess Plateau (6.05 ppb). Although the bias-corrected NH_3 concentrations in these regions ranked immediately after the Huang–Huai–Hai Plain, their values were approximately two times lower than those observed in the Huang–Huai–Hai Plain.

3.4 Estimation of spatial-temporal variations of NH_3 dry deposition across China

3.4.1 Simulation of spatial-temporal dry deposition velocities

To estimate NH_3 dry deposition flux across China, we first used the GEOS-Chem model to simulate NH_3 dry deposition velocities for the year 2015 (Figure 5a). Considering the high computational cost and limited temporal flexibility of the GEOS-Chem model, we adopted a hybrid modeling approach by training a random forest (RF) machine learning model on the GEOS-Chem model-based simulation results. This approach allowed us to extend the simulation to the full 2013–2023 period, while improving both spatial resolution and

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computational efficiency. The GEOS-Chem-derived 2015 dry deposition velocity served as the response variable for training the RF model.

The resulting RF-predicted dry deposition velocities for 2015 show high spatial agreement with the GEOS-Chem outputs (Figure 5b). Both models identify southern China as a hotspot for dry deposition velocity, likely due to the region's warm and humid conditions that facilitate gaseous NH_3 deposited onto ground surface. Additionally, southern China is a major rice-producing region where surface resistance in paddy fields is lower than in dryland fields, further enhancing dry deposition rates. Figure 5c shows the differences between the two model outputs, with over 99% of grid cells having discrepancies less than 0.1 cm s^{-1}, indicating strong consistency and validating the reliability of the RF model for long-term simulations. Using this trained model, we further simulated NH_3 dry deposition velocities from 2013 to 2023 at monthly averages.

3.4.2 The spatial-temporal variations of NH_3 dry deposition in China

With the corrected spatial-temporal NH_3 concentrations and simulated deposition velocities from 2013 to 2023, we derived the monthly grid-level NH_3 dry deposition flux for China. These were further aggregated to estimate flux and total deposition over different surface types (Figure 6, Figure S12, *SI*). Figure 6a illustrates the spatial distribution of NH_3 dry deposition flux average from 2013 to 2023. Distinct spatial differences are evident, where the eastern coastal regions exhibited significantly higher deposition flux than inland areas, with values higher than 1.8 g NH_3 m^{-2} yr^{-1}. Notably, the Huang–Huai–Hai Plain and the southwestern region of the Qinghai–Tibet Plateau emerged as prominent hotspots of NH_3 dry deposition, highlighting the substantial impact of intensive agricultural activities and industrial emissions. Elevated deposition rates were also observed in the southern Tibetan Plateau, driven by locally high NH_3 concentrations.

A trend analysis of dry deposition over the 11-year period (Figure 6b) shows statistically significant increases in deposition flux in eastern coastal areas (> 0.1 g m^{-2} yr^{-1}), likely reflecting rising NH_3 concentrations in these regions. In contrast, western China shows minimal change, with some areas even exhibiting slight declines. Unlike the NH_3 concentration trends,

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there is no region in western China displayed a statistically significant increase in dry deposition flux, emphasizing the spatial decoupling between emission intensity and deposition patterns in less industrialized regions.

The interannual variation of NH_3 dry deposition also exhibited significant spatial heterogeneity at the regional scale (Figure 6c and Table 4). The Huang-Huai-Hai Plain, characterized by persistently high NH_3 concentrations, recorded the highest area-specific dry deposition flux, reaching 1.06 g m^{-2} yr^{-1}—approximately twice the levels observed in other regions. MK trend analysis indicated a significant increasing trend in dry deposition flux across all regions except the Tibetan Plateau, where a weak downward trend was observed but was not statistically significant. The most pronounced increase was found in the Huang-Huai-Hai Plain, with an average annual increment of 0.05 g m^{-2} yr^{-1}, followed by the middle and lower reaches of the Yangtze River, at 0.03 g m^{-2} yr^{-1} (Table 4).

3.4.3 Comparisons of NH_3 concentration, dry deposition velocity and flux in different surface categories

In addition to meteorological factors, land cover types play a pivotal role in regulating dry deposition processes. In this section, we annually extracted NH_3 concentrations, dry deposition velocities, and dry deposition fluxes across different land cover categories. The analysis focused on four representative land-use types—urban, cropland, forest, and grassland—selected based on their distinct NH_3 emission characteristics (Figure 7; Table S3, *SI*). The average NH_3 concentrations, ranked from highest to lowest, were: urban (8.76 ppb), cropland (6.27 ppb), national average (6.01 ppb), grassland (5.72 ppb), and forest (3.76 ppb) (Figure 7a). Urban areas exhibited both the highest concentrations and the largest interannual variability, with a statistically significant upward trend ($p < 0.05$, $Z > 1.96$), increasing at an average rate of 0.39 ppb yr^{-1}. This trend is primarily attributed to anthropogenic sources such as vehicular emissions, as well as the urban heat island effect, which raises urban temperatures by 1–3°C—and occasionally by over 10°C—relative to surrounding rural areas. These elevated temperatures, further amplified by global warming, facilitate enhanced NH_3 volatilization.

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While NH_3 concentrations over grassland areas remained relatively stable throughout the study period, cropland regions exhibited a continuous upward trend, with the two trends intersecting in 2016 (Figure 7a), after which NH_3 concentrations in croplands exceeded those in grasslands. NH_3 emissions in grassland ecosystems are predominantly associated with livestock grazing, and the stabilization observed is likely attributable to the implementation of grazing restrictions and ecological restoration policies. In contrast, despite the introduction of fertilizer reduction policies in some agricultural areas, rising food demand driven by population growth has sustained or even increased fertilizer application, thereby contributing to the observed increase in cropland NH_3 levels. At the national scale, NH_3 concentrations exhibited a statistically significant upward trend, with an average increase of 0.1 ppb yr^{-1} (equivalent to an annual growth rate of 2.2%). Forested regions, which are minimally impacted by anthropogenic sources such as synthetic fertilizers and livestock emissions, maintained the lowest and most stable NH_3 concentrations, showing only a slight upward trend that may be linked to climate warming (Figure 7a; Figure 8).

Dry deposition velocities exhibited limited interannual variability across different land cover types. Forested areas recorded the highest average deposition velocity, likely attributable to greater surface roughness and enhanced canopy-induced turbulence, followed by urban and cropland regions (Figure 7b; Figure 8). The mean NH_3 dry deposition velocities for forest, urban, cropland, grassland, and the national average were 0.43, 0.42, 0.40, 0.32, and 0.36 cm s^{-1}, respectively. Mann–Kendall trend analysis revealed statistically significant increasing trends in urban and cropland areas, with annual rates of 0.0013 and 0.0012 cm s^{-1} yr^{-1}, respectively. Although forests maintained the highest mean velocity and exhibited a positive trend, the change was not statistically significant. At the national scale, deposition velocity showed a weak but consistent upward trend. In contrast, grassland areas experienced a slight decline in deposition velocity over the 11-year period, though this trend was not statistically significant.

Area-specific NH_3 dry deposition fluxes closely followed the spatial distribution of atmospheric concentrations across different land cover types (Figure 7c; Figure 8). Urban regions exhibited the highest mean flux (0.88 g m^{-2} yr^{-1}), followed by cropland areas (0.61 g m^{-2} yr^{-1}). Both

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urban and national average fluxes demonstrated statistically significant upward trends over the study period. The steepest increase was observed in urban areas, with a rate of 0.04 g m^{-2} yr^{-1}—approximately four times the national average—followed by croplands at 0.03 g m^{-2} yr^{-1}. Recent findings (Chen P et al., 2023) suggest that, although fertilizer application has been partially reduced under agricultural emission control policies, non-agricultural sources—such as industrial processes and transportation—have become the predominant contributors to NH_3 emissions in China, particularly concentrated in urban areas. This shift has contributed to elevated NH_3 concentrations and enhanced dry deposition fluxes in cities.

In contrast, forests and grasslands showed relatively stable fluxes, likely due to lower levels of anthropogenic disturbance. Nevertheless, a statistically significant increasing trend in forest deposition flux was detected, which may have important ecological implications. Sustained increases in NH_3 deposition could lead to adverse effects such as plant nutrient imbalances, biodiversity loss, and eutrophication of adjacent aquatic systems, potentially compromising forest health and long-term ecosystem stability. Furthermore, interannual variability in dry deposition was more pronounced in urban areas, reflecting the dynamic nature of urban development and emission variability, whereas cropland fluxes exhibited a more gradual trend in response to evolving fertilizer management practices.

Trends in total NH_3 dry deposition across different land cover types generally mirrored those of area-specific fluxes; however, absolute deposition values were modulated by the extent of each land category. Grasslands accounted for the largest share of total NH_3 dry deposition (1.23 Tg), followed by croplands (1.15 Tg), forests (0.92 Tg), urban areas (0.21 Tg), and a national total of 4.85 Tg. Over the 11-year study period, statistically significant upward trends in total dry deposition were observed at the national scale, as well as in cropland, forest, and urban areas, with annual increases of 0.10, 0.05, 0.03, and 0.01 Tg yr^{-1}, respectively. Although grasslands also exhibited an increasing trend, it was not statistically significant. Changes in total NH_3 deposition are driven not only by atmospheric concentrations and deposition velocities but also by land-use dynamics (Figure 7d; Figure 8). In particular, the continuous expansion of urban areas from 2013 to 2023 contributed substantially to the increasing trend in

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total urban NH_3 deposition. These findings highlight the importance of considering both biogeochemical processes and anthropogenic land-use changes in assessing long-term trends in reactive nitrogen deposition.

3.5 Simulation of NH_3 concentration and contribution factors analysis to decade's trend for both NH_3 concentration and deposition flux

In this section, we further investigated the key drivers of atmospheric NH_3 concentrations using a Random Forest (RF) regression model. Model performance was evaluated by comparing simulated NH_3 concentrations with observations for the period 2013–2023, showing good agreement (Figure 9). The RF model effectively captured the spatial variability of NH_3 concentrations, with deviations generally within ±0.1 ppb, indicating robust predictive capability. The input variables were categorized into two major groups: meteorological factors and anthropogenic emissions, including direct NH_3 emissions as well as co-emitted SO_2 and NOx species.

The RF model simulation results indicated that anthropogenic emissions were the dominant driver, accounting for approximately 77% of the variance in NH_3 concentrations, while meteorological conditions explained the remaining 23% (Figure 10). Among meteorological parameters, air temperature emerged as the most influential factor, whereas other variables (e.g., relative humidity, wind speed) exhibited minimal interannual variation and lower predictive importance. Analysis of ERA5 reanalysis data revealed a persistent warming trend over the past decade, with the annual mean surface temperature in 2023 being 8.4% higher than in 2013 (Figure S10, *SI*). Previous studies, such as Hu et al. (2020), reported an exponential relationship between NH_3 mixing ratios and temperature, with NH_3 concentrations increasing from 4 ppb to 19 ppb as temperature rises from 0°C to 10°C. The regional temperature sensitivity (Q_{10}) of NH_3 emissions was estimated to be approximately 2.5, indicating that continued warming will likely enhance NH_3 volatilization. This may further exacerbate nitrogen loss from agricultural systems and elevate NH_3 dry deposition to downwind ecosystems, potentially intensifying ecological risks such as eutrophication and biodiversity loss.

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Figure S13 illustrates multi-year emission trends of SO_2, NOx, and NH_3 derived from multiple emission inventories, including EDGAR and MEIC. Although observed atmospheric NH_3 concentrations have increased over the period 2013–2023, all inventories consistently indicate a slight decline in NH_3 emissions. This apparent contradiction suggests that the observed rise in NH_3 concentrations may be primarily driven by reduced emissions of acidifying species—namely SO_2 and NOx—which typically enhance NH_3 partitioning into the particulate phase. The reductions in SO_2 and NOx emissions may have suppressed their atmospheric reactions with NH_3, thereby decreasing the formation of particulate ammonium and leaving a greater fraction of NH_3 un-neutralized in the gas phase. This shift likely contributed to elevated ambient NH_3 concentrations, as reported in previous studies (Xu et al., 2019a; Liu et al., 2018; Liu et al., 2017a).

According to EDGAR data, national SO_2 and NOx emissions declined by approximately 20% from 2013 to 2022, following the implementation of the Air Pollution Prevention and Control Action Plan in 2013, which led to substantial reductions in these precursor gases. It is important to note that our Random Forest model does not account for atmospheric chemical processes involving the formation and partitioning of secondary inorganic aerosols, such as nitrate (NO_3^-), sulfate (SO_4^{2-}), and ammonium (NH_4^+). Therefore, for future investigations aiming to quantify the role of atmospheric chemistry in modulating NH_3 concentrations and deposition, the use of comprehensive atmospheric chemical transport models such as WRF-Chem or GEOS-Chem is strongly recommended. These models are capable of resolving multiphase reactions and the thermodynamic partitioning of NH_3 into the aerosol phase, thereby offering a more mechanistic understanding of NH_3 dynamics in response to co-emitted precursor changes.

To further elucidate the drivers of NH_3 dry deposition trends, we employed a logarithmic differential method to decompose the relative contributions of changes in NH_3 concentrations and deposition velocities across different land cover types (Figure 10; Table S4, *SI*). All variables were normalized to facilitate comparison of relative contributions. Results indicate that variations in NH_3 dry deposition fluxes were predominantly driven by changes in atmospheric NH_3 concentrations, accounting for approximately 70–80% of the total variation

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across all land types. Urban areas exhibited the lowest contribution from concentration changes (72.6%) and the highest from deposition velocity changes (27.4%), likely reflecting the more complex aerodynamic and surface resistance conditions in urban environments. In contrast, forested areas showed the highest concentration-driven contribution (81.2%), consistent with their relatively stable surface characteristics and low anthropogenic disturbance.

In summary, the observed increase in atmospheric NH_3 concentrations across China is largely attributable to the substantial reductions in SO_2 and NOx emissions. Concurrently, changes in NH_3 dry deposition fluxes are primarily driven by rising NH_3 concentrations, which are indirectly influenced by declining SO_2 and NOx levels. This inference is supported by consistent evidence from both satellite and ground-based monitoring networks, which document a marked decrease in SO_2 concentrations (Liu M. et al., 2019; Xi et al., 2021) alongside improvements in acid rain conditions.

China has made significant progress in controlling acidifying pollutants over the past decades. SO_2, a key precursor to acid rain formation, has been regulated since the 1980s, with acid rain management formally institutionalized in 1990. Between 1993 and 2021, the area affected by acid rain initially expanded before contracting, reaching a peak in 2015. Since 2006, the acidity of acid rain has steadily declined (Zhang et al., 2023; Zhao et al., 2010). Although China initiated acid rain control efforts approximately a decade later than Western countries, it has achieved more rapid mitigation through the implementation of stringent regulations and widespread adoption of advanced technologies.

Previous studies have indicated that optimizing fertilizer application and adjusting protein content in animal feed could potentially reduce NH_3 emissions by up to 30% without compromising agricultural yields or incurring additional costs (Zhang et al., 2020). In contrast, regulation of NH_3 emissions has lagged behind that of other pollutants. It was not until the implementation of the 2018 “Three-Year Action Plan for Winning the Blue-Sky Defense Battle” that agricultural NH_3 emissions were formally addressed. This plan emphasized enhanced recycling of livestock waste and measures to reduce NH_3 volatilization. Subsequently, the “14th

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Five-Year Plan for Energy Conservation and Emission Reduction" further targeted improvements in fertilizer and pesticide use efficiency, setting a goal to reduce NH_3 emissions from large-scale livestock operations in the Beijing-Tianjin-Hebei region by 5%. Although these recent policies have initiated efforts to mitigate NH_3 emissions, the rate of reduction remains substantially lower than that achieved for SO_2.

The IPCC Sixth Assessment Report (2023) indicates that global surface temperatures increased by approximately 1.1°C from 2011 to 2020 relative to pre-industrial levels (1850–1900). Continued greenhouse gas emissions are projected to drive further warming. Most Shared Socioeconomic Pathways (SSPs) scenarios forecast global temperature rise reaching 1.5°C by around 2040, with only the most stringent SSP1-1.9 scenario likely to limit warming below this threshold, necessitating global carbon neutrality by approximately 2055. In the context of future warming, we analyzed projected emissions of SO_2, NOx, and NH_3 under five SSP scenarios based on the Dynamic Projection Emission Coefficient (DPEC) inventory developed by Tsinghua University (Figure S14, *SI*). All scenarios indicate declining trends for these pollutants; however, NH_3 exhibits the smallest reduction, amounting to roughly two-thirds of the decreases projected for SO_2 and NOx. This discrepancy, combined with rising temperatures and decreasing acid gas emissions, is expected to further enhance atmospheric NH_3 concentrations. Consequently, despite ongoing mitigation efforts targeting NH_3, ambient NH_3 levels may continue to increase. To counteract the synergistic effects of warming and reductions in acid-neutralizing pollutants, more stringent NH_3 emission control policies will be required in China over the coming decades to effectively stabilize or reduce atmospheric NH_3 concentrations.

4 Comparison with previous studies and implications

To evaluate and contextualize atmospheric NH_3 concentrations and dry deposition in China relative to other global regions, we conducted a comprehensive literature review summarized in Table 5. This table integrates the findings of the present study with previous assessments of atmospheric NH_3 levels and dry deposition fluxes worldwide. The comparative analysis highlights considerable spatial variability, with NH_3 concentrations ranging from approximately 2 to 10 ppb and area-specific dry deposition fluxes spanning 0.06 to 1.00 g m^{-2}

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yr^{-1}. The values reported in this study are generally consistent with those documented in comparable geographic and climatic regions.

This study estimates the national average NH_3 concentration in China at 4.98 ppb and the corresponding dry deposition flux at 0.51 g m^{-2} yr^{-1}, and the results for each province of China were also displayed in Figure S15 (*SI*). The national average results closely align with those of Liu et al. (2020a), who employed IASI satellite retrievals and reported NH_3 concentrations of 4.15 ppb and dry deposition fluxes of 0.58 g m^{-2} yr^{-1}. The Tianjin megacity, Shandong province, Henan province, Hebei province and Beijing megacity ranked as the largest top 5 regions for NH3 concentration and dry deposition flux, where Tianjin and Beijing are located within North China Plain hotspots, and were largely influenced by atmospheric transport process from nearby agricultural fields. Compared to Liu et al. (2020a), our analysis extends the observation period and incorporates calibration against ground-based monitoring data, thereby achieving higher accuracy. Jia et al. (2016) estimated the global NH_3 dry deposition flux using empirical models based on ground station measurements, reporting a value of 0.68 g m^{-2} yr^{-1} for China.

In contrast, Xu et al. (2015), utilizing data from the National Nitrogen Deposition Monitoring Network (NNDMN), reported substantially higher values for China (10.65 ppb and 1.00 g m^{-2} yr^{-1}), likely reflecting sampling bias due to the predominance of monitoring sites in urban and agricultural regions characterized by elevated NH_3 emissions and underrepresentation of background locations, resulting in overestimation of national averages. Further evidence of spatial variability is provided by Hu et al. (2020, 2021), who documented significant differences in NH_3 concentrations and deposition rates between cropland and forested background sites, underscoring the critical influence of land use and emission sources on atmospheric NH_3 dynamics.

Overall, the synthesis of data summarized in Table 5 indicates that NH_3 concentrations in China generally range from 4 to 10 ppb, with corresponding dry deposition fluxes between 0.5 and 1.0 g m^{-2} yr^{-1}. The observed variability is primarily attributed to differences in observation periods, measurement methodologies, and spatial coverage. By comparison, the United States

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exhibits average NH_3 concentrations of approximately 2.65 ppb and dry deposition fluxes ranging from 0.07 to 0.3 g m^{-2} yr^{-1}, while Europe reports concentrations near 3.13 ppb and deposition fluxes between 0.1 and 0.3 g m^{-2} yr^{-1}. These findings highlight that both NH_3 concentrations and deposition fluxes in China are substantially higher than those reported for the United States, Europe, and global averages. Notably, Europe has integrated NH_3 control into its air pollution regulatory framework, resulting in measurable emission reductions in recent years. This experience underscores the importance of implementing more stringent NH_3 mitigation policies in China to effectively address the ongoing increases in atmospheric NH_3 concentrations and dry deposition fluxes.

Previous studies have typically examined either atmospheric NH_3 concentrations or dry deposition independently, with relatively few providing a comprehensive assessment integrating both components. This study addresses this gap by combining satellite-derived NH_3 concentrations with ground-based observations and utilizing the GEOS-Chem atmospheric chemistry transport model in conjunction with a machine learning-based Random Forest algorithm to simulate deposition velocities and fluxes. This integrated approach facilitates the generation of high-resolution, multi-year estimates of NH_3 dry deposition across China. The resulting dataset provides a robust scientific basis for improving national nitrogen management policies and offers valuable insights into regional and global nitrogen cycling processes.

5 Conclusions

This study presents a comprehensive analysis of the spatial distribution and temporal trends of atmospheric ammonia (NH_3) concentrations and dry deposition across China during 2013–2023. The key findings are as follows:

(1) The North China Plain exhibited persistently high NH_3 concentrations (>10 ppb), with significant annual increases in central and eastern regions (>0.4 ppb yr^{-1}). The largest seasonal increases occurred in summer (0.065 ppb yr^{-1}). NH_3 concentrations in 2023 were 14%–31% higher than in 2013 across all seasons. CrIS satellite retrievals were strongly correlated with in-situ measurements (R = 0.79), but are larger than later by a factor of about two.

(2) The spatial pattern of NH_3 dry deposition revealed a pronounced east-west gradient, with

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the highest flux in the North China Plain and Sichuan Basin, and a significant upward trend along the eastern coast (>0.1 g m^{-2} yr^{-1}). Over the 11-year period, NH_3 concentrations, deposition flux, and total deposition increased significantly in the land use types of urban, cropland, and forest ecosystems. Urban areas showed the highest concentration and deposition flux as well as the fastest growth rates, while grasslands exhibited the largest total deposition. (3) The national mean NH_3 concentration and dry deposition flux were estimated to be 4.98 ppb and 0.51 g m^{-2} yr^{-1}, respectively. In addition, our analysis indicated that anthropogenic emissions were the dominant driver, accounting for approximately 77% of the variance in NH_3 concentrations, while meteorological conditions explained the remaining 23%; 70%–80% of deposition changes were governed by atmospheric dynamics. These findings underscore the increasing NH_3 pollution across China and provide a critical scientific basis for informed nitrogen management within one of global largest NH_3 emission hotspots regions.

Data Availability: CrIS satellite retrievals of NH_3 were obtained from Environment and Climate Change Canada (ECCC) at https://hpfx.collab.science.gc.ca/~mas001/satellite_ext/cris/ (Shephard et al., 2015; 2020). Ground-based NH_3 measurements were sourced from Xu et al. (2019b), available at https://www.nature.com/articles/s41597-019-0061-2. NH_3 emission inventories were obtained from the Multi-resolution Emission Inventory for China (MEIC; http://meicmodel.org.cn/?page_id=560), the Emissions Database for Global Atmospheric Research (EDGAR v8.1; https://edgar.jrc.ec.europa.eu/dataset_ap81#p3), and the Dynamic Projection model for Emissions in China (DPEC; http://meicmodel.org.cn/?page_id=1917). Emission data for SO_2 and NOx were derived from the Inversed Emission Inventory for Chinese Air Quality (CAQIEI; https://www.scidb.cn/en/detail?dataSetId=81cc0de9c68b4a4981e2f295ac612fbf), the Air Benefit and Cost and Attainment Assessment System (ABaCAS; https://abacas-dss.com/abacasChinese/Default.aspx), and the Community Emissions Data System (CEDS; https://github.com/JGCRI/CEDS/). The MEIC and EDGAR inventories were used for both NH_3 and SO_2/NOx emissions. Meteorological data were obtained from the ERA5 reanalysis dataset provided by the European Centre for Medium-Range Weather Forecasts (ECMWF) at

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https://cds.climate.copernicus.eu/datasets/reanalysis-era5-single-levels. Agricultural fertilizer application data were sourced from the National Bureau of Statistics of China (https://www.stats.gov.cn/sj/ndsj/2024/indexch.htm). Agricultural zoning data were obtained from the Resource and Environmental Science Data Center (https://www.resdc.cn/Default.aspx), and land cover data were retrieved from the National Cryosphere Desert Data Center (https://www.ncdc.ac.cn/portal/metadata/9de270f3-b5ad-4e19-afc0-2531f3977f2f).

Supplement. The supplement related to this article is available online

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.

Author contributions: FS conducted the data analysis and wrote the draft under supervision of CH, CH designed the study and revised this paper, JS and XL conducted GEOS-Chem modeling, all other co-authors collected supporting data, read and approved the final manuscript.

Acknowledgments

Cheng Hu is supported by the National Science founding of China (grant nos. 42475125, 42105117, 42021004 and 41975143), this work was also supported by the National Key R&D Program of China (nos. 2019YFA0607202 and 2020YFA0607501); Jiangsu Science Foundation for Distinguished Young Scholar (No. BK20220055); The 333 Project of Jiangsu Province (No. BRA2017402); R&D Foundation of Jiangsu Province, China (No. BK20220020). Cheng Hu also thanks the founding support from Key Laboratory of Ecosystem Carbon Source and Sink, China Meteorological Administration (ECSS-CMA202403). We also Sincerely thank the support from Environment and Climate Change Canada (ECCC) CrIS group.

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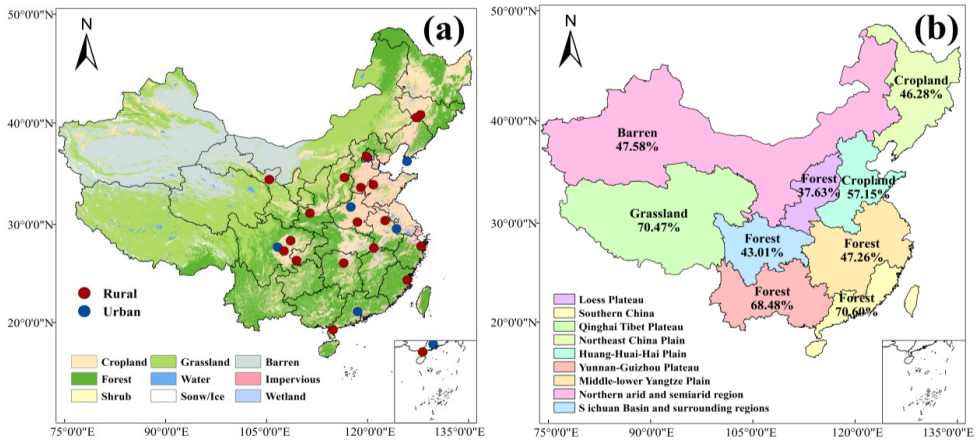

Figure 1. (a) Spatial distribution of land use types and NH_3 monitoring sites in China in 2022, (b) classification of China into nine major agroecological zones based on agricultural practices and climatic conditions.

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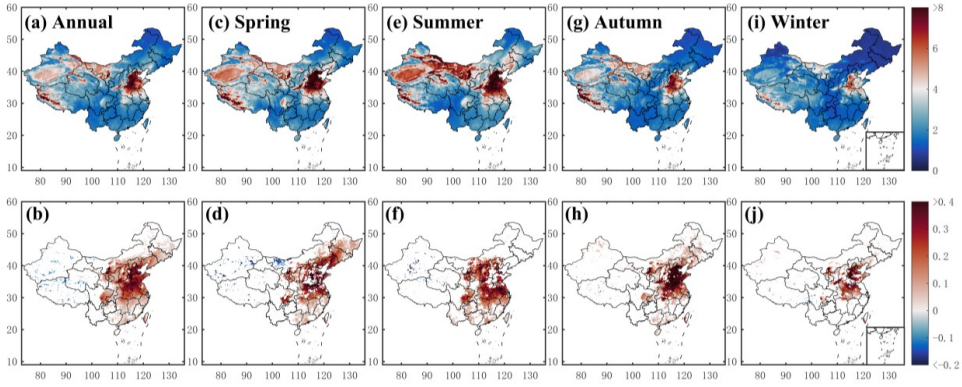

Figure 2. Spatial distribution and temporal trends of annual and seasonal NH_3 concentrations in China from 2013 to 2023 (Units: ppb for concentration; ppb yr^{-1} for trend).

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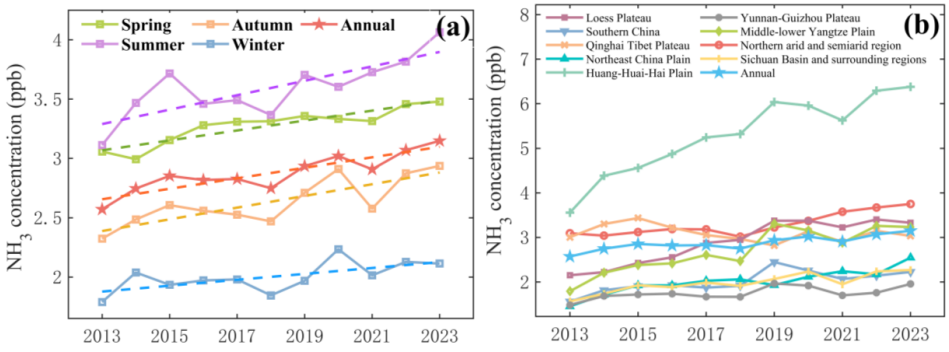

Figure 3. (a) Seasonal and (b) regional variations in NH_3 concentrations across China from 2013 to 2023 (Unit: ppb).

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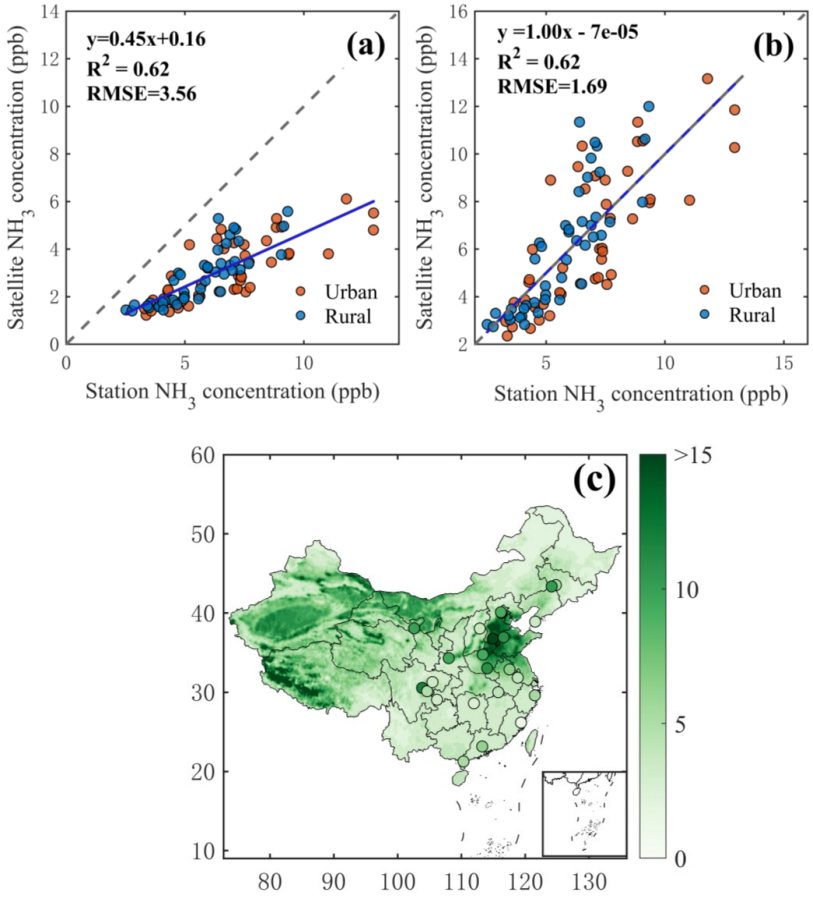

Figure 4. Comparison between satellite and ground-based NH_3 observations: (a) correlation between satellite-derived and ground-based NH_3 concentrations; (b) correlation after correction of satellite-derived NH_3 concentrations; (c) comparison of corrected satellite-derived and ground-based NH_3 concentrations in 2015 (Unit: ppb).

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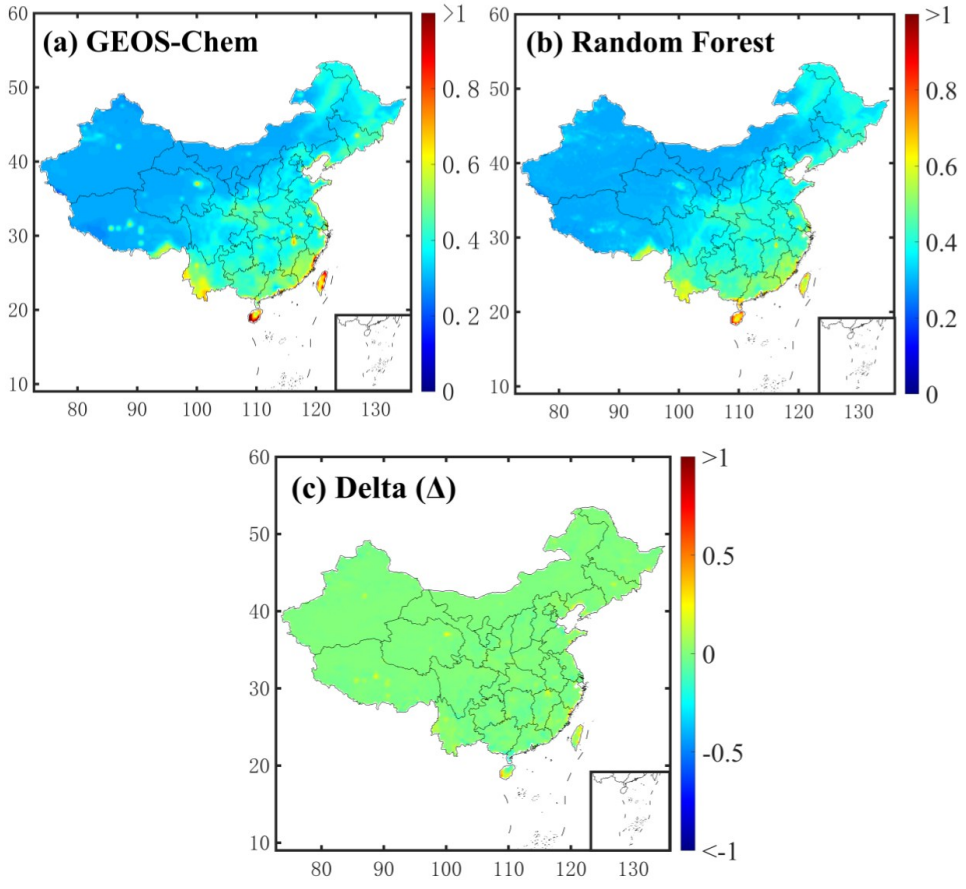

Figure 5. NH_3 dry deposition velocity in China in 2015: (a) GEOS-Chem simulation; (b) Random forest simulation; (c) Model difference (Unit: $cm \cdot s^{-1}$)

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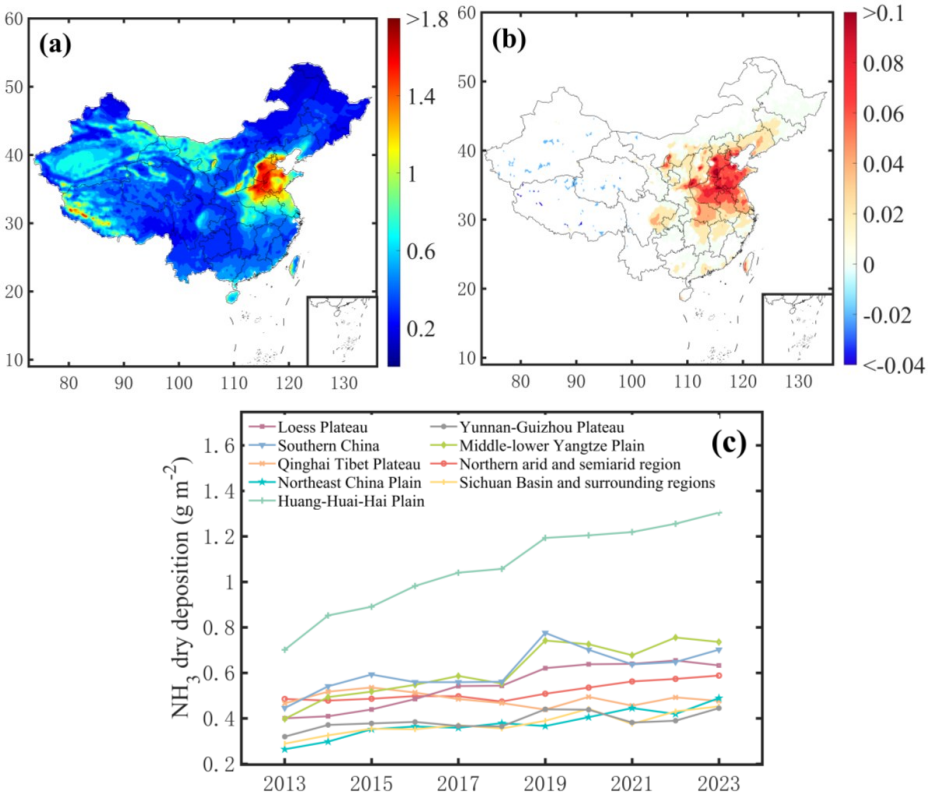

Figure 6. Spatial and regional trends in annual mean NH_3 dry deposition in China from 2013 to 2023: (a) spatial distribution of annual mean NH_3 dry deposition (Unit: $g \cdot m^{-2}$); (b) temporal trend of NH_3 dry deposition (Unit: $g \cdot m^{-2} \cdot yr^{-1}$); (c) interannual variation of NH_3 dry deposition across different regions (Unit: $g \cdot m^{-2}$).

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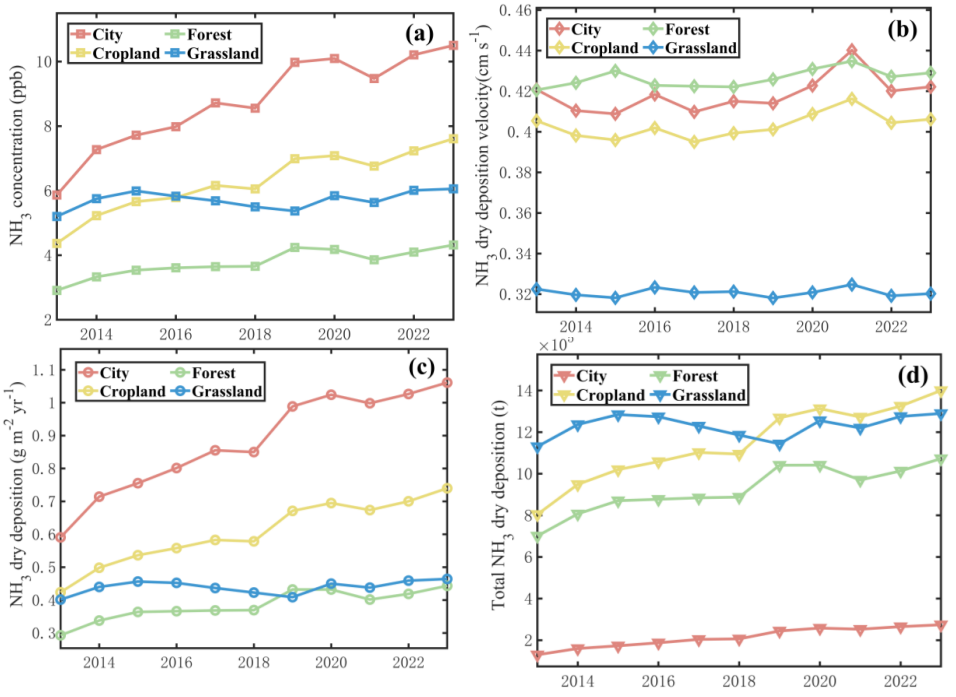

Figure 7. Trends in NH_3 concentration, dry deposition velocity, and dry deposition amount in China from 2013 to 2023: (a) trends in corrected NH_3 concentrations across different land surface types (Unit: ppb); (b) NH_3 dry deposition velocities over different land surface types (Unit: $cm \cdot s^{-1}$); (c) trends in NH_3 dry deposition flux per unit area over different land surface types (Unit: $g \cdot m^{-2}$); (d) interannual variation in total NH_3 dry deposition over different land surface types (Unit: t).

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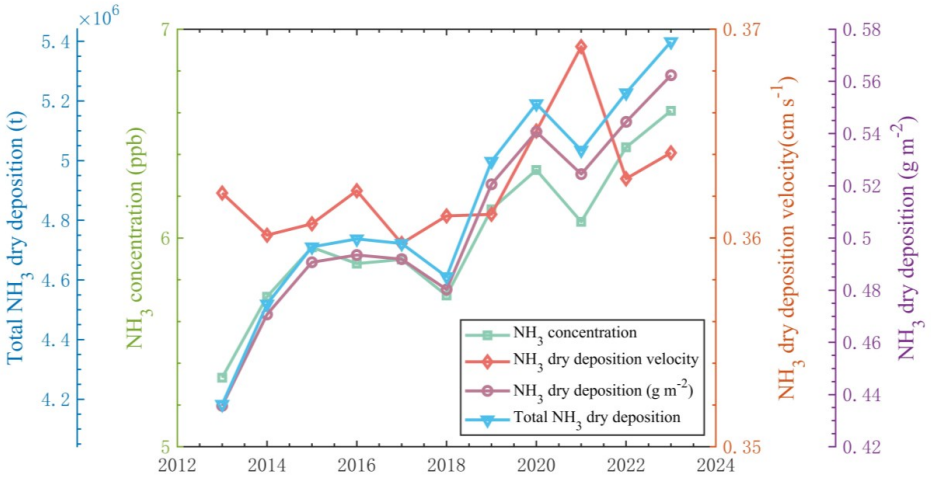

Figure 8. Annual changes in NH_3 concentration, dry deposition velocity, dry deposition flux and total dry deposition for China from 2013 to 2023;

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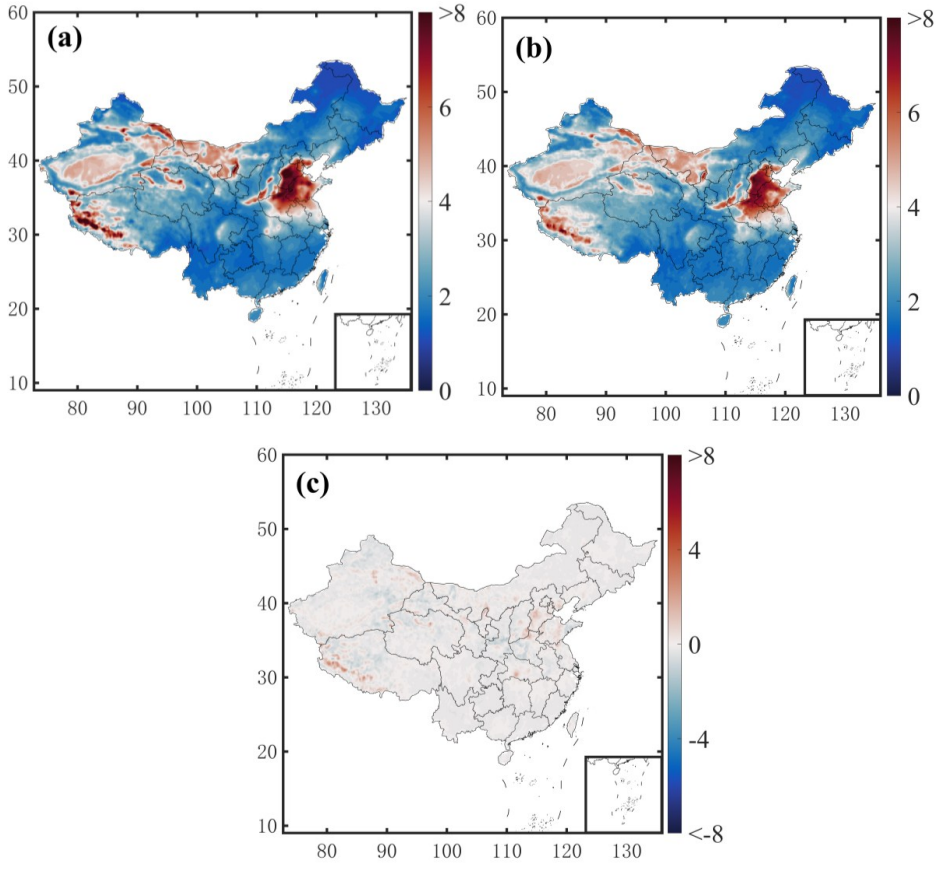

Figure 9. (a) Spatial distribution of satellite-based observation of NH_3 concentration for averages between 2013 and 2023, (b) simulation of NH_3 concentration by RF model for averages between 2013 and 2023, (c) difference between observation and model simulation, Units: ppb.

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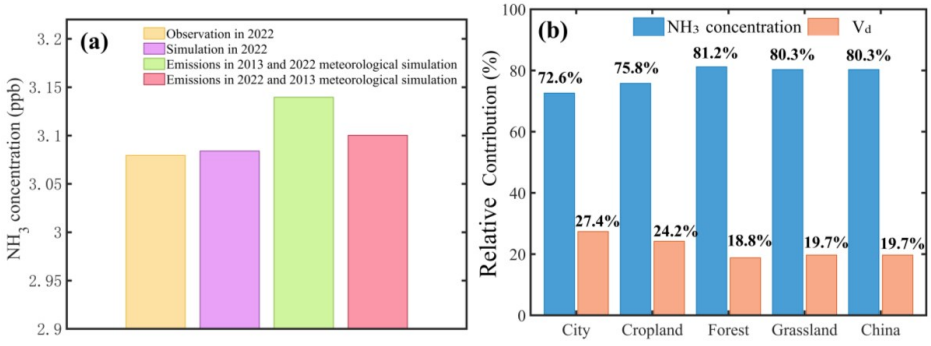

Figure 10. (a) The contribution of emissions and meteorological factors to the variation of NH_3 concentration; (b) The relative contribution of NH_3 concentration and dry deposition velocity to the dry deposition flux changes.

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Table 1. Dominant underlying surface types and their proportional area coverage in each defined region.

Agricultural zoning	Main underlying surface type	Area proportion
Northeast China Plain	Cropland	46.28%
Yunnan-Guizhou Plateau	Forest	68.48%
Northern arid and semiarid region	Barren	47.58%
Southern China	Forest	70.60%
Sichuan Basin and surrounding regions	Forest	43.01%
Middle-lower Yangtze Plain	Forest	47.26%
Qinghai Tibet Plateau	Grassland	70.47%
Loess Plateau	Forest	37.63%
Huang-Huai-Hai Plain	Cropland	57.15%

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Table 2. Annual and seasonal average NH_3 concentrations and their annual mean increase rates.

Season	NH_3 concentration (ppb)	Annual growth in NH_3 concentration (ppb yr^{-1})
Annual	2.88	0.045
Spring	3.28	0.039
Summer	3.59	0.065
Autumn	2.63	0.050
Winter	2.00	0.023

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Table 3. Average NH_3 concentration per unit area and annual mean increment and corrected NH_3 concentration in the nine major agricultural regions of China from 2013 to 2023.

Agricultural zoning	NH_3 concentration (ppb)	Annual growth in NH_3 concentration (ppb yr^{-1})	Corrected NH_3 concentration (ppb)
Huang-Huai-Hai Plain	5.29	0.24	11.36
Northern arid and semiarid region	3.29	0.08	6.93
Qinghai Tibet Plateau	3.09	-0.03	6.48
Loess Plateau	2.90	0.14	6.05
Middle-lower Yangtze Plain	2.70	0.13	5.62
Southern China	2.01	0.06	4.09
Northeast China Plain	2.01	0.08	4.09
Sichuan Basin and surrounding regions	1.98	0.06	4.02
Yunnan-Guizhou Plateau	1.75	0.03	3.52

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Table 4. Average NH_3 dry deposition per unit area and annual mean increment in the nine major agricultural regions of China from 2013 to 2023.

Agricultural zoning	Dry deposition of NH_3 (g m^{-2})	Annual growth of NH_3 dry deposition (g m^{-2} yr^{-1})	Agricultural zoning	Dry deposition of NH_3 (g m^{-2})	Annual growth of NH_3 dry deposition (g m^{-2} yr^{-1})
Huang-Huai-Hai Plain	1.064	0.054	Southern China	0.486	0.020
Northern arid and semiarid region	0.612	0.012	Northeast China Plain	0.389	0.018
Qinghai Tibet Plateau	0.611	-0.004	Sichuan Basin and surrounding regions	0.377	0.014
Loess Plateau	0.546	0.030	Yunnan-Guizhou Plateau	0.376	0.008
Middle-lower Yangtze Plain	0.517	0.034			

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Table 5. Comparison of global and regional NH_3 concentrations and dry deposition rates across different studies. note: All results have been standardized to uniform units.

Reference	Study period	Study region	NH_3 dry deposition (g m^{-2} yr^{-1})		NH_3 concentration (ppb)	
This study	2013-2023	China	City	0.88	City	8.76
			Forest	0.38	Forest	3.76
			Cropland	0.61	Cropland	6.27
			Grassland	0.44	Grassland	5.72
			China	0.51	China	4.98
Liu et al., 2020a	2008-2016	Global	0.17		--	
		China	0.58		Crop	8.04
					Urban	6.86
					Forest	4.66
					Grass	3.10
					Grass	3.37
					Mean	4.15
		Europe	0.36		Crop	4.00
					Urban	4.52
					Forest	3.32
					Grass	2.34
					Grass	1.87
					Mean	3.13
		US	0.26		Crop	4.38
					Urban	3.10
					Forest	2.51
					Grass	2.91
					Grass	1.87
					Mean	2.65
Jia et al., 2016	2005–2014	Asia (China)	0.29 (0.68)		--	
		North America	0.042 (0.078)		--	

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		(US)				
		Europe	0.11		--	
		Africa	0.32		--	
		South America	0.12		--	
		Oceania	0.037		--	
		Global land	0.18		--	
Kharol et al., 2018	2013 warm season (April–September)	North America	0.06-1.22		--	
		USA	0.27		--	
		Canada	0.18		--	
Zhang et al. 2012	2006–2008	US	0.11		--	
Liu et al., 2019	2008-2016	China	--		4.15 (0.39-22.90)	
		Europe	--		3.14 (0.07-16.58)	
		US	--		2.66 (0.24-18.52)	
Xu et al., 2015	2010-2014	China	1.00 (0.06-1.95)		10.65 (0.52-22.89)	
Phillips et al., 2006	1999 Summer	North Carolina	0.36		--	
Hu et al., 2020	November 2017	Tall-tower (100 m) observations in Minnesota	Forested lands	0.10-0.16	56 m	6.76
			Agricultural lands	0.41-0.62	100 m	6.64
Shao et al., 2019	October - November 2018	Nanjing	--		21.96±9.61	
Hu et al., 2021	2017-2019 warm season	US Corn Belt	Forested lands	0.054±0.0054, 0.059±0.011, 0.059±0.011	Forested lands	0.58±0.12, 0.71±0.14, 0.60±0.12
			Agricultural lands	0.77±0.16, 0.76±0.16, 0.77±0.16	Agricultural lands	6.87±1.4, 6.76±1.4, 6.48±1.3

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Van Der Graaf et al., 2018	2014 warm season	Europe	LOTOS-EUROS model	0.21	--
			IASI	0.27	