



Atmospheric NH₃ in urban Beijing: long-term variations and implications for secondary inorganic aerosol control

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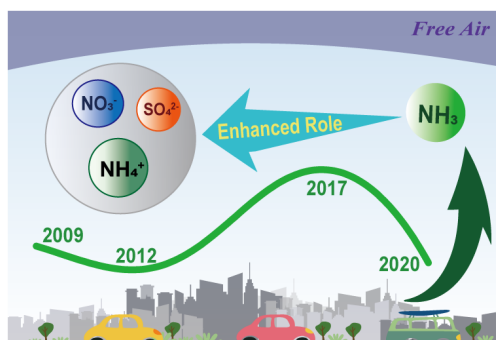
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Abstract. Ammonia (NH₃) has major effects on the environment and climate. In situ measurements of NH₃ concentrations taken between June 2009 and July 2020 at an urban site in Beijing were analyzed to study its long-term behavior, responses to meteorological conditions, and influences on the formation of secondary inorganic aerosols (SIAs). The 11-year average NH₃ mixing ratio was 26.9 ± 19.3 ppb (median 23.5 ppb). The annual average NH₃ mixing ratio increased from 2009 to 2017 by 50 % and then decreased by 49 % from 2017 to 2020. Notably, the long-term trend for NH₃ at the ground level did not align with the trends derived from satellite observations and emission estimates. The NH₃ concentration exhibited a stronger correlation with the daily variation in water vapor (H₂O) concentration than with air temperature. Thermodynamic modeling revealed the nonlinear response of SIAs to NH₃, with increased sensitivity when its concentration was reduced to 40 % of the initial level. Although reducing NH₃ concentrations can improve air quality during winter, controlling acid gas concentrations has a greater effect than controlling NH₃ concentrations on reducing SIA concentrations, until NH₃ and acidic gas concentrations are reduced below 80 % of their current levels. Nevertheless, the increased mass proportion of ammonium salts in SIAs during the observation period indicates that future control measures for NH₃ concentrations may need to be prioritized in Beijing.

Graphical abstract



1 Introduction

Excessive input of anthropogenic nitrogen into the environment can directly harm ecosystems and influence climate change (Charlson et al., 1991; Reay et al., 2008; Shadman et al., 2016). As the most abundant alkaline trace gas in the atmosphere (Meng et al., 2017), NH₃ interacts with the oxidized products of atmospheric acidic gases to form secondary aerosols, which considerably affect the radiative balance of the atmosphere and air quality (Fuzzi et al., 2015). Over the years, China has been committed to controlling air pollution and has effectively managed the emissions of primary pollutants such as sulfur dioxide (SO₂) and nitrogen oxide

(NO_x). However, particulate matter 2.5 (PM_{2.5}, particulate matter with a diameter less than 2.5 μm in size) pollution is still a severe problem. Existing studies on controlling SO₂ and NO_x emissions indicate that controlling NH₃ emissions is the most economically effective way to reduce PM_{2.5} concentrations (Gu et al., 2021; Pinder et al., 2008; Xie et al., 2022). However, the effectiveness of NH₃ reduction varies by region (M. Liu et al., 2019; Karydis et al., 2021), and there is still a debate regarding the efficacy of NH₃ reduction measures (Guo et al., 2018; Meng et al., 2022; Wei and Mohamed Tahrin, 2023).

Anthropogenic sources are the primary contributors to atmospheric NH₃ emissions (Olivier et al., 1998). In China, agricultural sources dominate, accounting for approximately 80 % of total emissions (Zhou et al., 2015). However, the contribution of non-agricultural sources in urban areas is considered significant. Studies indicate that over 30 % of NH₃ emissions observed in urban areas can be attributed to traffic (Elser et al., 2018; Gu et al., 2022a; Walters et al., 2022). Nevertheless, some research suggests that biogenic sources (primarily green spaces) predominate in urban areas and account for approximately 60 % of emissions (Teng et al., 2017), while the contribution from traffic sources is negligible (Yao et al., 2013). The complexity of urban NH₃ sources results in intricate variability in its atmospheric characteristics.

Long-term observations are important for analyzing the environmental impacts and control strategies of atmospheric NH₃. In Europe (Horváth and Sutton, 1998; Sutton et al., 2001; Horvath et al., 2009; den Bril et al., 2011; Lolkema et al., 2015; Tang et al., 2018), North America (Butler et al., 2016; Yao and Zhang, 2019; Yamanouchi et al., 2021), and Asia (Yamamoto et al., 1995, 1988; Saraswati et al., 2017), studies have been conducted on NH₃ variations over a period of 5 years or more. In most of these regions, NH₃ concentrations have either remained stable or exhibited an increasing trend. Satellite observations have detected rising global atmospheric NH₃ concentrations, influenced by reductions in acidic gas emissions, temperature increases, and the rising use of chemical fertilizers (Warner et al., 2017). In China, according to the monitoring results from the Nationwide Nitrogen Deposition Monitoring Network (NNDMN), NH₃ concentrations at 12 urban sites and 43 rural sites increased by approximately 80 % from 2011 to 2018 (Wen et al., 2020). Satellite data analysis by Dong et al. (2023) indicated a significant increase (~ 32 %) in NH₃ vertical column densities in China from 2008 to 2019. In the North China Plain, a hotspot for global NH₃ emissions, Luo et al. (2020) found a rapid increase in urban NH₃ concentration from 2011 to 2018. Wen et al. (2024) found a 26 % decrease in Beijing NH₃ concentrations from August 2005 to August 2020 and a 50 % increase from January 2005 to January 2020. To date, long-term ground-based observations of atmospheric NH₃ at high temporal resolution are relatively rare in China, and the contrasting trends between NH₃ emissions and satellite- and

in situ-measured concentrations in urban areas have not been fully explored.

The present study examined high-temporal-resolution NH₃ observations at the surface in urban Beijing from 2009 to 2020. Using data from emission inventories, satellite observations, meteorological elements, concentrations of various types of atmospheric pollutants, and particle ion composition, the present study aimed to obtain the characteristics of long-term variations in NH₃, influencing factors, and the contributions of NH₃ to particle formation in the atmosphere of Beijing. Analyzing long-term NH₃ observations helps to understand how changes in NH₃ concentrations have affected atmospheric pollution in the past. This knowledge is crucial for predicting future atmospheric pollution and formulating effective environmental policies. Furthermore, it provides a scientific basis and reference for developing future NH₃ control strategies.

2 Materials and methods

2.1 Data

Between June 2009 and July 2020, data on continuous on-line measurements of NH₃ concentrations were collected in Haidian District, Beijing (39°95' N, 116°32' E; Fig. S1 in the Supplement). From June 2009 to September 2017, data were collected from an observational site located on the third floor of a building within the premises of the China Meteorological Administration (CMA). Subsequently, the observation site was relocated to the 14th floor of the Science and Technology Building of Minzu University of China (MUC), which is less than 1 km away from the previous location, located just across the road from it. The ground-floor elevations of both buildings are 56 m above sea level, and the observation heights above the ground are 10 m on the 3rd floor and 56 m on the 14th floor. Both observation sites are surrounded mostly by urban roads, office spaces, residential areas, and parks, and no large-scale industrial sources of NH₃ were located near the site at the time of study.

Beginning in June 2009, NH₃ concentration monitoring was conducted using an EC9842 NO_x / NH₃ analyzer (Ecotech, Australia). Starting in April 2015, additional NH₃ measurements were simultaneously taken using an NH₃ Economical Ammonia Analyzer (EAA; Los Gatos Research, USA). From May 2016 onward, only the NH₃ EAA was used. The EC9842 NO_x/NH₃ analyzer employs gas-phase chemiluminescence to continuously analyze NH₃, NO_x, and N_x concentrations; its detection limit is less than 2 ppb and data record time is 1 min. The instrument was subject to weekly zero and span checks to identify potential analyzer faults and response drift. Multipoint calibrations were typically performed every month, and data were corrected on the basis of the multipoint calibrations. The NH₃ EAA features a low detection limit of less than 0.2 ppb and a maximum drift of 0.2 ppb within 24 h, with a time resolution of 50 s, and it

utilizes off-axis integrated cavity output spectroscopy technology. At the CMA site, the air had been drained into an air-conditioned room with a 4.5 m long Teflon line, and the inlet height was 1.8 m above the rooftop (about 12 m above ground level). At the MUC site, air was introduced from outside the sealed window through a borehole, with the air inlet extending 20–30 cm outside the window. Since the site is on the 14th floor, the air outside the building flows smoothly. To maintain data comparability, NH₃ standard gases, which had been traceable to a uniform standard, were used as measurement references. The comparison result of the two instruments can be found in Zhang et al. (2021), in which the two instruments exhibited a considerable correlation, with a correlation coefficient of 0.949 ($n = 5316$, $p < 0.01$) and slope s of 0.999 ± 0.005 .

During data analysis, minute-level data were converted into hourly average data. Throughout the observation period, a total of 40 692 and 46 917 valid hourly average data points were obtained from the EC9842 and EAA instruments, respectively, resulting in a total of 13 420 datasets being obtained simultaneously through measurements on the two instruments. These two sets of results exhibited a significant correlation ($N = 13\,420$, slope = 1.09, $R = 0.95$, $p < 0.05$), and the parallel observations from the two analyzers were generally consistent (Fig. S2). The NH₃ observation data were finalized by averaging the synchronized data.

Furthermore, NH₃ satellite observation data were obtained through the Metop-A satellite's Infrared Atmospheric Sounding Interferometer (IASI) remote sensing product. These data had a spatial resolution of $12 \times 12 \text{ km}^2$ and were collected monthly (Van Damme et al., 2017). In the present study, daytime satellite NH₃ data from June 2009 to April 2020 were used. The average NH₃ satellite observation results for Beijing were calculated using data for the region spanning 36.5 to 42.5° N in latitude and 113.5 to 118.5° E in longitude. The trend for satellite observation values obtained at the grid point at the location of the monitoring station closely matched the trend for the average observation values collected for this region (Fig. S3). NH₃ emission inventory data for Beijing (from June 2009 to December 2017) are presented in Fig. S4, comparing NH₃ emissions from Beijing and its surrounding areas (Huang et al., 2012; Kang et al., 2016). Meteorological data collected between June 2009 and February 2012 were obtained from the Beijing Capital International Airport station. From March 2012 to April 2020, meteorological data were sourced from the Haidian Meteorological Station. The temperature and relative humidity data acquired from the two stations exhibited a high level of correlation (Fig. S5). Absolute humidity was calculated using the acquired temperature and relative humidity data. Data for other pollutants such as PM_{2.5}, SO₂, and NO₂ were acquired from the Wanliu Monitoring Station in Haidian District, Beijing. These monitoring data were collected between 2 April 2014 and 11 July 2020. Figure S6 provides additional details of these data.

In the present study, offline sampling of PM_{2.5} was conducted on the rooftop of the School of Pharmacy at Minzu University in China. Atmospheric samples were collected twice daily, specifically from 06:00 to 17:00 (all times are given in local time throughout the paper; daytime sampling) and from 18:00 to 05:00 on the following day (nighttime sampling). The sampling periods were from 8 to 21 September 2018 (autumn), 6 to 21 November 2018 (autumn), 1 to 21 January 2019 (winter), 3 to 21 March 2019 (spring), 8 to 15 May 2019 (spring), and 8 to 21 June 2019 (summer). The collected PM_{2.5} samples on filters were subsequently sent to the Chinese Academy of Meteorological Sciences for chemical analysis of ion components (Na⁺, SO₄²⁻, NH₄⁺, NO₃⁻, Cl⁻, Ca²⁺, K⁺, and Mg²⁺), from which 184 datasets were obtained. Additionally, data from the study by Hu et al. (2014) that spanned 5 May to 30 November 2009 and data from the study of Wu et al. (2019) that spanned 15 to 23 December 2016 were used in the present study as references for monitoring PM_{2.5} components within the premises of the China Meteorological Administration.

2.2 Data analysis methods

2.2.1 Long-term trend analysis

Long-term trends of atmospheric NH₃ were obtained using ensemble empirical mode decomposition (EEMD) (Wu and Huang, 2009). This method adaptively decomposes a signal into a series of intrinsic modal functions (IMFs) from high to low frequencies. It separates oscillation or trend components of varying scales from the original signal. EEMD integrates the advantages of wavelet analysis and augments the empirical mode decomposition (EMD) method by introducing white noise. This enhancement effectively mitigates the mode mixing problem inherent in the EMD method. EEMD demonstrates greater stability in decomposing nonlinear and non-stationary data series, enabling the accurate extraction of genuine signal variations (Qian et al., 2011). Currently, EEMD has been used in studies on air-quality trend analysis (Yao and Zhang, 2016; Fu et al., 2020; Wang et al., 2022; Wang and Zhang, 2023). In the present study, the EEMD was performed using the Rlibeemd package of the R programming language (Luukko et al., 2016).

2.2.2 Thermodynamic modeling

The ISORROPIA II model is mainly used to simulate the physical state and concentration of inorganic components of the aerosol system at thermodynamic equilibrium. A distinct advantage of the ISORROPIA II model over other thermodynamic models is the inclusion of the K⁺, Ca²⁺, and Mg²⁺ ions in the calculations, and taking these components into account significantly improves the accuracy of the model simulations (Allen et al., 2015). Additionally, the high precision and computational efficiency of the ISORROPIA II model have been widely demonstrated (Fountoukis and Nenes,

2007). To assess the sensitivity of sulfate, nitrate, and ammonium (SNA) to changes in precursor concentrations, the present study employed the ISORROPIA II thermodynamic equilibrium, version 2.3 (<http://isorroopia.epfl.ch>, last access: 8 April 2023). The model was run in “forward + metastable” mode, taking inputs such as temperature (in kelvins), relative humidity (up to 1), and concentrations of particulate components (SO₄²⁻, Cl⁻ + HCl, NO₃⁻ + HNO₃, NH₄⁺ + NH₃, Na⁺, K⁺, Ca²⁺, and Mg²⁺) expressed in μg m⁻³ for calculations.

3 Results and discussion

3.1 Long-term variations in NH₃

From June 2009 to July 2020, the hourly average mixing ratio of atmospheric NH₃ in Beijing was 26.9 ± 19.3 ppb (median 23.5 ppb). Table S1 summarizes results from various NH₃ monitoring studies conducted in urban areas. The results of the present study are basically consistent with the annual NH₃ mixing ratio averages that have been observed in urban Beijing by other researchers through optical instruments (Gu et al., 2022a, 2022b; Pu et al., 2020; Sun et al., 2023; Wang et al., 2019). As a densely populated country with intensive agricultural activities, China contains several areas that are major global hotspots for atmospheric NH₃ concentration (L. Liu et al., 2019; Van Damme et al., 2018). The monitoring results of the present study indicate that the overall NH₃ mixing ratio in Beijing is lower than that in Delhi (Saraswati et al., 2019; Singh and Kulshrestha, 2014) but considerably higher than those in other developed cities such as New York, Toronto, and Rome (Chatain et al., 2022; Nguyen et al., 2021; Park et al., 2021; Perrino et al., 2002; Phan et al., 2013; Zbieranowski and Aherne, 2012; Zhou et al., 2019). Even within China, the NH₃ mixing ratio in Beijing is higher relative to that in Shanghai, which is also a megacity (i.e., the NH₃ mixing ratio in Shanghai is less than one-third of that in Beijing), and only a few cities in northern China have mixing ratios comparable to that in Beijing (Cao et al., 2009; Chang et al., 2019; Huang et al., 2021; Pan et al., 2018). The primary reasons for this phenomenon are the frequent agricultural activities and the presence of highly alkaline soils in the North China Plain, where Beijing is located (Ju et al., 2009).

Due to significant data gaps from January 2013 to June 2013 and from May 2017 to August 2017, the period from 2009 to 2020 was divided into three segments for linear regression analysis (Fig. S7). From June 2009 to January 2013, the observed hourly average atmospheric NH₃ mixing ratio showed a decreasing trend ($R = -0.23$, $p < 0.05$, slope = -0.01); from June 2013 to May 2017, the NH₃ mixing ratio increased ($R = 0.04$, $p < 0.05$, slope = 0.22×10^{-2}); and from September 2017 to July 2020, the NH₃ concentration exhibited a decreasing trend again ($R = -0.03$, $p < 0.05$, slope = -1.42×10^{-3}). Similarly to in situ observations, the satellite observations of NH₃ concentration showed

a decreasing trend from June 2009 to January 2013 ($R = -0.19$, $p < 0.05$, slope = -3.88×10^{-4}) and an increasing trend from June 2013 to May 2017 ($R = 0.12$, $p < 0.05$, slope = 3.65×10^{-4}) but differed from in situ atmospheric NH₃ trends as it continued to rise from September 2017 to July 2020 ($R = 0.23$, $p < 0.05$, slope = 1.17×10^{-3}).

To further analyze the long-term trends of the atmospheric NH₃ concentration, the present study referred to the findings of Vu et al. (2019) and used meteorological factors to construct a random forest model for imputing missing values. The computed time series for the atmospheric NH₃ concentration is presented in Fig. S8. The complete dataset obtained through EEMD was used to characterize the changes in atmospheric NH₃ concentrations in Beijing (Fig. 1). The NH₃ mixing ratio initially exhibited a slight decrease but started to increase in 2012 and peaked in 2017, subsequently declining. From 2009 to 2017, the NH₃ mixing ratio increased by 50 %, but by 2020, the NH₃ mixing ratio had decreased by 49 % from its peak in 2017. A comparison of monthly average NH₃ concentrations obtained from satellite observations revealed that prior to 2018, the trend for the surface NH₃ mixing ratio was similar to that observed by satellites, exhibiting a decline followed by an increase in atmospheric NH₃ concentrations. However, starting in 2018, these two trends diverged, with satellite observations indicating a continued increase in NH₃ concentrations, while the surface NH₃ mixing ratio exhibited a decreasing trend. The monitoring results of this study were compared with NH₃ monthly concentrations observed by NNDMN in Beijing from April 2011 to December 2015 (Xu et al., 2019) and from January 2005 to August 2020 (Wen et al., 2024) (Fig. S9). From April 2011 to December 2015, both the NH₃ mixing ratio observed in this study ($R = 0.27$, $p < 0.05$) and the satellite-observed concentrations ($R = 0.28$, $p < 0.05$) exhibited increasing trends, while the NNDMN station did not show a significant trend ($R = 0.16$, $p > 0.05$). The NNDMN station observations from January 2009 to August 2020 were significantly correlated with this study's observations ($R_{\text{Aug}} = 0.66$, $p < 0.05$; $R_{\text{Jan}} = 0.65$, $p < 0.05$), but neither the present study's observations nor the NNDMN observations were significantly correlated with satellite-observed NH₃ concentrations. Satellite observations showed a strong correlation between NH₃ concentrations in the Beijing urban area and the Beijing–Tianjin–Hebei region (Fig. S3). However, measurements by Zhang et al. (2020) at five stations in Beijing indicated that four stations had lower NH₃ concentrations in 2017 (winter) than in 2020 (winter + spring), while one station had higher concentrations in 2017 than in 2020, indicating variability in observation results even within the same city. Due to the short atmospheric lifetime, low transport altitude, high dry deposition rate, limited transport distance, and abundance of atmospheric NH₃, its complex temporal and spatial characteristics contribute to the complexity of NH₃ variations (Asman and van Jaarsveld, 1992; Nair and Yu, 2020). Satellite observations are limited by the observation height and spa-

tial resolution, which may mask variations in local surface NH₃ concentrations. Additionally, differences between the present study's observations and satellite observations may also be due to changes in the monitoring location and observation height in September 2017. However, tower observations conducted by the Institute of Atmospheric Physics, Chinese Academy of Sciences (6.7 km from the present study's site), in the urban area showed only slight variations within a 300 m altitude range (Wang et al., 2019; Zhang et al., 2019). Therefore, the change in observation altitude may have had a limited impact on the change in NH₃ mixing ratio trends after 2017.

3.2 Influences on variation characteristics of NH₃

NH₃ emissions directly affect the variations in atmospheric NH₃ concentrations. The emission inventory data (Fig. 1) indicate that NH₃ emissions in Beijing remained stable from 2009 to 2014, peaking in 2012. After 2014, NH₃ emissions in Beijing rapidly decreased, declining by 25 % from 2014 to 2017. However, during this period of declining emissions, the NH₃ mixing ratio in Beijing exhibited an increasing trend. Similar phenomena have been reported by studies conducted outside of China. For instance, in Scotland, NH₃ emissions decreased by approximately 15 % from 1990 to 2003, whereas atmospheric NH₃ concentrations increased (Friedman and Schwartz, 2011). In Hungary, NH₃ emissions were estimated to have decreased by 50 % from 1983 to 1993; however, NH₃ concentrations exhibited a slight upward trend during this monitoring period (Horvath et al., 2009). A possible reason for these differences between NH₃ emissions and concentrations could be the significant reduction in the concentrations of SO₂ and NO_x, which reduced the amount of atmospheric NH₃ neutralized by acid gases (Fu et al., 2017; Lachatre et al., 2019; Liu et al., 2018; Yu et al., 2018). Over the past 2 decades, Beijing has implemented a series of strict measures to control air pollution and has achieved considerable success (United Nations Environment Programme, 2019). The concentrations of SO₂, NO₂, CO, PM₁₀, and PM_{2.5} in Beijing all exhibited decreasing trends; in particular, the concentration of SO₂ decreased by 88 % from 2009 to 2020 (Fig. 2).

To discuss the influence of chemical loss on the annual increase in NH₃ concentrations, the present study referred to research by Yao and Zhang (2019), assuming that NH₄⁺ is uniformly distributed in the urban area of Beijing and that changes in NH₄⁺ concentrations directly affect atmospheric NH₃ concentrations on a 1 : 1 basis. By calculating the change in NH₄⁺ concentration relative to the baseline year, we adjust the atmospheric NH₃ concentrations. The present study set 2009 as the baseline year, using the annual average NH₄⁺ concentration observed by Cheng (2021) in the urban area of Beijing to calculate the adjusted NH₃ concentrations from 2009 to 2017. The calculations show (Fig. S10) that, overall, the original NH₃ concentration in 2017 was

50 % higher than in 2009 and the adjusted NH₃ concentration was 46 % higher. Therefore, changes in chemical losses have a limited impact on the increased trend of NH₃ concentrations, and the discrepancy between NH₃ concentrations and emission trends may be due to imperfections in the emission inventory.

Various meteorological factors can influence the atmospheric NH₃ concentration. Among the factors identified, temperature has been reported to be positively correlated with NH₃ concentration. An increase in temperature can increase soil NH₃ emissions, leading to the equilibrium shift of particulate NH₄NO₃ toward gaseous NH₃, which increases NH₃ concentration (Behera et al., 2013; Li et al., 2014). During the observation period, the temperature in Beijing followed the seasonal sequence of summer (being the warmest) followed by spring, autumn, and winter, and the rankings of NH₃ mixing ratios across the seasons were consistent with the trend. Other studies conducted in temperate regions of the Northern Hemisphere have reported similar findings (Liu et al., 2021; Shon et al., 2012; Wang et al., 2018). The interannual trends for temperature and NH₃ mixing ratios across multiple seasons (Fig. 3) revealed that temperature remained stable in summer and autumn over the years; when calculated in kelvins, the average seasonal temperatures exhibited interannual variation coefficients of 0.42 % in spring, 0.15 % in summer, 0.17 % in autumn, and 0.51 % in winter. For the two seasons of summer and autumn, no significant correlation was identified between the annual average NH₃ mixing ratio and the variations in temperature over the years. After 2014, the annual average temperature in spring remained stable, whereas the NH₃ mixing ratio gradually decreased, possibly because of a reduction in agricultural activities. A weak positive correlation was identified between the annual average NH₃ mixing ratio and temperature only in winter, and the significant increase in winter temperature from 2013 to 2014 could have led to the high NH₃ mixing ratios in the winter of 2014.

Our investigation examined the correlations among daily temperature, absolute humidity, and diurnal fluctuations in atmospheric NH₃ concentrations throughout an observation period of 4058 d (screening criteria for effective dates: $p < 0.05$ and ≥ 18 effective hours per day; the p values were adjusted using the Benjamini–Hochberg method; Benjamini and Hochberg, 1995). We observed that temperature exhibited both positive (45 %) and negative (55 %) correlations with the NH₃ mixing ratio, with these two categories each accounting for nearly half of the valid observation days. However, on most days (i.e., 93 % of the valid observation days), absolute humidity was positively correlated with the NH₃ mixing ratio (Fig. S11). Overall, the average daily variations in NH₃ mixing ratios in Beijing in spring, summer, and autumn indicated a significant negative correlation with the temperature ($R_{\text{spring}} = -0.93$, $R_{\text{summer}} = -0.72$, $R_{\text{autumn}} = -0.76$, $p < 0.01$). The NH₃ mixing ratio was positively cor-

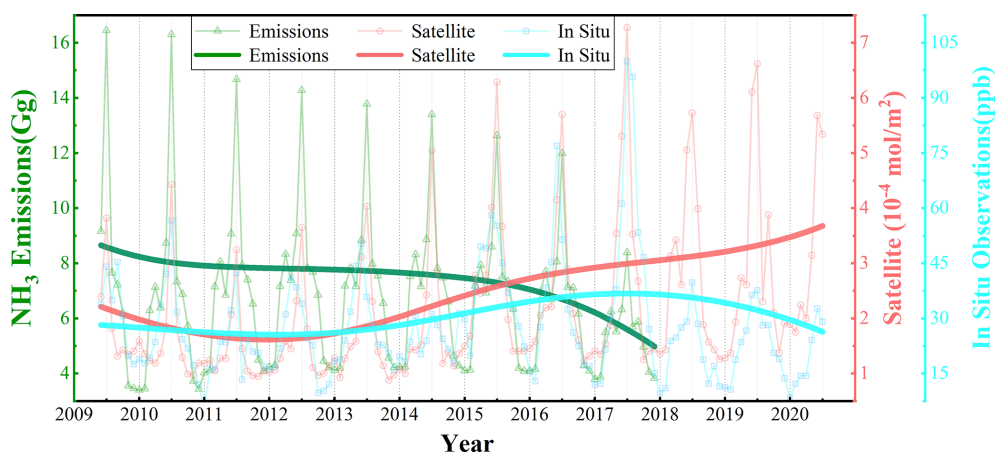


Figure 1. Monthly averages of surface observations and satellite inversions of NH₃ concentrations and total NH₃ emissions in Beijing from June 2009 to July 2020 (fine light line) and trends pertaining to changes (thick solid line).

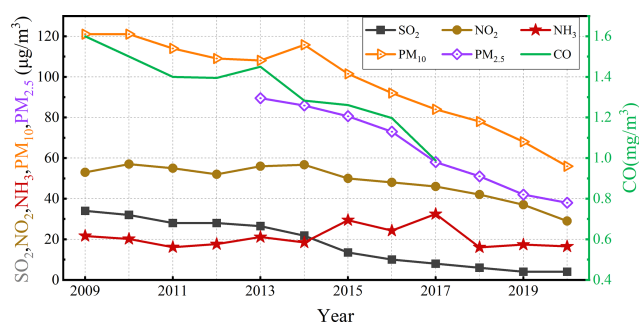


Figure 2. Annual average concentrations of atmospheric NH₃ and five other air pollutants in Beijing. The measurement unit is mg m⁻³ for CO concentration and µg m⁻³ for all other pollutants (air pollutant data were retrieved from the Beijing Environmental Bulletin website: <http://sthjj.beijing.gov.cn/bjhrb/index/xxgk69/sthjlyzwg/1718880/1718881/1718882/>, last access: 8 April 2023).

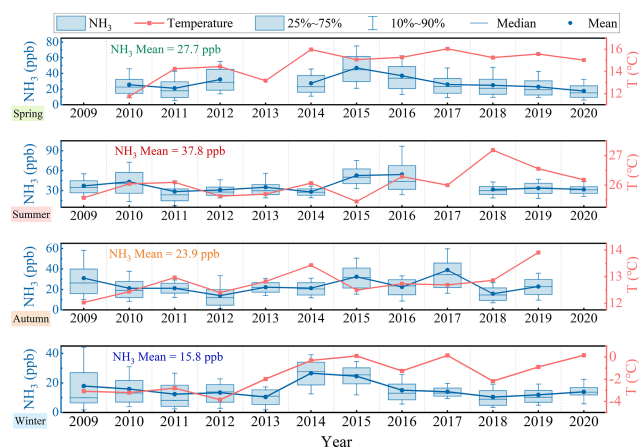


Figure 3. Interannual variations in the mean NH₃ mixing ratio and mean temperature in Beijing for each season.

related with absolute humidity ($R_{\text{spring}} = 0.80$, $R_{\text{summer}} = 0.50$, $R_{\text{autumn}} = 0.67$, $R_{\text{winter}} = 0.49$, $p < 0.01$).

Several studies have suggested that temperature plays a pivotal role in driving diurnal variations in atmospheric NH₃ concentrations (Clarisse et al., 2021; Langford et al., 1992). However, the present study shows that NH₃ concentrations are significantly influenced by temperature across seasonal changes (Fig. 3); in terms of diurnal patterns, the days showing positive and negative correlations between NH₃ concentrations and temperature each constitute nearly half of the effective observation days (Fig. S11). The mean diurnal variations in different seasons typically exhibit lower concentrations during the day and are higher at night in spring, summer, and autumn (Fig. S12). This difference suggests that correlations observed on a seasonal (climatic scale) tend to obscure lower-frequency data relationships, with daily variations in NH₃ concentrations being more influenced by the transition from day to night (meteorological or weather scale). This highlights the complex factors affecting NH₃ concentrations in the urban areas of Beijing and underscores the importance of high temporal resolution in observations. Several studies have noted a reduction in NH₃ concentrations during daylight hours (Buijsman et al., 1998; Sharma et al., 2014; Gu et al., 2022b; Lan et al., 2021). Increased temperatures during the day promote the volatilization of NH₃, but lower daytime concentrations may result from higher wind speeds and more favorable mixing conditions, whereas at night, NH₃ tends to accumulate within a shallower boundary layer (Buijsman et al., 1998). The diurnal variation in the boundary layer height in Beijing exhibits a single-peak pattern, rising rapidly from 06:00 to 08:00, reaching its peak between 14:00 and 15:00, then declining sharply, and stabilizing after 18:00 to 20:00 (Fig. S12). During the daytime, NH₃ concentrations are influenced by a combination of temperature (which promotes emissions) and changes in the boundary layer height (which causes dilution), with the

valley value of NH₃ concentrations lagging behind the peak times of boundary layer height and temperature. Moreover, during spring, summer, and autumn, the continuous decline in NH₃ concentrations during the daytime indicates that vertical mixing transport has a limited impact on atmospheric NH₃ in the urban areas of Beijing. In winter, the evaporation of dew or frost in the early morning leads to a rapid increase in H₂O and NH₃ concentrations (Wentworth et al., 2016), while the effects of afternoon temperature and vertical mixing dilution are comparable, keeping NH₃ concentrations relatively stable.

Several studies have reported a high correlation between the NH₃ mixing ratio and humidity. Previous research has shown that NH₃ can be significantly affected only by sharp changes in humidity, and a new balance requires tens of minutes to be re-established. Averaging minute-level data over 1 h can smooth the effects caused by variations in humidity. Notably, parallel observations in urban and suburban Beijing found that a positive correlation between daily NH₃ and H₂O concentration variations was only significant in urban areas (Lan et al., 2021). Gu et al. (2022b) reported that daily variations in NH₃ concentrations in urban Beijing, as measured by the Picarro ammonia analyzer and ChemComb, were consistent. These results suggest that atmospheric NH₃ cannot be explained solely by the influence of H₂O effects on the instruments. Additionally, Sun et al. (2023) discovered a positive correlation between atmospheric NH₃ concentrations and relative humidity (RH) in Beijing and a negative correlation in Shanghai. In rural northern China, He et al. (2020) also observed a strong correlation between NH₃ and RH, which they attributed to dew evaporation. At present, the relationship between the NH₃ mixing ratio and the water vapor concentration requires further clarification.

In summary, the results suggest that temperature plays a pivotal role in driving the seasonal variations in atmospheric NH₃ concentrations throughout a given year. However, in the long term, the influence of temperature and other meteorological factors may be masked. Regarding diurnal variations, our analysis revealed that a single-day increase in temperature did not consistently lead to a direct elevation in atmospheric NH₃ mixing ratios on most days. Conversely, atmospheric water vapor mixing ratios exhibited a consistently positive correlation with NH₃ mixing ratios throughout the day. Notably, the day-to-day variations in meteorological factors remained consistent across the years, whereas the variation in diurnal NH₃ differed across different years and seasons (Fig. S13). Therefore, the conclusion is that diurnal fluctuations in the atmospheric NH₃ concentration are not solely determined by meteorological factors. In recent years, scholars have been increasingly studying the contribution of traffic sources to urban NH₃ concentrations. Gu et al. (2022b) confirmed that vehicle exhaust emissions during winter in Beijing lead to the occurrence of morning peaks in NH₃ concentrations. Nonetheless, our results indicated that atmospheric NH₃ concentrations did not consistently peak in the morning

throughout the observation period, even when high concentrations of traffic emissions were present. Furthermore, the morning peaks for atmospheric NH₃ concentrations tended to occur earlier relative to those for CO, which are influenced by traffic emissions (Fig. S13).

Studies have comprehensively explored the influence of wind direction and wind speed on the pollutant mixing ratios in Beijing, and they have reported that southerly (S) winds transport a high concentration of pollutants to Beijing, leading to the accumulation of NH₃ in the city. Conversely, the winds from the north by west (NW) facilitate the dispersion and dilution of atmospheric NH₃ in Beijing (Lin et al., 2011; Meng et al., 2017). Figure S14 presents the wind rose diagrams for atmospheric NH₃ concentrations in various seasons and at various wind speeds. Under near-calm wind conditions (wind speed (*ws*) ≤ 1.5 m s⁻¹), the prevailing winds across all seasons were predominantly in the north-east and east-northeast directions, and NH₃ mixing ratios did not vary significantly because of the wind direction, indicating that local emissions had the most pronounced effect on atmospheric NH₃ concentrations. At low wind speeds (1.6 m s⁻¹ ≤ *ws* ≤ 3.3 m s⁻¹), the predominant wind direction varied across seasons, with southwesterly winds prevailing in spring and summer and northerly winds dominating in autumn and winter. At higher wind speeds (*ws* > 3.4 m s⁻¹), the predominant wind direction was NW in spring, autumn, and winter but southerly in summer. In general, changes in the prevailing wind direction did not significantly influence NH₃ mixing ratios across various wind sectors. However, in specific wind sectors, such as the west by south (WS) sector in spring, the east by south (ES) sector in summer and autumn, and the south by east (SE) sector in winter, higher wind speeds tended to lead to lower NH₃ mixing ratios (Fig. S15). Notably, the decline in NH₃ mixing ratios was more pronounced in wind sectors affected by NW winds, indicating that strong winds, particularly those from the NW direction, had a significant cleansing effect on NH₃ in Beijing. Conversely, southerly winds, and sometimes specific wind directions, contributed to NH₃ accumulation.

3.3 Influence of NH₃ on secondary inorganic aerosol formation

The present study investigated the role of atmospheric NH₃ in the formation of secondary inorganic aerosols (SIAs) in Beijing by analyzing the relationship between NH₃ and SNA concentrations during the observation period. According to the study of Wei et al. (2023) conducted between 2013 and 2020, the SNA concentrations in Beijing exhibited a significant downward trend. However, the proportion of SNA in PM_{2.5} (mass concentration) did not change substantially during this period. Table S2 lists the proportions of various SNA components in PM_{2.5} (mass concentration) recorded in urban areas of Beijing for the years 2009, 2016, 2018, and 2019. In the summer and autumn of 2009, SO₄²⁻ accounted for more

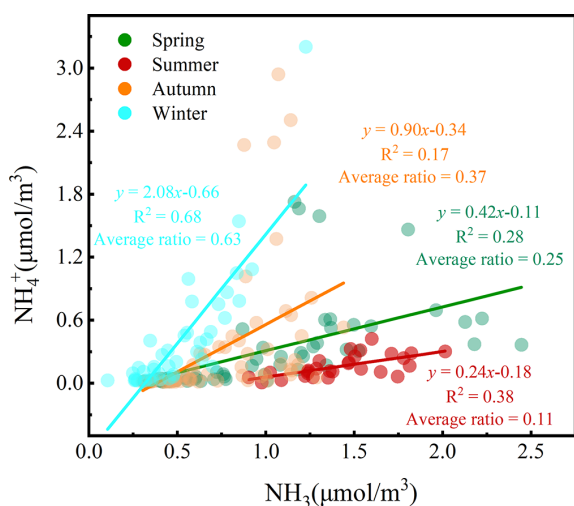


Figure 4. Correlation between gaseous NH₃ and fine particulate ion NH₄⁺.

than 50 % of SNA content, considerably exceeding the concentrations of NO₃⁻ and NH₄⁺. However, by 2016, except for the summer season when SO₄²⁻ was still the predominant component, NO₃⁻ became the dominant component of the SNA mass concentration. Over time, the proportion of NH₄⁺ in the SNA mass concentration increased across multiple seasons. Wen et al. (2024) and Cheng (2021) have also observed this phenomenon in urban Beijing. These findings indicate the necessity of controlling NH₃ and NO_x concentrations to mitigate future PM_{2.5} pollution.

Figure 4 depicts the relationship of NH₄⁺ in fine particulates to atmospheric NH₃. Overall, a positive correlation was identified between NH₄⁺ and NH₃, indicating that variations in the concentration of the precursor gas NH₃ influenced the formation of NH₄⁺. An increase in the NH₃ concentration led to a higher concentration of NH₄⁺ in fine particulate matter, and this effect was most pronounced in winter, during which the correlation between NH₄⁺ and NH₃ was the strongest ($R^2 = 0.68$, $p < 0.01$) and the average molar concentration ratio of NH₄⁺ to NH₃ was the highest. The seasonal differences in the response of aerosol NH₄⁺ to atmospheric NH₃ may mainly be caused by variations in meteorological conditions, in addition to those in precursor gases of SNA. For example, low temperature and high humidity promote the conversion of gaseous NH₃ to particulate NH₄⁺ (Wang et al., 2015). Thus, winter meteorological conditions may increase the formation of NH₄⁺ in fine particulate matter, exacerbating fine particulate pollution and haze formation.

To gain further insights, the ISORROPIA II thermodynamic equilibrium model was employed to simulate and analyze the sensitivity of SNA in PM_{2.5} to changes in precursor concentrations in each season. Concentrations of SO₄²⁻ + H₂SO₄ (TS), HNO₃ + NO₃⁻ (TN), and NH₃ + NH₄⁺ (TA) were increased or reduced by up to 20 %, and the changes

in simulated concentrations relative to the baseline (without perturbation) were calculated. The simulation results revealed a strong correlation between the simulated and observed NH₃ concentrations ($R_{\text{spring}} = 0.89$, $R_{\text{summer}} = 1.00$, $R_{\text{autumn}} = 0.97$, $R_{\text{winter}} = 0.98$, $p < 0.01$), indicating the reliability of the above observation-based results. The simulation results (Table S3) indicated that NH₄⁺ was the component least sensitive to changes in the TA concentration in spring, autumn, and winter, suggesting that atmospheric NH₃ was not the limiting reactant for the generation of (NH₄)₂SO₄ and NH₄NO₃ in these seasons during the observation period. The responses of NH₄⁺ to changes in TA and TN concentrations were less apparent in summer because NH₄⁺ was mainly bound to SO₄²⁻ rather than to NO₃⁻ during this hot season, in which the high temperature was unfavorable for the generation and retention of NH₄NO₃. Furthermore, the increase in TS had an over-proportional or nearly (winter) proportional perturbation effect on NH₄⁺, indicating the presence of sufficient NH₃ in the atmosphere of Beijing throughout the year. Su et al. (2021) have also suggested that the acidic components in the atmosphere in Beijing were sufficiently neutralized. Therefore, a relatively small reduction (20 %) in NH₃ abundance seems not to be able to significantly lower the SNA levels in Beijing.

When NH₃ was abundant, the sensitivity of SNA content to changes in TA within the ± 20 % range was low (within ± 2 %, Table S3). Under such conditions, changes in TS and TN concentrations had much larger perturbation effects on SNA concentrations. However, the winter SNA concentrations mostly responded linearly to changes in TS and TN concentrations but nonlinearly to changes in the TA ones (Fig. 5). If the TA concentrations, which were more pronounced in 2019, were reduced by 60 %, the rate of decrease in SNA content would have accelerated considerably. By contrast, the effect of changes in the TS concentration decreased after its reduction of over 40 %. In a previous study based on nationwide measurements and simulation, Meng et al. (2022) suggested that SNA content in China can be more effectively controlled by reducing the concentrations of acidic gases (SO₂ and NO_x) in the atmosphere than by reducing the concentration of NH₃ by the same percentage. Additionally, Zheng et al. (2022) discovered that the joint control of SO₂ and NO_x emissions is still the preferred method for reducing SNA concentrations in central China, except when acidic gas emissions are well controlled and the environmental chemical balance tends to favor the effective control of NH₃. Therefore, under the current atmospheric conditions, controlling acidic gas emissions is still a priority for reducing the PM_{2.5} concentration in Beijing. Nevertheless, the cost of emissions reduction also increases with the progress of controlling SO₂ and NO_x emissions. Although they provide similar abatement benefits, the cost of reducing NH₃ is only 10 % of that required to reduce NO_x (Gu et al., 2021). Thus, reducing NH₃ emissions should be prioritized as a means of improving future air quality in Beijing.

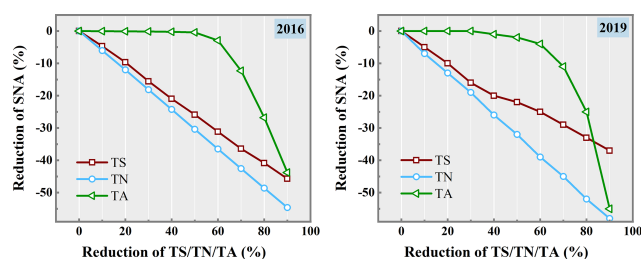


Figure 5. ISORROPIA predictions of the percentage reduction in SNA mass concentration based on winter observations (charts display the percentage reduction in SNA plotted against the percentage reduction in TS, TN, and TA concentrations).

4 Conclusions

Over the 11 years analyzed, the NH₃ concentration in urban Beijing initially increased by 50 % in the first 8 years but subsequently decreased by 49 % in the following 3 years. In particular, the annual average concentration of NH₃ in 2020 was 24 % lower than that in 2009. The trend for NH₃ mixing ratios did not align with those for annual NH₃ emissions and the increasing trend indicated by satellite-based NH₃ monitoring data. These discrepancies highlight the complexity of NH₃ sources and removal processes in urban areas, which implies there are further challenges in performing atmospheric NH₃-related modeling and implementing future emission reduction strategies.

The long-term trend in NH₃ in urban Beijing was not significantly influenced by meteorological factors such as temperature. However, the seasonal variations in NH₃ mixing ratios were strongly influenced by temperature, with higher temperatures corresponding to higher NH₃ mixing ratios during warmer seasons. Regarding daily variations, NH₃ mixing ratios exhibited both positive and negative correlations with temperature but consistently exhibited a positive correlation with absolute humidity on most days. Across the observation years, the daily variations in NH₃ concentrations did not exhibit a consistent pattern across different seasons. In some cases, the patterns were even entirely opposite. By contrast, various meteorological factors and the daily variation patterns of other common air pollutants were mostly consistent across different years and seasons. Consequently, the factors influencing atmospheric NH₃ concentrations appeared to be more complex compared with those influencing other common air pollutants.

The concentrations of various PM_{2.5} ion components in Beijing for the years 2009, 2016, 2018, and 2019 indicate that, apart from during the summer season, the SNA content in Beijing was mainly dominated by sulfate (nearly 50 %). Furthermore, the proportion of ammonium in SNA content increased over time. An analysis of the neutralization levels of major acidic gases and a modeling analysis of perturbation indicated that an excessive concentration of NH₃ was

maintained throughout the year in Beijing. The findings of the present study suggest that, even though the concentrations of SO₂ and NO_x in Beijing have decreased substantially over the past 2 decades, SIA formation is more sensitive to acid gases than to NH₃. Therefore, reducing acidic gas emissions is still a primary focus for controlling fine-particulate-matter pollution in Beijing. Given that the trends in urban atmospheric NH₃ concentrations do not align with emissions trends, clarifying the relationship between them and identifying the sources of NH₃ in Beijing will play a crucial role in effectively reducing atmospheric NH₃ concentrations in the city.

In the present study, atmospheric NH₃ concentrations in urban Beijing were continuously monitored over a long period with high temporal resolution. However, it should be noted that there are potential limitations of surface monitoring in representing urban or regional trends due to the uneven distribution of atmospheric NH₃ sources and the lack of vertical information. Similarly, with monitoring at a single site, it is necessary to verify whether the response measures are broadly applicable across the entire Beijing urban area. This will require further observational research on atmospheric NH₃ in urban Beijing in the future. In addition, existing studies have demonstrated that emission inventories have underestimated atmospheric NH₃ emissions in the Beijing urban area (Xu et al., 2023), and the assessment results have varied across emission inventories (Chen et al., 2023). Additionally, given the limited research years of current emission inventories, the observed differences between long-term trends in monitored NH₃ concentrations and NH₃ emissions require continued attention in the future.

Data availability. For access to datasets, please contact Weili Lin (linwl@muc.edu.cn).

Supplement. The supplement related to this article is available online at: <https://doi.org/10.5194/acp-24-9355-2024-supplement>.

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