Regional Transport Increases Ammonia Concentration in Beijing, China

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Abstract: To elucidate the critical factors influencing the ammonia (NH₃) concentration in Beijing, this study combined observational analyses, backward trajectory calculations, and meteorology–chemistry coupled simulations to investigate the variations in the NH₃ concentration from 11 May to 24 June, 2015. A significant positive correlation was found between the NH₃ and PM₂.₅ concentrations in Beijing. By examining the relationships between meteorological parameters and the NH₃ concentration, both near-surface temperature and relative humidity showed positive correlations with the NH₃ concentration. The higher NH₃ concentrations were usually associated with the warming of the upper atmosphere. Distinct wind directions were noted during the days of the top and bottom 33.3% NH₃ concentrations. The top 33.3% concentrations were primarily related to southwesterly winds, while the bottom ones were associated with westerly and northerly winds. Since there are strong NH₃ emissions in the southern plains adjacent to Beijing, the regional transport induced by the southerly prevailing winds would increase the NH₃ concentration in Beijing significantly. From 23 to 25 May, more than one third of NH₃ in Beijing was contributed by the southerly transport processes. Thus, joint efforts to reduce NH₃ emissions in the whole Beijing–Tianjin–Hebei region are necessary to regulate the NH₃ concentration in Beijing.

Keywords: regional transport; WRF-Chem; backward trajectory analysis; thermal stratification

1. Introduction

Being the most abundant basic/alkaline gas species in the atmosphere, ammonia (NH₃) plays an important role in determining the overall acidity of precipitation, cloud water, and airborne particulate matter [1,2]. The largest source of NH₃ emission is agriculture, including animal husbandry and NH₃-based fertilizer applications [2]. Sutton et al. estimated that 57% of global atmospheric NH₃ was emitted from crop and livestock production in 2008 [1]. Once emitted into the atmosphere, NH₃ undergoes complex atmospheric processes such as transformation due to reactions, transport associated with winds, and wet and dry deposition [3].

During the past decades, the rapid development of economy and urbanization in China have led to poor air quality and high PM₂.₅ (particle matter with an aerodynamic diameter less than 2.5 µm) loading in most cities, particularly in the densely populated Beijing–Tianjin–Hebei region [4,5]. Among the aerosol precursors, several studies have pointed out the importance of NH₃ [6–9], which acts as a limiting species in the formation of secondary inorganic aerosols (SIA). In the atmosphere, NH₃ can neutralize ambient acidic species, such as sulfuric acid (H₂SO₄) and nitric acid (HNO₃), to form ammonium salts, which are the dominant inorganic compounds in the ambient PM₂.₅ [2,10]. These reaction pathways link NH₃ to aerosol pollution and its subsequent impacts on human health and climate change [11,12]. It was found that the mass of secondary sulfate, nitrate, and ammonium...
accounted for 25–60% of the total PM$_{2.5}$ concentration [13,14]. The total NH$_3$ emission in China in 2015 was estimated to be 15.6 ± 0.9 Tg N yr$^{-1}$ [6]. Such a high emission rate makes NH$_3$ become one of the critical species related to the air pollution issues in China. Long-term measurements of NH$_3$ in Shanghai of China demonstrated the important role of the gas-to-particle conversion of NH$_3$ on PM$_{2.5}$ formation [15], and the ammonium accounted for about 10% of the PM$_{2.5}$ concentration measured in Shanghai [15]. The gas-to-particle conversion of NH$_3$ was also observed to lead to a significant increase of ammonium during haze events in Beijing [16,17]. The chemical composition analysis of PM$_{2.5}$ in Beijing indicated that the ammonium accounted for 11% of the total defined PM$_{2.5}$ components during the heavily polluted days [16,17]. In addition, the atmospheric deposition of NH$_3$ into terrestrial and aquatic ecosystems can cause adverse environmental effects, such as soil acidification, eutrophication of water bodies, and even plant biodiversity reduction [2,18–21]. Although the environmental impacts of NH$_3$ in China have been recognized [10,13], until now, the NH$_3$ emissions have not been regulated by the Chinese government; by contrast, stringent measures have been taking to control the emissions of sulfur dioxide (SO$_2$) and nitrogen oxide (NO$_X$) [22]. Since the balance between SO$_2$, NO$_X$, and NH$_3$ emissions will define cation- or anion-limited regimes of inorganic aerosol formation [23,24], it is suggested to consider the regulation of NH$_3$ emissions as a possible measure to mitigate the heavy aerosol pollution in some regions of China [24].

The atmospheric NH$_3$ concentration in Beijing is not only contributed by local emission sources (e.g., traffic exhaust, husbandry), but can also be influenced by adjacent agricultural emissions from surrounding croplands (Figure 1) under certain synoptic conditions [25,26]. Based on one-year observations of the vertical distribution of NH$_3$ at the 325-m meteorological tower in Beijing, Zhang et al. found that air masses from the agricultural regions contributed most to the high NH$_3$ concentration measured [27]. At a rural site in the Beijing–Tianjin–Hebei region, Meng et al. also observed that the increased NH$_3$ concentration was associated with specific wind directions, and suggested that the transport of NH$_3$ emitted from agricultural regions was critical to the air quality in the downstream regions [25]. Although the regional transport of NH$_3$ in the Beijing–Tianjin–Hebei region has been documented, the potential source region to Beijing and to what extent the regional transport influences the NH$_3$ concentration in Beijing are not yet clearly understood.

To identify the potential source regions, and quantitatively evaluate the impacts of regional transport on the NH$_3$ concentration in Beijing, this study combines observational analyses, backward trajectory calculations, and meteorology–chemistry coupled simulations to systematically investigate the transport processes of NH$_3$ in the Beijing–Tianjin–Hebei region. The rest of this paper is organized as follows. In Section 2, the methods and data are described. In Section 3, the impacts of regional transport on NH$_3$ concentration in Beijing are analyzed. Finally, the main findings are summarized in Section 4.
Figure 1. Maps of (a) Weather Research and Forecasting model coupled with chemistry (WRF-Chem) simulation domain and anthropogenic NH$_3$ emissions in May, 2016, and (b) the land use types in Beijing and its adjacent regions. The location of Beijing is denoted by the red lines in Figure 1a, and the locations of the NH$_3$ monitoring site, PM$_{2.5}$ monitoring site, and meteorological site in Beijing are marked by the blue cross, red dot, and black plus in Figure 1b, respectively. The dashed rectangle in Figure 1a indicates the regions using the emission zero-out method in the WRF-Chem simulation.

2. Methods and Data

To understand the variations in NH$_3$ concentration in Beijing, hourly NH$_3$ concentration from 11 May to 24 June in 2015 was measured in the Beijing Municipal Environmental Monitoring Center (116.32° E, 39.93° N; marked by the blue cross in Figure 1b) using a differential optical absorption spectroscope (DOAS, Anhui Landun Photoelectron Co. Ltd., Tongling, Anhui, China). The DOAS, an open-path monitoring technique, is based on the wavelength dependent absorption of light over a specified light path [28]. It is often utilized in the UV and visible parts of the electromagnetic spectrum. The absorption lines and bands of gas molecules in this part of the spectrum are caused by electronic transitions and their shapes only weakly depend on temperature and pressure [28]. Regarding NH$_3$, it has a strong absorption band with narrowband features in the UV part from 170 to 220 nm [29], and the DOAS system can monitor its concentration with a high accuracy of 0.04 µg m$^{-3}$ for a total light path of 100 m [30]. In this study, both the transmitter and retro-reflector were placed 20 m high above the ground [30]. The detailed information of the NH$_3$ measurement in the Beijing Municipal Environmental Monitoring Center can be found in the previous study of Cheng et al. [30]. The PM$_{2.5}$ concentration in Beijing (116.31° E, 39.97° N; marked by the red dot in Figure 1b) was also collected from the China National Environmental Monitoring Center. Besides, both the ground-level and upper-air meteorological observations in the southern Beijing (116.47° E, 39.80° N, marked by the black plus in Figure 1b) were obtained. The ground-level meteorological parameters were recorded hourly, while the sounding balloons were launched twice a day at 08:00 and 20:00 local time (LT = UTC + 8h). In total, 45-day measurements in Beijing were collected, including the near-surface NH$_3$ and PM$_{2.5}$ concentrations, 2-m temperature (T), 2-m relative humidity (RH), precipitation and the vertical profiles of the potential temperature (PT) and horizontal winds.

Through analyzing these observational data, the relationships between the meteorological conditions and NH$_3$ concentration in Beijing were elucidated, and then a typical pollution episode (23–25 May) associated with southerly prevailing winds was selected and simulated using the Weather Research and Forecasting model coupled with chemistry (WRF-Chem) [31]. The simulation domain was centered in the Beijing–Tianjin–Hebei region with a horizontal resolution of 11 km, covering an area of 106–126° E in longitude and 31–45° N in latitude (Figure 1a). In the vertical dimension, 33 vertical layers were set from the 10-hPa level, of which 15 layers were set below 2 km above ground-level (AGL). The physics parameterization schemes used in the WRF-Chem simulation included the Noah land surface scheme [32], Yonsei University (YSU) boundary layer scheme [33], rapid radiative transfer model for general circulation (RRTMG) long-/short-wave radiation scheme [34],
Betts–Miller–Janjic cumulus scheme [35], and WRF single-moment-5-class microphysics scheme [36]. Regarding the chemical processes, the RADM2-MADE/SORGAM mechanism [37–39] was used with the Multi-resolution Emission Inventory for China (MEIC), and the dry and wet deposition schemes were turned on. The MEIC is the most updated and widely used emission data available for the studied region, which is developed and provided by Tsinghua University [40]. The WRF-Chem simulation was initialized at 20:00 LT on 21 May, and ran for 100 h until 00:00 LT on 26 May. The first 28 h were considered as a spin-up period. The initial and boundary conditions of the meteorological variables were configured using the 5th generation of atmospheric reanalysis produced by European Centre for Medium-Range Weather Forecasts (ERA5), and the initial and boundary conditions of the chemical variables were derived from the output of a global chemical transport model [41]. In the rest, the simulation using these configurations is referred to as the baseline (BASE) run.

To identify the potential source regions, for each day of the selected episode, 24-h air mass backward trajectories were calculated using the hybrid single particle Lagrangian integrated trajectory model (HYSPLIT, developed by the Air Resource Laboratory of the United States National Oceanic and Atmospheric Administration, College Park, Maryland, US). The model has been widely applied to identify the air mass origins and pathways that reach the receptor sites [42]. The ending point was set using the location of the NH$_3$ monitoring site (116.32° E, 39.93° N) with a height of 100 m AGL, and the ending time was 18:00 LT of each day. According to those backward trajectories, a sensitivity numerical experiment was designed using the emission zero-out method [26,43], in which all the anthropogenic emissions in the possible source region were set to zero. Except for the emission configuration, the other settings of the sensitivity experiment used were the same settings of the BASE run. The sensitivity experiment is referred to as the EXP run hereafter. Thereby, the contributions of regional transport to the NH$_3$ concentration in Beijing can be roughly estimated as the simulation differences between the BASE and EXP runs. In the rest of this paper, the simulation results presented and discussed are derived from the BASE run unless otherwise noted.

To examine the effects of emission control measures on the aerosol pollution in Beijing, two more sensitivity simulations (i.e., SIM-1 and SIM-2) were conducted using the same settings of the BASE run except for the emissions. In SIM-1, the emissions of SO$_2$ and NO$_X$ in the whole simulation domain were reduced by 50%. In SIM-2, the changes in SO$_2$ and NO$_X$ emissions were the same as those in SIM-1, and the emissions of NH$_3$ were also reduced by 50%.

3. Results and Discussion

3.1. Relationships between NH$_3$ Concentration and Meteorological Parameters

The time series of the daily NH$_3$ concentration from 11 May to 24 June in Beijing is shown in Figure 2a. The peak daily NH$_3$ concentration was 34.6 µg m$^{-3}$ on 17 May, and the minimum concentration was 2.0 µg m$^{-3}$ on 19 May. During the studied period, there were in total 12 rainy days in Beijing (Figure 2c). To isolate the complex impacts of precipitation on air pollution [44], only the measurements of the rest 33 days under dry conditions were further analyzed in this study. After excluding the rainy days, the relationships between the NH$_3$ concentrations and near-surface meteorological parameters in Beijing were examined (Figure 2b,c). It was found that the daily NH$_3$ concentration was positively correlated with the near-surface T and RH, with correlation coefficients of 0.49 and 0.65, respectively. The link between the vertical thermal stratification and ground-level NH$_3$ concentration in Beijing was also investigated based on the sounding data at 20:00 LT. The higher NH$_3$ concentrations in Beijing were usually associated with the warming of the upper atmosphere (Figure 2d), such as the episodes that occurred around 25 May and 1 June. As a result, a significant positive correlation was found between the 1500-m PT and NH$_3$ concentration in Beijing ($R = 0.72$). Such a relationship can be explained by the development of the boundary layer and its impact on the pollutants’ vertical dispersion. The warming aloft can increase the thermal stability, which would suppress the development of the boundary layer to some extent, as well as the vertical dispersion.
and dilution of pollutants [5,45,46]. Besides, we compared the NH\textsubscript{3} concentrations with the PM\textsubscript{2.5} concentrations, and a significant positive correlation (R = 0.75) was found (Figure 2a). It can be explained by the day-to-day variations in the meteorological conditions that caused the simultaneous increase or drop in the NH\textsubscript{3} and PM\textsubscript{2.5} concentrations. On the other hand, NH\textsubscript{3} is also intricately linked to the secondary formation of aerosols [22–24], which is relevant to the acidity (pH), liquid water content and HNO\textsubscript{3} level [47,48].

### Table 1

| Date (Month-Day) | NH\textsubscript{3} (µg m\textsuperscript{-3}) | PM\textsubscript{2.5} (µg m\textsuperscript{-3}) | T (°C) | RH (%) | PT 1500 (K) | WD 800 (°) |
|-----------------|---------------------------------|-------------------------------|--------|--------|-------------|------------|
| 05-17           | 34.6                             | 22.9                          | 64     | 53     | 304.6       | 217        |
| 05-30           | 25.5                             | 24.5                          | 56     | 302.0  | 183         | 183        |
| 05-27           | 24.4                             | 26.2                          | 51     | 303.5  | 231         | 231        |
| 05-26           | 28.0                             | 27.3                          | 45     | 307.9  | 217         | 217        |
| 05-25           | 29.2                             | 26.9                          | 44     | 307.9  | 215         | 215        |
| 05-24           | 28.9                             | 25.8                          | 48     | 305.4  | 220         | 220        |
| 05-16           | 27.6                             | 27.4                          | 50     | 305.4  | 267         | 267        |
| 05-16           | 27.6                             | 27.4                          | 50     | 305.4  | 267         | 267        |
| 05-11           | 33.3                             | 28.2                          | 53     | 308.9  | 209         | 209        |
| 05-31           | 25.4                             | 26.7                          | 49     | 308.3  | 214         | 214        |

#### Figure 2

Daily variations in (a) NH\textsubscript{3} and PM\textsubscript{2.5} concentrations, (b) 2-m temperature (T), (c) 2-m relative humidity (RH), and rainfall, (d) potential temperature vertical profile in Beijing from 11 May to 24 June, 2015. There were in total 12 rainy days during the studied period, and the correlation coefficients (R) between the NH\textsubscript{3} concentration and different variables in Beijing under dry conditions are given for each panel, including the PM\textsubscript{2.5} concentration, T, RH and 1500-m PT.

To further understand the factors leading to the increased NH\textsubscript{3} concentrations in Beijing, Figure 3 and Table 1 compare the different characteristics associated with the top and bottom 33.3% daily NH\textsubscript{3} concentrations under dry conditions. The top 33.3% NH\textsubscript{3} concentrations in Beijing were within 22.4–34.6 µg m\textsuperscript{-3}, significantly higher than the bottom 33.3% concentrations (2.0–16.6 µg m\textsuperscript{-3}). When the top 33.3% concentrations happened, the average 2-m T and 2-m RH in Beijing were 26.0 °C and 52%, respectively, significantly warmer and moister than those associated with the bottom 33.3% concentrations (i.e., 23.1 °C and 35%). At the upper levels, the thermal difference between the days associated with the top and bottom 33.3% concentrations was larger; the average values of the 1500-m PT were 305.6 K and 300.7 K, respectively. In short, the warmer and moister conditions and appearance of strong thermal inversion aloft in Beijing favor the accumulation of NH\textsubscript{3}.

In addition, distinct wind directions could be found based on the wind measurements at 800-m AGL (close to the 925-hPa level). The top 33.3% NH\textsubscript{3} concentrations in Beijing were primarily in relation to the southwesterly winds (181–225°), while the bottom 33.3% NH\textsubscript{3} concentrations were corresponding to the westerly and northerly winds (226–360°). Since there are strong NH\textsubscript{3} emissions in the southern plains adjacent to Beijing (Figure 1a), it is hypothesized that the regional transport of NH\textsubscript{3} induced by the southerly prevailing winds is critical to the increased NH\textsubscript{3} concentration in Beijing. To quantitatively evaluate the impact of regional transport, the episode from 23 to 25 May associated
with the southwesterly prevailing winds was simulated using WRF-Chem and further analyzed in the Section 3.2.

### Table 1. NH$_3$ concentrations, 2-m T, 2-m RH, 1500-m PT and 800-m wind direction (WD) in Beijing during the dry conditions from 11 May to 24 June, 2015. Except for that the PT and WD are presented according to the soundings at 20:00 LT, the other variables are derived from the daily mean values.

| Date (Month-Day) | NH$_3$ (µg m$^{-3}$) | T (°C) | RH (%) | PT$_{1500}$ (K) | WD$_{800}$ (°) |
|------------------|----------------------|--------|--------|-----------------|---------------|
| Days with Top 33.3% Daily NH$_3$ Concentrations |
| 05-17            | 34.6                 | 22.9   | 64     | 304.6           | 217           |
| 05-23            | 22.6                 | 24.5   | 47     | 303.8           | 208           |
| 05-24            | 28.9                 | 25.8   | 48     | 305.4           | 220           |
| 05-25            | 29.2                 | 26.9   | 44     | 307.9           | 215           |
| 05-26            | 28.0                 | 27.3   | 45     | 307.9           | 217           |
| 05-27            | 24.4                 | 26.2   | 51     | 303.5           | 231           |
| 05-30            | 25.5                 | 24.5   | 56     | 302.0           | 183           |
| 05-31            | 25.4                 | 26.7   | 49     | 308.3           | 214           |
| 06-01            | 33.3                 | 28.2   | 53     | 308.9           | 209           |
| 06-16            | 27.6                 | 27.4   | 50     | 305.4           | 267           |
| 06-24            | 29.9                 | 25.2   | 64     | 303.3           | 218           |
| Days with Bottom 33.3% Daily NH$_3$ Concentrations |
| 05-11            | 7.8                  | 13.5   | 64     | 291.2           | 337           |
| 05-14            | 9.6                  | 22.3   | 27     | 301.3           | 205           |
| 05-15            | 9.3                  | 21.2   | 32     | 298.8           | 235           |
| 05-18            | 11.5                 | 23.7   | 37     | 300.4           | 345           |
| 05-19            | 2.0                  | 23.0   | 15     | 297.1           | 348           |
| 05-20            | 5.8                  | 24.1   | 23     | 301.4           | 242           |
| 05-21            | 13.8                 | 23.0   | 34     | 302.7           | 229           |
| 06-02            | 16.6                 | 27.2   | 30     | 305.0           | 353           |
| 06-03            | 15.1                 | 25.4   | 36     | 304.6           | 234           |
| 06-12            | 9.5                  | 25.0   | 38     | 301.7           | 302           |
| 06-14            | 14.1                 | 25.9   | 52     | 303.9           | 184           |
concentrations, (b) 2-m T, (c) 2-m RH, and (d) 1500-m PT. The central box represents the values from the lower to upper quartile (25th to 75th percentile). The vertical line extends from the 10th percentile to the 90th percentile value. The middle solid line represents the median, and the dot represents the mean value. In (e), wind rose diagram based on the 800-m horizontal wind associated with the top 33.3% (in blue) and bottom 33.3% (in red) NH₃ concentrations in Beijing are shown. Except for that the 1500-m PT and 800-m wind are shown according to the soundings at 20:00 LT, the other variables are derived from the daily mean values.

### 3.2. Case Study on the Regional Transport of NH₃ to Beijing using WRF-Chem

In this section, the simulation results were first validated against the observations, and then the impact of regional transport on the NH₃ concentration in Beijing was presented. Figure 4 shows the time series of the observed and simulated NH₃ concentrations, PM₂.₅ concentrations, 2-m Ts, and 2-m RHs in Beijing from 23 to 25 May. Although the model tends to underestimate the daytime NH₃ and PM₂.₅ concentrations in Beijing, both the diurnal pattern (peaking in the early morning and reaching the minimum in the afternoon) and the daily variation were generally well reproduced. The correlation coefficients between the observed and simulated NH₃ and PM₂.₅ concentrations in Beijing were 0.73 and 0.70, respectively (Figure 4a,b). Good agreements also can be found between the observations and simulations of T and RH (Figure 4c,d). In addition to the near-surface parameters, the simulated vertical structures of PT and horizontal wind over Beijing were also compared with the sounding data (Figure 5). The gradual warming of the upper air and the strengthening of southerly winds in Beijing from 23 to 25 May were accurately simulated by the model. Overall, these generally good model performances (Figures 4 and 5) provide a basis to use the simulation results to investigate the transport process during the studied period.

![Figure 4](image_url)

**Figure 4.** Time series of observed and simulated (a) NH₃ concentrations, (b) PM₂.₅ concentrations, (c) 2-m Ts, and (d) 2-m RHs in Beijing from 23 to 25 May, 2015. The correlation coefficients (R) between observations and simulations are presented for each panel.

To identify the possible source regions, 24-h backward trajectories ending at the NH₃ monitoring site in Beijing were simulated using the HYSPLIT. With a high pressure located over the south seas at the 925-hPa level, there was a southeast-to-northwest pressure gradient across Beijing from 23 to 25 May, which supported the southerly prevailing winds towards Beijing (Figure 6). Influenced by such synoptic conditions, the backward trajectories during those three days all passed through the south part of Hebei province. Based on these trajectories and the spatial distribution of NH₃ emissions (Figures 1 and 6), we roughly delimited a potential source region in the southern plains adjacent to Beijing (114.3–118.0° E, 35.8–39.5° N). The EXP simulation was conducted using the emission zero-out configuration in the potential source region. Figure 7 presents the spatial distribution of the ground-level NH₃ concentration in the Beijing–Tianjin–Hebei region at 18:00 LT during the selected episode. The NH₃ concentration in the center of Beijing was around 12 µg m⁻³, significantly lower than those in the southern upstream plains, in which the NH₃ concentrations were greater than 20 µg m⁻³.
on 24 and 25 May (Figure 7a–c). By calculating the difference between the BASE and EXP runs, the contributions of the potential source region to the NH₃ concentrations in the Beijing–Tianjin–Hebei region were illustrated in Figure 7d–f. At 18:00 LT, the transport process can increase the NH₃ concentration in the center of Beijing by 5–7 µg m⁻³. Averaging the simulation results of each day, the daily NH₃ concentrations in the center of Beijing were calculated, which were 15.1 µg m⁻³ on 23 May, 15.0 µg m⁻³ on 24 May, and 15.7 µg m⁻³ on 25 May (Figure 8), and the southern plains could contribute 5.4 µg m⁻³ (36%), 6.1 µg m⁻³ (41%), and 5.3 µg m⁻³ (34%), respectively. These results indicate that when the Beijing–Tianjin–Hebei region is influenced by the southerly prevailing winds, the regional transport plays an important role in the NH₃ concentration in Beijing. Thus, joint efforts to reduce NH₃ emissions in the whole Beijing–Tianjin–Hebei region are necessary to regulate the NH₃ concentration in Beijing.

![Figure 5.](image_url)

**Figure 5.** Vertical structures of (a) the observed and (b) the simulated PTs and horizontal winds in Beijing from 23 to 25 May, 2015. Both simulations and observations are shown with an interval of 12-h. At the top of Figure 5a, the correlation coefficients (R) between simulations and observations are presented.

Besides, to understand the impacts of NH₃ emissions on the PM₂.₅ pollution, we compared the simulated SIA concentrations in the Beijing–Tianjin–Hebei region under different emission scenarios (i.e., BASE, SIM-1 and SIM-2). By halving the emissions of SO₂ and NOₓ (Figure 9b), the near-surface SIA concentration in Beijing can decrease by 14.3 µg m⁻³ (41%). Comparing the results of SIM-2 with those of SIM-1, it was found that the control of NH₃ emissions could result in an additional 13% reduction in the SIA concentration in Beijing (Figure 9c). Similar results have also been found by Fu et al. [24]. Thus, enforcing the NH₃ emissions control in conjunction with other gas pollutants can benefit the PM₂.₅ pollution mitigation [2,10,24]. It should be noted that the link between NH₃ and aerosol pollution is quite complicated [23,24,47,48], and here we just presented some results of simplistic numerical experiments, which warrants further studies. To develop the effective emission control policies, the framework that explicitly considers the pH and liquid water content proposed by Nenes et al. [47] can be used to determine the chemical domains of the sensitivity of the aerosol concentration to the NH₃ and HNO₃ levels.
than those in the southern upstream plains, in which the NH$_3$ concentrations were greater than 20 ppbv. Thus, enforcing the NH$_3$ emissions control in conjunction with other gas pollutants can result in a significant reduction in the SIA concentration in Beijing (Figure 9c). Similar results have also been found by Fu et al. [24].

Based on these trajectories and the spatial distribution of NH$_3$ emissions, such synoptic conditions, the backward trajectories during those three days all passed through the southern part of Hebei province. The transport plays an important role in the NH$_3$ concentration in Beijing. Thus, joint efforts to reduce the NH$_3$ emissions could result in an additional 14.3% decrease in the SIA concentration in Beijing.

To develop the effective emission control strategies for PM$_{2.5}$ pollution, we need to understand the impacts of NH$_3$ emissions on PM$_{2.5}$ pollution. We compared the PM$_{2.5}$ concentrations in Beijing derived from the BASE run and sensitivity experiment (EXP). The location of the NH$_3$ monitoring site in Beijing is marked by the blue cross, and 24-h backward trajectories arriving in the site are shown as the green lines.

The high-pressure systems in the southern seas are denoted by the text “H”.

**Figure 6.** Spatial distributions of 925-hPa geopotential height (GH) and horizontal winds in the simulation domain at 1800 LT on (a) 23 May, (b) 24 May, and (c) 25 May, 2015, derived from the atmospheric reanalysis of European Centre for Medium-Range Weather Forecasts. The location of the NH$_3$ monitoring site is marked by the blue cross, and 24-h backward trajectories arriving in the site are shown as the green lines. The dashed rectangle adjacent to Beijing indicates the regions using the emission zero-out method in the WRF-Chem simulation.

**Figure 7.** Spatial distribution of the simulated near-surface NH$_3$ concentrations and winds at 1800 LT on (a,d) 23 May, (b,e) 24 May, and (c,f) 25 May, 2015. The mountainous regions are denoted by the grey shadings. The top panels show the simulation results of the baseline (BASE) run, and the bottom panels present the differences between the BASE run and sensitivity experiment (EXP). The location of the NH$_3$ monitoring site in Beijing is indicated by the blue cross.

**Figure 8.** Bar plot showing the daily NH$_3$ concentrations in Beijing derived from the BASE run and contributed by the southern plain regions (BASE-EXP).
4. Conclusions

In this study, the influence of regional transport on the NH$_3$ concentration in Beijing was systematically investigated by combining observational analyses, backward trajectory calculations, and meteorology–chemistry coupled simulations. The NH$_3$ concentration was measured in the Beijing Municipal Environmental Monitoring Center from 11 May to 24 June, 2015.

A statistically significant positive correlation was found between the NH$_3$ and PM$_{2.5}$ concentrations in Beijing. The relationships between the NH$_3$ concentrations and meteorological parameters in Beijing were also examined. The daily NH$_3$ concentration was positively correlated with the near-surface $T$ and RH. The higher NH$_3$ concentrations in Beijing were usually associated with the warming of the upper atmosphere. The warming aloft can increase the thermal stability, which would suppress the development of the boundary layer to some extent, as well as the vertical dispersion of pollutants. Further, distinct wind directions could be found during the days of the top and bottom 33.3% NH$_3$ concentrations. The top 33.3% NH$_3$ concentrations in Beijing were primarily in relation to the southwesterly winds (181–225°), while the bottom 33.3% NH$_3$ concentrations were corresponding to the westerly and northerly winds (226–360°). Since there are strong NH$_3$ emissions in the southern plains adjacent to Beijing, the regional transport of NH$_3$ induced by the southerly prevailing winds plays an important role in the NH$_3$ concentration in Beijing.

From 23 to 25 May, with a high pressure located over the south seas at the 925-hPa level, there was a southeast-to-northwest pressure gradient across Beijing, which supported the southerly prevailing winds towards Beijing. As a result, the NH$_3$ emitted from southern Hebei could be easily transported to Beijing, leading to a higher NH$_3$ concentration in Beijing. Thus, joint efforts to reduce NH$_3$ emissions in the whole Beijing–Tianjin–Hebei region are necessary to regulate the NH$_3$ concentration in Beijing. Besides, enforcing the NH$_3$ emissions control in conjunction with the ongoing stringent control of SO$_2$ and NO$_x$ emissions can further mitigate the aerosol pollution in Beijing.

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