Rapid SO$_2$ emission reductions significantly increase tropospheric ammonia concentrations over the North China Plain

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Abstract. The North China Plain has been identified as a significant hotspot of ammonia (NH$_3$) due to extensive agricultural activities. Satellite observations suggest a significant increase of about 30% in tropospheric gas-phase NH$_3$ concentrations in this area during 2008–2016. However, the estimated NH$_3$ emissions decreased slightly because of changes in Chinese agricultural practices, i.e., the transition in fertilizer types from ammonium carbonate fertilizer to urea, and in the livestock rearing system from free-range to intensive farming. We note that the emissions of sulfur dioxide (SO$_2$) have rapidly declined by 60% over recent few years. By integrating in situ measurement datasets, multi-year NH$_3$ emission inventories, and chemical transport model simulations, we demonstrate that the increases in NH$_3$ can be almost entirely attributable to this rapid SO$_2$ emission reduction. The annual average sulfate concentrations decreased by about 50%, which significantly weakened the formation of ammonium sulfate and increased the average proportions of gas phase NH$_3$ within the total NH$_3$ column concentrations from 26% (2008) to 37% (2016). Both the decreases in sulfate and increases in NH$_3$ concentrations...
show highest values in summer, possibly because the formation of sulfate aerosols is more sensitive to SO₂ emission reductions in summer than in other seasons.

1 Introduction

Ammonia (NH₃) is considered the most important alkaline gas in the atmosphere. On both a global and regional scale, NH₃ is mostly emitted from agricultural activities, mainly including fertilization and livestock industry (Bouwman et al., 1997). Gas-phase NH₃ can react with ambient sulfuric and nitric acids to form ammonium sulfate/bisulfate and ammonium nitrate aerosols (SNA), which constitute a significant fraction of atmospheric fine particles (PM₂.₅) associated with potential human health impacts (Pope et al., 2009; Seinfeld and Pandis, 2006). Ammonia and ammonium (NH₄⁺) is ultimately deposited back to the earth surface, contributing to acid deposition and eutrophication (Asman, 1998; Behera et al., 2013; Pozzer et al., 2017).

As a major agricultural country, China is the world’s largest emitter of NH₃, the amount of which (~10 Tg) exceeds the sum of those in Europe (~4.0 Tg) and North America (~4.0 Tg) (Huang et al., 2012; Bouwman et al., 1997; Paulot et al., 2014). Fertilizer application and livestock manure management contribute to nearly 90% of China’s NH₃ emissions (Huang et al., 2012; Zhang et al., 2018). Until now, NH₃ emission has not been regulated by the Chinese government, although it may be a potentially important contributor to haze pollution in China.

The North China Plain (the spatial definition of this area is illustrated in Fig. S1) is a hotspot of NH₃ loadings, as revealed by satellite detection and in situ ground measurements (Clarisse et al., 2009; Pan et al., 2018). Interestingly, satellite observations over the past decade have shown an increase in tropospheric columns of gaseous NH₃ in this area (Warner et al., 2017). But no quantitative studies have been performed to explain it. A long-term bottom-up inventory indicated that NH₃ emissions in China have displayed a slightly decreasing tendency. During 2006–2016, ammonium bicarbonate for crop fertilization was replaced by urea fertilizer (its fraction of application increasing from 60 to 90% of all nitrogen fertilizers), and the traditional free-range livestock system was replaced by intensive animal rearing system (i.e., raising livestock in confinement at a high stocking density) in the livestock industry (increasing from 21% in 2006 to 48% in 2016; shown in
Table S1. These changes in agricultural practices have lowered the volatilization rates of NH$_3$ (Kang et al., 2016).

Through the widespread use of the flue gas desulfurization in power plants since 2000s in China, SO$_2$ emissions have gradually decreased (Lu et al., 2011; Li et al., 2010). Li et al. (2017) found it was reduced by 70% from the peak year (around 2006) to 2016 based on satellite observations and bottom up methods. Specifically, the initiation of the “Action Plan for Air Pollution Prevention and Control” (referred to as the national “Ten Measures for Air”) since 2013 resulted in a rapid reduction of about 50% over recent few years, from ~30 Tg in 2012 to ~14 Tg in 2016 according to the Multi-resolution Emission Inventory for China (MEIC). To our knowledge, such a strong decrease in SO$_2$ emissions is only found in China. In contrast, emissions of nitrogen oxides (NO$_x$) in MEIC peaked around 2012 with only a moderate decrease of ~20% from 2012 to 2016 (Liu et al., 2016).

Here, we hypothesize that the rapid SO$_2$ emission reduction is the reason for the increase in tropospheric NH$_3$ concentrations over the North China Plain. To verify this, we first used observation datasets from the ground and space to infer the relationship between the trends in NH$_3$ and SO$_2$ concentrations. A comprehensive long-term NH$_3$ emission inventory, developed by our recent studies based on bottom-up methods, was also used to demonstrate the inter-annual variations of NH$_3$ emissions in this region. Then, we performed multi-year simulations with Weather Research and Forest model coupled with Chemistry (WRF-Chem) to test the impact of changes in SO$_2$ emissions on tropospheric NH$_3$ concentrations in terms of the magnitude and seasonal variation.

2 Methods

2.1 Observations datasets

Observations from space and ground stations were used in this study. Tropospheric vertical column densities (VCDs) of NH$_3$ were derived from the measurements of Infrared Atmospheric Sounding Interferometer (IASI) onboard MetOp-A (Van Damme et al., 2015; Clarisse et al., 2009; Van Damme et al., 2017). We determined the annual averages of NH$_3$ column concentrations over the North China Plain on a 0.25° × 0.25° grid during 2008–2016, based on the relative error weighting mean method (Van Damme et al., 2014). The monthly NH$_3$ concentrations were measured using passive NH$_3$ diffusive
samplers (Analysts, CNR-Institute of Atmospheric Pollution, Roma, Italy) from September 2015 to August 2016 at 11 sites over Northern China (Pan et al., 2018). The SO\textsubscript{2} VCDs were provided by the ozone monitoring instrument (OMI) measurements to test the trend of SO\textsubscript{2} concentrations. They were derived from the daily level 3 data set OMSO2e, released by the NASA Goddard Earth Sciences Data and Information Services Center. Besides, daily PM\textsubscript{2.5} were sampled by quartz-fiber filters at an urban atmosphere environment monitoring station in Peking University (39.99°N, 116.3°E) of Beijing, China since 2013. The major water-soluble inorganic compounds (e.g., NH\textsubscript{4}+, NO\textsubscript{3}−, and SO\textsubscript{4}²−) were analyzed by ion-chromatography.

### 2.2 WRF-Chem simulations

In this study, the simulations with Weather Research and Forecast Model coupled Chemistry (Grell et al., 2005) version 3.6.1 (WRF-Chem) were conducted for the domain of North China Plain for the years 2008, 2010, 2012, 2014, 2015, and 2016 (referred to as Run_08–16). We ran the model with a horizontal resolution of 30 × 30 km and 24 vertical layers, extending from the surface to 50 hPa. The initial and boundary meteorological condition was derived from 6-h National Centers for Environmental Prediction reanalysis data. The detailed model configuration were described in our previous study (Huang et al., 2014). The anthropogenic emissions, including power plant, industrial, residential, and vehicle emissions, were derived from the MEIC (Multi-resolution Emission Inventory for China; available online at: http://www.meicmodel.org/) database. The MEIC data show that the annual SO\textsubscript{2} emissions in North China Plain were cut by about 60%, from 990 Gg in 2008 to 418 Gg in 2016.

### 2.3 NH\textsubscript{3} emission inventory

A high-resolution NH\textsubscript{3} emission inventory (1km×1km, month) was developed by our research group based on the bottom-up method. The emission factors were parameterized with regional farming practices, ambient temperature, soil pH and wind speeds etc. The full details can be found in our previous studies (Kang et al., 2016; Huang et al., 2012; Huo et al., 2015). The inventory had similar spatial features with recent satellite observations (Van Damme et al., 2014), and its amount is close to the emission estimated by the inversion model using ammonium wet deposition data (Paulot et al., 2014). Recent modeling
results also showed its good performance by comparing with ammonium observations in China (Huang et al., 2015). The inventories covered the period from 1980 to 2016 and considered the inter-annual variability in activity levels and agricultural practices. In the North China Plain, the increasing use of urea fertilizer (from 4.5 and 5.2 million tons) but decreased ammonium bicarbonate (from 1.5 to 0.4 million tons) led to a 20% reduction in NH₃ emissions from fertilizer application during 2008–2016 (Table S1). Meanwhile, although the number of some livestock animals increased slightly, the proportion of intensive animal rearing system increased to nearly half of the livestock industry in 2016, compared to 28% in 2008. The increased livestock animals raised but more effective manure management resulted in the livestock emissions in North China Plain remaining almost constant (around 115 Gg per year) (Table S1).

3 Results and Discussions

3.1 Trends in emissions and concentrations of NH₃ vs. SO₂

According to the Infrared Atmospheric Sounding Interferometer measurements, the North China Plain showed the highest VCDs of NH₃ in China, which mostly ranged from 15 to 30 × 10¹⁵ molecules/cm² during 2008–2014, and increased to above 30 × 10¹⁵ molecules/cm² in 2015 and 2016 (Fig. S1). We found the annual NH₃ column concentrations increased significantly (P < 0.05) over the North China Plain between 2008 and 2016 (Fig. 1a). The average tropospheric NH₃ columns first fluctuated between 2008 and 2013, and then rapidly increased from 21 × 10¹⁵ molecules/cm² in 2013 to 27 × 10¹⁵ molecules/cm² in 2016. It showed an overall increase of 30%, or an average annual increase of 0.9 × 10¹⁵ molecules cm⁻² yr⁻¹. Seasonally, the increase in NH₃ columns was more pronounced in summertime (June–August, JJA), with an annual increase rate of 1.8×10¹⁵ molecules/cm² between 2008 and 2016, which was much higher than in other seasons (< 1×10¹⁵ molecules/cm²).

In contrast to the trends in tropospheric NH₃ concentrations, the NH₃ emissions (developed based on the methods by Kang et al. (2016)) first experienced a decreasing tendency from 2008 to 2011 (300 Gg in 2009 to 275 Gg in 2011), and then remained constant at around 280 Gg during 2011–2016 over the North China Plain, representing a slight decrease (Fig. 1b). As aforementioned, the changes in fertilizer use and livestock rearing practices have lowered NH₃ emission rates. Overall,
the NH$_3$ emissions cannot track the upward trend of tropospheric NH$_3$ concentrations.

During 2008–2016, SO$_2$ column concentrations were subject to a dramatic decline ($P < 0.01$) due to a 60% decrease in SO$_2$ emissions (derived from the MEIC database). The annual mean SO$_2$ VCDs reduced from $14 \times 10^{15}$ molecules/cm$^2$ (2008) to $4 \times 10^{15}$ molecules/cm$^2$ (2016), showing a percent reduction of nearly 70%. Especially during 2012–2016, the decreases in SO$_2$ emissions and VCDs accelerated owing to the implementation of the "Action Plan for Air Pollution Prevention and Control" by the Chinese government. In situ ground measurements in a typical urban station in the North China Plain indicated that the annual average sulfate concentration (SO$_4^{2-}$) in PM$_{2.5}$ decreased by 35% (2013–2016) along with rapid SO$_2$ reductions, which was accompanied by a 33% decrease of particulate ammonium (NH$_4^+$) (Fig. 1b). The decrease in SO$_4^{2-}$ during summertime (JJA) reached 60%, which was much higher than in other seasons.

3.2 Simulations of increasing trend in NH$_3$ columns

We first evaluated WRF-Chem model results against measurements of surface NH$_3$ concentrations available in North China Plain as well as the satellite-retrieved NH$_3$ columns. The simulated monthly averaged surface NH$_3$ concentrations at 11 in situ stations (mean + standard deviation: $13.5 \pm 6.8$ μg/m$^3$) generally agreed with corresponding observations ($13.4 \pm 9.7$ μg/m$^3$) with a correlation coefficient of 0.57. More than 70% of the comparisons differed within a factor of two (Fig. 2).

Both simulations and observations show high NH$_3$ concentrations of about 30 μg/m$^3$ in warm seasons (March-October) due to enhanced NH$_3$ volatilization and frequent fertilization activities, and lower values (mostly < 15 μg/m$^3$) in other months (Fig. 3). Spatially, the hotspot of NH$_3$ was mainly concentrated in Hebei, Shandong and Henan provinces, which had the most intensive agricultural productions over China and thus emitted considerable gas-phase NH$_3$ into atmosphere. We note that the simulated NH$_3$ concentrations were underestimated by about a factor of two in wintertime (January, February, and December). Recently, NH$_3$ emission from the residential coal and biomass combustion for heating is considered to be a potentially important source of NH$_3$ in suburban and rural areas during wintertime (Li et al., 2016), but it has been not fully included in our bottom-up inventory, which could be responsible for such deviation between the model and observations.

The simulated NH$_3$ VCDs, calculated by integrating NH$_3$ molecular concentrations from the surface level to top troposphere, was consistent with observed NH$_3$ columns of 2016 on the magnitude and spatial-temporal patterns, although
the winter results were underestimated (Fig. S2). Both IASI measurements and the WRF-Chem simulation showed high annual mean NH$_3$ column concentrations in Hebei, Shandong and Henan provinces, reaching above $30 \times 10^{15}$ molecules/cm$^2$. Moreover, we also evaluated the modelled SNA concentrations using the filter-based PM$_{2.5}$ samples at an urban atmospheric monitoring station in North China Plain during 2014–2016 (Fig. S3). The model generally reproduced the observed SNA concentrations, with small annual mean bias for sulfate (−2%) and ammonium (−13%) and a relatively large bias for nitrate (−24%). Overall, the model performed well in modelling the concentrations in tropospheric NH$_3$ as well as secondary inorganic aerosols, which provides high confidence for the following interpretation of the NH$_3$ increases.

The model successfully reproduced the observed increasing trend in NH$_3$ columns over the North China Plain during 2008–2016 (Fig. 4). The modelled NH$_3$ columns were systemically lower than the measurements because the relative error weighting mean method would bias a high result due to the smaller relative error in a larger column (Van Damme et al., 2014; Whitburn et al., 2016). Similar to IASI observations, an increase of 25% in NH$_3$ columns was found in the simulations between 2012 and 2016, and the SO$_2$ columns averaged over the North China Plain decreased by 50% in this period, both of which were close to the measurements.

To verify our hypothesis, we replaced SO$_2$ emissions during 2010–2016 by those in 2008, and repeated the simulations (referred to as Run_10_S08 to Run_16_S08). It was noticeable that under these conditions, the increasing trend of NH$_3$ column concentrations disappeared, and even a slight decrease took place (Fig. 4). The largest difference was found in 2015 and 2016, when the annual NH$_3$ columns were reduced by about 40%, or $10 \times 10^{15}$ molecules/cm$^2$, corresponding to the 60% reduction in SO$_2$ emissions between 2008 and 2016. These tests support our hypothesis that the rapid SO$_2$ emission reductions led to the increased NH$_3$ levels during 2008–2016.

### 3.3 Influence of SO$_2$ emission reductions on tropospheric NH$_3$ concentrations

As we indicated above, SO$_4^{2−}$ was observed to be decreasing over recent years in response to the reductions of SO$_2$ emissions. This was also reproduced by our simulations, which showed that the annual average sulfate concentrations decreased by almost 50% in the lower troposphere. This decreasing trend was especially pronounced after 2013 owing to the much effective SO$_2$ emission reductions. Given that the vapor pressure of H$_2$SO$_4$(g) is practically zero over atmospheric
particles, atmospheric $\text{SO}_4^{2-}$ is predominately in the particle phase and can combine with $\text{NH}_3$ available in air, forming sulfate salts (mostly ammonium sulfate/bisulfate) (Seinfeld and Pandis, 2006). Since North China Plain is typically under rich $\text{NH}_3$ regimes, $\text{SO}_4^{2-}$ is mainly in the form of ammonium sulfate (Meng et al., 2011; Huang et al., 2017); and the aforementioned $\text{SO}_4^{2-}$ reductions would therefore increase atmospheric $\text{NH}_3$ concentrations by driving the phase state of $\text{NH}_3$ from particulate to gaseous.

By assuming that a 1 mol decrease in simulated $\text{SO}_4^{2-}$ would lead to a 2 mol increase in ambient gaseous $\text{NH}_3$ in this region, the average annual increase in the tropospheric $\text{NH}_3$ columns due to the reductions of $\text{SO}_4^{2-}$ was estimated to be approximately $1.5 \times 10^{15}$ molecules cm$^{-2}$ yr$^{-1}$ over North China Plain during 2008–2016, which is comparable with or higher than the simulated results from Run_08 to Run_16, as well as the IASI observations ($0.9 \times 10^{15}$ molecules cm$^{-2}$ yr$^{-1}$). By neglecting the deposition processes, we found that the rapid $\text{SO}_2$ emission reduction of 50% from 2012 to 2016 could result in a 55% increase in the $\text{NH}_3$ columns, compared to that of 30% recorded by IASI observations. Overall, the estimation results confirmed that the increasing trend of $\text{NH}_3$ can be entirely attributable to the $\text{SO}_2$ emission reductions.

We compared the spatial patterns of decreased $\text{SO}_4^{2-}$ and increased $\text{NH}_3$ between 2008 and 2016 (Run_08 – Run_16). Large reductions of $6–10 \times 10^{15}$ molecules/cm$^2$ in annual averages of sulfate columns were concentrated in Hebei, Shandong and Henan provinces, the area subject to high $\text{SO}_2$ loadings and stringent emission controls (Fig. 5a). Meanwhile, the simulated increases in $\text{NH}_3$ columns reached more than $8 \times 10^{15}$ molecules/cm$^2$ in most parts of the North China Plain (Fig. 5b), and were comparable with those observed by the IASI ($8–16 \times 10^{15}$ molecules/cm$^2$). In addition, we found that $\text{NH}_3^+$ concentrations have decreased with a similar magnitude to the increases in gas-phase $\text{NH}_3$ levels between Run_08 and Run_16. The proportion of $\text{NH}_3$ in the total ($\text{NH}_3 + \text{NH}_3^+$) increased on average from 26% in 2008 to 37% in 2016 over North China Plain. Figure 5c, d illustrated that without the large $\text{SO}_2$ emission reductions between 2008 and 2016 (i.e., replacing $\text{SO}_2$ emissions in 2016 by those in 2008, Run_08 – Run_16_S08), the sulfate columns partly increased, and correspondingly the $\text{NH}_3$ columns remained constant or decreased by about $5 \times 10^{15}$ molecules/cm$^2$ (~20% relative to the 2008 level) in parts of the North China Plain. Thus, the increase in the tropospheric $\text{NH}_3$ columns was the result of a transition in $\text{NH}_3$ phase partitioning, which was associated with the decreased formation of ammonium sulfate due to $\text{SO}_2$ emission reductions.
The seasonal variations in $\text{SO}_4^{2-}$ decreases and $\text{NH}_3$ increases were almost consistent (Fig. 6). We can see that the reduction of sulfate column concentrations between the Run_08 and Run_16 reached $1.3 \times 10^{15}$ molecules/cm$^2$ in summer (JJA), which was about three times larger than in other seasons. The corresponding percent reductions ranged from 15% in DJF to 36% in JJA. Considering that the $\text{SO}_2$ emission reductions were uniform throughout the year, this seasonal pattern was likely attributed to the conversion efficiency of $\text{SO}_2$ to $\text{H}_2\text{SO}_4$. Our simulations showed that a 1 mol decrease in $\text{SO}_2$ corresponded to an approximately 0.7 mol decrease in particulate sulfate in summer over North China Plain, but the values dropped to below 0.4 in other seasons. It is known that the photochemical oxidation of $\text{SO}_2$ by OH radical is most active in summertime due to high atmospheric oxidizing capacity, and it dominates the formation of $\text{SO}_4^{2-}$, which could make the response of $\text{SO}_4^{2-}$ concentrations to $\text{SO}_2$ emission reductions more sensitive (Paulot et al., 2017; Huang et al., 2014). The comparison of modelled $\text{NH}_3$ columns also showed a markedly higher increase in summer months than during other seasons, driven by the variations in $\text{SO}_4^{2-}$. Furthermore, by comparing the model results between the Run_16 and Run_16_S08 cases, we found that without considering the $\text{SO}_2$ emission reductions, the seasonal increases in $\text{NH}_3$ columns and decreases in $\text{SO}_4^{2-}$ concentrations disappeared.

In addition, we noted that the simulated particulate nitrate ($\text{NO}_3^-$) concentrations appear to increase in the North China Plain between 2008 and 2016 despite a 23% reduction in $\text{NO}_x$ emissions (Fig. S4). The in situ measurements in Beijing indicated that the $\text{NO}_3^-$ concentrations fluctuated during 2013–2016. It implied that the $\text{NO}_x$ emission reduction could not be responsible for the increase in $\text{NH}_3$. We also tested the effects of meteorological conditions on $\text{NH}_3$ variations by a simulation with meteorological fields in 2016 and anthropogenic emissions in 2012 (Run_16_E12). Compared to the Run_12 case, we found the change in meteorological fields (2012 vs. 2016) had a negligible influence on $\text{NH}_3$ concentrations in most of North China Plain. Although temperature increase was reported to partly contribute to the positive trend of $\text{NH}_3$ (Warner et al., 2017; Fu et al., 2017), our simulations indicated that the overall meteorological factors could not explain the observed significant increase tropospheric $\text{NH}_3$ concentrations over North China Plain.

4 Conclusion

In this study, we found that an increase in tropospheric $\text{NH}_3$ columns observed over the North China Plain during 2008–2016
was not caused by the increase in NH$_3$ emission, which actually displayed a slightly decreasing tendency. Neither meteorological conditions nor NOx emissions could explain it. Our work strongly indicates that the rapid SO$_2$ emission reductions (60%) from 2008 to 2016 were responsible for almost the entire NH$_3$ increases. The SO$_2$ emissions reduction decreased SO$_4^{2-}$ concentrations by about 50% in the lower troposphere, which reduced the formation of ammonium sulfate particles and consequently increased the average proportions of gas phase NH$_3$ from 26% (2008) to 37% (2016) within the total NH$_3$ column concentrations. The transition in the NH$_3$ phase state from particulate to gaseous was more pronounced in summertime than in other seasons, due to a more sensitive response of SO$_4^{2-}$ concentrations to SO$_2$ emission reductions.

Given the on-going stringent controls on SO$_2$ emissions in China, a continued increase in NH$_3$ concentrations is anticipated if NH$_3$ emissions are not well-regulated. The increased tropospheric NH$_3$ levels may have a significant impact on air pollution and nitrogen deposition in China. For instance, the elevated NH$_3$ would facilitate ammonium nitrate formation based on the aerosol thermodynamic equilibrium and negatively impact PM$_{2.5}$ control. That is supported by the fact that NO$_3^-$ concentrations remain high in Northern China and have become increasingly important in contributing to PM$_{2.5}$ pollution (Wen et al., 2018; Li et al., 2018), despite a moderate NO$_x$ emission reduction. The increased proportion of gas-phase NH$_3$ within the total can increase ammonium-nitrogen deposition since gas-phase ammonia deposits more rapidly than particle ammonium. This may alter the spatial pattern of regional nitrogen deposition with higher levels of NH$_3$ deposited near emission sources. These effects are important for human and ecosystem health and need to be investigated in future studies.

Data availability. NH$_3$ vertical column density data are freely available through the AERIS database: http://iasi.aeris-data.fr/NH3/. The SO$_2$ vertical column density retrieved from the Ozone Monitoring Instrument is available from Level-3 Aura/OMI Global OMSO2e Data Products released by NASA Goddard Earth Science Data and Information Service Center (https://disc.sci.gsfc.nasa.gov/). Anthropogenic emissions in industry, power plants, transportation, and residential sectors are obtained from Multi-resolution Emission Inventory for China (MEIC, http://www.meicmodel.org/).

Author contributions. Y.S., M.H., and T.Z. designed the study. Z.W. and M.H. conducted in situ measurements of aerosol
chemical compositions. Y.P. conducted in situ measurements of gas-phase ammonia concentrations. Q.Z. developed the MEIC emission database. M.L. and X.H. contributed to the development of ammonia emission inventory. M.L., X.H., Y.S., S.W., L.Z and T.Z. analyzed data. M.L. led the writing with input from all co-authors.

Competing interests. The authors declare that they have no conflict of interest.

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References


Figure 1. (a) Inter-annual trends of SO$_2$ and NH$_3$ VCDs averaged over North China Plain from 2008 to 2016. (b) Inter-annual trends of emissions of SO$_2$ and NH$_3$ from 2008 to 2016, and mean concentrations of PM$_{2.5}$ sulfate and ammonium derived from in situ measurements at an urban station (Beijing, 39.99° N, 116.3° E) in North China Plain from 2013 to 2016.
Figure 2. Comparison of modelled gaseous NH$_3$ concentrations with corresponding monthly measurements of NH$_3$ from Sep. 2015 to Aug. 2016. The 1:2 and 2:1 dashed lines are shown for reference and the Pearson correlation coefficient is shown inset.
Figure 3. Spatial distribution of modelled surface NH$_3$ concentrations (μg/m$^3$) and in situ measurements over North China Plain from September, 2015 (201509) to August, 2016 (201608). The location of the North China Plain can be found in Fig. S1.
Figure 4. Trends in the annual averages of observed and simulated NH$_3$ columns. The red stars denote the simulated NH$_3$ columns under the 2008 SO$_2$ emissions levels (i.e., Run_10_S08 to Run_16_S08).
Figure 5. The differences between Run_08 and Run_16 (a, b), and between Run_08 and Run_16_S08 (c, d). A-F in Figure 3a denote Beijing, Tianjin, Hebei, Shanxi, Shandong, and Henan Provinces, respectively.
Figure 6. Seasonal patterns of simulated $\text{SO}_4^{2-}$ (a) and $\text{NH}_3$ (b) columns for Run_08, Run_16, and Run_16_S08 (the simulation for 2016 with $\text{SO}_2$ emissions in 2008) cases. MAM, JJA, SON and DJF represent spring (Mar., Apr. and May), summer (Jun., Jul. and Aug.), autumn (Sep., Oct. and Nov.) and winter (Dec. Jan. and Feb.) months for this region.