The nonlinear response of fine particulate matter pollution to ammonia emission reductions in North China

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Abstract

Recent Chinese air pollution actions have significantly lowered the levels of fine particulate matter (PM2.5) in North China via controlling emissions of sulfur dioxide (SO2) and nitrogen oxides (NOx) together with primary aerosols, while the emissions of another precursor, ammonia (NH3), have not yet been regulated. This raises a question that how effective the NH3 emission controls can be on the mitigation of PM2.5 pollution along with the reduction of SO2 and NOx emissions. Here we use a regional air quality model to investigate this issue focusing on the PM2.5 pollution in North China for January and July 2015. We find that the efficiency of the PM2.5 reduction is highly sensitive to the NH3 emission and its reduction intensity. Reductions in the population-weighted PM2.5 concentration (PWC) in the Beijing–Tianjin–Hebei region are only 1.4–3.8 µg m⁻³ (1.1%–2.9% of PM2.5) with 20%–40% NH3 emission reductions, but could reach 8.1–26.7 µg m⁻³ (6.2%–21%) with 60%–100% NH3 emission reductions in January 2015. Besides, the 2015–2017 emission changes (mainly reduction in SO2 emissions) could lower the PM2.5 control efficiency driven by the NH3 reduction by up to 30% for high NH3 emission conditions, while lead to no change or increase in the efficiency when NH3 emissions become low. NOx emission reductions may enhance the wintertime PM2.5 pollution due to the weakened titration effect and can be offset by simultaneously controlling NH3 emissions. Our results emphasize the need to jointly consider NH3 with SO2 and NOx emission controls when designing PM2.5 pollution mitigation strategies.

1. Introduction

Fine particulate matter (particle with aerodynamic diameter less than or equal to 2.5 µm; also referred as PM2.5) not only poses serious harm to human health but also adversely influences atmospheric environment (Li et al. 2014, Gao et al. 2017, Hou et al. 2019). In recent years, the North China Plain has experienced severe PM2.5 air pollution and drawn worldwide attention (Huang et al. 2014, Zhang et al. 2015). To abate the PM2.5 air pollution, the Chinese government has implemented the ‘Action Plan on Prevention and Control of Air Pollution’ in 2013 and ‘Three-year Action Plan Fighting for a Blue Sky’ in 2018 (Chinese State Council 2013, 2018, Zhang et al. 2019a). As a consequence, the annual mean PM2.5 concentration in the Beijing–Tianjin–Hebei (BTH) region has decreased from 106 µg m⁻³ in 2013 to 64 µg m⁻³ in 2017 (MEE 2016, 2018). However, the latter value is still much higher than the China’s National Ambient Air Quality Standard of 35 µg m⁻³, which calls for more stringent emission control measures.

Ammonia (NH3) is the main alkaline gas in the ambient atmosphere and plays a critical role in nitrogen deposition and haze pollution.
(Wang et al. 2013, Zhang et al. 2015, Pan et al. 2018). It first reacts with sulfuric acid (H$_2$SO$_4$, typically produced by the oxidation of SO$_2$) to form ammonium sulfate aerosol, and excessive NH$_3$ then reacts with nitric acid (HNO$_3$, produced from the oxidation of NO$_2$) to form ammonium nitrate aerosol. These secondary inorganic aerosols (SIAs, including sulfate, nitrate, and ammonium) account for 30%–50% of PM$_{2.5}$ in eastern China (Zhao et al. 2013, Huang et al. 2014, Sun et al. 2016). Depending on the abundance of NH$_3$ in the air, the formation of SIA can be considered as the NH$_3$-poor condition (when there is insufficient NH$_3$ to neutralize H$_2$SO$_4$) or the NH$_3$-rich condition (when there is NH$_3$ to further neutralize HNO$_3$) (Seinfeld and Pandis 2006). When NH$_3$ is too excessive, the formation of nitrate becomes HNO$_3$-limited, and most NH$_3$ remains gaseous (Xu et al. 2019). The availability of NH$_3$ also significantly modulates liquid aerosol pH and then affects the heterogeneous production of secondary aerosols (Zheng et al. 2020).

Clean air actions in China have implemented a series of emission control measures mainly targeting fuel combustion induced emissions of SO$_2$, NO$_x$ (NO + NO$_2$), and primary aerosols (Zhang et al. 2019a). Regional NH$_3$ emissions are dominantly from agricultural activities (i.e. fertilizer application and livestock manure management) (Zhang et al. 2018) and have not yet been regulated in China (Fu et al. 2017, Zheng et al. 2018). The recent ‘Three-year Action Plan Fighting for a Blue Sky’ called for agricultural NH$_3$ emission controls but without a specific reduction target (Chinese State Council 2018). Atmospheric chemistry modelling studies indicated that controlling agricultural NH$_3$ emissions would significantly decrease aerosol nitrate in North China (Han et al. 2020) in particular during severe wintertime events (e.g. a decrease of SIA by ∼21% from 40% reduction of the NH$_3$ emissions in North China found by Xu et al. (2019)), while thermodynamic calculations suggested that >50% reduction of the NH$_3$ emissions was required to effectively reduce the SIA levels in this region (Guo et al. 2018, Song et al. 2019). The discrepancy can be largely induced by the accuracy of the NH$_3$-emission estimates. In addition, the effectiveness of NH$_3$ emission controls on PM$_{2.5}$ along with the rapid changes of the SO$_2$ and NO$_x$ emissions in North China remains undetermined.

To address these issues, we use a regional air quality model combined with our recent developed Chinese agricultural NH$_3$ emission inventory (Zhang et al. 2018). We conduct a series of model simulations to quantify the effectiveness of NH$_3$ emission controls on the PM$_{2.5}$ pollution under different NH$_3$ emission reduction conditions as well as under different SO$_2$ and NO$_x$ emission conditions (e.g. considering the 2015–2017 SO$_2$/NO$_x$ emission changes).

2. Methodology and data

2.1. The WRF-Chem model

The Weather Research and Forecasting (WRF) version 3.6.1 model coupled with Chemistry (WRF-Chem) is employed to simulate the meteorology and atmospheric chemistry. The modeling framework is configured with two domains (figure S1 (available online at stacks.iop.org/ERL/16/034014/mmedia)) using 161 (east–west) × 171 (south–north) and 150 (east–west) × 159 (south–north) grid cells at 27 km and 9 km horizontal resolutions, respectively. The outer domain covers China and its adjacent areas, and the inner domain covers North China (110º–120º E and 35º–43º N; figure S1) where this study focuses on. The National Center for Environmental Prediction Final (FNL) Analysis data with 1º spatial resolution and 6 h temporal resolution are used for the initial and lateral meteorological boundary conditions. The meteorological fields are re-initiated every 2 d using the FNL Analysis data to prevent the simulated meteorology drifting away from the actual conditions, so that they are nearly the same for all our simulation scenarios (table 1) with slight differences likely driven by the coupling of chemistry and meteorology. The chemical initial and boundary conditions are from the outputs of the global chemical transport model MOZART-4 (Emmons et al. 2010).

Our simulations use the gas-phase Carbon-Bond Mechanism Z mechanism (Zaveri and Peters 1999) coupled with a four-bin sectional (with dry diameters of 0.039–0.156, 0.156–0.625, 0.625–2.5, and 2.5–10.0 μm) Model for Simulating Aerosol Interactions and Chemistry aerosol scheme (Zaveri et al. 2008). Formation of sulfate aerosol in the model accounts for gas-phase oxidation SO$_2$, and aqueous-phase oxidation of SO$_2$ by H$_2$O$_2$ and O$_3$ in clouds. We include the heterogeneous sulfate formation reactions on particle surface based on Chen et al. (2016) to improve the

### Table 1. Emission settings in the WRF-Chem simulation scenarios.

| Simulation scenario | Description |
|---------------------|-------------|
| Base                | The 2015 emission conditions, also referred as S1R0 |
| S1RN ($N = 20/40/60/100$) | NH$_3$ emission is reduced by 20%, 40%, 60%, 80% and 100%, respectively. |
| S2RN ($N = 0/20/40/60/100$) | Similar to S1RN, but further reduces the NO$_x$ and SO$_2$ emissions from the levels of 2015 to those of 2017 in North China. |
| S3RN ($N = 0/20/40/60/100$) | Similar to S2RN, but further reduces NO$_x$ emissions by 20% in North China. |
model simulation of SIA. The NH$_3$ and HNO$_3$ gas-aerosol equilibrium is determined by the gas-particle partitioning module Adaptive Step Time-Split Euler Method (Zaveri et al. 2008). We increase the anthropogenic organic carbon (OC) emissions by a factor of 4 in July to account for secondary organic aerosols in the model (Sun et al. 2012) as our model configuration does not consider online secondary organic aerosols (SOA) formation. This SOA assumption shall not affect our analyses because the chemistry of SIA and SOA is uncoupled in the model.

The model physical settings include the Morrison double-moment microphysics scheme (Morrison et al. 2009), the Grell-3 cumulus scheme (Grell et al. 2002), the Rapid Radiative Transfer Model long-wave radiation scheme (Mlawer et al. 1997), the Goddard short-wave radiation scheme (Chou et al. 1994), the Yonsei University planetary boundary layer scheme (Hong et al. 2006), the revised MM5 (fifth-generation Mesoscale Model) Monin–Obukhov surface layer scheme, and the Unified Noah land-surface model (Chen and Dudhia 2001). A single-layer Urban Canopy Model is used to explicitly simulate the urban areas (Kusaka et al. 2001). We have further updated the land use types with the 2015 Moderate Resolution Imaging Spectroradiometer Land Cover Type (MCD12Q1) Version 6 data product (https://lpdaac.usgs.gov/products/mcd12q1v006/).

Anthropogenic emissions are from the 2015 Multi-resolution Emission Inventory for China (MEIC, www.meicmodel.org/) and the 2010 MIX inventory for regions outside mainland China (Li et al. 2017), except for Chinese agricultural NH$_3$ emissions that are from Zhang et al. (2018) with updated statistics for the year 2015. Biogenic emissions are calculated online using the Model of Emissions of Gases and Aerosols from Nature (Guenther et al. 2006). Biomass burning emissions are from the Fire Inventory from the NCAR (Wiedinmyer et al. 2011). Figure S2 shows the spatial distributions of NH$_3$, SO$_2$, and NO$_x$ emissions over North China in January and July 2015, and table S1 summarizes the emission totals. Our estimates of anthropogenic NH$_3$ emissions in North China are 0.11 Tg month$^{-1}$ in January and 0.25 Tg month$^{-1}$ in July. Compared with the MEIC NH$_3$ emissions, our estimates are about 1% lower in January and 44% higher in July. Our study does not consider bi-directional NH$_3$ fluxes (Bash et al. 2013), and treats emission and deposition as separate processes. This may affect model SIA simulation (Zhu et al. 2015), and needs to be evaluated in future work.

We conduct a series of WRF-Chem simulations as summarized in Table 1. First, the baseline simulation (Base, also denoted as the S1R0 scenario) includes the emissions described above and can be evaluated with observations. Second, a group of sensitivity simulations (S1RN) by reducing anthropogenic NH$_3$ emissions over North China by 20%, 40%, 60%, 80%, and 100%, respectively (denoted as S1RN scenarios, N = 20/40/60/100). The differences in PM$_{2.5}$ concentrations between S1R0 and S1RN then estimate the effects of NH$_3$ emission reductions. Third, a group of sensitivity simulations (S2RN, N = 0/20/40/60/100), similar to S1RN, but reduces anthropogenic SO$_2$ emissions by 40% (S2RN, N = 0/20/40/60/100) and anthropogenic NO$_x$ emissions by 8% in North China to reflect emission changes from 2015 to 2017 (Zheng et al. 2018; figure S3). Fourth, another group of sensitivity simulations, similar to S2RN, but further reduces anthropogenic NO$_x$ emissions in North China by 20% (S3RN, N = 0/20/40/60/100) reflecting potential NO$_x$ emissions reduction in the next few years (Liu et al. 2016). For all simulations, a winter month (January) and a summer month (July) in 2015 are simulated after 3 d spin-up for initialization. We find a longer spin-up time of 10 d only slightly change the model simulations.

2.2. Meteorology and surface measurements
For model evaluation, meteorological observations including 10 m wind direction (WD10), 10 m wind speed (WS10), 2 m air temperature (T2), and 2 m relative humidity (RH2) in January and July 2015 at 36 stations in North China are collected from National Climatic Data Center (https://ncdc.noaa.gov/isd/data-access). Hourly observations of surface PM$_{2.5}$ concentrations at 39 stations in North China are obtained from the Ministry of Ecology and Environment of China (http://106.37.208.233:2035/).

Monthly NH$_3$ concentrations at seven sites from the Ammonia Monitoring Network in China (AMoN-China; Pan et al. 2018) are used to evaluate our NH$_3$ emission inventory in North China. We use NH$_3$ measurements from AMoN-China conducted during 01–31 January and 15–31 July 2015. We use measurements of PM$_{2.5}$ components, including sulfate (SO$_4^{2-}$), nitrate (NO$_3^-$), ammonium (NH$_4^+$), OC, and black carbon in January and July 2015 at Beijing (39.94° N, 116.38° E) and Tianjin (39.09° N, 117.31° E) obtained by the Institute of Atmospheric Physics. Hourly model results are sampled at the grids covering the stations. Correlation coefficient (R) and mean bias (MB) between observations and model results are calculated.

3. Results
3.1. Observed and simulated surface pollutant concentrations
Evaluations of model simulated meteorological variables (WD10, WS10, T2, and RH2) are shown in figure S4. The spatial patterns of simulated meteorological variables are overall in good agreement with observations in North China. Figure 1 shows the time series and spatial distributions of observed and base-simulated PM$_{2.5}$ concentrations over North China in January (figures 1(a) and (b)) and
Figure 1. Observed and WRF-Chem base simulated surface PM$_{2.5}$ concentrations over North China in January and July 2015. The left panels show time series of hourly observations (black dots) and model results (red lines) by averaging 39 stations in North China. The right panels show spatial distributions of observed (circles) and simulated (contours) monthly mean concentrations. Mean observed values (OBS) and corresponding model results (MOD), and their correlation coefficients (temporally and spatially) are shown inset.

The comparisons of PM$_{2.5}$ components with measurements at Beijing and Tianjin are shown in figure S5. The WRF-Chem base simulation in general captures the magnitudes and variations of observed surface PM$_{2.5}$ concentrations in both January and July with R values of >0.60 in January and >0.33 in July. The MB is small (2.6 µg m$^{-3}$) in January and relatively large in July (−9.1 µg m$^{-3}$). Evaluations with measurements of PM$_{2.5}$ components show that the model simulated SIA concentrations are biased low by 10%–40% in July, especially for sulfate. The reasons why our model biases are larger in July than January are unclear, and may reflect uncertainties in emissions and aerosol processes. The implemented heterogeneous sulfate formation herein (Chen et al 2016) perhaps needs further enhancements in summer. Model simulated RH$_2$ fields over North China also show larger negative biases in July than January (figure S4).

To further illustrate the effect of NH$_3$ emission changes on surface concentrations, we show in figures 2(e) and (f) changes in BTH regional mean gaseous NH$_3$, aerosol NH$_4^+$, and total reduced nitrogen (NH$_x$ = NH$_3$ + NH$_4^+$) as we gradually reduce anthropogenic NH$_3$ emissions in North China (i.e. S1R$^{sc}$ scenarios). When we begin to decrease NH$_3$ emissions (reductions <40%), surface NH$_3$ concentrations decrease rapidly while NH$_4^+$ concentrations decrease much slower, reflecting NH$_3$-excessive conditions with current emissions. However, under large emission reductions (>60%), changes in NH$_4^+$ concentrations become faster than NH$_3$ concentrations. The different responses of gaseous NH$_3$ and aerosol NH$_4^+$ to emission reductions suggest changes in their partitioning and thus the lifetime of NH$_x$ as gaseous NH$_3$ has a shorter lifetime than aerosol NH$_4^+$. Regionally, the changes balance each other, leading a close-to-linear response of NH$_x$ concentrations to NH$_3$ emission reductions for both months. This is consistent with previous studies that suggest atmospheric NH$_x$ is a better indicator of NH$_3$ emissions than NH$_3$ or NH$_4^+$ alone in the US (Pinder et al 2006, Zhang et al 2012).
Figure 2. Observed and simulated surface NH₃ concentrations in North China for the periods of 01–31 January (top panels) and 15–31 July (bottom panels) 2015. The left panels show comparison of measurements (circles) with the base simulation, and central panels show comparisons with sensitivity simulations (with NH₃ emissions in North China reduced by 20% for January and increased by 20% for July). Regional mean observed values (OBS) and corresponding model results (MOD), and their correlation coefficients are shown inset. The right panels show changes in Beijing–Tianjin–Hebei (BTH) mean gaseous NH₃ (blue lines), aerosol ammonium (NH₄⁺, red lines), and total reduced nitrogen (NHₓ = NH₃ + NH₄⁺, black lines) when NH₃ emissions in North China are decreased for January and July 2015.

3.2. Response of PM₂.5 pollution to NH₃ emission reductions
The sensitivity simulations with perturbed NH₃ emissions allow us to assess the responses of air pollution to NH₃ emission reductions. Figure 3 shows changes in surface PM₂.₅ concentrations as we gradually reducing NH₃ emissions in North China in January 2015. To describe the saturation of atmospheric NH₃, we follow previous studies (Song et al. 2018, Xu et al. 2019) and define the excess NH₃ (in unit of µg m⁻³) as the differences in NHₓ and required NH₃ to meet ionic equilibrium using the formula below:

\[
\text{Excess NH}_3 = \text{Total NH}_x - \text{required NH}_3. \tag{1}
\]

\[
\text{Total NH}_x = 17 \times \left( \frac{[\text{NH}_4^+]}{18} + \frac{[\text{NH}_3]}{22.4} \right). \tag{2}
\]

\[
\text{Required NH}_3 = 17 \times \left( \frac{[\text{SO}_4^{2-}]}{48} + \frac{[\text{NO}_3^-]}{62} + \frac{[\text{Cl}^-]}{35.5} + \frac{[\text{HNO}_3]}{22.4} + \frac{[\text{HCl}]}{22.4} - \frac{[\text{Na}^+]}{23} \right) \tag{3}
\]

where [NH₄⁺], [SO₄²⁻], [NO₃⁻], [Cl⁻], and [Na⁺] are the mass concentrations (in unit of µg m⁻³) of these ions, and [NH₃], [HNO₃], and [HCl] are gas mixing ratios (ppb) converting to molar unit with the value of 22.4 L mol⁻¹ at the standard atmospheric condition.

As shown in figure 3, changes of mean PM₂.₅ concentration in January become much more distinct with stronger NH₃ emission reductions in the region. The first 20% NH₃ emission reduction would only decrease PM₂.₅ in Beijing by 1.6 µg m⁻³ and by 1.4 µg m⁻³ in BTH. The values increase to 8.3 µg m⁻³ in Beijing and 7.0 µg m⁻³ in BTH with 60% NH₃ emission reductions, and 20.8 µg m⁻³ in Beijing and 20.4 µg m⁻³ in BTH when all NH₃ emissions are turned off. The largest PM₂.₅ responses shift towards the southern Hebei province where PM₂.₅ concentrations are particularly high (figure 1(a)). Such non-linear responses can be largely explained by the derived excess NH₃ in each scenario. As also shown in figure 3, NH₃ is highly saturated in the southern Hebei province in the base condition and scenarios with small NH₃ emission reductions, and thus the SIA portion of PM₂.₅ is insensitive to NH₃ emissions. We find similar results for July but with smaller PM₂.₅ decreases under strong NH₃ emission reductions than those in January (figure S7).

Figure 4 summarizes the changes of mean concentrations of PM₂.₅ and its components in BTH as driven by NH₃ emission changes in North China for January and July 2015. The decreases of PM₂.₅ concentration associated with NH₃ emission reductions
Figure 3. (Top panels) January mean changes in surface PM$_{2.5}$ concentrations due to NH$_3$ emission reductions in North China estimated as the differences between the base simulation and S1Rn scenarios with NH$_3$ emissions reduced by N% ($N = 20/40/60/80/100$). (Bottom panels) Excess NH$_3$ concentrations in January as estimated by the formula described in the text for the base simulation and S1Rn scenarios ($N = 20/40/60/80$). Regional mean values in Beijing and in BTH are shown inset.

Figure 4. Effectiveness of NH$_3$ emission reductions in North China on BTH regional mean surface PM$_{2.5}$ pollution in January (top panels) and July (bottom panels) 2015. The left panels show BTH geometric mean PM$_{2.5}$ (orange lines), sulfate (red shading), ammonium (green shading), and nitrate (blue shading) levels. The central panels show reductions in monthly mean (black lines), minimum (blue dashed lines), and maximum (red dashed lines) PM$_{2.5}$ concentrations. The right panels show changes in population-weighted PM$_{2.5}$ concentration (PWC) together with sulfate, ammonium, and nitrate contributions. Numbers inset are their values ($\mu g \text{ m}^{-3}$) in the base simulation.

follow a power exponential function in January leading to small PM$_{2.5}$ changes with small NH$_3$ emission reductions. The responses in July are closer to a linear function, reflecting a stronger sensitivity to NH$_3$ due to greater HNO$_3$ availability in summer than in winter. The mean BTH PM$_{2.5}$ in July would be decreased by 1.6/5.3/11.1 $\mu g \text{ m}^{-3}$ with 20%/60%/100% NH$_3$ emission reductions in North China. The PM$_{2.5}$ components in both months show that sulfate has minor changes but nitrate can be substantially decreased with reducing NH$_3$ emissions, as also found by Han et al (2020). The small decreases in sulfate concentrations under strong NH$_3$ emission reductions in January are caused by slightly lower sulfate formation on aerosol surface under these scenarios in the model. We find stronger PM$_{2.5}$ responses in heavy pollution episodes in both months. As shown in figure 4, for the heavy pollution episodes, defined as the highest 5% PM$_{2.5}$ concentrations, their values can be decreased by 4.5/24.2/64.4 $\mu g \text{ m}^{-3}$ when NH$_3$ emissions in North China are reduced by 20%/60%/100% in January. By contrast, the cleanest 5% PM$_{2.5}$ concentrations have insignificant change associated with NH$_3$ emission reductions.
Figure 4 also shows the responses of PM$_{2.5}$ PWC in North China as a metric more relevant to human health using population data from the Gridded Population of the World version 4 (GPWv4) dataset (CIESIN 2018). PWC values show similar but larger responses than the regional geometric means. When NH$_3$ emissions in North China are reduced by 20%–40%, monthly mean BTH PWC could be reduced by 1.4–3.8 µg m$^{-3}$ (1.1%–2.9% of PWC) in January and 1.8–3.6 µg m$^{-3}$ (4.3%–8.7% of PWC) in July. When NH$_3$ emissions are reduced by 60%–100%, BTH PWC would be reduced by 8.1–26.7 µg m$^{-3}$ (6.2%–21% of PWC) in January and 5.9–13 µg m$^{-3}$ (14%–32% of PWC) in July, illustrating PM$_{2.5}$ air quality improvements we can achieve by the NH$_3$ emission controls under the 2015 emission condition.

The analyses above have emphasized strong non-linear responses of PM$_{2.5}$ concentrations to NH$_3$ emission changes in North China. To better quantify their effectiveness, we further calculate the efficiency of NH$_3$ emission controls based on the sensitivity simulations as $\beta_1 = \frac{\Delta PWC}{PWC} / \Delta E$, where $\Delta PWC / PWC$ is the relative change of PWC and $\Delta E$ is the relative change of NH$_3$ emissions in North China, denoting the relative response of PWC in percentage to 1% reduction in NH$_3$ emissions under each NH$_3$ emission scenario. We also calculate the absolute efficiency $\beta_2 = \Delta PWC / \Delta E$, describing changes in PM$_{2.5}$ per unit mass change in NH$_3$ emissions as shown in figure S8. We find for the 2015 emission condition, the BTH mean $\beta_1$ efficiencies in January increase from 0.055%/% in the base condition to 0.48%/% (a factor of 8.7 higher) when NH$_3$ emissions are reduced by 80%. The $\beta_1$ efficiencies in July also indicate a non-linear response, yet much weaker than January, with values of 0.22%/% for the base condition and 200% higher (0.65%/%) when NH$_3$ emissions are 80% lower.

The effect of NH$_3$ emission reduction on PM$_{2.5}$ air pollution is thus highly sensitive to its emission estimate. A lower NH$_3$ emission estimate over North China would present a higher efficiency of NH$_3$ emission controls for mitigating PM$_{2.5}$ air pollution in this region. Xu et al. (2019) reported a 40% decrease in aerosol nitrate with NH$_3$ emissions over North China reduced by 40% in winter. This efficiency is higher than our estimate, which may be attributed to a lower wintertime NH$_3$ emission (Kang et al. 2016) in their study (figure S2). Future work is required to accurately constrain the NH$_3$ emissions.

3.3. Effects of NO$_x$ and SO$_2$ emission changes

We now quantify the influence of NO$_x$ and SO$_2$ emission reductions on the efficiency of NH$_3$ emission controls. This can be estimated by comparing the S1RN with S2RN and S3RN scenarios. As described above, S1RN scenarios reflect NH$_3$ emission reductions for the 2015 emission condition, S2RN scenarios reflect the 2017 condition, and S3RN scenarios further consider 20% NO$_x$ emission reduction. The differences of S2R0 minus Base and S3R0 minus S2R0 then estimate, respectively, the impacts of 2015–2017 SO$_2$/NO$_x$ emission changes and 20% further NO$_x$ emission reductions. The 2015–2017 emission change has led to decreases in BTH PM$_{2.5}$ in both months, mainly driven by the SO$_2$ emission reductions (figure S9), while with further 20% NO$_x$ emission reductions the BTH regional mean PM$_{2.5}$ would increase by 1.0 µg m$^{-3}$ in January (figure S9). Reducing North China NO$_x$ emissions alone in winter would increase ozone levels due to weakened titration and further enhance the formation of secondary aerosols, as recently found during the COVID-19 pandemic (Huang et al. 2020).

Figure 5 shows the changes in BTH PWC and $\beta_1$ efficiency of NH$_3$ emission reduction in the S1RN, S2RN, and S3RN scenarios for January and July 2015. We notice that the effects of 2015–2017 SO$_2$/NO$_x$ emission reductions (i.e. ~40% reduction in SO$_2$ emissions and ~8% reduction in NO$_x$ emissions) on BTH PWC improvements (4.6 µg m$^{-3}$ in January and 4.9 µg m$^{-3}$ in July) are comparable to 40%–60% NH$_3$ emission reduction in 2015. The maximum BTH PWC reductions as can be achieved by NH$_3$ emission controls are 26.7 µg m$^{-3}$ in January and 13.2 µg m$^{-3}$ in July for the S1RN scenarios, and 27.6 µg m$^{-3}$ in January and 9.4 µg m$^{-3}$ in July for the S3RN scenarios. The much larger differences between S1RN and S3RN in July (13.2 vs 9.4 µg m$^{-3}$) than January (26.7 vs 27.6 µg m$^{-3}$) are mainly driven by the different responses of PM$_{2.5}$ to the 20% NO$_x$ emission reduction. We can see that the impacts of the further 20% NO$_x$ emission reduction on BTH PWC (as contributed by decreases in nitrate) in July become smaller with lower NH$_3$ emissions.

Changes in SO$_2$ and NO$_x$ emissions can thus affect the efficiency of NH$_3$ emission reduction on PM$_{2.5}$ pollution. The 2015–2017 emission changes have generally decreased $\beta_1$ efficiencies (figure 5), for no NH$_3$ emission reduction scenarios, from 0.055%/% to 0.038%/% (30% reduction) in January and from 0.22%/% to 0.19%/% (14% reduction) in July. SO$_2$ emission controls, in one way, decrease the formation of ammonium sulfate aerosol, causing NH$_3$ in the air being more saturated, and in another way, enhance the formation of nitrate aerosol when more gaseous NH$_3$ is available. The net effects as can be seen in figure 5 are decreases in the efficiency under high NH$_3$ emission conditions, and no change or increase for low NH$_3$ emission conditions. The additional 20% NO$_x$ emission reduction would further suppress $\beta_1$ efficiencies in July by decreasing HNO$_3$ availability but increase them in January, reflecting the enhanced nitrate formation due to NO$_x$ emission reduction in BTH winter as discussed above. Our results indicate that starting NH$_3$ emission controls at a stage with high SO$_2$/NO$_x$ emissions will be more effective for PM$_{2.5}$ air pollution regulation, and a joint
Figure 5. Changes in BTH PWC (left panels) and NH$_3$ emission reduction efficiency (right panels) in the S1RN scenarios (black lines; the 2015 emission condition), S2RN scenarios (green lines; the 2017 emission condition), and S3RN scenarios (orange lines, the 2017 emission condition with NO$_x$ emission further reduced by 20%) in January (top panels) and July (bottom panels). BTH PWC changes are relative to the base-simulated results (i.e. S1R0). PWC values for S1R0, S2R0, and S3R0 are given inset.

NO$_x$ and NH$_3$ emission control in winter will be more effective than controlling NO$_x$ alone.

4. Conclusions

In summary, we have shown strong nonlinear responses of the PM$_{2.5}$ air pollution to NH$_3$ emission controls in North China. Using three sets of model simulations testing NH$_3$ emission reductions under different SO$_2$/NO$_x$ emission conditions, we find that under the current emission condition, changes in the PM$_{2.5}$ concentration in North China associated with NH$_3$ emission reductions follow a power exponential function in January. The BTH PWC in January would only decrease by 1.4–3.8 µg m$^{-3}$ (1.1%–2.9% of PM$_{2.5}$) when NH$_3$ emissions in North China were reduced by 20%–40%, but the decreases would reach 8.1–26.7 µg m$^{-3}$ (6.2%–21% of PM$_{2.5}$) with 60%–100% NH$_3$ emission reductions. Such nonlinearity reflects a switch of NH$_3$-excessive to NH$_3$-limited conditions for SIA, in particular, nitrate formation. The PM$_{2.5}$ changes in July also show a nonlinear response, but the nonlinearity is much weaker than in January.

As SO$_2$ emissions in North China have substantially reduced over 2015–2017, we find that lowered the efficiency of NH$_3$ emission controls on the PM$_{2.5}$ air pollution in both winter and summer by up to 30% for high NH$_3$ emission conditions, but lead to no change or increase in the efficiency when NH$_3$ emissions are low. Future reduction of NO$_x$ emissions may partly enhance the PM$_{2.5}$ pollution in BTH winter due to the weakened titration effect, and can be offset by jointly controlling NH$_3$ emissions.

The Chinese government has implemented a series of active clean air actions in recent years (Zheng et al 2018, Zhang et al 2019a). China’s anthropogenic emissions were estimated to decrease by 62% for SO$_2$ and 17% for NO$_x$ over 2010–2017, while NH$_3$ emissions slightly increased by 1% due to the absence of NH$_3$ emission controls (Zheng et al 2018). In the next 5–10 years, changes in SO$_2$ emissions might level off as the power plants have operated with ultralow emission standards, while NO$_x$ emissions will become stringently controlled to ensure further air quality improvements (Zheng et al 2018). Such future SO$_2$/NO$_x$ emission changes would increase the efficiency of NH$_3$ emission controls in winter. Our results emphasize the need to jointly consider emission reductions of SO$_2$, NO$_x$, and NH$_3$ for mitigating SIA air pollution. In addition to the air quality effect, NH$_3$ emission controls can also lead to
other environmental benefits, such as reducing nitrogen deposition and water pollution (Guo et al 2020), which should be considered in current environmental strategies.

Data availability statement

The data that support the findings of this study are available upon reasonable request from the authors.

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