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High efficiency of livestock ammonia emission controls on alleviating particulate nitrate during a severe winter haze episode in northern China

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Abstract

Although nitrogen oxide (NOx) emission controls have been implemented for several years in northern China, recent observations show particulate nitrate (NO_3^-) is becoming increasingly important during haze episodes. In this study, we find that particulate NO_3^- formation would easily become NH_3-limited under severe haze conditions, enhancing its sensitivity to NH_3 emission controls. Furthermore, improved manure management of livestock husbandry could reduce 40% of NH_3 emissions (currently 100 kiloton per a month) in winter of northern China. Under this emission reductions scenario, simulations from the thermodynamic equilibrium model (ISORROPIA-II) and the Weather Research and Forecast model coupled chemistry (WRF-Chem) all show that particulate NO_3^- could be reduced by approximately 40% during a typical severe haze episode (averagely from 40.8 to 25.7 μg/m^3). Our results indicate that reducing livestock NH_3 emissions would be highly effective to reduce particulate NO_3^- during severe winter haze events.

1 Introduction

In northern China (including Beijing, Tianjin, Hebei, Shandong, Shanxi and Henan), severe haze pollution events occur frequently during wintertime, with the concentration of $PM_{2.5}$ (particles with an aerodynamic diameter less than 2.5 μm) reaching hundreds of micrograms per cubic meter (Wang et al., 2015;Zheng et al., 2015;Elser et al., 2016). In severe haze events, secondary inorganic aerosol (SIA) plays a crucial role in haze formation, accounting for 30–77% of $PM_{2.5}$ (Huang et al., 2014). In October 2014, four extreme haze episodes were reported in North China Plain (NCP), with the concentrations of $PM_{2.5}$ exceeding 400 μg/m^3, and the concentrations of SNA (sulfate, nitrate, and ammonium) and particulate nitrate (NO_3^-) at this time exceeding 190 and 90 μg/m, respectively(Yang et al., 2015).

To mitigate severe fine particle pollution, the Chinese government has been taking strong measures to control SO_2 and NO_x emissions. It has been reported that SO_2 emissions in China have been reduced by 75% since 2007 (Li et al., 2017a). Meanwhile, particulate sulfate has also been found to decrease since 2005 (Geng et al., 2017). Liu et al. (2017a) found that NOx emissions in 48 Chinese cities decreased by 21% from 2011 to 2015. Unfortunately, in recent years, no obvious decreasing trend in the concentration of particulate NO_3^- had been observed in northern China (Zhang et al., 2015;Li et al., 2017b) and the ratio of ammonium nitrate (NH_4NO_3) to SNA increased continuously during severe haze events (Li et al., 2017d;Yang et al., 2017).

It was reported in recent studies that the large atmospheric NH_3 emissions in northern China have made particulate NO_3^- become more important, and it will reduce the effectiveness of existing $PM_{2.5}$ control strategies through SO_2 and NO_X emission reductions (Wang et al., 2013;Fu et al., 2017). However, the effectiveness of controlling NH_3 emissions in reducing particulate NO_3^- during severe winter haze events has not been reported.

In this study, we firstly compile a comprehensive NH_3 emission inventory for northern China in winter of 2015, and estimate the NH_3 emission reductions by improving manure management. Then, the ISORROPIA-II and WRF-Chem models are used to investigate the

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effectiveness of NH_3 emission reductions on alleviating particulate NO_3^- during a severe haze episode. Finally, the molar ratio of observational data is used to explore the particulate NO_3^- reductions efficiency during the wintertime.

2 Methods and Materials

2.1 Observational data

Hourly time-resolution aerosol and gas measurements were conducted at the Peking University urban atmosphere environment monitoring station (PKUERS) (39.991N, 116.313E) in Beijing in December 2015 and December 2016. A commercialized semi-continuous In-situ Gas and Aerosol Composition (IGAC) Monitor was used to measure the concentrations of water-soluble ions (e.g., NH_4^+, SO_4^{2-}, NO_3^-, Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-) in $PM_{2.5}$ and inorganic gases (e.g., NH_3, HNO_3, HCI). Relative humidity (RH) and temperature were observed at 1-min resolution at the same site. The quality assurance and control for the IGAC was described in Liu et al. (2017b). A typical severe haze episode occurred during the 6 to 10 in December 2015, with daily average concentrations of $PM_{2.5}$ exceeding 150 μg/m^3 for three days ($PM_{2.5}$ data are from China National Environmental Monitoring Centre). The average RH and temperature in this haze event were 60.9 ± 11.4% and 276.5 ± 1.4 K. The south wind was dominant with wind speed mostly less than 3 m/s. The average concentrations of particulate NO_3^-, NH_4^+ and SO_4^{2-} were 39.8 ± 14.7 μg/m^3, 27.7 ± 8.6 μg/m^3 and 42.4 ± 16.0 μg/m^3, respectively. The ratios of particulate NO_3^- concentrations to SNA were 36.5 ± 4.0%.

2.2 NH_3 emission inventory

A comprehensive NH_3 emission inventory in 2015 at a monthly and 1 km × 1 km resolution is developed based our previous studies (Huang et al., 2012;Kang et al., 2016). A diverse range of sources, including both agricultural (livestock manure and chemical fertilizer) and non-agricultural sectors (e.g., traffic, biomass burning etc.) were fully considered. Recent studies documented that our results agreed well with the satellite measurements by Infrared Atmospheric Sounding Interferometer (IASI) (Van Damme et al., 2014) and Tropospheric Emission Spectrometer (TES), and inverse model results by using ammonium (NH_4^+) wet deposition data (Paulot et al., 2014;Zhang et al., 2018). According to our inventory, the estimated NH_3 emission amount in northern China was 100 kiloton in December 2015. The largest source was livestock waste (57.0% of the total emissions), following by vehicle (12.2%), chemical industry (8.8%), biomass burning (5.4%), waste disposal (4.0%), synthetic fertilizer applications (2.4%) and other minor sources (9.1%). The proportion of chemical fertilizer is very small due to the limited fertilization activity in winter.

2.3 ISORROPIA-II and WRF-Chem models

The thermodynamic equilibrium model, ISORROPIA-II (Fountoukis and Nenes, 2007), being used to determine the phase state and composition of an NH_4^+- SO_4^{2-} - NO_3^- - K^+ - Ca^{2+} - Mg^{2+} - Na^+ - Cl^- - H_2O aerosol system with its corresponding gas components in thermodynamic equilibrium, was used to investigate the response of particulate NO_3^- to NH_3 emission reductions. Using measurements of water-soluble ions, T and RH from PKUERS as inputs, ISORROPIA-II can avoid the inherent uncertainty in estimates of

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emission inventories, pollutant transport, and chemical transformation. In this study, ISORROPIA-II was run in the "forward mode" and assuming particles are "metastable" with no solid precipitates, which is due to the relatively high RH range observed during this haze event (RH = 60.9 ± 11.4%).

We assess the performance of ISORROPIA-II by comparing measured and predicted particulate NO_3^-, NH_4^+ and gaseous HNO_3, NH_3. An error metric, the mean bias (MB), is used to quantify the bias (the description of MB is shown below Figure S1). The predicted particulate NO_3^-, NH_4^+ and NH_3 agree well with the measurements and the value of R^2 are 0.99, 0.94 and 0.84, respectively (Figure S1). The MB is only 1.0 ± 1.1 μg/m^3, 0.3 ± 1.3 μg/m^3 and -1.8 ± 1.6 μg/m^3, respectively. However, the model performs poorly on HNO_3, with an R^2 of only 0.06 and a MB of -1.0 ± 1.1 μg/m^3. This is because particulate NO_3^- is predominantly in the particle phase (the mass ratio of particulate NO_3^- to the total nitric acid (TN = NO_3^- + HNO_3) was 99.2 ± 1.9%), small errors in predicting particulate NO_3^- are amplified in HNO_3 predicting. Since the MB of HNO_3 is much smaller than the observed particulate NO_3^- (39.8 ± 14.7 μg/m^3) and NH_4^+ (27.7 ± 8.6 μg/m^3), this bias have little influence on simulating the efficiency of particulate NO_3^- reductions.

In the real atmosphere, changes in the level of total ammonia (TA = NH_4^+ + NH_3) can affect the lifetime of TN (Pandis and Seinfeld, 1990). This is because the gaseous HNO_3 has a faster deposition rate in the atmosphere than particulate NO_3^-, and reductions in NH_4^+ may prompt particulate NO_3^- partitioning into the gas phase. In such a case, the concentration of TN would not remain constant but decrease. In order to consider these, we use the Weather Research and Forecast Model coupled Chemistry (WRF-Chem) model (ver. 3.6.1) to investigate the effect of NH_3 emission controls on particulate NO_3^- formation in the regional scale. The simulations were performed for the severe haze event during 6 to 10 December 2015. The modeling domain covered the whole northern China with horizontal resolution of 25 km and 24 vertical layers from surface to 50 hPa. The initial meteorological fields and boundary conditions were taken from the 6 h National Centers for Environmental Prediction (NCEP) global final analysis with a 1° × 1° spatial resolution. The inorganic gas-aerosol equilibrium was predicted by Multicomponent Equilibrium Solver for Aerosols (MESA) in WRF-Chem(Zaveri et al., 2005;Zaveri et al., 2008). The Carbon-Bond Mechanism version Z (CBMZ) photochemical mechanism and Model for Simulating Aerosol Interactions and Chemistry (MOSAIC) aerosol model were used in this study(Fast et al., 2006). Anthropogenic emissions from power plants, industrial sites, residential locations, and vehicles were taken from the Multi-resolution Emission Inventory for China (MEIC; available at www.meicmodel.org).The WRF-Chem model could approximately reproduce the temporal variations of inorganic aerosol components in this haze event (Figure S2).

3 Results

3.1 High potential reduction of wintertime NH_3 emissions in northern China

Livestock husbandry accounts for the largest proportion of NH_3 emissions in winter of northern China (approximately 60%), which is mainly caused by the poor manure management. There are three main animal-rearing systems in China: free-range, grazing and intensive. On the one hand, the proportion of intensive livestock husbandry in China

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is only about 40%, far lower than that of developed countries. As a result, the widespread free-range and grazing animal rearing systems contribute more than half of the total livestock NH_3 emissions due to lacking manure collection and treatment (Kang et al., 2016). On the other hand, there were no relevant regulations about storage and application of manure for intensive farms in China in the past few decades. This causes most livestock farms also lack necessary measures and facilities for manure collection and storage (Chadwick et al., 2015). Meanwhile, most of the solid fraction of manure is applied to crops without any treatment and the liquid fraction is often discharged directly (Bai et al., 2017).

Due to the current poor manure management in China, the improved manure management may have great potential for NH_3 emission reductions from livestock husbandry (Wang et al., 2017). The improved manure management mainly includes three phases: in-house handling, storage and land application (Chadwick et al., 2011). According to previous studies, for in-house handling, regularly washing the floor and using slatted floor or deep litter to replace solid floor could both reduce NH_3 emissions by more than 50% (Monteny and Erisman, 1998;Hou et al., 2015). For storage, covering slurry and manure could reduce NH_3 emissions by about 50%-70% (Hou et al., 2015;Wang et al., 2017). For land application, cultivating the soil surface before application or incorporation and injection could both reduce NH_3 emissions by more than 50% (Sommer and Hutchings, 2001;Hou et al., 2015).

Based on the above research results, the livestock NH_3 emission reductions strategies applied in this study include the following steps. Firstly, the proportion of intensive livestock production was raised from 40% to 80% in our NH_3 emission inventory model. In our model, the animals in free-range and grazing animal rearing systems are assumed to live outdoors for half a day, and the improved manure management is only effective for indoor animals. Therefore, increasing the proportion of intensive livestock production is conducive to better manure management (Hristov et al., 2011). Secondly, the ratios of NH_3 emission reductions mentioned above were multiplied by NH_3 emission factors in three phases of manure management: 50% reduction at in-house handling, 60% (average value of 50% and 70%) reduction at storage and 50% reduction at land application. With these measures, we estimate that the NH_3 emission factors for the livestock in China could be comparable to those in Europe and the USA (shown in Table S1). Meanwhile, the NH_3 emission model predicted that the livestock NH_3 emissions were reduced by 60% (from 57 to 23 kiloton), causing approximately 40% reduction in total NH_3 emissions. Spatially, NH_3 emissions decreased significantly in Hebei, Henan and Shandong, where the livestock NH_3 emissions accounted for a large proportion of the total (shown in Figure S3).

3.2. Simulations of NO_3^- reduction due to NH_3 emission controls

In the ISORROPIA-II simulation, 40% reduction of TA was used to reflect the effects of reducing NH_3 emissions by 40%. In this haze event (from 6 to 10 December, 2015), the mean concentration of particulate NO_3^- decreased from 40.8 to 25.7 $\mu g/m^3$ (a 37% reduction). In addition, the peak hourly concentration of NO_3^- decreased from 81.9 to 30.7 $\mu g/m^3$ (a 63% reduction) (shown in Figure 1). The fundamental thermodynamic processes of TA reductions on decreasing particulate NO_3^- are explained below. Firstly, we found that NH_3 was quite available to react with HNO_3 in the thermodynamic equilibrium system, because NH_3 was 6.6 ± 3.8 $\mu g/m^3$ while HNO_3 was only 0.4 ± 1.1 $\mu g/m^3$. Secondly, almost all of particulate NO_3^- condensed into aerosol phase (the mass ratio of particulate NO_3^- to

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TN was 99.2 ± 1.9%) under such low temperature conditions (276.5 ± 1.4 K). Thirdly, the NH_3-HNO_3 partial pressure production (Kp) was as low as about 0.1 ppb^2 (calculated from ISORROPIA-II outputs, depending not only on temperature and RH but also sulfate concentration). The value of K_P would remain constant, if the temperature, RH and sulfate concentration remained unchanged. In general, NH_4NO_3 was not easy to volatilize into gas phase under these circumstances.

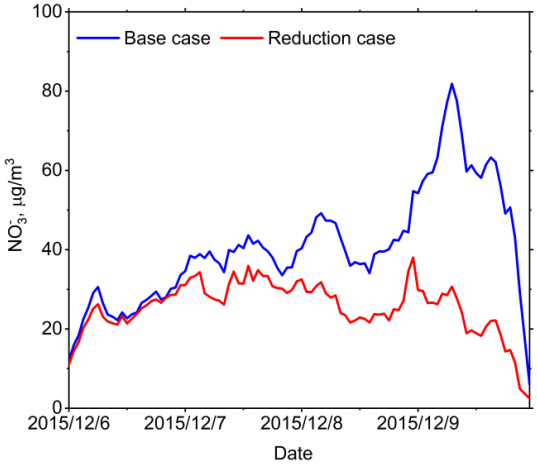

Figure 1. A comparison of particulate nitrate (NO_3^-) between the base (blue line) and emission reductions cases (red line) simulated by the ISORROPIA-II model in this severe haze episode.

When TA was reduced by 40%, the average mass concentration of gaseous NH_3 decreased from 6.6 to 0.01 μg/m^3 (from 8.8 ppb to 0.05 ppb). In order to keep the value of K_P constant in the thermodynamic equilibrium state, the reductions of NH_3 increased HNO_3, which shifted the particulate NO_3^- partitioning toward the gas phase. Hence, when NH_3 in gas phase was almost completely depleted, HNO_3 increased from 0.4 to 15.5 μg/m^3 (from 0.1 ppb to 5.6 ppb), leading to a reduction of particulate NO_3^- from 40.8 to 25.7 μg/m^3 (a 37% reduction). Meanwhile, NH_4^+ also decreased from 27.9 to 20.6 μg/m^3 and there was almost no change in sulfate level (decreased from 39.7 to 39.3 μg/m^3), with only trace amount of NH_4HSO_4 produced. This indicated that the reduction of particulate NH_4^+ and NO_3^- was mainly due to the reduction of NH_4NO_3.

The above process could also be explained by the interactions between aerosol acidity and gas-particle partitioning of HNO_3. Guo et al. (2018) used the S curves to demonstrate the relationship between aerosol pH and HNO_3 partitioning in the United States, Europe and China. The results showed that when TA was reduced to a certain extent, aerosol pH began to decline, prompting the particulate NO_3^- volatilizing into gas phase. However, using aerosol pH as an indicator of the sensitivity of particulate NO_3^- to TA has some limitations. On the one hand, it may not be suitable for low temperature or low relative humidity conditions. Because under these conditions, the estimation of aerosol pH would

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become inaccurate (Fountoukis et al., 2009). On the other hand, recent studies showed many factors could cause bias in aerosol pH prediction. For instance, Vasilakos et al. (2018) found that non-volatile cations (K^+, Na^+, Ca^{2+} and Mg^{2+}) in the fine mode could cause bias in the aerosol pH and HNO_3 partitioning prediction. Silvern et al. (2017) found that particles coated by organic material might retard the uptake of NH_3, which may also cause bias in modeling aerosol acidity and HNO_3 partitioning. In general, studying the process of aerosol acidity affecting particulate NO_3^- formation still requires more work to do, especially sensitivity tests, to unravel the potential effects of other factors.

We also conducted WRF-Chem simulations to quantify the impacts of NH_3 emission controls on particulate NO_3^- regionally. A 60% reduction in livestock NH_3 emissions was used as an emission reductions scheme and Figure 2 shows the spatial distribution of particulate NO_3^- under the base case and the emission reductions case. The spatial distribution of particulate NO_3^- was mainly concentrated in most parts of Henan (HN) and Hebei (HB), with the average concentration over 30 μg/m^3 (included in the blue box shown in Figure 2a). The highest particulate NO_3^- concentrations, more than 60 μg/m^3, were mainly located in central south of Hebei and northern Henan. In the emission reductions case, the mean concentration of particulate NO_3^- decreased from 34.2 to 20.7 μg/m^3 (a 40% reduction) in the range of the blue box. In addition, the sulfate concentration slightly changed from 28.1 to 24.3 μg/m^3, and $PM_{2.5}$ concentration dropped from 161.7 to 139.3 μg/m^3. The largest reductions in particulate NO_3^- were mainly located in the central north of Henan and central Hebei, where the percentage reduction was generally more than 60% (shown in Figure 2b). In these regions, severe haze events occurred frequently due to their large emissions of air pollutants, including NH_3 (Wang et al., 2014;Zhao et al., 2017). The contrast of figure 2a and 2b shows that particulate NO_3^- had been effectively reduced, especially in high concentration areas. The reason is explained in Sect. 3.3.

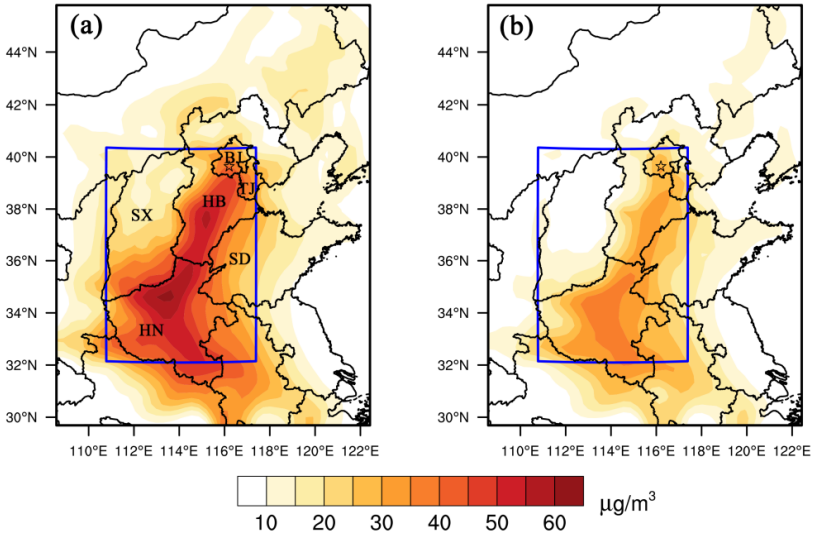

Figure 2. (a) Spatial distribution of particulate NO_3^- concentrations in northern China predicted by WRF-Chem from 6 to 10 December, 2015, for **(a)** the base case, and **(b)** the

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emission reductions case. The scope of this study focuses on the blue box, including Beijing (BJ), Tianjin (TJ), Hebei (HB), Shanxi (SX), Shangdong (SD) and Henan (HN).

3.3 The particulate NO_3^- reduction efficiency during the wintertime

The sensitivity of particulate NO_3^- to NH_3 is often determined by the availability of ambient NH_3, which can be represented by the observable indicator (Seinfeld and Pandis, 2006). In this study, we use the observed molar ratio (R) of TA to the sum of sulfate, total chlorine and TN minus Na^+, K^+, Ca^{2+} and Mg^{2+} to represent the availability of ambient NH_3 and predict the sensitivity of the particulate NO_3^- to changes in TN and TA.

$$R = \frac{TA}{2SO_4^{2-} + NO_3^- + HNO_3(g) + Cl^- + HCl(g) - 2Ca^{2+} - Na^+ - K^+ - 2Mg^{2+}} \quad (1)$$

The accuracy of R was examined by constructing the isopleths of particulate NO_3^- concentrations as a function of TN and TA (shown in Figure 3). The NO_3^- concentration was constructed by varying the input concentrations of TA and TN from 0 to 200 μg/m^3 in increments of 10 μg/m^3 independently in ISORROPIA-II, while using the observed average value for the other components. Over a range of temperatures (273–283 K) and RHs (30–90%), the dashed line of R = 1 divides each isopleth into two regions with tiny bias, which indicates that R can be used to qualitatively predict the response of the particulate NO_3^- to changes in concentrations of TN and TA.

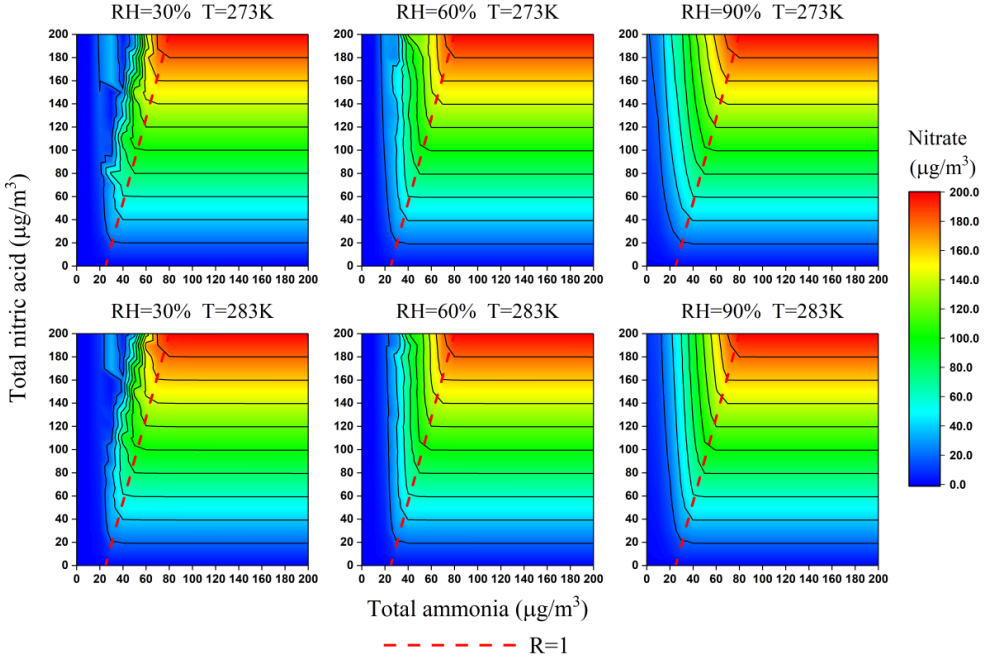

Figure 3. Isopleths of the particulate NO_3^- concentration (μg/m^3) as a function of TN and TA under average severe haze conditions in winter. The concentration of SO_4^{2-}, Cl^-, K^+,

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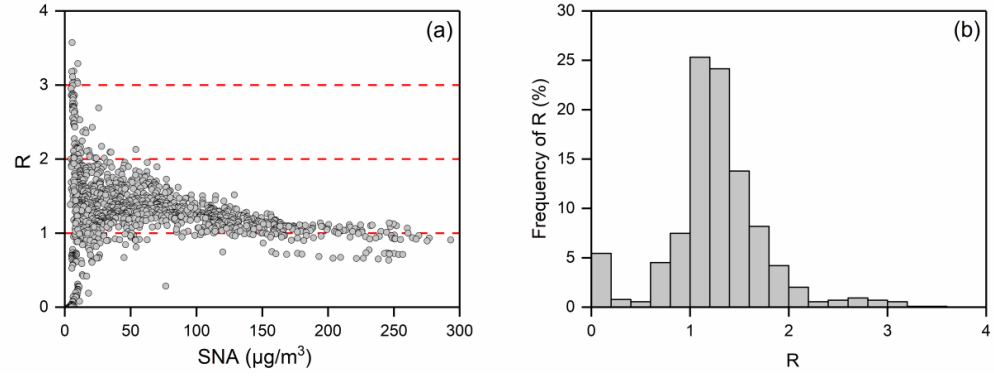
286 Ca^{2+}, Na^{+}, and Mg^{2+} was 60.2, 9.3, 0.56, 0.04, 0.75, and 0.03 μg/m^3, respectively. Values
287 are averages from all severe hazes during the observation period.

288

289 In the right side of the dashed line (R > 1), particulate NO_3^- formation is HNO_3-limited.
290 The NH_3 is surplus and almost all particulate NO_3^- exists in the aerosol phase. The TA
291 reductions mainly reduce NH_3, with negligible effects on particulate NO_3^-. By contrast,
292 particulate NO_3^- formation is NH_3-limited in the left of the dashed line (R < 1). There is
293 less NH_3 present in the gas phase, and TA reductions could reduce particulate NO_3^-
294 efficiently. For example, when the concentrations of TN and TA are 100 and 50 μg/m^3 (RH
295 = 60 % and T =273 K), the concentration of particulate NO_3^- is about 100 μg/m^3 and the
296 value of R is close to one (typical observational values during the severe haze in this study).
297 In such cases, if TA were reduced by 50% to 25 μg/m^3, the particulate NO_3^- would be
298 significantly reduced from 100 to 20 μg/m^3, an 80% reduction.

299 Under the typical winter conditions in northern China, the value of R was generally
300 greater than one and gradually declining with the increase in SNA concentrations (shown
301 in Figure 4a). When the concentration of SNA is greater than 150 μg/m^3, the values of R
302 become close to and frequently lower than one. This indicated that particulate NO_3^-
303 formation would easily become NH_3-limited under severe haze conditions when NH_3
304 emissions were reduced. In general, particulate NO_3^- will be reduced effectively by a 40%
305 reduction of NH_3 emissions in the condition that the value of R is less than 1.4 (shown in
306 Figure S4). This situation accounts for 68.1% of the entire December (shown in Figure 4b).
307 It should also be noted that the particulate NO_3^- can be insensitive to a 40% reduction in
308 NH_3 emissions when the value of R is greater than 1.4 (shown in Figure S4). This situation
309 mainly occurs in relatively clean days (the concentration of SNA is less than 75 μg/m^3),
310 accounting for only 31.9% of the entire December (shown in Figure 4a and 4b). Overall,
311 reducing 40% of NH_3 emissions could effectively reduce the levels of particulate NO_3^-
312 under typical winter haze conditions in northern China.



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314 **Figure 4.** (a) The observed molar ratio (R) and the concentrations of SNA in PKUERS
315 in December 2015 and December 2016. (b) The frequency of R during the same period.

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Improved manure management could reduce 40% of NH_3 emissions in winter of northern China. For a 40% reduction of TA in the atmosphere, ISORROPIA-II predicts that particulate NO_3^- could be reduced by 37% for the haze event (from 40.8 to 25.7 $\mu g/m^3$). When NH_3 emissions are reduced by 40%, the WRF-Chem simulation shows that particulate NO_3^- concentration could be reduced effectively throughout the whole region (the mean concentration of particulate NO_3^- was reduced from 34.2 to 20.7 $\mu g/m^3$, a 40% reduction), especially in the area with high particulate NO_3^- concentration (more than 60%). The molar ratio (R) of winter observational data in northern China shows that as the concentration of inorganic salts increases, the excess NH_3 in the atmosphere is decreasing, and particulate NO_3^- becomes more sensitive to NH_3 emission reductions. In general, controlling livestock NH_3 emissions could be an effective measure to reduce NH_3 levels and limit particulate NO_3^- formation.

The observed R provides a simple method to rapidly estimate the efficiency of NH_3 emission reductions on the particulate NO_3^- reductions, which can avoid the shortage of the air quality model, especially the uncertain estimates of meteorology. However, it also has some limitations, such as requiring accurate measurements of water-soluble ions and gaseous components. In addition, the accuracy of R needs to be examined in more detail for specific pollution and meteorological conditions. Therefore, the observed indicator and air quality models should be used in a complementary way to assess the effectiveness of NH_3 emission controls strategies.

NO_X emission controls could be a more direct and effective way to reduce the particulate NO_3^- than NH_3 emission reductions. However, in northern China, the target of NO_X emission reductions is only about 25% in the 13th Five-Year Plan (2016-2020). Furthermore, the previous study has shown that NO_X emission reductions in the U.S could be much costly to control $PM_{2.5}$ than reductions in NH_3 emission (Pinder et al., 2007). Due to the dominance of extensive livestock farming and the lack of emission controls policies, NH_3 emission reductions in China may be easier and cost less than the U.S. Therefore, in order to control $PM_{2.5}$ pollution more effectively in northern China, NH_3 emission controls are urgently needed. In addition, a comprehensive cost-effectiveness analysis of NH_3 emission control strategies in China is also much needed in the future.

It should be noted that the NH_3 emission reductions have plenty of significant environmental implications. On the one hand, it could increase the particle acidity to increase the solubility of transition metals in aerosol phase, which is related to aerosol oxidative stress and toxicity (Fang et al., 2017;Longo et al., 2016). Meanwhile, metal mobility could affect photosynthesis productivity (Duce and Tindale, 1991;Li et al., 2017c) and oxygen levels in the ocean (Ito et al., 2016) by changing nutrient distributions. Furthermore, particle strong acidity has also been directly linked to adverse respiratory effects (Schlesinger, 2007;Ward et al., 2002). On the other hand, anthropogenic NH_3 emission controls can decrease excess reactive nitrogen inputs to earth ecosystems, which could alleviate many adverse ecological effects including soil acidification, plant biodiversity reduction, and eutrophication (Bouwman et al., 2002;Stevens et al., 2004;Bowman et al., 2008). All these environmental impacts in China require further research.

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