Our point-by-point responses are provided below. The referees' comments are italicized.

Response to Referee #1

Referee: Xu et al. applied a model analysis, and found "High efficiency of livestock ammonia emission controls on alleviating particulate nitrate during a severe winter haze episode in northern China". The research topic is of extreme importance for adding scientific knowledge and supporting policy-makers on ammonia controls from livestock sector. This finding (based on real-time IGAC measurements and atmospheric modeling) provides strong evidence of the importance of livestock NH_3 mitigation (combined with NOx and SO_2 emission reductions) in improving air quality in this intensive agricultural and industrial region. Nevertheless, several statements & discussions are needed to be clarified in this manuscript. I suggest the manuscript to be published in ACP after proper revisions as below.

Response: We would like to thank the referrer for your detailed and constructive comments. Please see our point-by-point reply below.

Referee: 1. General.

While this paper could be useful as a theoretic support of ammonia emission controls on alleviating particulate matters, however, the authors should express their new findings (e.g. the detailed analysis of the equilibrium between …) clearly in the revision. Because it is not surprising that a reduction in NH_3 emission alleviates particulate matter (e.g. $PM_{2.5}$) pollution (see Wu Y. et al., 2016; Wu S.-Y. et 23 al., 2008; Backes et al., 2016; Pinder et al., 2007).

Response: Accepted. There are three new findings in our study. 1. During severe winter haze episodes, the particulate NO_3^- formation is NH_3-limited, resulting in its high sensitivity to NH_3 emission reductions. 2. Livestock NH_3 emission controls is a very efficient way to alleviate particulate NO_3^- pollution during severe winter hazes. 3. Improved manure management in livestock husbandry could effectively reduce total NH_3 emissions by 40% (from 100 kiloton to 60 kiloton) in winter of northern China, which would lead to a reduction of particulate NO_3^- by about 40% (averagely from 40.8 to 25.7 $\mu g/m^3$) during severe haze conditions. As you suggested, we reworded in the revised manuscript.

Revision: (Page 12, Line 381-388) "In this study, we found that during severe winter haze episodes, the particulate NO_3^- formation is NH_3-limited, resulting in its high sensitivity to NH_3 emission reductions. Meanwhile, livestock NH_3 emission controls is a very efficient way to alleviate particulate NO_3^- pollution during severe winter hazes. The estimations showed that the improvements in manure management of livestock husbandry could effectively reduce total NH_3 emissions by 40% (from 100 kiloton to 60 kiloton) in winter of northern China. It would lead to a reduction of particulate NO_3^- by about 40% (averagely from 40.8 to 25.7 μg/m^3) during severe haze conditions."

Referee: 2. Methodology.

The use of WRF model did not reproduce the temporal variations of inorganic aerosol components in this haze event (Figure S2 in the supporting information)*. As shown in Fig. S2, the correlation between the observations and simulations was relatively low, but the authors did not show this value deliberately. Due to such low accuracy of the WRF to simulate the inorganic aerosol components, how can the authors draw such strong conclusions based an unconvincing simulations? I suggest the authors validate their simulations using the observations, make some improvements of the simulation ability, and discuss the potential biases of the simulations; or alternatively, discuss the uncertainties of the simulation results in the discussions section. This is important because it's the fundamental base for your conclusions.*

Response: Accepted. As you suggested, we improved our model performance and added discussions of the simulation biases and their impacts in Section 2.2 and 3.3, respectively. Averagely, the observed and simulated NO_3^-, NH_4^+, SO_4^{2-} and TA are, respectively: (1) NO_3^-, 39.8 ± 14.7 μg/m^3 versus 39.1 ± 15.6 μg/m^3; (2) NH_4^+, 27.7 ± 8.6 μg/m^3 versus 26.5 ± 11.7 μg/m^3; (3) SO_4^{2-}, 42.4 ± 16.0 μg/m^3 versus 39.7 ± 20.8 μg/m^3 and (4) TA, 34.6 ± 8.5 μg/m^3 versus 32.1 ± 11.0 μg/m^3. The MB of these four species are -0.7, -1.2, -2.7 and -2.5 μg/m^3, respectively. Simulated particulate NO_3^-, NH_4^+, SO_4^{2-} and TA approximately agreed with the measurements.

In fact, we used 1-hr resolution measurements to compare with the simulations. The severe hazes often happened in stagnant conditions, in which the turbulent diffusion is weak and the winds almost keep calm. In this situation, it is very difficult for chemical transport models (like WRF-Chem) to describe the local atmospheric stability or diffusion processes very well (Steeneveld et al., 2006;Steeneveld, 2014). Moreover, the uncertainty in emissions could not be neglected. These factors make it difficult for chemical transport models to reproduce the temporal variations of inorganic aerosol components very well at hourly resolution (Li et al., 2016).

The simulation biases may affect the simulation of particulate NO_3^- reductions efficiency. Based on our results in Section 3.3, particulate NO_3^- reduction efficiency is determined by the availability of ambient NH_3 (represented as R in this study). Correspondingly, the influence of simulation biases on particulate NO_3^- reduction efficiency simulation mainly depends on the simulation biases of R. During the simulation case, the average simulated value of R is 1.3, which is equivalent to the observed value (1.3). Since WRF-Chem has a good estimation of the availability of ambient NH_3, its estimation of the efficiency of particulate NO_3^- reductions is reliable. Therefore, the conclusions drawn in Sect 3.2 are reliable.

Revision: (Page 5, Line 185-193) "The performance of WRF-Chem is evaluated by comparing measured and simulated NO_3^-, NH_4^+, SO_4^{2-} and TA. Specifically, the observed and simulated values are, respectively: (1) NO_3^-, 39.8 ± 14.7 μg/m^3 versus 39.1 ± 15.6 μg/m^3; (2) NH_4^+, 27.7 ± 8.6 μg/m^3 versus 26.5 ± 11.7 μg/m^3; (3) SO_4^{2-}, 42.4 ± 16.0 μg/m^3 versus 39.7 ± 20.8 μg/m^3 and (4) TA, 34.6 ± 8.5 μg/m^3 versus 32.1 ± 11.0 μg/m^3. The MB of these four species are -0.7, -1.2, -2.7 and -2.5 μg/m^3, respectively. Simulated particulate NO_3^-, NH_4^+, SO_4^{2-} and TA approximately agreed with the measurements (Figure S2). There are still some simulation biases that may affect the simulation of particulate NO_3^- reductions efficiency. This is discussed in detail in Sect 3.3."

(Page 12, Line 365-369) "Based on the above analysis, the influence of WRF-Chem simulation biases on particulate NO_3^- reduction efficiency simulation mainly depends on the simulation bias of R. During the simulation case, the average simulated value of R is 1.3, which is equivalent to the observed value (1.3). Since WRF-Chem has a good estimation of the availability of ambient NH_3, its estimation of the efficiency of particulate NO_3^- reductions is reliable."

Referee: 3. Form and structure.

There are well known heterogeneities in the NH_3 emission datasets that would need to be discussed in detail (refer to Zhang et al, 2018, Agricultural ammonia emissions in China reconciling bottom-up and top-down estimates. Atmospheric Chemistry and Physics, 18: 339-355).

Response: Accepted. As you suggested, we added more descriptions about the heterogeneities in the NH_3 emission datasets in Sect 2.2.

Revision: (Page 4, 127-141) "Another method for estimating NH_3 emissions is the inverse modeling method, which provides top-down emission estimates through optimizing comparisons of model simulations with measurements. For example, Paulot et al. (2014) used the adjoint of a global chemical transport model (GEOS-Chem) and data of NH_4^+ wet deposition fluxes to optimize NH_3 emissions estimation in China. Zhang et al. (2018a) applied TES satellite observations of NH_3 column concentration and GEOS-Chem to provide top-down constraints on NH_3 emissions in China. Their estimates are 10.2 Tg a^{-1} and 11.7 Tg a^{-1} respectively, which are close to our results (9.8 Tg a^{-1}) (Paulot et al., 2014;Zhang et al., 2018a). The accuracy of this method relies on many factors, such as the accuracy of initial conditions, the emission inventories, meteorological inputs, reaction rate constants, and deposition parameters in the chemical transport model. Errors of these parameters could cause biases in the top-down estimation of NH_3 emissions. In addition, measurements of NH_3 or NH_4^+ used in this method, including surface and satellite date, are usually sparse in spatial coverage and have uncertainties, which will also affect the estimation of NH_3 emissions."

In the authors' estimates, the livestock NH_3 emission is in general lower than 1.8 kg NH_3 ha-1 (180 kg NH_3 km^{-2}) (Fig. S3). It is such low livestock NH_3 emission in northern China in December. Is it right? And why such low livestock NH_3 emission have so big impact on particular matters? I wonder if the unit of NH_3 emission is kg NH_3 ha^{-1} $month^{-1}$?

Response: Yes, the correct unit is kg NH_3 km^{-2} $month^{-1}$. Figure S4 has been revised.

The authors had good measurements dataset of the inorganic aerosol components during in December 2015 and December 2016. Unfortunately, it is very surprising that the authors made a conclusion based the simulation data rather than their measurements. If the authors want to make a strong conclusion that livestock ammonia emission controls on alleviating particulate nitrate during a severe winter haze, they should first show what they has gained from the two time periods of December 2015 and December 2016 ***regarding the measurements of inorganic aerosol components as well as their estimates of livestock NH_3 emissions****? Again, the simulation results are unacceptable for inorganic aerosol components from the two time periods of December 2015 and December 2016. The conclusion should be based on their measurements work. At least, their simulations should be finely validated with their observations.*

Response: Firstly, in fact, our conclusions are mainly based on measurements. In the ISORROPIA-II simulation, the input data are all the observation data and we show the comparison between observed and simulated particulate NO_3^- after TA reductions. In addition, the analysis of the availability of ambient NH_3 in Section 3.3 is also based entirely on observations. In the WRF-Chem simulation, because we needed to show the particulate NO_3^- reductions regionally, we calculated the change of simulated value of particulate NO_3^- before and after NH_3 emission reductions.

Secondly, our observations and NH_3 emission inventory have been described in detail in section 2.1 and 2.2. The importance of particulate NO_3^- in SNA and the dominant role of livestock in NH_3 emissions are pointed out. Furthermore, from lines 249 to 259, we made a conclusion that the richness of NH_3 leads to the stability of NH_4NO_3 in the atmosphere by calculating the NH_3-HNO_3 partial pressure production (Kp) and analyzing the phase state and composition of pollutants. This conclusion directly linked high NH_3 emissions to high particulate NO_3^- concentrations, which is also based entirely on observations.

Thirdly, as you suggested, we discussed the simulation biases and their impacts in Section 2.2 and 3.3, respectively. See the previous reply for details.

Specific comments:

Introduction

1. *line 66-71 these review introductions are very lacking, and numerous studies on this topic have been ignored by the authors, which I have given several of them above. It is impossible for the reader to judge what the merits are of the current paper without ploughing through the recent literature, which as pointed out before is not properly reviewed.*

Response: Accepted. As you suggested, we added more review introductions to highlight the importance and innovation of our research.

Revision: (Page 2-3, Line 48-88) "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 and SIA (secondary inorganic aerosol) accounting for more than 50% of $PM_{2.5}$ (Zheng et al., 2016;Tan et al., 2018). To mitigate fine particle pollution, the Chinese government has been taking strong measures to control SO_2 emissions (http://www.gov.cn/zwgk/2011-12/20/content_2024895.htm). Since 2007, SO_2 emissions have been reduced by 75% in China (Li et al., 2017). Consequently, the particulate sulfate concentration have also been declining continuously in the past decade (Geng et al., 2017).

Although NO_x emissions in 48 Chinese cities decreased by 21% from 2011 to 2015 (Liu et al., 2017a), unfortunately, no obvious decreasing trend for particulate NO_3^- had been observed in northern China during recent years (Zhang et al., 2015). In October 2015, a severe haze episode was reported in North China Plain (NCP), with the hourly peak concentration of particulate NO_3^- exceeding 70 μg/m^3 (Zhang et al., 2018b). Even in November 2018, during a heavy haze episode in northern China, the hourly peak concentration of $PM_{2.5}$ still exceeded 289 μg/m^3, of which particulate NO_3^- accounted for 30% (http://www.mee.gov.cn/xxgk2018/xxgk/xxgk15/201811/t20181116_674022.html).

Another way to alleviate the particulate NO_3^- pollution is to control NH_3 emissions. Previous studies were performed to demonstrate the necessity of NH_3 emissions abatement in reducing $PM_{2.5}$ concentrations in the United States (Pinder et al., 2007;Tsimpidi et al., 2007;Pinder et al., 2008;Wu et al., 2016) and Europe (de Meij et al., 2009;Bessagnet et al., 2014;Backes et al., 2016). Recently, a feature article pointed out that NH_3 could be key to limiting particulate pollution (Plautz, 2018). In contrast with low particulate matter pollution levels in the United States and Europe, what we are facing in northern China is the extremely high particulate NO_3^- pollution especially happened in severe winter haze events.

Although Fu et al. (2017) proposed that the NH_3 emission controls are urgently required in China, the effectiveness of NH_3 emissions mitigation to alleviate the particulate NO_3^- peaks during severe winter haze episodes was seldom reported. Only Guo et al. (2018b) used a thermodynamic model to estimate the sensitivity of particulate NO_3^- to TA (sum of ammonia and ammonium) during one winter haze episode in Beijing. In their study, the atmospheric chemistry simulations based on NH_3 emission controls scenario were lacking to demonstrate the regional effects.

To alleviate severe particulate NO_3^- pollution in northern China is urgent, the study on the effectiveness by NH_3 emission controls is necessary. 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 effectiveness of NH_3 emission reductions on alleviating particulate NO_3^- during a severe haze episode. The molar ratio based on observations is used to explore the efficiency of particulate NO_3^- reductions during the severe haze conditions in wintertime."

Methods

1. *Line 83: the authors said the measurements were conducted in December 2015 and December 2016. Why are the results of December 2016 not shown in the paper, and why the validation was only performed in December 2015 (Fig. S2)?*

Response: In section 3.3, the analysis of the molar ratio (R) have included all observations of December 2015 and 2016. Figure S2 shows the validation of the WRF-Chem simulation during the haze episode (from 6 to 10, December 2015), since WRF-Chem does not simulate other periods.

2. *Line 86: HCl (rather than HCI).*

Response: Accepted. Revised at line 97.

3. *Line 96-110: The validation of the livestock NH_3 emission products should be described in detail.*

Response: Accepted. As you suggested, we added more descriptions about the validation of the livestock NH_3 emission products in Section 2.2.

Revision: (Page 4, 120-141) "In the past few years, our inventory has been compared with many studies to prove its reliability. For example, the spatial pattern of NH_3 emissions calculated in our inventory agreed well with the distribution of the NH_3 column concentrations in eastern Asia retrieved from the satellite measurements of Infrared Atmospheric Sounding Interferometer (IASI) (Van Damme et al., 2014). Specially, our estimation of livestock NH_3 emissions in China is comparable to the results of Streets et al. (2003) and Ohara et al. (2007).

Another method for estimating NH_3 emissions is the inverse modeling method, which provides top-down emission estimates through optimizing comparisons of model simulations with measurements. For example, Paulot et al. (2014) used the adjoint of a global chemical transport model (GEOS-Chem) and data of NH_4^+ wet deposition fluxes to optimize NH_3 emissions estimation in China. Zhang et al. (2018a) applied TES satellite observations of NH_3 column concentration and GEOS-Chem to provide top-down constraints on NH_3 emissions in China. Their estimates are 10.2 Tg a^{-1} and 11.7 Tg a^{-1} respectively, which are close to our results (9.8 Tg a^{-1}) (Paulot et al., 2014;Zhang et al., 2018a)."

Results

1. *Line 61: "On the one hand, the proportion of intensive livestock husbandry in China is only about 40%, far lower than that of developed countries". What's the proportion of intensive livestock husbandry in developed countries (90% or 100%)? At least, a reference should be given here.*

Response: Accepted. Related reference has been added.

Revision: (Page 5, 200-202) "… is only about 40%, far lower than that of developed countries (Harun and Ogneva-Himmelberger., 2013). As a result, the widespread free-range and grazing animal rearing …"

2. *Lines 165-170: these statements are very biased since their study timespan concerned the winter time (December), while the N application commonly occurred in spring or summer. The authors should focus on the timespan of their study, and avoid overstatements of their findings.*

Response: We agree with this comment. The studies quoted here are to show the backwardness of current livestock management in China. For winter, the emission reduction measures mainly focus on in-house handling and storage, since land application mainly occurs in spring and summer. To avoid ambiguity, we deleted this sentence.

Revision: (Page 6, Line 207-209) "… 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).~~"

3. Lines 171-197: Again these statements are overstated. Actually, the authors just make a very subjective reduction in livestock NH_3 emissions, and then drive the WRF model using the reduced livestock NH_3 emission.

Response: We cited more articles about exploring livestock NH_3 emission controls in in-house handling and storage during winter. These studies show that even under low temperature conditions in winter, the NH_3 emission reduction measures in in-house handling and storage are still very effective. Therefore, the proportions of NH_3 emission reductions used in our NH_3 emission inventory are reasonable. In addition, we removed the proportion of NH_3 emission reductions in land application due to the lack of appropriate references. In fact, in our NH_3 emission inventory, the NH_3 emissions from manure land application only account for 5% of the NH_3 emissions from livestock in winter. Therefore, the removal of this part of emission reductions has little effect on the overall emission reduction ratio (Total NH_3 emission reductions can still reach 40%). The changes are as follows:

Revision: (Page 6, Line 213-215) "phases: in-house handling, storage and land application (Chadwick et al., 2011). For winter, the emission reduction measures mainly focus on in-house handling and storage, since land application mainly occurs in spring and summer."

(Page 6, Line 220-223) "emissions by about 50%-70% (Balsari. et al., 2006;Petersen et al., 2013;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).~~"

(Page 6, Line 231-233) "emission reductions mentioned above were multiplied by NH_3 emission factors in two phases of manure management: 50% reduction at in-house handling and 60% (the average value of 50% and 70%) reduction at storage. With these measures…"

4. *Lines 199-200: In the ISORROPIA-II simulation, 40% reduction of TA was used to reflect the effects of reducing NH_3 emissions by 40%. This process is also very subjective and has no explanation at all why the authors adopted this value. At least the author should give reference to support this process. In fact, there are numerous subjective descriptions in the main text, and it's hard to specify all of them and prove them validate.*

Response: Accepted. As you suggested, we cited some relevant studies that used this method. We also used WRF-Chem to examine this method. Results showed that there was little difference between NH_3 emission reductions and TA reductions (40% versus 40.7%). ISORROPIA-II is a box model, which calculates the thermodynamic equilibrium between aerosol phase and gas phase. It will redistribute NH_4^+ and NH_3 into aerosol phase and gas phase when TA changes. In fact, chemical transport models (e.g., WRF-Chem) also have a similar thermodynamic equilibrium calculation process when NH_3 emissions decreases. We added following sentences to Section 3.2.

Revision: (Page 6, Line 242-245) "This approach has been used in many previous studies (Blanchard and Hidy, 2003; Vayenas et al., 2005). However, in the real atmosphere, the reductions of NH_3 emission are not always equal to the reductions of TA due to the regional transmission. Their differences are discussed in the WRF-Chem simulation."

(Page 8, Line 298-301) "Correspondingly, TA decreased by 40.7% (from 17.2 μg/m^3 to 10.2 μg/m^3), very close to the reductions of NH_3 emission (40%). This indicates that it is reasonable to use TA reductions to represent NH_3 emission reductions in the ISORROPIA-II simulation."

Discussions

1. *Lines 319-336: All these were already shown in results part, but were again repeated in the discussions. I suggest the authors re-organize the discussions sector in order to summarize their results completely, also for better comparison to some latest references.*

Response: Accepted. We re-organized the discussions sector as you suggested.

Revision: (Page 11, Line 381-396) "In this study, we found that during severe winter haze episodes, the particulate NO_3^- formation is NH_3-limited, resulting in its high sensitivity to NH_3 emission reductions. Meanwhile, livestock NH_3 emission controls is a very efficient way to alleviate particulate NO_3^- pollution during severe winter hazes. The estimations showed that the improvements in manure management of livestock husbandry could effectively reduce total NH_3 emissions by 40% (from 100 kiloton to 60 kiloton) in winter of northern China. It would lead to a reduction of particulate NO_3^- by about 40% (averagely from 40.8 to 25.7 μg/m^3) during severe haze conditions.

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) (http://www.gov.cn/zhengce/content/2017-01/05/content_5156789.htm). Due to the dominance of free-range animal rearing systems and the lack of emission controls policies, livestock NH_3 emission reductions in China could be practicable. In order to control $PM_{2.5}$ pollution more effectively in northern China, measures to improve manure management in livestock urgently need to be implemented."

References

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