Review of the manuscript by Xu et al

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

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. The most important finding is that 40% of ammonia emission mitigation could lead to almost the same reduction in particulate nitrate in the North China Plain in winter season. 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.

Major comments

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. PM2.5) pollution (see Wu Y. et al., 2016; Wu S.-Y. et al., 2008; Backes et al., 2016; Pinder et al., 2007).

Refs mentioned:

Y. Wu, B. Gu, J. W. Erisman, S. Reis, Y. Fang, X. Lu, X. Zhang, PM2.5 pollution is substantially affected by ammonia emissions in China. Environmental Pollution 218, 86-94 (2016).

S.-Y. Wu, J.-L. Hu, Y. Zhang, V. P. Aneja, Modeling atmospheric transport and fate of ammonia in North Carolina—Part II: Effect of ammonia emissions on fine particulate matter formation. Atmospheric Environment 42, 3437-3451 (2008).

A. M. Backes, A. Aulinger, J. Bieser, V. Matthias, M. Quante, Ammonia emissions in Europe, part II: How ammonia emission abatement strategies affect secondary aerosols. Atmospheric Environment 126, 153-161 (2016).

R. W. Pinder, P. J. Adams, S. N. Pandis, Ammonia Emission Controls as a Cost-Effective Strategy for Reducing Atmospheric Particulate Matter in the Eastern United States. Environmental Science & Technology 41, 380-386 (2007).

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.

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). 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} $month^{-1}$?

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.

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.

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)?.
2. Line 86: HCl (rather than HCI).
3. Line 96-110: The validation of the livestock NH_3 emission products should be described in detail.

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.

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.

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.

4. Lines 199-200: In the ISORROPIA-II simulation, 40% reduction of TA was used to reflect the effects of reducing NH3 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.

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.