Response to Referee #1

General comments (overall quality):

The paper addresses an important issue relating to atmospheric pollution with ammonia, namely the interaction with SO_2 emissions. The material is highly relevant to the subject matter covered by the journal and the results represent a useful contribution to knowledge concerning the interaction between tropospheric ammonia and SO_2 emissions. The level of English in the manuscript is fulfilling and the length of the submission seems appropriate. The recommendation is for publication of the paper.

Response: We would like to thank the referee for the encouragements and providing the insightful suggestions, which indeed help us to improve the manuscript.

Specific comments (individual scientific questions/issues):

The manuscript states that intensive farming results in lower volatilization rates of NH_3. This may be true, but I associate intensive farming with an increased number of livestock, which has the opposite effect, i.e. more livestock increases ammonia emissions. Already in the abstract it would be interesting to get an indication of how the number of livestock has changed over time, as this is an important factor when it comes to emissions of ammonia. It would be interesting to get an indication of the increase (in percentage) on page 5, row 5, that states: The number of some livestock increased slightly.

Response: Accepted. There are an increased number of livestock animals during 2008−2016. We have shown the detailed information in Table S1. As suggested by the referee, we added the quantitative data in the revised manuscript.

The intensive systems are characterized by lower NH_3 emission factors than the free-range (traditional animal rearing system in rural area) (Huang et al., 2012; Kang et al., 2016; EEA). The increased number of animals will produce more manure in farms, but applying intensive farming in livestock industry lowers the volatilization rates of NH_3 per animal. In this work, we find that the transition from the free-range to the intensive farming in the Chinese livestock industry offset the effect of increased animals on the NH_3 emissions. The resulting livestock NH_3 emissions in northern China do not show a significant trend during this period.

Revision: (Page 5, Line 23-30) "On the other hand, the number of some major livestock increased (Beef −20%, Dairy +39%, Goat −23%, sheep +55%, Pig +18%, and Poultry +19%; see Table S1 for details), while the proportion of intensive animal rearing systems rises to nearly half of the

livestock industry in 2016, compared to only 28% in 2008 (Table S1). The intensive systems are characterized with more effective livestock manure management in favor of lower volatilization rates of NH_3 (Kang et al., 2016). The transition from the free-range to the intensive in livestock animal rearing offset the effect of increased animals on the NH_3 emissions, thereby resulting in the annual livestock emissions in the North China Plain almost constant (around 1.2 Tg yr^{-1}). Overall, the decreasing NH_3 emissions cannot track the upward trend of tropospheric NH_3 concentrations."

In the background or discussion, it would be interesting to read more about other similar studies (outside of China), relating to the result in this study, e.g. Aneja et al (2003), Agricultural ammonia emissions and ammonium concentrations associated with aerosols and precipitation in the southeast United States, or Ferm & Hellsten (2012), Trends in atmospheric ammonia and particulate ammonium concentrations in Sweden and its causes.

Response: Accepted. We cited those related papers in the background and discussions parts in the revised manuscript.

Revision: (Page 3, Line 1-3) "Several studies have proposed that reduction in SO_2 emissions or NO_x emissions is an important factor in determining the increase in atmospheric NH_3 concentrations on the global and region scales (Warner et al., 2017; Yu et al., 2018; Saylor et al., 2014)."

(Page 10, Line 12-23) "Interestingly, increasing trends of gas-phase NH_3 in the atmosphere have also been observed in the last twenty years in the Midwest of the United States and Western Europe by satellite retrievals and ground measurements (Saylor et al., 2015; Warner et al., 2017; Ferm and Hellsten, 2012). The marked decreases in SO_2 and NO_x emissions were largely responsible for these increases, as confirmed by the corresponding trends of particulate sulfate and nitrate concentrations. Warner et al. (2017) infer that SO_2 emission reduction in China may be a leading cause of the increased NH_3. More recently, Yu et al. (2018) quantified the contributions of the acid gases on the trends of NH_3, and found that emissions of SO_2 contributed to 2/3 and NO_x to 1/3 of the change in NH_3 over the United States from 2001 to 2016. In this work, we demonstrate that the rapid reduction in SO_2 emissions was responsible for the increase in NH_3 over the North China Plain during 2008−2016, while other potential pathways (NH_3 emissions, NO_x emissions, and meteorological conditions) decreased its concentrations by approximately 13% for this period."

The authors state that "the increase in ammonia concentrations was highest in summer". However I lack some reasoning regarding seasonal variations in ammonia emissions (e.g. more fertilization of the fields, and higher temperatures in

summertime). It would be useful also to mention this in the discussion and its implications on the result.

Response: The NH_3 emissions in the North China Plain are concentrated in spring and summertime due to frequent fertilization activities and higher temperature, which facilitates the volatilization rates of NH_3. We added the description about the seasonal distribution of NH_3 emissions in the North China Plain in the Methods of the revised manuscript.

However, the main finding of our study is that the rapid reduction in SO_2 emissions in China strongly reduced the concentrations of ammonium sulfate aerosols and transfers NH_3 from particle to gas phases. Therefore, the seasonal feature of the increase in gaseous NH_3 concentrations was mainly determined by that of sulfate concentrations rather than the NH_3 emissions. Both observations and simulation indicate that the concentrations of particulate sulfate demonstrated highest decreases in summer, thereby causing high increase in NH_3.

Revision: (Page 4, Line 28-30; Page 5, Line 1-3) "It shows distinct seasonal feature in NH_3 emissions over the North China Plain. There are 75% of annual NH_3 emissions released in spring and summer months (March-September), during which intensive agricultural fertilization and elevated ambient temperature facilitate the volatilization rates of NH_3. In this study, to integrate our NH_3 inventory into WRF-Chem simulations, we adopted a diurnal profile with 80% of the NH_3 emissions in the daytime, following previous studies (Zhu et al., 2015; Paulot e al., 2016; Asman, 2001)."

(Page 9, Line 1-4) "The seasonal variations in SO_4^{2-} decreases and NH_3 increases were consistent (Fig. 6). We can see that the reduction of sulfate column concentrations between the Run_08 and Run_16 reached 1.3 × 10^{15} molecules/cm^2 in summer (JJA), which was about three times larger than in other seasons. The corresponding percent reductions ranged from 15% in DJF to 36% in JJA. As aforementioned, the long-term observations of $PM_{2.5}$ in Beijing also confirmed the highest decrease of sulfate in summer."

Technical corrections

Page Row 1 24 Remove "s" in "increases"

Response: Accepted. We remove it.

Revisions: (Page 1, Line 27) "we demonstrate that this large SO_2 emission reduction is responsible for the NH_3 increase"

2 14 Consider changing "NH3 emission has" to "NH3 emissions have"

Response: Accepted. We change it.

Revisions: (Page 2, Line 17-18) "Until now, NH_3 emissions have not been regulated by the Chinese government, although they serve as an important contributor to haze pollution in China."

3 2 Change "2000" to either "year 2000" or only "2000"

Response: Accepted. We change it.

Revisions: (Page 3, Line 3-4) "Through the widespread use of the flue gas desulfurization in power plants since 2006 in China, SO_2 emissions have gradually decreased."

4 13 Change "were" to "was"

Response: Accepted. We change it.

Revisions: (Page 4, Line 20) "A high-resolution NH_3 emission inventory (1km ×1km, month) was developed based on the bottom-up method."

5 5 Remove "animals" 5 6 Change "system" to "systems" 5 7 Change "The increased livestock animals raised but more effective...." to "Despite increased livestock numbers, more effective...."

Response: Accepted. We reword these statements.

Revisions: (Page 5, Line 23-30) "On the other hand, the number of some major livestock increased (Beef −20%, Dairy +39%, Goat −23%, sheep +55%, Pig +18%, and Poultry +19%; see Table S1 for details), while the proportion of intensive animal rearing systems rises to nearly half of the livestock industry in 2016, compared to only 28% in 2008 (Table S1). The intensive systems are characterized with more effective livestock manure management in favor of lower volatilization rates of NH_3 (Kang et al., 2016). The transition from the free-range to the intensive in livestock animal rearing offset the effect of increased animals on the NH_3 emissions, consequently resulting in the annual livestock emissions in the North China Plain being almost constant (around 1.2 Tg per year)."

6 17 Change "hotpot" till "hotspot", and change "had" to "have" 6 18 Consider changing "over" to "in" and "into the atmosphere". We noted...."

Response: Accepted. We reword the statement.

Revisions: (Page 6, Line 21-23) "Spatially, the hotspot of NH_3 was mainly concentrated in Hebei, Shandong and Henan provinces, which have the most intensive agricultural productions in China and thus emit considerable gas-phase NH_3 into the atmosphere."

6 20 Consider adding "s" to emission, i.e. "emissions"

Response: Accepted. We reword it.

Revisions: (Page 6, Line 25) "Recently, NH_3 emissions from the residential coal and biomass combustion for heating are considered to be a potentially important source of NH_3 in suburban and rural areas during wintertime"

6 21 Consider changing to "it has not been fully included...."

Response: Accepted. We change it.

Revisions: (Page 6, Line 27) "it has not been fully included in our bottom-up inventory."

7 16 Change "disappeard"

Response: Accepted. We reword the statement.

Revisions: (Page 7, Line 18-19) "It was noticeable that under these conditions, the increasing trend of NH_3 column concentrations was not observed."

Figures Not consistent when it comes to the units, sometimes writing "µg/m3" and sometimes "_g m-3", please consistently use the latter.

Response: Accepted. We use the unit of µg m^{-3} in the whole manuscript.

Revisions:

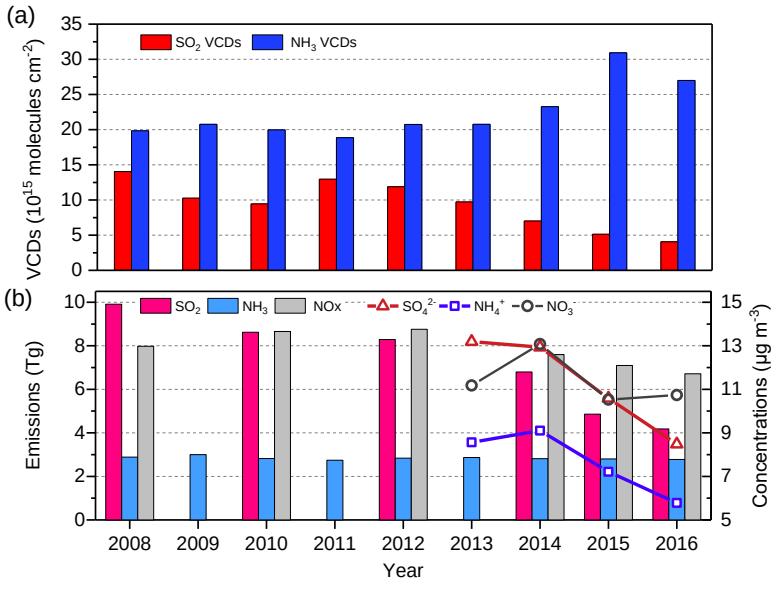

Figure 1. (a) Inter-annual trends of SO_2 and NH_3 VCDs averaged over North China Plain from 2008 to 2016. (b) Inter-annual trends of emissions of SO_2 NH_3, and NO_x in the North China Plain from 2008 to 2016, and annual mean concentrations of $PM_{2.5}$ sulfate, ammonium, and nitrate derived from measurements at an urban station (Beijing, 39.99 °N,

116.3 °E) in North China Plain from 2013 to 2016.

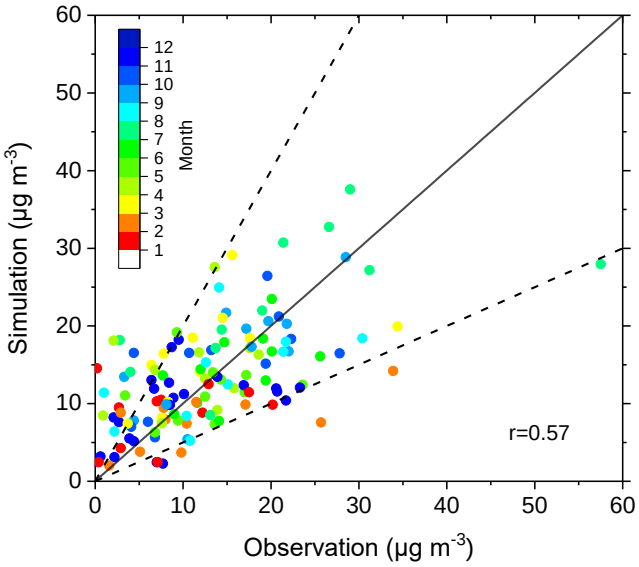

Figure 2. Comparison of modelled gaseous NH_3 concentrations with corresponding monthly measurements of NH_3 from September 2015 to August 2016. The 1:2 and 2:1 dashed lines are shown for reference and the Pearson correlation coefficient (r) is shown inset

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