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Interactive comment on "Numerical analysis of the impact of agricultural emissions on $PM_{2.5}$ in China using a high-resolution ammonia emissions inventory" *by* Xiao Han et al.

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China is one of the largest agricultural countries in the world. The NH3 emissions from agricultural activities in China, such as fertilizer and husbandry, farmland ecosystems, livestock waste, crop residue burning and fuel wood combustion, significantly affect regional air quality and horizontal visibility by contribution to secondary inorganic aerosols. In the manuscript, the air quality modeling system RAMS-CMAQ (regional atmospheric modeling system-community multiscale air quality), coupled with the ISAM (integrated source apportionment method) module is applied to capture the contribution of NH3 emitted from total agriculture (Tagr) in China. It explores that the annual

average contribution of Tagr NH3 to PM2.5 mass burden in China was 14-18%. Specific to the PM2.5 components, Tagr NH3 provided a major contribution to ammonium formation (87.6%) but a tiny contribution to sulfate (2.2%). Though the Tagr NH3 only contributed 10.1% of nitrate under current emissions scenarios, the reduction of nitrate could reach 98.8% upon removal of the Tagr NH3 emissions. The results are meaningful, but the explanation for these phenomenon was not enough. I recommend the manuscript to be accepted after some minor revisions, and detail some issues below. Major points:

1. The most important gas in this manuscript was NH3, but there are no NH3 in Figure 2 in comparing between the modeled and observed results.

R: Thanks for this comment. However, NH3 is not included in the conventional observation species in China at present. Therefore, it is hard to collect available observation data of NH3 mass concentration for model evaluation directly. Most of the available information was derived from the published research paper. In Han et al. (2017; Modeling dry deposition of reactive nitrogen in China with RAMS-CMAQ. Atmos. Environ.), the simulated NH3 by RAMS-CMAQ has been compared with the observations from many studies in detail, including the multi-year observation results with Nationwide Nitrogen Deposition Monitoring Network and the seasonal variation characteristics from Pan et al. (2012; Wet and dry deposition of atmospheric nitrogen at ten sites in Northern China. Atmos. Chem. Phys.). In this paper, we also compare the simulation results with the value and seasonal variation at several stations from Pan et al. (2018) and Zhang et al. (2018) (Line 200-211). We kindly hope these content could reflect the reasonability of modeled NH3.

2. Why is the NH3 contribution to nitrate small under "rich NH3" conditions and large in "poor NH3" environments? What is the internal logical relationship?

R: Thanks for this comment. In fact, the detail discuss about "rich NH3" and "poor NH3" can be found in Wang et al. (2011; Impact Assessment of Ammonia Emissions on

Inorganic Aerosols in East China Using Response Surface Modeling Technique). The results of RSM (Response Surface Modeling) in their study shows that the change of NO3- mass concentration is very sensitive to the emission level of NH4+ and performs as nonlinear relationship. The reduction of NH3 emissions can play a significant role in reducing the mass concentration of NO3- under NH3-poor condition. However, there will be excess NH3 in the atmosphere under NH3-rich condition, and these excess NH3 could neutralizes more nitric acid even in the case of emission reduction. Thus, the effect of emission reduction is not significant under NH3-rich condition. In addition, the SO2 will compete for NH3 and prevent the generation of NH4NO3 under NH3-poor condition because the reaction between H2SO4 and NH3 takes precedence over the one between HNO3 and NH3. Oppositely, SO2 should be benefit for the formation of NO3- (especially in summer) under NH3-rich condition according to the calculation of Wang et al. (2011). This should be a reason why the effect of NH3 emission control is not obvious in the case of NH3-rich condition as well.

3. The study period is January, April, July, and October, but only the modeled and observed results in January and July are compared in Figure A1, A2, A3 and A4.

R: Thanks for this comment. We added the comparison of meteorological factors in April and October. Please check if it is appropriate.

4. The author thinks that the obvious deviation between the observed and modeled SO2 in January may be a systemic underestimation due to the lack of emission intensity in this month. Did the lack of emission intensity only appear in SO2? Why are SO2 and NO2 underestimated and PM2.5 overestimated?

R: Thanks for this comment. The monthly mean observation data were used in the submitted version. However, we would like to provide more details about the evaluation. Thus, the hourly observation data from the China National Environmental Monitoring Centre were collected and compared with simulation results. The scatter plots (Figure 2) were replaced and the comparison of SO2, NO2 and PM2.5 in January, April,

July, and October at six sites were presented, and the statistical summary of the comparisons and related discussion were modified (Line 186-198). Please check if it is appropriate.

5. How much NH3 is removed in Figure 7? And it's more intuitive to use a negative value for reduction.

R: Thanks for this comment. Here the emission of NH3 from all agricultural sources were removed. For detail information, please see the percentage shown in Figure A6 which we added. In addition, the horizontal distribution of the PKU-NH3 emission inventory can be viewed in Kang et al. (2016) (Kang et al., 2016: High-resolution ammonia emissions, High-resolution, ammonia 1980, 2012.). On the other hand, the Figure 7 was modified. Please check if it is appropriate.

6. Why do the trend of the decrease in ammonium mass concentration accelerate while NH3 emissions is less than 20%?

R: Thanks for this comment. Here the simulation scenario was conducted for each emission reduction of 10% so that the acceleration should appear between 20% and 30%. In fact, it can be found that the accelerated decline mainly started when the emission reduction exceeds 50%. Therefore, we could deduce that the accelerated decline should be emerged gradually with NH3 emission reduction. This feature indicates that the formation of NH4+ should be nonlinear with NH3 emission intensity as well. The reason may also be related to the complex neutralization reaction among sulfate, nitrate and ammonium. The consumption of NH3 should become more sufficient when the mass concentration of NH3 is lower. Thus, the variation of ammonium is more sensitive under low NH3 mass burden.

7. What is the horizontal distributions of the contribution percentage of NH3 emissions to ammonium, nitrate and sulfate mass concentration, respectively? Which aerosol determines the horizontal distributions of SNA mass concentration? Why is the horizontal distributions of NH3 emissions different with the horizontal distributions of the

contribution percentage of NH3 emissions to SNA mass concentration?

R: Thanks for this comment. The horizontal distributions of NH3 emission contribution to sulfate, nitrate and ammonium is shown in Figure R1, and ammonium provided the major contribution to SNA (Table 4 also presented related information). In addition, Figure 6 shows the horizontal distributions of contribution percentage which may not follow the distribution pattern of mass concentration. For example, it can be seen that the agricultural NH3 emission generally provided more than 90% contribution to ammonium over China in January as shown in Figure R1. Therefore, the contribution ratio should differ from the horizontal distribution pattern.

Minor points: 1. In Figure 6 and Figure 7, it should be the horizontal distributions in January, April, July, and October. 2. In Line 226, it should be “Since NH3 concerns mainly with secondary inorganic aerosols (SNA): sulfate, nitrate, and ammonium formation”. 3. In line 269, what is “TA NH3 emission”? 4. In Line 833, should is it “The regional percent (%) of Tagr NH3 contribution”?

R: Thanks for the comments. All error points were modified.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2019-1128/acp-2019-1128-AC1-supplement.pdf

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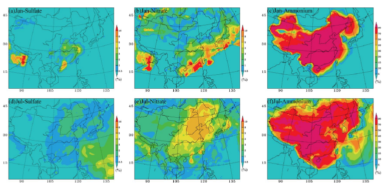

Figure R1 The horizontal distributions of the contribution percentage of NH_3 emissions to sulfate, nitrate and ammonium mass burden (%) in January and July.

Fig. 1. Figure R1 The horizontal distributions of the contribution percentage of NH3 emissions to sulfate, nitrate and ammonium mass burden (%) in January and July.