General comments:

This study presents an optimized ammonia emission inventory with a focus on fertilizer application relevant sources over mainland China in 2016. The approach adopted in this study considers the time variability of fertilizer application and thus enables this inventory accurate capture the ammonia emissions over time. To illustrate the capability of this emission inventory against existing emission inventory (MEIC developed by Tsinghua University), the authors evaluate the emission inventory by comparing modelled NH3 concentrations by WRF-Chem with ammonia inventory in this work and MEIC. Further, the model performance is validated with NH3 observations from AMoN-China and IASI NH3 column. This paper is well-written and presents results that would be interesting to the air quality modeling community. However, I have several concerns that the authors should consider when revising the manuscript, as listed below. I recommend this work to be published after the following comments are adequately addressed.

Response: We sincerely thank the reviewer for providing valuable and constructive comments. The detailed responses are listed as follows.

Major comments:

1) While the authors illustrated that large uncertainties remain in the NH3 emissions from agriculture sector thus motivates the improvement in NH3 emission inventory, I think vehicular NH3 emissions co-emitted with nitrogen oxides (NOx) are also important sources for NH3. This additional NH3 source has been always underestimated in emission inventory (such as MEIC). Although this work is focused on agriculture ammonia emissions, I would suggest the authors briefly discuss this in the Introduction because this work is entitled "ammonia emission inventory in China".

Response: Thanks for your suggestion. we added some sentences to state the importance of NH_3 emissions from the transportation sectors in the Introduction.

"Of course, certain non-agricultural sources of NH_3 are also important. For example, vehicular emissions contain both NH_3 and NO_x and may have a more effective pathway to particle formation, particularly in urban areas (Farren et al., 2020; Chang et al., 2016). The amount of NH_3 emitted by the transportation sector has not been well quantified, and is generally thought to be underestimated (Meng et al., 2017; Farren et al., 2020). "

2) In Sec. 2.4, the uncertainty assessment of NH3 emission inventory established in this work based on the Monte Carlo method is missing. Please provide the uncertainties for each sub-sector if possible.

Response: We are grateful for this critical comment. We added one sub-section (Section 3.4.1) to discuss the uncertainty of NH_3 emission based on the Monte Carlo simulation.

"3.4.1 Uncertainty

Uncertainty in the estimated NH_3 emissions results from both the activity level and EF input data. We ran 10000 Monte Carlo simulations to estimate the range of NH_3 emissions from each source with a 95% confidence interval. The estimated total NH_3 emission range was 10.5-16.0 Tg. The 95% confidence intervals of fertilizer application, livestock waste, and others ranged from -20.5% to

64.41%, -23.0% to 37.1% and -42.9% to 62.4% (Fig. 6). Due to the large amounts of NH_3 emitted by fertilizers and livestock waste, the uncertainty of total NH_3 emissions is mainly caused by the uncertainties of these two sources. The uncertainty of fertilizer application was slightly greater than that of livestock waste. The emission factors, especially the corrected EF, were the largest contributors to the uncertainties of fertilizer application emissions. Additionally, it is clear that NH_3 emissions from other sources exhibited the largest uncertainty (-42.9% to 62.4%), mainly due to the high degree of uncertainty resulting from the many sub-sources, such as −77.1% ~ 96.9% of the transportation sector and −79.4% ~ 122.7% of the industrial sector. In comparison, the emissions from other sources were relatively small; hence, the large uncertainties of other sources did not have a significant impact on the uncertainty of total NH_3 emissions.

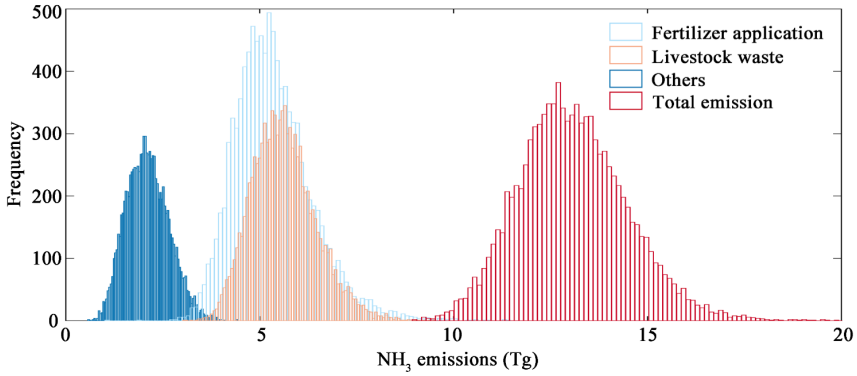

Fig. 6. Uncertainties of NH_3 emissions sourced from fertilizer application, livestock waste, and others."

3) The authors employed the WRF-Chem model for performing numerical simulations over eastern China and compared with model outputs driven by MEIC and IASI satellites. However, I could not find the detailed configurations of WRF-Chem simulations (initial and boundary conditions for WRF-Chem, physical parameterizations, chemical mechanisms, etc). I suggest the authors add a section in the supplementary providing the configurations of WRF-Chem.

Response: We sincerely thank the reviewer for this suggestion. We added one section (Section S1) to state the configurations of WRF-Chem model in the supplementary materials.

"**Section S1 WRF-Chem model configuration**

A fully coupled online Weather Research and Forecasting with Chemistry model (WRF-Chem v3.7) is used to evaluate the accuracy of different NH_3 emission inventories. The WRF-Chem model is designed to cover most parts of North, East, Central, and South China at the horizontal resolutions of 27 km (Fig.2). The vertical dimension is resolved by 46 full sigma levels, with 18 layers located in the bottom 2 km for finer resolution in the planetary boundary layer; the height of the first layer averaged over the analyzed region is about 30 m.

Meteorological initial and lateral boundary conditions used in the WRF-Chem model are taken from the NCEP (National Center for Environmental Prediction) (Final) Operational Global Analysis data with a spatial resolution of 1° × 1°. The forecasts from the MOZART-4 global chemical transport model are processed to provide the chemical initial and boundary conditions for the WRF-Chem model (Emmons et al., 2010).

Air pollutants emissions (including SO_2, NO_x, CO, CO_2, NMVOC, BC, OC, $PM_{2.5}$, and PM_{10})

of 2016 were obtained from Multi-resolution Emission Inventory for China (MEIC) (http://meicmodel.org/), with the horizontal resolution of 0.25° (Li et al., 2017). The emission rate of each species for each hour is based on Gao et al. (2015). The biogenic emissions are calculated online using the MEGANv2.04 (Model of Emission of Gases and Aerosol from Nature v2.04) model (Guenther et al., 2006). Biomass-burning emissions are obtained from the GFEDv3 (Global Fire Emissions Database v3) (Randerson et al., 2005). Dust emissions and sea salt emissions are calculated online using algorithms proposed by Shao (2004) and Gong et al. (1997), respectively.

The Carbon Bond Mechanism Z (CBM-Z) is selected as the gas-phase chemical mechanism (Zaveri and Peters, 1999), and the full 8-bin MOSAIC (Model for Simulating Aerosol Interactions and Chemistry) aerosol module with aqueous chemistry is used to simulate aerosol evolution (Zaveri et al., 2008). The photolysis rates are calculated by the Fast-J scheme (Wild et al., 2000). Aerosol radiation is simulated using RRTMG (Rapid Radiative Transfer Model for GCMs) for both shortwave (SW) and longwave (LW) radiation (Zhao et al., 2011). Other major physical parameterizations used in this study are listed in Table S5.

Table S5 Parameterizations used in the WRF-Chem model

Options	WRF-Chem
Microphysics option	Purdue Lin Scheme
Longwave radiation option	RRTMG Scheme
Shortwave radiation option	RRTMG Scheme
Surface layer option	Revised MM5 Monin-Obukhov Scheme
Land surface option	Unified Noah land-surface model
Urban canopy model	Single-layer UCM Scheme
Boundary layer option	YSU Scheme
Cumulus option	Grell 3D ensemble Scheme
Photolysis scheme	Fast-J
Dust scheme	Shao_2004
Chemistry option	CBMZ
Aerosol option	MOSAIC

”

4) In the WRF-Chem simulations with NH3 emission inventory developed in this work and MEIC, I wonder whether other air pollutants emissions (such as SO2, NOx, VOCs) are both based upon MEIC. If it is, please clarify it.

Response: Yes, emissions of other air pollutants in 2016 (including SO_2, NO_x, CO, CO_2, NMVOC, BC, OC, $PM_{2.5}$, and PM_{10}) are both based upon MEIC in the WRF-Chem simulations. We added one sentence to clarify it in section 2.4.

“Emissions of other air pollutants in 2016 (including SO_2, NO_x, CO, CO_2, NMVOC, BC, OC, $PM_{2.5}$, and PM_{10}) were obtained from MEIC (http://meicmodel.org/), with a horizontal resolution of 0.25° (Li et al., 2017b).”

5) For readers not familiar with the China geography and the location of each province, providing a map with province names marked would be valuable.

Response: Thanks for your kindly suggestion. We marked the province names in the Fig. 2.

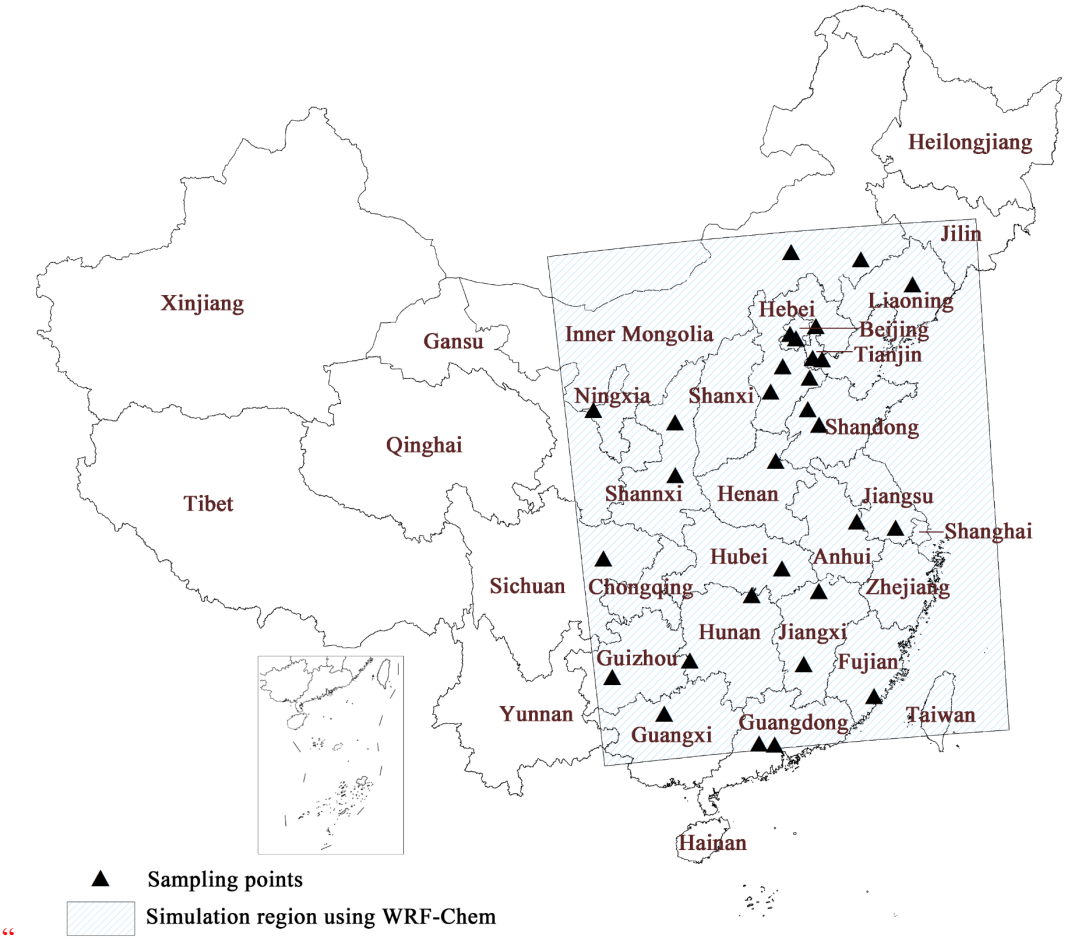

Fig. 2. WRF-Chem simulation domain and sampling points (Ammonia Monitoring Network, China).”

6) In Sec. 3.4, it seems the WRF-Chem model with optimized NH_3 emission inventory yields better performance in July while large bias still existed in January as compared with the MEIC driven WRF-Chem model. Could you be more specific on the reasons for the discrepancy between the simulated NH_3 concentrations and ambient measurements?

Response: We sincerely thank the reviewer for this suggestion. As suggested, we added one paragraph in Section 3.4.3 to explain the possible reasons for the discrepancy between the simulated NH_3 concentrations and ambient measurements in January and October.

“In general, our inventory exhibited better spatial accuracy than other inventories that utilize WRF-Chem and observations, although January and October showed a relatively large bias between measured and simulated data. The NH_3 concentrations were low in these two months. We found that the accuracy of the simulation increased as the concentration of atmospheric NH_3 increased. The mean values of IASI NH_3 VCDs in the simulation domain in January and October were 7.2×10^{15} and 8.7×10^{15} molec/cm^2, respectively, significantly lower than the mean values of 14.9×10^{15} and 19.4×10^{15} molec/cm^2 in April and July. In January and October, the relatively low NH_3 emissions (32.5% and 54.6% of July emissions) combined with the short lifetime of NH_3 and uncertainties in gaseous NH_3 and aerosol NH_4^+ partitioning pose a challenge to the chemical mechanisms of the WRF-Chem model, making it more difficult for the model to fully capture the heterogeneity of NH_3 concentration. Besides, the relatively large uncertainty of the IASI VCDs could also contribute to

inconsistency between simulated and observed concentrations (Van Damme et al., 2017; Chen et al., 2020). Zhang et al. (2018) and Zhao et al. (2020) obtained similar results which indicated that the correlations between measured and simulated NH_3 data were lower in January and October than during the other two months."

7) I think Figure 6(a) is misleading with the erroneous higher concentrations of NH3 in January than July due to the different scales in colormap. Please use the same color scale for both January and July. Otherwise, it is misleading to the readers.

Response: According to your advice, we used the same color scale for both January, April and July in Fig.7 (the original Fig.6).

8) Conclusion: This section would be stronger if a discussion on future work that the community should consider toward improving the representation of ammonia emissions in chemical transport models (For example, investigating the impacts of optimized ammonia emission inventory on simulating $PM_{2.5}$ concentrations and aerosol pH changes).

Response: We sincerely thanks for your constructive suggestion. As suggested, we illustrated the potential applications of our improved ammonia emission inventory in the Conclusion.

"We believe that the improved NH_3 emission inventory can be used in future research, to simulate atmospheric aerosol formation, investigate the influence of NH_3 emission on $PM_{2.5}$ mass burden and aerosol pH changes, develop targeted NH_3 reduction strategies to further improve air quality, and explore the atmospheric N cycle process."

Specific comments:

1) Line 17: "differences"-->"variability". "observed" -->"identified".

Response: We have corrected it.

2) Line 84: "huge"-->"considerable".

Response: Corrected.

3) Line 215: "30 samples" should be "30 sampling stations".

Response: Corrected.

4) Line 223: "represented"-->"contributed".

Response: Corrected.

5) Line 223: The 13.1 Tg is derived from Kong et al., (2019). Thus, there is a lack of citation for Kong et al., (2019).

Response: Thanks for pointing it out, we added this citation.

6) Table 2: I suggest the authors include the ammonia emissions quantified in this work in Table 2 as well.

Response: We added the NH_3 emission value of this study in Table 2.

7) Line 277: Please further clarify the reason why located in north China is relevant to elevated ammonia emissions.

Response: We added more details to explain the reasons for the high NH_3 emission density in the North China Plain.

"These provinces are all concentrated in the North China Plain (NCP, which includes the above six provinces and Beijing), an area that features well-developed crop farming and animal husbandry. The NCP contains a large amount of high-quality arable land, and the farms in this area produce 34.47% of China's major farm products (NBS, 2017c). The fertilization of crops emits large amounts of NH_3. The soil in this area is alkalescent (the average pH value is 7.15), which further increases the NH_3 volatilization. Additionally, the NCP produces as much as 28.16 million tons of pork and beef per year, accounting for 32.5% of the total (NBS, 2017c), contributing to higher NH_3 emissions in the North China Plain."

8) Line 313: Please rephrase this sentence for read.

Response: We have revised this sentence as follows:

"Unlike some pollutants (such as $PM_{2.5}$ and BC) which exhibit higher emissions in winter, NH_3 emissions tended to be high in summer and low in winter."

9) Line 324: Delete "it".

Response: Deleted.

10) Line 349: Delete "Thus".

Response: Deleted.

11) Line 359: The spatial resolution for MEIC is 0.25°×0.25°.

Response: Corrected.

12) Figure S2: Small typo for the caption of subplot for Others.

Response: Corrected.

13) Some figures are hard to read due to the small font size. Please consider improving them.

Response: Thanks for your critical comment. As suggested, we enlarged the font size and improved the resolution of Fig. 4, Fig. 7 and Fig. 8.

14) The manuscript is hard to follow in several places but that could be addressed with thorough language editing.

Response: The manuscript has been edited via professional editing service to reduce grammatical errors.