Responses to Anonymous Referee #2

During the past years, $PM_{2.5}$ pollution have been reduced substantially, while the occasionally occurred heavy $PM_{2.5}$ episodes and its driving forces still need to be explored. One of non-common sense hotspots is the role of NH_3. The relative importance of traffic sources to NH_3 emissions is still under debate.

Base on emission measurement data throughout different cities in China, this study developed high-quality traffic emission inventory of vehicular NH_3 emissions. This work can give a better insight into the absolute value and relative importance of vehicular NH_3 emissions in different regions, seasons and population densities in China. According to the results, they show that the significant role of on-road NH_3 emissions in populated areas have been underappreciated, which is quite important in terms of the atmospheric chemistry and air quality implications.

Overall, this manuscript is well organized and presented with some new insights on NH3 emissions and its contribution, thus, I think this paper is suitable for publishing in ACP after well addressing the following comments, questions and suggestions.

1. As shown in Fig. 2, NH_3 emissions from gasoline vehicles have already declined since 2010 while emissions from diesel vehicles grew significantly since 2014. What's the most possible trend in total on-road NH_3 emissions in the near future under the join effects of vehicle growth and turnover?

The figure below shows the possible trend in total on-road NH_3 emissions in the near future under the join effects of vehicle growth and fleet turnover (impacts of COVID19 are not considered). Evolution of China's vehicle fleet in future is predicted based on the methodology in Wu et al (Wu et al., 2017). Total vehicular NH_3 emissions will reach the peak around 2020. NH_3 emissions from gasoline vehicles will keep decreasing in the next 5 years, while those from diesel vehicle also start to decrease with the implements of China VI emission standard since Jul 2021.

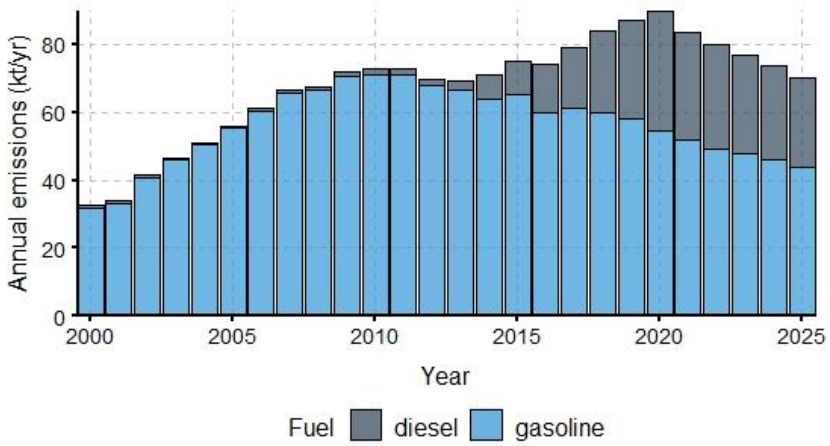

Fig R1. Annual vehicular NH_3 emissions by fuel type in China, 2000-2025.

2. I witnessed an overall higher level of EFs estimated from remote sensing than dynamometer measurements in Fig. 1. What's the possible reason? Some discussion should be added.

Dynamometer measurements are conducted for limited vehicles under the type-approval test cycles with controlled environmental condition. While remote sensing measurements always involve a huge number of vehicles and a complex mix of factors affecting vehicle emissions, such as fleet composition, driving conditions, environmental impacts, etc. As shown in Fig R1 and R2 of my reply to referee comment 1, NH_3 emissions are highly affected by ambient temperature and driving speed. Most of the remote sensing references mentioned in this paper contain measurements under cold temperature and highway driving modes. Also, remote sensing is easily affected by air turbulences that mix the plume emissions of various vehicle categories. Thus, the overall higher level of EFs estimated from remote sensing may result from the impacts of various ambient environment and driving conditions, as well as interferences from high-emitters among the fleets or other vehicle types.

We have added this discussion in the manuscript (line 150-153).

3. Why the shares of vehicular NH_3 emissions in total anthropogenic NH_3 emissions show a large difference between the urban areas Shanghai and Beijing? More detailed discussion is required to quantitatively address the uncertainty (e.g., NH_3 emissions from residential or industrial sectors).

NH_3 emission data from other anthropogenic sources used in this study were obtained from the updating works of Zheng et al (Zheng et al., 2019). The shares of vehicular NH_3 emissions in total anthropogenic NH_3 emissions in Beijing and Shanghai are 8.91% and 7.33% in 2019, respectively. As shown in the figure below, NH_3 emissions from residential and traffic sector are similar in Beijing and Shanghai, while industrial emissions are neglectable. The higher share of vehicular NH_3 emissions in Beijing is mainly result from lower agricultural emission share (i.e., livestock productions and fertilizer applications). The differences in agricultural emissions result from the differences in ambient temperature, type of fertilizer, the application rate of fertilizer and number of breeding stock in two cities.

Uncertainties in NH_3 emissions from other sectors may be brought by activity data and emission factors. Activity data from other emission sources were obtained from authoritative statistics in China to minimize the uncertainties (Zheng et al., 2019). Based on the methodology of uncertainty analysis from Dong et al (Dong et al., 2010) and Fu et al (Fu et al., 2015), the uncertainty ranges of industrial and residential emissions are -83%~127% and -58%~66%, respectively.

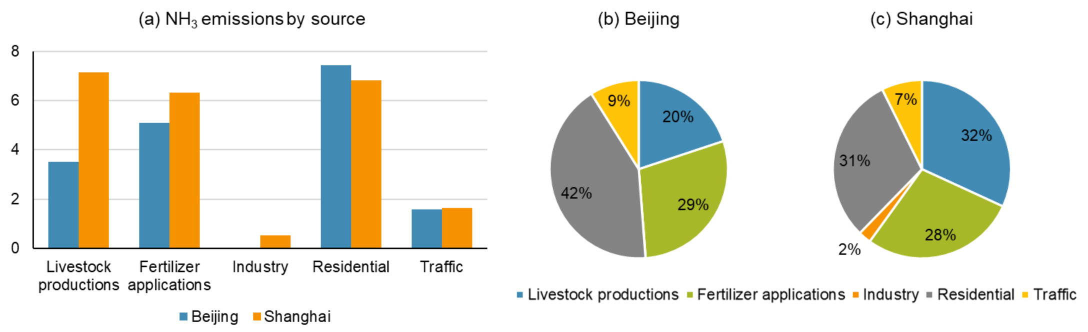

Fig R2. NH_3 emission contributions by sources in Beijing, and Shanghai in 2019.

4. This paper mainly focused on emission inventory but didn't reach to the real impacts on air quality. I understand that the scope of this paper may not be stretched further, but still wonder whether there's any evidence for the air quality impacts of on-road NH3 emissions in urban areas?

In another paper that has been accepted by EST recently, we conducted air quality simulations in Beijing and Shanghai, China, to access the impacts of on-road NH_3 emissions on air quality in urban areas. The results show that local vehicular NH_3 emissions could be responsible for approximately 3% of urban $PM_{2.5}$ concentrations during wintertime, and the contributions could be much higher during polluted periods (~3 μg/m^3) due to the high transformation ratio from the gaseous NH_3 to particle-phase ammonium.

5. The significance, shortage and implications of this study is suggested to be added in the Conclusions sections.

We have added discussions about the major limitation of this study in the Conclusions section, that we didn't account for the impacts of driving conditions on vehicular NH_3 emissions due to the lack of detailed traffic monitoring data in national wide (Line 256-261).

Reference:

Dong, W., Xing, J., and Wang, S.: Temporal and spatial distribution of anthropogenic ammonia emissions in China: 1994-2006, Huan jing ke xue= Huanjing kexue / [bian ji, Zhongguo ke xue yuan huan jing ke xue wei yuan hui "Huan jing ke xue" bian ji wei yuan hui.], 31, 1457-1463, 2010.
Fu, X., Wang, S., Ran, L., Pleim, J., Cooter, E., Bash, J., Benson, V., and Hao, J.: Estimating NH3 emissions from agricultural fertilizer application in China using the bi-directional CMAQ model coupled to an agro-ecosystem model, Atmospheric Chemistry and Physics Discussions, 15, 745-778, 10.5194/acpd-15-745-2015, 2015.
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Zheng, H., Zhao, B., Wang, S., Wang, T., Ding, D., Chang, X., Liu, K., Xing, J., Dong, Z., Aunan, K., Liu, T., Wu, X., Zhang, S., and Wu, Y.: Transition in source contributions of PM2.5 exposure

and associated premature mortality in China during 2005–2015, Environment International, 132, 105111, https://doi.org/10.1016/j.envint.2019.105111, 2019.