Responses to Anonymous Referee #3

This study provides a comprehensive vehicular NH3 emission model with useful insight into spatial and temporal variations of vehicular NH3. The important role of NH3 emissions from vehicles in urban areas with higher population densities is highlighted, which could have important implications for PM2.5 and haze events. Overall the paper is well written and I recommend publication if the comments below can be addressed.

- Section 2.1. Please clarify how the NH3 emission factors were obtained. For gasoline vehicles, was NH3 measured directly or predicted based on correlation with MCE? Further information on sample sizes and whether the data represents a wide range of driving conditions is needed. What are the uncertainties associated with the NH3 emission factors?

For gasoline vehicles, NH_3 EFs were not measured directly, but predicted based on the correlation between NH_3 EFs and MCE. Original measurements of NH_3 emissions and the derivation of relationship between NH_3 and MCE (calculated based on CO and CO_2 EFs) are detailed in Huang et al (Huang et al., 2018). CO and CO_2 EFs under basic driving conditions were obtained from EMBEV model, the archetype model for China's National Emission Inventory Guidebook (Zhang et al., 2014). For diesel vehicles, NH_3 EFs were derived based on measurement data from a fleet of heavy-duty diesel vehicles (HDDVs) (China III to China V) using PEMS and dynamometer (He et al., 2020).

Having the EFs under basic driving conditions, we also established speed correction modules to justify the discrepancy between real-world NH_3 EFs and the basic driving condition. For gasoline vehicles, the speed correction curve was established according to the correlations between NH_3 emissions and VSP (Huang et al., 2018) (see Fig R1 for speed corrections for LDGVs). For diesel vehicles, the speed correction curves were fitted based on average NH_3 EFs tested under different driving conditions. Hence, it's highly possible to quantify the impacts of various driving conditions such as traffic congestion on vehicular NH_3 emissions if real-world speed monitoring data are available. However, the national NH_3 emission inventory in this study was established based on provincial-level statistical data but not link-level traffic profiles due to the lack of detailed traffic monitoring data in national wide. Thus, the EFs used in this study are those under basic driving conditions. To address the possible impacts of driving conditions on vehicular NH_3 emissions, we have added a discussion in the manuscript (Line 256-261).

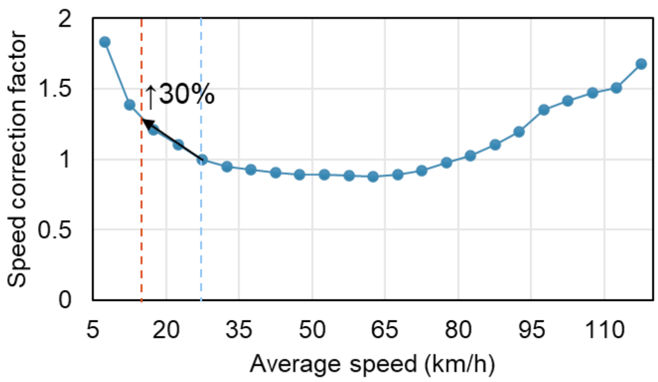

Fig. R1 Speed correction curve for NH_3 EF of LDGVs with average speed from 5 to 120 km/h relative to the basic driving condition (25~30 km/h).

As for the uncertainties in NH_3 EFs, we referred to the error bars of NH_3 emission measurements from various studies (Table S2 and S3 in SI) to estimate the uncertainty ranges of gasoline and diesel vehicles under different emission standards, shown as below.

Table R1. Uncertainty ranges of NH_3 emission factors.

Vehicle types	Emission standards	Uncertainty ranges
LDGV	Euro/China 2	4%
LDGV	Euro/China 3	27%
LDGV	Euro/China 4	25%
LDGV	Euro/China 5	33%
LDGV	Euro/China 6	38%
HDDV	Without SCR	52%
HDDV	SCR-equipped	81%
HDDV	SCR+AMOX	45%

- Line 94 - 95 explains that NH3 emission factors of other diesel vehicles were calculated based on the relative fuel consumptions compared with HDDVs. It would be useful to highlight any limitations of this approach. It is also stated that the NH3 emissions varied significantly among tested HDDVs. How did you account for this?

This study estimated EFs of other diesel vehicles based on the relative fuel consumptions compared with HDDVs due to the lack of measurement data. This approach has obvious limitations and can be improved if more measurement data are available. Nevertheless, HDDVs accounted for 89.8% of the total NH_3 emissions from diesel vehicles in 2019. Thus, the uncertainties brought by EFs of other diesel vehicles are limited.

We have added a discussion about the limitations of the estimation of EFs for other diesel vehicles (Line 261-264).

- Many findings e.g. total vehicular NH3 (32.8 kt to 87.1 kt NH3 from 2000-2019), proportions of NH3 in different provinces (e.g. 8.91%) will be affected by the uncertainties in the NH3 emission factors. Provide estimates of uncertainty associated with these statistics.

Based on the estimated uncertainties of NH_3 EFs (Fig R1), trends of fleet average NH_3 EFs for gasoline and diesel vehicles with uncertainty ranges are show in Fig R2. We have replaced Fig S3 with the figure below.

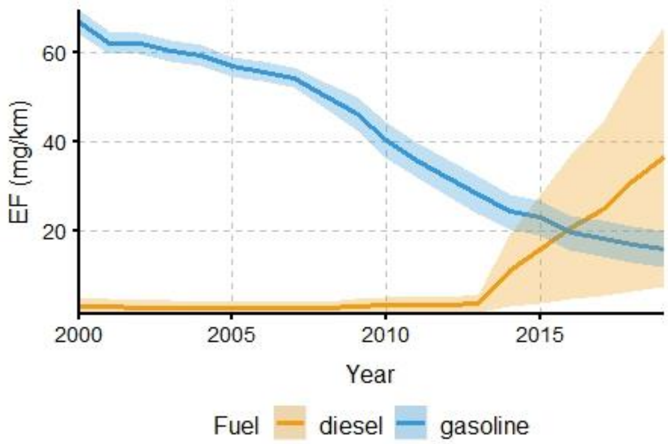

Fig.R2 Trends of fleet average NH_3 EFs for gasoline and diesel vehicles in China, 2000-2019. Shadows show the uncertainty ranges.

We calculated the corresponding uncertainty in total emissions based on the uncertainty ranges in emission factors, shown as below. The annual vehicular NH_3 emissions increased from 32.8±1.7 kt/yr to 87.1±37.5 kt/yr from 2000 to 2019 in China. Proportions of vehicular NH_3 emission in Beijing and Shanghai are 8.91±3.83% and 7.33±3.15%, respectively. We have added uncertainty ranges in results in the manuscript (Line 163-164, 183-185) and replaced Fig 2 with the figure below.

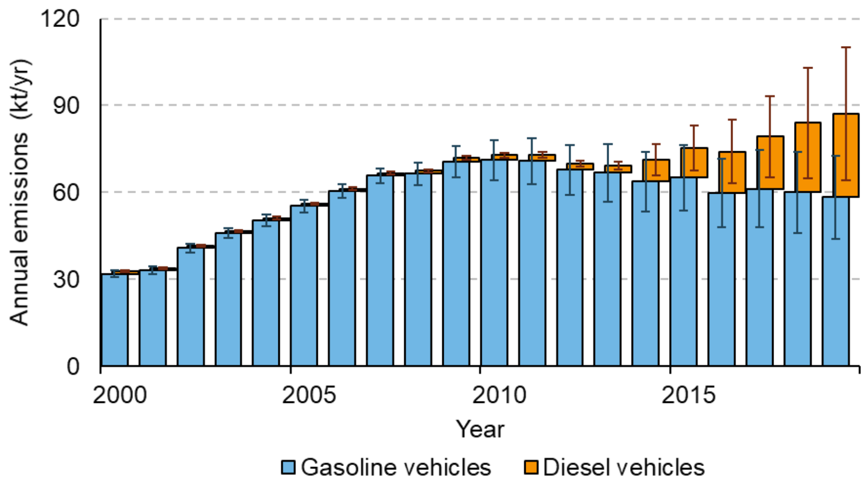

Fig. R3 Annual vehicular NH_3 emissions by fuel type in China with uncertainty ranges, 2000-2019.

- Does the compilation of gridded NH3 emission inventories account for any effects of different traffic conditions?

The impacts of traffic conditions were not considered in compilation of the gridded NH_3 emission inventory due to the lack of detailed traffic monitoring data in national wide. We have addressed this limitation in conclusion section (Line 256-261).

- Figure 1. The authors should refer to the SI, which explains how g/kg EFs have been converted to mg/km. It is useful to explain potential reasons for observed differences. For example, the derivations of mg/km emissions from remote sensing have not been adjusted to account for different driving conditions / fuel consumption, whilst dynamometer measurements may be lower than on-road emissions. Farren 2020 (ES&T) could be useful for mg/km NH3 EFs.

A fleet-averaged rather than a time-specific fuel consumption (g/s) was used to convert the mg/kg EFs to mg/km, thus the derivations of mg/km EFs from remote sensing have not been adjusted to account for different driving conditions / fuel consumption. We have added this explanation in the manuscript (line 150-154).

- Section 3.1. The literature suggests NH3 emissions from gasoline vehicles can increase as vehicles deteriorate / vehicle mileage increases. Do the trends consider this effect, which may be particularly important in the future if gasoline car ownership is increasing? It would also be useful to state the proportion of the proposed increase in NH3 from diesel vehicles that can be attributed to HDDVs and therefore how this may change with implementation of China VI.

The deviations in NH_3 EFs of gasoline vehicles caused by deterioration were aggregated into various emission standards in our model framework. NH_3 EFs under a certain emission standard vary with different model years. Thus, the trends in Fig 1 have considered the effects of deterioration.

We have provided a prediction of NH_3 emission trends in the near future in response to RC2. 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.

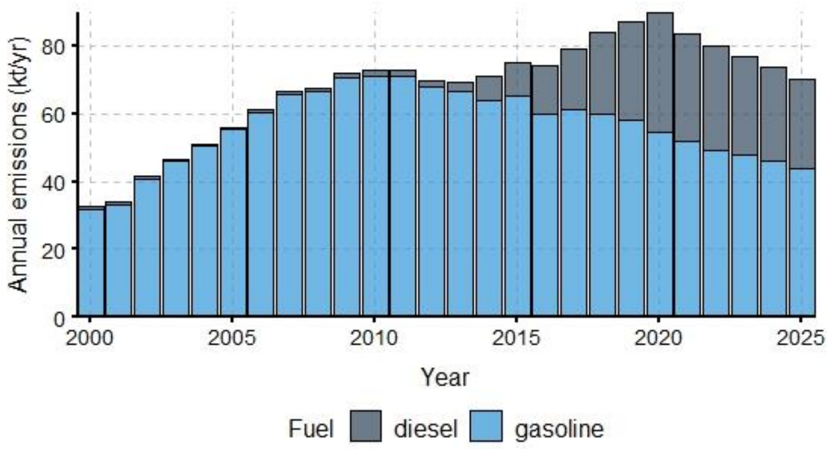

Fig R4. Annual vehicular NH_3 emissions by fuel type in China, 2000-2025.

- Conclusion. This study provides useful insight into vehicular NH3 emissions. It is recommended that the conclusions address the limitations of this study and how this could be improved in the future to better understand the air quality impacts of vehicular NH3.

We have added discussions about the major limitations of this study in the Conclusions section (Line 256-264). Firstly, impacts of driving condition were not included in this study. For urban areas with complex driving conditions and easily affected by traffic congestion, vehicular NH_3 emissions can be further enhanced. It's important to address the impacts of traffic conditions on vehicular NH_3 emissions in urban areas if real-world speed monitoring data is available in future works. Secondly, we estimated EFs of other diesel vehicles based on the relative fuel consumptions compared with HDDVs due to the lack of measurement data. This approach has obvious limitations and can be improved if more measurement data are available.

Technical corrections:

- Use of informal language e.g. line 41 'What's more', line 154, line 176.
- Line 144: 'The monthly variations compare well'
- Line 168: 'might be probably controlled' - be more specific
- Line 198: 'among various population densities.'
- Line 207: should this be 20,000 person/km2?

It's 2000 person/km^2 for sure. 2000 person/km^2 is higher than the population density of most of cities in China.

- Line 236: 'more severe'
- Line 244: 'Euro 7/VII vehicles comply

Technical corrections are modified accordingly.

Reference:

He, L. Q., Zhang, S. J., Hu, J. N., Li, Z., Zheng, X., Cao, Y., Xu, G., Yan, M., and Wu, Y.: On-road emission measurements of reactive nitrogen compounds from heavy-duty diesel trucks in China, Environmental Pollution, 262, 114280, 10.1016/j.envpol.2020.114280, 2020.

Huang, C., Hu, Q., Lou, S., Tian, J., Wang, R., Xu, C., An, J., Ren, H., Ma, D., Quan, Y., Zhang, Y., and Li, L.: Ammonia Emission Measurements for Light-Duty Gasoline Vehicles in China and Implications for Emission Modeling, Environmental Science & Technology, 52, 11223-11231, 10.1021/acs.est.8b03984, 2018.

Wu, Y., Zhang, S. J., Hao, J. M., Liu, H., Wu, X., Hu, J., Walsh, M. P., Wallington, T. J., Zhang, K. M., and Stevanovic, S.: On-road vehicle emissions and their control in China: A review and outlook, Science of the Total Environment, 574, 332-349, 10.1016/j.scitotenv.2016.09.040, 2017.

Zhang, S., Wu, Y., Wu, X., Li, M., Ge, Y., Liang, B., Xu, Y., Zhou, Y., Liu, H., Fu, L., and Hao, J.: Historic and future trends of vehicle emissions in Beijing, 1998–2020: A policy assessment for the most stringent vehicle emission control program in China, Atmospheric Environment, 89, 216-229, 10.1016/j.atmosenv.2013.12.002, 2014.