Responses to Anonymous Referee #1

General comments

In this work, the authors established a comprehensive vehicular NH3 emission model and compiled a gridded on-road NH3 emission inventory with high spatial (3 km × 3 km), and temporal (monthly) resolutions for mainland China using published NH3 emission factors of motor vehicles and their relevant impact factors. With this high-resolution emission inventory, vehicular NH3 emissions during the period of 2000-2019 were estimated. The authors showed that vehicular NH3 emissions could exceed agricultural emissions in the densely populated areas, especially for the extreme populous megacities such as Beijing and Shanghai. Although this conclusion is not unexpected, the paper gives a quantifiable and reliable result, which is valuable for future study. The paper is overall well written. I still have some doubts about the uncertainties of the vehicular NH3 emission inventory and some minor questions as listed below. I recommend publication after these issues are addressed.

Specific comments

1. Line 100: "Bottom-up estimation of long-term vehicular NH3 emissions". My major concern here is whether the authors have considered the additional impacts of the enhancement of driving conditions caused by traffic congestion in densely populated areas on vehicular NH3 emission factors? Or maybe the authors can discuss some uncertainties caused by this factor in the consequent sections of the text. After all, this paper focuses on the importance of NH3 emissions from motor vehicles in densely populated areas.

We have established a speed correction module in NH_3 EF model to justify the discrepancy between real-world NH_3 EFs and the basic driving condition according to the correlations between NH_3 emissions and VSP (Huang et al., 2018) (see the figure below for speed corrections for LDGVs). 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. As shown in Fig R1, the NH_3 EFs of LDGVs under traffic congestion (with average speed ~15 km/h) is about 30% higher than basic driving conditions (with average speed 25~30 km/h). NH_3 EFs also increase significantly under aggressive highway driving cycles (> 90 km/h).

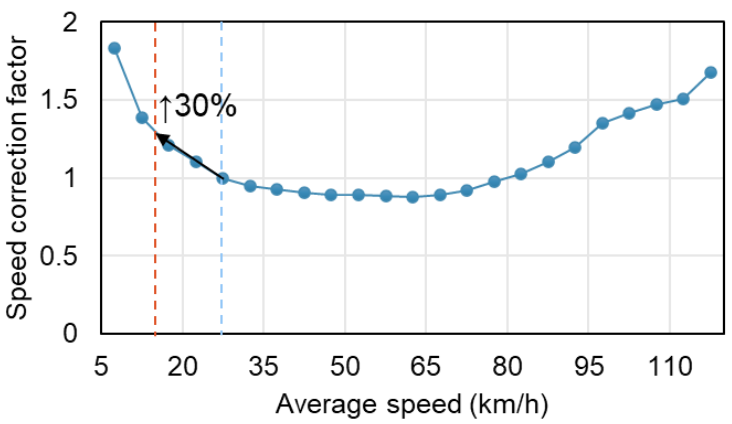

Fig. R1 Speed correction curve for NH_3 EF of LDGVs with average speed from 5 to 120 km/h relative to the basic driving condition (25~30 km/h).

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 (shown as below).

Line 256-261: Note that the impacts of driving condition were not included in this study. It's well documented that LDGV NH_3 emissions are strongly dependent on driving conditions (Huang et al., 2018). Higher LDGV NH_3 emissions are found under both low-speed (Farren et al., 2021) and aggressive highway driving cycles (Huang et al., 2018). For urban areas easily affected by traffic congestion, vehicular NH_3 emissions can be further enhanced. It's important to quantify the impacts of traffic conditions such as traffic congestion on vehicular NH_3 emissions in urban areas if real-world speed monitoring data is available in future researches.

2. Line 190: I suggest that the temporal distributions of vehicular NH3 emissions can be moved to the main text, because this topic is one of the novelties of this study. It would be better if the authors could provide a set of temperature-depended NH3 emission factor correction factors for reference.

We have introduced the ambient temperature corrections for NH_3 EFs in section 2.1 in the manuscript. To better address the novelty about temporal distributions of vehicular NH_3 emissions, the figure below illustrates the monthly variations in NH_3 EFs for LDGVs in Beijing and Shanghai generated from the NH_3 EF model. We have added this plot in the manuscript (Fig 4).

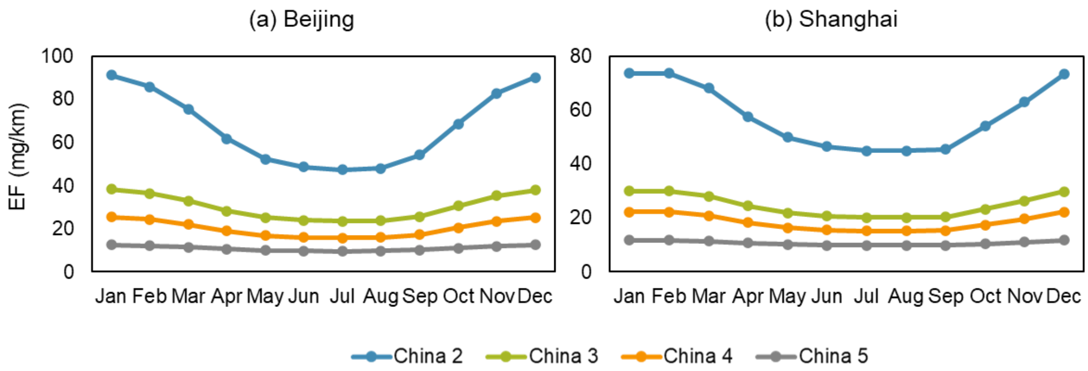

Fig. R2 Monthly NH_3 EFs for LDGVs of various emission standards in (a) Beijing, and (b) Shanghai in 2019.

3. Line 196: If possible, I suggest that the authors could add more discussions on NH3 emissions from residential sources. According to Figure S7, Beijing and Shanghai also have a relatively high proportion of NH3 emissions from residential sources. Their emissions in each season are even higher than those from motor vehicles, and their emissions, if not unexpected, should also be mainly concentrated in densely populated areas.

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). Residential NH_3 emissions mainly include human excrement and domestic fuel combustion, and are mostly attributed to human activities in rural residential areas for megacities like Beijing and Shanghai (Zheng et al., 2019). Thus, even with high emission contributions in the whole city, residential emissions may not be as influential as traffic emissions in urban areas. We have added this discussion in the manuscript (line 218-222).

4. Line 219: Due to the lack of measurements of vehicular NH3 emission factors, NH3 emission inventory still has large uncertainty on the whole. Especially for diesel vehicles, abnormal urea uses and different SCR control strategies will affect its NH3 emission factor. Therefore, it is suggested that the authors could discuss more on the uncertainty of vehicular NH3 emission inventory.

We referred to the error bars of NH_3 emission measurements from various studies (Table S2 and S3) to estimate the uncertainty ranges of gasoline and diesel vehicles under different emission standards, shown as below. For diesel vehicles, the large uncertainty ranges in SCR-equipped EFs have involved the impacts of abnormal urea uses and different SCR control strategies.

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%

Trends of fleet average NH_3 EFs for gasoline and diesel vehicles with uncertainty ranges are show in Fig R3. We have replaced Fig S3 with the figure below.

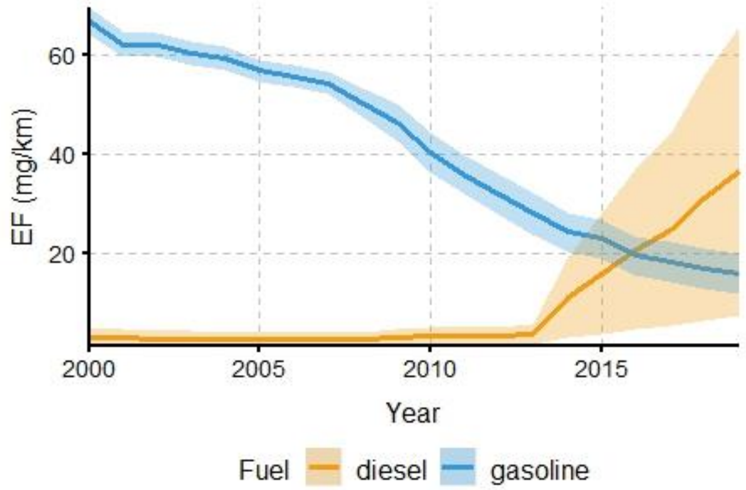

Fig.R3 Trends of fleet average NH_3 EFs for gasoline and diesel vehicles in China, 2000-2019. Shadows show the uncertainty ranges.

We calculated the corresponding uncertainty in total emissions based on the uncertainty ranges in emission factors, shown as below. We have added discussions about uncertainty of vehicular NH_3 emission inventory in the manuscript (Line 163-164) and replaced Fig 2 with the figure below.

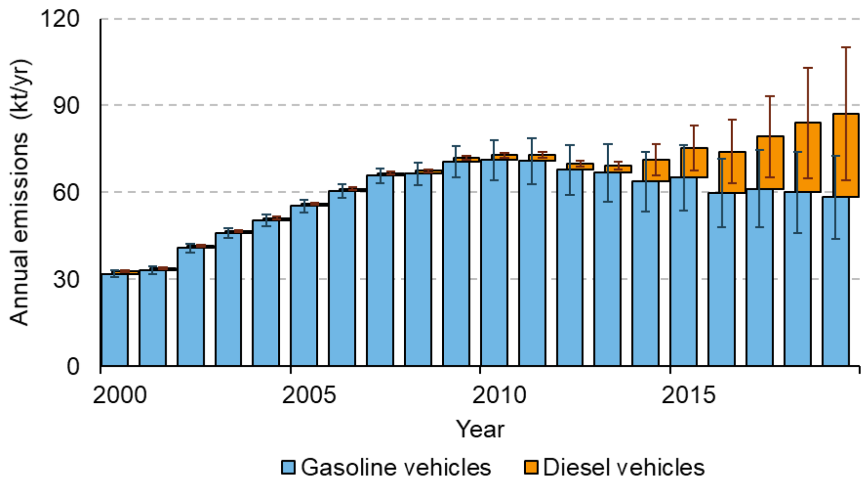

Fig. R4 Annual vehicular NH_3 emissions by fuel type in China with uncertainty ranges, 2000-2019.

References:

Farren, N. J., Davison, J., Rose, R. A., Wagner, R. L., and Carslaw, D. C.: Characterisation of ammonia emissions from gasoline and gasoline hybrid passenger cars, Atmospheric Environment: X, 11, 10.1016/j.aeaoa.2021.100117, 2021.

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