Reply to comments on "High-resolution mapping of vehicle emissions of atmospheric pollutants based on large-scale, real-world traffic datasets" by Daoyuan Yang et al.

"Black" means the comments from reviewer and "Blue" text are our responses.

We are deeply grateful to the reviewers of this paper for the very helpful comments. These comments are fully understood by the authors and individually responded to. Our responses to the comments are listed below, and major changes in the revised manuscript are also quoted in the response document.

Reply to comments from Anonymous Referee #2:

(1) The paper was difficult to read in places. It would benefit from careful proof reading by a native English speaker.

We have revised the specific comments by avoiding non-scientific wordings (i.e., according to Comment 5). We used the ACS Authoring Service to ensure the language quality (Certificate ID: 905A-3009-F9DC-30B3-E4EE). Before the final publication, further professional editing will be also conducted by the ACP editorial staff.

(2) It would be useful if the results were put into context in terms of impacts on human or ecosystem health. For example, in the abstract what is the significance of 'high' carbon monoxide and hydrocarbon emissions from traffic during rush hour. Do the CO and HC concentrations exceed air quality guidelines, and if so by how much, and how important is this?

The reviewer raised an important point regarding the potential risk of certain pollutants during traffic rush hours. In China, the National Ambient Air Quality Standards (NAAQS) set daily and hourly average CO concentration limits at 4 mg/m^3 and 10 mg/m^3. One-year observation records in 2013 for the most polluted site in Beijing (i.e., a traffic site near the South Third Ring Road) indicated that its maximum hourly concentration was 1.9 mg/m^3. Therefore, we conclude that the direct health impact from traffic CO emissions in Beijing would be not significant.

However, there is no such monitoring network in Beijing for certain HC species (e.g., particularly for mobile source air toxics, MSATs). For the reviewer's information, the average concentration of benzene in the urban area of Beijing was 1.7 $\mu g/m^3$ from a ten-day field study in the summer of 2012 (Li et al., 2014). This level was lower than the current annual limits in Japan (3 $\mu g/m^3$) and Europe (5 $\mu g/m^3$).

Li, L. et al., 2014, Pollution characteristics and health risk assessment of benzene homologues in ambient air in the northeastern urban area of Beijing, China. *J. Environ. Sci. 26* (1), 214-223.

(3) It appears that the NO_X concentrations were estimated using a dispersion model without chemistry. If that is the case, the modeling estimation neglects losses of NO_X via conversion to nitrate aerosol and HNO_3. If so, the computed NO_X concentrations are upper limits. Addition of clarification and an estimate of the overestimation if appropriate are needed.

The reviewer understood correctly that we didn't include detailed atmospheric chemistry in the dispersion process of vehicular NO_X emissions. In the comparison with ground-level observations, we only included the daytime NO_X chemistry (NO-NO_2-O3) to estimate the in-situ NO concentrations for the AQ sites.

Due to the lack of measurement data for city-scale NO_Z concentrations, we relied on the atmospheric simulations to further quantify the potential impact from ignoring the production of daytime NO_Z. We referred to air quality simulation results conducted by using the WRF/CMAQ system on the 4-km scale (i.e., using the emission inventory data from Zheng et al., 2019). The results indicate that the daytime NO_Z concentration would be 4.4 ppbv, which approximately 10% of concurrent NO_X concentrations (see Page 10, Line 263 to 266). However, we are currently limited to further estimate the exclusive NO_Z contribution from vehicular emissions in Beijing, which requires more advanced source apportionment tool.

Zheng, H., et al., 2019. Development of a unit-based industrial emission inventory in the Beijing–Tianjin–Hebei region and resulting improvement in air quality modeling. *Atmos. Chem. Phys.*, 19(6), 3447-3462.

"We acknowledge that the daytime concentration of other reactive oxides of nitrogen (i.e., NO_Z, including HNO_3 and HONO) could be approximately 10% of concurrent NO_X concentrations by analyzing the air quality simulation outputs of Zheng et al. (2019). Further studies would be needed to couple dispersion and advanced atmospheric chemistry to better resolve urban pollution."

(4) The results are based on traffic in 2013. My understanding is that there has been a large increase in the fleet size, and substantial decreases in the emissions from new vehicles, since 2013. These factors presumably offset to some degree. Discussion of the emission changes since 2013 resulting from changes to the vehicle fleet is needed.

Beijing has adopted a series of license control and driving restriction polities to control excessively rapid growth in vehicle population. The total vehicle population during 2013-2017 only increased by 8%, which was much lower than the previous growth before 2010. Meanwhile, the Beijing Traffic Research Institute publishes annual traffic reports for the urban areas, which released annual-average urban traffic congestion by using an overall traffic index (from 0, least congested, to 10, most congested; different from the congestion index reported in the manuscript). The annual index values for 2013 to 2017 all ranged within 5.5 to 5.7, representing quite stable traffic conditions over the past years.

On the other hand, the fleet-average emission factors did receive significantly reductions due to the newly implemented emission standards (China 5/V) and subsidized scrappage of older vehicles (Zhang et al., 2014). For example, compared with 2013, fleet-average emissions factors of CO, THC and NO_X for light-duty gasoline vehicles were reduced by ~40%. With updated emission factors and actual traffic data for inter-city expressways in 2017, and scaled vehicle speeds according to the average traffic index, we estimate that daily emissions in 2017 weekdays are 523 tons for CO, 62.5 tons for THC, 256 tons for NO_X, 8.33 tons for $PM_{2.5}$ and 4.18 tons for BC, respectively. The emission reductions (~30% for CO and THC, and 20%~25% for NO_X, $PM_{2.5}$ and BC) relative to the 2013 levels would be majorly attributed to the improvements in average emission factors (see Page 8, Line 199 to 204).

Beijing Transport Institution (BTI), Beijing Transportation Annual Report (in Chinese), 2018. Available at http://www.bjtrc.org.cn/JGJS.aspx?id=5.2&Menu=GZCG.

Zhang, S., et al. 2014. Historic and future trends of vehicle emissions in Beijing, 1998–2020: A policy assessment for the most stringent vehicle emission control program in China. *Atmos. Environ.*, 89, 216-229.

"The recent traffic monitoring data indicate the overall congestion in the urban area has not changed significantly, which is owing to the stringent restrictions on the registration of new vehicles in Beijing (BTI, 2018). On the other hand, average emission factors have decrease significantly due to the implementation of newer emission standards and the subsidized scrappage of older vehicles. As a result, we estimated that the total daily emissions would be 523 tons for CO, 62.5 tons for THC, 256 tons for NO_X, 8.33 tons for $PM_{2.5}$ and 4.18 tons for BC, respectively, in 2017. The significant reductions are primarily attributed to the improvements in average vehicle emission factors."

(5) There are several examples of the use of non-scientific language in the paper. Terms such as "vivid", "soaring", "dramatically", "massive", "flooded", and "tales" should be replaced by more quantitative scientific text. The claim that "Beijing is a microcosm of other megacities" does not make sense.

Revisions are made in the revised manuscript.

Reply to comments from Anonymous Referee #3:

I've seen a variation of this paper before and must say that this version has much improved. The paper presents comprehensive analysis of on-road vehicle emissions in the metropolitan Beijing road network using a link based emission model, which is validated by an air dispersion modeling approach (note that there is no direct way of validating emission results). The four scenarios represent different policies that may affect the vehicle emission level in Beijing and thus have important policy implications. Overall I recommend the paper be considered for publication with this journal.

We appreciate the positive comments from the reviewer.