

Interactive comment on “Gasoline aromatic: a critical determinant of urban secondary organic aerosol formation” by Jianfei Peng et al.

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We thank the referee for his/her careful and critical review of our paper. The following are our responses to the referee's comments.

1. This study investigates the effect of aromatic content on the secondary aerosol production from photochemical processing of gasoline vehicle exhaust. The authors observed a large increase in the SOA production when fuel with higher aromatic content is used. In view of this, the authors suggest that regulations on the gasoline aromatic content would introduce 'unexpected benefit on air quality in urban area'. As such suggestions might help to develop and implement future regulatory plans, it is necessary to take both environmental consequences and economic factors into account. That is,

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for the different types of fuels used, are they producing the same amount of energy or driving distance? The authors are suggested to normalize the reported SOA yields values by the total driving mileage during one test cycle, to be more illustrative to evaluate the influence of aromatic content in the fuel on the PM emissions.

Response: the referee made an important point here. During our vehicle experiments, the same vehicle using different fuels were performed on the chassis dynamometer to run the same driving cycle (with the same distance). The average fuel consumption per unit distance using F1, F2 and F3 fuels were 0.113, 0.112 and 0.113 L km⁻¹, respectively, indicating no difference in fuel economy among the three fuels. The SOA production per unit distance were for 6.3 mg km⁻¹ and 2.1 mg km⁻¹ for F3 and F2 fuel, respectively, when OH exposure was 12 equivalent photochemical hours. These values have been added in the manuscript at line 201 “The average fuel consumption per unit distance using F1, F2 and F3 fuels were 0.113, 0.112 and 0.113 L km⁻¹, indicating no difference in fuel economy among the three fuels”, and at line 209 “The average SOA production at 12 equivalent photochemical-hours using F3 fuel was 76 mg kg-fuel⁻¹ (6.3 mg km⁻¹), equivalent to 3 times of that using F2 fuel (25 mg kg-fuel⁻¹, 2.1 mg km⁻¹).”

2. The SOA formation potential and OH reactivity of aromatic compounds are among the highest achieved in chamber experiments simulating SOA production from a variety of anthropogenic and biogenic precursors. The authors may refer to any global SOA production models, like CMAQ, for the SOA yields used from a selection of VOCs in the model mechanism. It is not surprising that increasing the aromatic content leads to enhanced SOA production from gasoline exhaust. My concern is that the authors did not provide sufficient evidence to support the causal relationship between the observed increase in SOA yield and the increasing amount of aromatic content. As shown in Table S3 in the supplementary materials, less than 50% fraction of the gasoline has been identified, including mostly olefin and aromatics. What if the unidentified carbon mass really contributes to the SOA production, and variations in the recipe of these un-

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known species in different types of fuel are the main drivers to the observed changes in SOA production? These unresolved carbons might include long-chain alkanes and alkenes that have been demonstrated to constitute a large fraction of gasoline emissions (Gentner et al. PNAS, 2012). The authors need to ensure that for the three types of fuels used, the aromatic content is the only variable and the rest of the carbon mass stays constant. This is the prerequisite for the further examination on the contribution of aromatics in gasoline fuels to SOA production.

We agree with the referee that it is important to make sure that aromatic content is the only difference for F2 and F3 fuels. This is why we used F2 fuel to blend F3 fuel. As discussed in part 2.2 of manuscript, F3 fuel was blended from 80% of F2 fuel, 15–20% of refinery reformat stream with high aromatic content and very small amount of octane and n-heptane to keep the same octane level. This means that at least 80% of the mass was the same for F2 and F3 fuel. Besides, to response to the referee's concern, we have added the molecular information for all three fuels as Table S4, which clearly exhibits that the concentrations of most long-chain alkanes and alkenes in F2 fuel were higher than those in F3 fuel, but the concentrations of aromatics were much higher in F3 fuel. Thus, if the long-chain alkanes and alkenes in gasoline fuel were more important than the aromatics on SOA formation, the SOA using F2 fuel should be higher than using F3 fuel. In contrary, we found SOA were much more using F3 fuel, which could only be explained by the higher SOA formation efficiency for fuel aromatics.

Minor comments 3. Page 2, Line 49: The vapor pressure of benzoic acid falls into the semi-volatility range. The authors may refer to Schwantes et al. ACP (2017) for an example.

We thank the reviewer for reminding this. The sentence has been revised as “. . . leading to the formation of a variety of semi- or low-volatile species (e.g., benzoic acid) (Zhang et al., 2015; Schwantes et al., 2017)”

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4. Page 2, Line 51: Please change 'exhibited' to 'shown'.

Thanks. It has been revised.

5. Page 3, Line 64-70: Please change 'underwent' to 'subject to'. Change to 'condition' to 'conditions'. Change 'under strong oxidizability conditions' to 'with high OH exposure'. The oxidation capacity in this study equals to one or two days of ambient OH exposure and does not necessary represent the high OH exposure cases that were reported in literatures (e.g., Lambe et al., ACP, 2015).

We thank the referee for pointing out these mistakes. They have been corrected. Also, to response to the referee's concern, we changed the sentence as "SOA formation experiments from engine exhausts were carried out with relatively high OH exposure compared to ambient conditions to obtain the SOA production potential."

6. Page 3, Line 75: Change 'emission' to 'emissions'.

Thanks. It has been corrected.

7. Page 4, Line 116: Delete 'were conducted'.

Thanks. It has been deleted.

8. Page 5, line 129: Delete 'of' in front of 'relative humidity'.

Thanks. It has been deleted.

9. Page 5, Line 140: Delete 'continually tracked'.

Thanks. It has been deleted.

10. Page 6, Line 164: the OH concentration unite should be 'molec cm-3' or 'molecule cm-3'.

Thanks. It has been corrected.

11. Page 8, Line 226: 'Administration.'? References need to be cited here.

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References have been added here.

12. Page 8, Line 213: Change 'reproducible' to 'reproducibility'.

Thanks. It has been corrected.

13. Page 9, Line 247: Please provide evidence for the conclusion that 'SOA formation from C10-aromatics, alkenes and alkanes is found to be negligible'. In contrast, there have been a number of studies showing significant SOA production from photooxidation of alkanes and alkenes (e.g., Loza et al., ACP, 2014; Matsunaga and Ziemann, 2010).

We apologize for such misleading. We understand that alkenes and alkanes can be important SOA precursors in the atmosphere. Here, we want to express that based on our VOCs measurement, the reacted alkenes and alkanes with more than 7 carbons were much lower compared with aromatic VOCs over the experimental period. We have modified the paragraph as "The SOA precursors here included benzene, toluene, C8-aromatics, C9-aromatics and styrene, which were measured by PTR-MS during each experiment. The contributions of the alkenes and alkanes (7-11 carbons) to SOA formation in our experiments were also estimated using Equ 1 based on the off-line GC-MS measurement. Results showed that the measured alkenes and alkanes (7-11 carbons) only accounted for approximately 4% of the total predicted SOA concentration (Fig. S6) due to the low emission factors as well as the small reacted proportion of these species inside the chamber." Additionally, we have provided more information on the SOA formation estimation in the Supplementary information (Fig. S6) to avoid this misleading.

14. Page 10, Line 271: Change 'with not' to 'without'.

Thanks. It has been corrected.

15. Page 10, Line 278: Change 'continuous' to 'continuously'.

Thanks. It has been corrected.

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16. Page 17, Figure 1: The unite for OH exposure should be 'molec cm⁻³ hr'.

We thank the reviewer for pointing out this mistake. The figure has been revised.

17. Page 18, Figure 2: SOA production from F1 is missing in subfigure (b).

In this study, we focus on the effect of gasoline aromatics on SOA production. This effect can be clearly found by the usage of F2 and F3 fuels, which show similar features between each other except aromatic contents. Therefore, we did not conduct any engine exhaust experiments using the F1 fuel. That's why there is no F1 fuel in Fig. 2b

18. Page 19: Figure 4: Please provide the data source for the emission factors for total hydrocarbon and total other NMHCs. Are they direct measurements from experiments? If so, instruments and methods for these measurements need to be given. It is difficult to differentiate these different hues of green color. Suggest to revise this figure for better visualization.

We thank the referee for the suggestion. The figure is revised. Also, we have added the following sentence "The total hydrocarbons (THC) were measured by vehicle emissions testing system, HORIBA, Ltd.), and the total aromatics and non-methane hydrocarbons (NMHCs) were measured by offline GC-MS" in the figure capture.

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