

Response to the Reviewer

We appreciate the reviewer for his/her constructive criticisms and valuable comments, which were of great help in improving the quality of the manuscript. We have revised the manuscript accordingly and our detailed responses are shown below. All the revision is highlighted in the revised manuscript.

Referee 2 Comments

General comments:

The manuscript by Wu et al. gave an overview of S/IVOCs and their contributions to SOA formation based on the model simulation. The authors improved the model setup parameters and reduced the uncertainty of simulation. The improved model simulation was used to evaluate the effect of S/IVOCs and key anthropogenic S/IVOCs to SOA formation. And the results also showed the potential area of S/IVOCs and sources.

Overall, the manuscript is well organized and within the scope of Atmospheric Chemistry and Physics. I have some minor comments about the manuscript. After addressing the concerns, I would recommend this manuscript for publication.

Reply: Many thanks for the reviewer's encouraging words. We have addressed all of the comments/suggestions in the revised manuscript. Detailed responses to the individual specific comment/suggestion are as follows.

Specific comments:

R2.1. Page 5 line 19. What are the POA factors and OM/OC ratios of different sources? Could you please provide the detailed information about them?

Reply: Sorry for the unclear expressions. It meant that POA emission factors and OM/OC ratios for different source categories, which included the industry, on-road and off-road mobile sources, residential sources, dust and biomass burning. To provide a clearer description, the text has been revised as followed:

“[...] POA emission factors for different source categories (*e.g.*, industry, on-road and off-road mobile sources, residential sources, dust, and biomass burning) were

obtained from POC (primary organic carbon) emission factors using source-specific ratios of OM/OC (mass ratios of organic matter to organic carbon) [...]"

For details, please refer to Lines 11-14, Page 6 in the revised manuscript.

R2.2. Page 8 line 15. Please clarify the definition of SI-SOA.

Reply: Thanks for pointing this out. The definitions of V-SOA and SI-SOA have been provided as followed:

“V-SOA (SOA formed by the oxidation of VOCs-traditional SOA precursors emitted from varied anthropogenic and biogenic sources) and SI-SOA (SOA formed by the oxidation of S/IVOCs-untraditional SOA precursors emitted from anthropogenic sources).”

For details, please refer to Lines 16-19, Page 9 in the revised manuscript.

R2.3. Page 9 line 6. I wonder where the OH rate constants come from. Please explain it.

Reply: The reviewer’s comment is highly appreciated. The OH reaction rate constant (k_{OH}) of $0.57 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ in the 2-species VBS was reduced by a factor of 7 from that of the 9-species VBS ($4 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$), which was assumed to be ~50% higher than that of a typical large saturated *n*-alkane as suggested by previous studies (Atkinson and Arey, 2003; Robinson et al., 2007), in order to align the SOA predictions between 2-species and 9-species VBS schemes.

The above description has been provided in the revised manuscript (Page 10, Lines 10-15).

R2.4. Page 12 line 4. From Fig. 2, I find that the dust and industry contributions to Zhaoqing and Shenzhen are similar. Do you consider the uncertainty when estimate the emission inventories?

Reply: Thanks for the reviewer's comment. The magnitudes of S/IVOC emissions from dust and industry in Zhaoqing and Shenzhen were similar, but the contributions were different. Dust contributed about 21.1% (3.6 Gg) and 7.4% (3.6 Gg) to the S/IVOC emissions in Zhaoqing and Shenzhen, and industry contributed about 22.8% (3.9 Gg) and 8.8% (4.3 Gg), respectively. The contributions of dust and industry to S/IVOC emissions in Shenzhen were smaller than those in Zhaoqing, attributable to the dominance of on-road mobile S/IVOC emissions in Shenzhen (81.3%, 39.6 Gg) because of the dense traffic (Pan et al., 2015). As Shenzhen and Zhaoqing have much less industrial point sources than cities located in the southeastern PRD such as southern Guangzhou and Foshan (Pan et al., 2015), their corresponding industrial S/IVOC emissions were also less. There were relatively higher S/IVOC emissions from road fugitive dust and lower emissions from building construction dust in Zhaoqing than those in Shenzhen because of shorter road lengths and more developed construction industries in Shenzhen (GSY, 2011; Peng et al., 2013), resulting in similar magnitudes of S/IVOC emissions from dust in these two cities. The above description for similar magnitudes and different contributions of S/IVOC emissions from dust and industry in Zhaoqing and Shenzhen has been provided in the revised manuscript.

For details, please refer to Lines 12-24, Page 13 in the revised manuscript.

On the other hand, we indeed have considered the uncertainty when estimate the emission inventory of S/IVOCs. As Table 4 in the revised manuscript showed, the uncertainties in S/IVOC emissions from dust and industry ranged from -84% to 235% and from -97% to 386% at 95% confidence interval, respectively. And the ratios of E_{IVOCs}/E_{POA} used in calculating the S/IVOC emissions for these two source categories were the key sources of uncertainties in the emission estimates.

For details of the uncertainty analysis, please refer to Section 3.2 and Table 4 in the revised manuscript.

R2.5. Page 15 line 9. “The simulation results of SOA formation” will be better.

Reply: Thanks for the suggestion. We have revised manuscript accordingly (Line 4, Page 17).

R2.6. Have you tried to screen out the dominant species among S/IVOCs which contribute mostly to SOA formation in PRD region?

Reply: Thanks for the reviewer’s great comment. As the fact that S/IVOCs in the model was treated as a ‘bulk species’ rather than the individual species, it is unable to figure out the dominant species among S/IVOCs that contribute mostly to SOA formation in PRD region. To better understand the roles of individual S/IVOCs in SOA formation, future work by using different models and source apportionment

results would be conducted.

Technical corrections:

R2.1. Page 1 line 15. “emissions” may be “emission”.

Reply: Sorry for the mistake. It has been revised accordingly (Line 14, Page 1).

R2.2. Page 2 line 12. “secondary organic aerosols” should be “secondary organic aerosol”.

Reply: Sorry for the mistake. It has been revised accordingly (Line 12, Page 2).

R2.3. Page 2 line 13. Please add some refs. About the SOA contribution to PM_{2.5}.

Reply: Sorry for the mistake. References have been added and the description of SOA as the key component of PM_{2.5} has been revised in the revised manuscript as followed:

“As the key component, secondary organic aerosol (SOA) accounts for 20–80% of organic aerosol (OA), while OA accounts for 20%–90% of fine particulate matter (PM_{2.5}) (Kanakidou et al., 2005; Carlton, Wiedinmyer and Kroll, 2009; Zhang et al., 2007, 2013).”

For details, please refer to Lines 12-14, Page 2 in the revised manuscript.

R2.4. Page 2 line 22. The ref. “Guo et al. et al.,” should be “Guo et al.,”. Please revise it.

Reply: Sorry for the mistake. It has been revised accordingly (Line 24, Page 2).

R2.5. Page 12 line 7. I think you have miswritten the figure number. Fig. 4 may be Fig. 3? Please check it.

Reply: Sorry for the mistake. The figure number has been revised through the manuscript (Lines 26-27, Page 13).

R2.6. Page 12 line 14. The discussion order is weird. Fig.3a is after Fig. 3c. And Fig.3b is after Fig. 3d. Please reorganize the discussion about spatial distribution of S/IVOCs.

Reply: Thanks for the suggestion. We have reorganized the discussion about the spatial distribution of S/IVOCs in the revised manuscript accordingly. For details, please refer to Lines 2-15, Page 14 in the revised manuscript.

R2.7. Page 14. In my opinion, I think the whole paragraph on this page is discussing the S/IVOCs emission inventory and comparisons with another study. So maybe an addition of a section title (3.3 ...) here will be better.

Reply: Thanks for the constructive suggestion. We have added a section title “3.3 Comparison with other emission inventory” in Line 23, Page 15.

Reference

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