

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 1 Comments

General comments:

The authors developed a high resolution emission inventory of semi-volatile and intermediate volatility organic compounds (S/IVOCs) for the Pearl River Delta region, and then evaluated the impacts of anthropogenic S/IVOCs on secondary organic aerosols (SOA) by a regional chemical transport model. The primary emissions and chemical degradation mechanisms of S/IVOCs are among the key knowledge gaps in better understanding and predicting the SOA formation. Thus this study is useful for future modelling studies of SOA in the PRD region. I would recommend that this manuscript can be considered for publication after the following specific comments being addressed.

Reply: We thank the reviewer's positive comments and helpful suggestions. 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:

R1.1. Introduction: a brief introduction of S/IVOCs at its first appearance should be helpful for the readership to better understand the context of this study. For example, what compounds do the S/IVOCs include? What are their major sources? Etc.

Reply: Thanks for the helpful suggestion. We have added the following text to Section

1 to give a brief introduction of S/IVOCs in the revised manuscript:

“[...] To date, S/IVOCs are found to mainly include straight chain and branched alkanes with carbon numbers > 12 , alkylcyclohexanes, unsubstituted and substituted polycyclic aromatic hydrocarbons (PAHs), alkylbenzenes, cyclic and polycyclic aliphatic material (Zhao et al., 2015; Li et al., 2018; Drozd et al., 2019). However, a vast majority of S/IVOC mass still have not been speciated at the molecular level, which are defined as an unresolved complex mixture (UCM) (Jathar et al., 2012; Zhao et al., 2015; Drozd et al., 2019). Incomplete combustion, such as the combustion of fossil fuel, especially vehicle exhaust has been reported to be a large contributor to S/IVOC emissions in developed regions (May, Presto, et al., 2013a, 2013b; Ots et al., 2016; Khare and Gentner, 2018). Recent studies have also shown that consumer products and commercial or industrial products, processes, and materials are significant sources of unspciated S/IVOCs (Czech et al., 2016; Khare and Gentner, 2018). On the other hand, biogenic S/IVOCs have recently been demonstrated to have a non-negligible impact on SOA formation, but very few measurements have been reported on their emissions (Palm et al., 2016, 2017). [...]”

For details, please refer to Lines 7-19, Page 4 in the revised manuscript.

R1.2. Section 2.1: it is not clear if this S/IVOCs emission inventory only include one ‘bulk species’ or include individually many S/IVOC compounds for model use? I also wonder if there are biogenic sources of S/IVOCs. If so, the authors may need elaborate that this study mainly focused on anthropogenic emissions.

Reply: The reviewer’s comment is highly appreciated. In this study, we used source-specific linear scaling factors between emission factors of S/IVOCs and POA to establish the S/IVOCs emission inventory, where S/IVOCs has been lumped into a “bulk species” as suggested by previous studies (Hodzic et al., 2010; Shrivastava et al., 2011; Koo et al., 2014). The total S/IVOCs emission was not split into emission of the individual S/IVOC species because of the lack of accurate identification of S/IVOCs

from different emissions (Jathar et al., 2012; Zhao et al., 2015; Drozd et al., 2019). Therefore, our S/IVOCs emission inventory treated S/IVOCs as one ‘bulk species’ for model use. On the other hand, even though previous studies have identified that biogenic sources could emit certain amounts of S/IVOCs, their contributions to SOA formation were insignificant in developed regions where anthropogenic emissions dominated (Palm et al., 2016, 2017; Khare and Gentner, 2018). To describe the emissions of S/IVOCs and their contributions to SOA formation more clearly, the text has been revised as followed:

“[...] anthropogenic emissions and the chemical mechanisms of S/IVOCs have been incorporated into different models [...].”

“[...] It should be noted that this study mainly focused on anthropogenic S/IVOCs and their roles in SOA formation in the PRD region as anthropogenic S/IVOCs were found to have much greater contributions to SOA formation than biogenic S/IVOCs in developed regions (Palm et al., 2016, 2017; Khare and Gentner, 2018).”

For details, please refer to Lines 20-21, Page 4, Lines 26-27, Page 5 and Lines 1-2, Page 6 in the revised manuscript.

R1.3. Section 3.1: I am curious that dust is a source of S/IVOCs. It would be helpful if the authors elaborate more about this source by several sentences.

Reply: Thanks for the reviewer’s comment. In our emission inventory, dust mainly includes road fugitive dust and building construction dust. Particles containing many toxic metals and organic contaminants such as PAHs and long-chain alkanes from various sources (*e.g.*, weathered materials of street surfaces, automobile exhaust, lubricating oils, gasoline, diesel fuel, tire particles, construction materials and atmospherically deposited materials), can be deposited on roads and construction sites, which are known as road fugitive dust and building construction dust (Takada et al., 1990; Rogge et al., 1993; Chen et al., 2012). Furthermore, dust is a large source of POA at urban locations, and S/IVOCs are frequently co-emitted with POA (Zheng et al., 2012; Shrivastava et al., 2013; van Drooge and Grimalt, 2015). Indeed, S/IVOCs,

such as *n*-alkanes (C₁₉–C₃₉) and PAHs have been identified in dust samples, confirming that dust could be a source of S/IVOCs (Takada et al., 1990; Rogge et al., 1993; Schefuß et al., 2003; Dong and Lee, 2009). Therefore, it is reasonable that dust is a source of S/IVOCs.

The above description for the fact that dust is a source of S/IVOCs has been provided in the revised manuscript. For details, please refer to Lines 1-11, Page 9 in the revised manuscript.

R1.4. P1, L13: change “the Pearl River Delta (PRD)” to “the PRD”, as you have already defined the PRD in Line 12.

Reply: Sorry for the mistake. It has been revised in the revised manuscript (Line 13, Page 1).

R1.5. P1, L15: emission factors of POA. . .

Reply: Sorry for the mistake. It has been revised in the revised manuscript (Lines 14-15, Page 1).

R1.6. P5, L16-17: I suggest the authors to provide a brief description of the definition of parameters used in this Equation (1) here, so that the readers do not need to refer to the supplement.

Reply: Thanks for the suggestion. The description of the definition of parameters used in Equation (1) has been moved from the supplement to the main text. For details, please refer to Lines 7-9, Page 6 in the revised manuscript.

R1.7. P9, L13-14: the same as the above comment.

Reply: Thanks for the suggestion. The description of the definition of parameters used in Equation (4) and (5) has been removed from the supplement to the main text. For

details, please refer to Lines 15-23, Page 10 in the revised manuscript.

R1.8. P10, L6: the roles of S/IVOCs in the formation of SOA. . .

Reply: Sorry for the mistake. It has been revised in the revised manuscript (Line 13, Page 11).

R1.9. P15, L13: in my opinion, WQS should be a regional receptor site in the PRD region other than a regional background site, as it is generally located at the downwind of city clusters in winter monsoon season.

Reply: We agreed with the reviewer that WQS is generally located at the downwind of city clusters of PRD in winter monsoon season. Indeed, in winter monsoon season, air masses could bring air pollutants from the center areas of PRD to the WQS site, making WQS site as a regional receptor site of PRD and it was frequently used to investigate the characteristics of air pollutants in PRD region (Guo et al., 2009; Ding et al., 2012). Therefore, the text has been revised as followed:

“In this study, daily measured concentrations of SOA at the WQS site in Guangzhou, a receptor site of the PRD region during autumn and winter seasons, were used to evaluate the model performance on the simulation of SOA (Ding et al., 2012). The monitoring data of this site could represent the regional air pollution in the PRD [...]”
For details, please refer to Lines 6-9, Page 17 in the revised manuscript.

R1.10. Table 3: I suggest moving this table to the supplement.

Reply: Thanks for the reviewer’s comment. Table 3 has been moved to the supplement accordingly.

R1.11. Is this newly developed emission inventory can be used by the modelling community? If so, how can it be accessed? A comment on this should be given in the

data availability.

Reply: Thanks for pointing this out, to provide the access for the developed emission inventory, a description has been added as followed:

“The underlying research data and the newly developed emission inventory of S/IVOCs in this study are available to the community and can be accessed by request to Xuemei Wang (eciwxm@jnu.edu.cn) of Jinan University.”

For details, please refer to Lines 5-7, Page 22 in the revised manuscript.