Anonymous Referee #1, 22 May 2022

General comments
This paper develops a high-resolution emission inventory of IS-VOCs over China and simulates organics over the Yangtze River Delta (YRD) region using the CMAQ model. The influence of difference IS-VOC sources are investigated. The paper is well written and interesting as the influence of the different sources of organics are carefully studied. However, the novelties of this study should be better highlighted, probably by adding a section in the introduction about previous work on IS-VOC emissions in China and aboard. Also, this paper seems similar to the paper of Li et al. Environmental Pollution 2022 (see reference in specific comments below). How do the set up and findings compare to the paper of Li et al. (2022)? I suppose the IS-VOC inventories are done differently, but this should be explained.

Re: Thanks a lot for the reviewer's valuable comment. We are deeply regret that we are not able to learn from the work of Li et al. (2022) since we just submitted our work when they published the study. We have carefully read the paper of Li et al. (2022), which have done a good job simulating SOA formation and source contributions in summer China. However, we believe our study still differs from Li et al. (2022) in the following three respects. First, as the reviewer has mentioned, our study developed a high-resolution I/SVOCs emission inventory by scaling the I/SVOCs emissions from VOCs or POA emissions (depending on their dominant pollutants) and determining their volatility bins based on previous measurements of specific sources. Compared with the unified POA emission scaling method adopted by Li et al. (2022), our inventory provided more detailed I/SVOC emissions and volatility distributions of each specific source. Regrettably, the paper of Li et al. (2022) didn't provide their I/SVOCs emissions so we could not make any comparison with them. Second, in Li et al. (2022), each S/IVOC surrogate is oxidized by OH, with the saturation vapor pressure reduced by one order of magnitude. In our paper, I/SVOCs undergo OH oxidation with four products, and the mass yields are derived based on chamber experiments for I/SVOCs emitted from mobile sources (see Lu et al., 2021). Third, our modeling results refined the SOA contribution of specific sources, which is of great significance for the subsequent control of organic aerosols, but was not covered in the paper of Li et al. (2022). In addition, due to the differences in the treatment of I/SVOCs emissions inventories, the contribution of I/SVOCs emissions to SOA in our study is also very different. In their study, S/I-SOA accounted for only 8.8%, while in our study it can reach 41% in summer. Of course, we will make specific explanations and supplements on these differences in the following reply. Based on the reviewer's comments, we have added a section in the introduction about previous work on I/SVOC emissions in China and aboard.

Changes in manuscript:
1. Introduction:

I/SVOC emission inventories have been developed and applied into air quality models over the past decade. Most of them were estimated by applying different scaling factors based on their relationship with POA, volatile organic compounds (VOCs), or some proxies like naphthalene (Pye and Seinfeld, 2010; Shrivastava et al., 2011; Jathar et al., 2017; Wu et al., 2019, 2021; Li et al., 2020, 2022; Ling et al., 2022). Yet in practice, a same scaling factor was applied to most of the sources in previous studies due to the lack of measurements on I/SVOC emission factors. For example, except

biomass burning (0.75–1.5), Wu et al. (2019) utilized scaling factors of 8–30 for all of the other emission source categories, which was estimated based on the measurements of on-road mobile source. Li et al. (2020) assumed scaling factors of 1.5 for on-road mobile source, and 0.34–1.5 for the other sources, such as industrial and residential sources, which were much lower than the estimations in Wu et al. (2020). Huang et al. (2021) have tried emission factor method to quantify the I/SVOC emissions, yet the results were 60% lower than the scaling factor method, far from catching the measured amount of SOA. Obviously, roughly estimating I/SVOC emissions using one or two emission profiles as surrogates for all emission sources will create large uncertainties.

Recent studies have successively determined the volatility distribution, chemical composition, and emission factors of I/SVOCs from mobile sources, including gasoline and diesel vehicles, non-road diesel machinery, marine vessel, and aircraft (Presto et al., 2011; Cross et al., 2013; Zhao et al., 2015, 2016b; Huang et al., 2018; Qi et al., 2019; Drozd et al., 2019). I/SVOC emission profiles have been reported for nonmobile-sources as well, including coal combustion, wood-burning, cooking, fuel evaporation, and industrial and residential volatile chemical products (Huffman et al., 2009; Gentner et al., 2012; May et al., 2013; Koss et al., 2018; McDonald et al., 2018; Cai et al., 2019; Drozd et al., 2021), making the quantification of I/SVOC emissions and their involvement in air quality models possible.

In China, SOA has been emerging as an important contributor to air pollution. Field observations reveal that OA dominates (30%) the $PM_{2.5}$ concentrations in most parts of China (Tao et al., 2017; Liu et al., 2018), among which the SOA contributes up to 80% of OA during haze pollution (Huang et al., 2014; Ming et al., 2017; Li et al., 2021). SOA formation in China has already been examined in several modeling studies. They found that by considering the POA aging and I/SVOCs oxidation in the models, which is realized by the coupling of VBS scheme, the formation and evolution of SOA can be much better simulated compared to the results of the two-product SOA modeling framework (Zhao et al., 2016a; Wu et al., 2019; Li et al., 2020; Yao et al., 2020; Huang et al., 2021). Chang et al. (2022) developed a full-volatility organic emission inventory with source-specific I/SVOC emission profiles for China, which have greatly improved the model performance on SOA concentrations. However, large gaps still exist between the observed and modeled SOA. Studies on high-resolution I/SVOC emission inventory for more specific sources are highly needed.

New references:

Chang, X., Zhao, B., Zheng, H., Wang, S., Cai, S., Guo, F., Gui, P., Huang, G., Wu, D., Han, L., Xing, J., Man, H., Hu, R., Liang, C., Xu, Q., Qiu, X., Ding, D., Liu, K., Han, R., Robinson, A. L., and Donahue, N. M.: Full-volatility emission framework corrects missing and underestimated secondary organic aerosol sources, One Earth, 5, 403–412, 2022.

Jathar, S. H., Woody, M., Pye, H. O. T., Baker, K. R., and Robinson, A. L.: Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions, Atmos. Chem. Phys., 17, 4305–4318, 2017.

Li, J., Han, Z., Wu, J., Tao, J., Li, J., Sun, Y., Liang, L., Liang, M., and Wang, Q.: Secondary organic aerosol formation and source contributions over east China in summertime, Environ. Pollut., 306, 119383, 2022.

Ling, Z., Wu, L., Wang, Y., Shao, M., Wang, X., and Huang, W.: Roles of semivolatile and intermediate-volatility organic compounds in secondary organic aerosol formation and its implication: A review, J. Environ. Sci., 114, 259–285, 2022.

Pye, H. O. T., Seinfeld, J. H.: A global perspective on aerosol from low-volatility organic compounds, Atmos. Chem. Phys., 10, 4377–4401, 2010.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmos. Chem. Phys., 11, 6639–6662, 2011.

Specific comments:
Introduction
•*L77 « SOA concentration is substantially lower than that measured in the atmosphere ». This is not always the case with the VBS method, and simulated SOA tends even to be too high, see Lane et al. (2008) for example.*
Lane, T. E., Donahue, N. M., and Pandis, S. N.: Simulating secondary organic aerosol formation using the volatility basis-set approach in a chemical transport model, Atmos. Environ., 42, 7439–7451, https://doi.org/10.1016/j.atmosenv.2008.06.026, 2008.

Re: Thanks for the comments. This statement is indeed inappropriate. We have rewritten this sentence to be: "the model simulated SOA concentration still has large gaps with that measured in the atmosphere"

•*L78-81 Another shortcoming in the modelling of OA with the VBS 1D is the lack of representation of the hydrophilic properties of OA. VBS 1D assumes SOA condenses onto an organic phase, whereas SOA may also condense on an aqueous phase, see Kim et al (2011) for example*
Kim Y., Couvidat F., Sartelet K. and Seigneur C. (2011), Comparison of different gas-phase mechanisms and aerosol modules for simulating particulate matter formation. J. Air Waste Manage. Assoc, 61, 1218-1226, doi:10.1080/10473289.2011.603999.

Re: Thanks for the comments. We have supplemented this statement in the introduction as follows. "Another shortcoming in the modelling of OA with the VBS 1D is the lack of representation of the hydrophilic properties of OA, which assumes SOA condenses onto an organic phase, whereas SOA may also condense on an aqueous phase (Kim et al., 2011)"

New references:
Kim, Y., Couvidat, F., Sartelet, K., and Seigneur, C.: Comparison of different gas-phase mechanisms and aerosol modules for simulating particulate matter formation, J. Air Waste Manage., 61, 1218–1226, 2011.

•*L88 : please define IVOC, SVOC. How are they quantify ? Which range of volatility ?*

Re: Thanks for reminding. We have supplemented the definition of IVOCs and SVOCs in the revised manuscript as follows. "IVOCs refer to organic compounds with effective saturation

concentrations between 10^3 to 10^6 μg·m^{-3} at 298 K and 1 atm, while SVOCs refer to organic compounds with effective saturation concentrations between 10^{-1} to 10^3 μg·m^{-3} at 298 K and 1 atm."

•L128 : « I/SVOC emission profiles have not been taken into account in previous studies. » This is not correct, they do have been taken into account in numerous studies. See the review of Ling et al. (2022) and other papers below.

Zhenhao Ling, Liqing Wu, Yonghong Wang, Min Shao, Xuemei Wang, Weiwen Huang, Roles of semivolatile and intermediate-volatility organic compounds in secondary organic aerosol formation and its implication: A review, Journal of Environmental Sciences, Volume 114, 2022, Pages 259-285, https://doi.org/10.1016/j.jes.2021.08.055.

Over the States : Jathar, S. H., Woody, M., Pye, H. O. T., Baker, K. R., and Robinson, A. L.: Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions, Atmos. Chem. Phys., 17, 4305–4318, https://doi.org/10.5194/acp-17-4305-2017, 2017.

Over Europe : K. Sartelet, S. Zhu, S. Moukhtar, M. André, J.M. André, V. Gros, O. Favez, A. Braselli, M. Redaelli, Emission of intermediate, semi and low volatile organic compounds from traffic and their impact on secondary organic aerosol concentrations over Greater Paris, Atmospheric Environment, Volume 180, 2018, Pages 126-137, https://doi.org/10.1016/j.atmosenv.2018.02.031.

Over China : Jie Li, Zhiwei Han, Jian Wu, Jun Tao, Jiawei Li, Yele Sun, Lin Liang, Mingjie Liang, Qin'geng Wang, Secondary organic aerosol formation and source contributions over east China in summertime, Environmental Pollution, Volume 306, 2022, 119383, https://doi.org/10.1016/j.envpol.2022.119383.

Re: Thanks for the comments. We have deleted this statement and included the reviews of previous work on I/SVOC emission profiles in the revised manuscript.

Changes in manuscript:

1. Introduction:

I/SVOC emission inventories have been developed and applied into air quality models over the past decade. Most of them were estimated by applying different scaling factors based on their relationship with POA, volatile organic compounds (VOCs), or some proxies like naphthalene (Pye and Seinfeld, 2010; Shrivastava et al., 2011; Jathar et al., 2017; Wu et al., 2019, 2021; Li et al., 2020, 2022; Ling et al., 2022). Yet in practice, a same scaling factor was applied to most of the sources in previous studies due to the lack of measurements on I/SVOC emission factors. For example, except biomass burning (0.75–1.5), Wu et al. (2019) utilized scaling factors of 8–30 for all of the other emission source categories, which was estimated based on the measurements of on-road mobile source. Li et al. (2020) assumed scaling factors of 1.5 for on-road mobile source, and 0.34–1.5 for the other sources, such as industrial and residential sources, which were much lower than the estimations in Wu et al. (2020). Huang et al. (2021) have tried emission factor method to quantify the I/SVOC emissions, yet the results were 60% lower than the scaling factor method, far from catching the measured amount of SOA. Obviously, roughly estimating I/SVOC emissions using one or two emission profiles as surrogates for all emission sources will create large uncertainties.

Recent studies have successively determined the volatility distribution, chemical composition, and emission factors of I/SVOCs from mobile sources, including gasoline and diesel vehicles, non-road diesel machinery, marine vessel, and aircraft (Presto et al., 2011; Cross et al., 2013; Zhao et al., 2015, 2016b; Huang et al., 2018; Qi et al., 2019; Drozd et al., 2019). I/SVOC emission profiles have been reported for nonmobile-sources as well, including coal combustion, wood-burning, cooking, fuel evaporation, and industrial and residential volatile chemical products (Huffman et al., 2009; Gentner et al., 2012; May et al., 2013; Koss et al., 2018; McDonald et al., 2018; Cai et al., 2019; Drozd et al., 2021), making the quantification of I/SVOC emissions and their involvement in air quality models possible.

In China, SOA has been emerging as an important contributor to air pollution. Field observations reveal that OA dominates (30%) the $PM_{2.5}$ concentrations in most parts of China (Tao et al., 2017; Liu et al., 2018), among which the SOA contributes up to 80% of OA during haze pollution (Huang et al., 2014; Ming et al., 2017; Li et al., 2021). SOA formation in China has already been examined in several modeling studies. They found that by considering the POA aging and I/SVOCs oxidation in the models, which is realized by the coupling of VBS scheme, the formation and evolution of SOA can be much better simulated compared to the results of the two-product SOA modeling framework (Zhao et al., 2016a; Wu et al., 2019; Li et al., 2020; Yao et al., 2020; Huang et al., 2021). Chang et al. (2022) developed a full-volatility organic emission inventory with source-specific I/SVOC emission profiles for China, which have greatly improved the model performance on SOA concentrations. However, large gaps still exist between the observed and modeled SOA. Studies on high-resolution I/SVOC emission inventory for more specific sources are highly needed.

In this study, taking the Yangtze River Delta (YRD) region, including Jiangsu, Zhejiang, Anhui provinces and Shanghai city, as a pilot, we established a high-resolution source specific I/SVOC emission inventory. We then applied the newly established inventory into CMAQ v5.3 to evaluate the contributions of I/SVOC emissions to SOA formation by comparing the results with the observation data collected in the region. Furthermore, we also run the model in different scenarios to quantify the seasonal contributions of different sources to POA and SOA formation in the YRD region.

New references:
Chang, X., Zhao, B., Zheng, H., Wang, S., Cai, S., Guo, F., Gui, P., Huang, G., Wu, D., Han, L., Xing, J., Man, H., Hu, R., Liang, C., Xu, Q., Qiu, X., Ding, D., Liu, K., Han, R., Robinson, A. L., and Donahue, N. M.: Full-volatility emission framework corrects missing and underestimated secondary organic aerosol sources, One Earth, 5, 403–412, 2022.

Jathar, S. H., Woody, M., Pye, H. O. T., Baker, K. R., and Robinson, A. L.: Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions, Atmos. Chem. Phys., 17, 4305–4318, 2017.

Li, J., Han, Z., Wu, J., Tao, J., Li, J., Sun, Y., Liang, L., Liang, M., and Wang, Q.: Secondary organic aerosol formation and source contributions over east China in summertime, Environ. Pollut., 306, 119383, 2022.

Ling, Z., Wu, L., Wang, Y., Shao, M., Wang, X., and Huang, W.: Roles of semivolatile and intermediate-volatility organic compounds in secondary organic aerosol formation and its implication: A review, J. Environ. Sci., 114, 259–285, 2022.

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compounds, Atmos. Chem. Phys., 10, 4377–4401, 2010.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmos. Chem. Phys., 11, 6639–6662, 2011.

Materials and methods
The authors state that they establish a new IS-VOC emission inventory. However, there is only little detail about it. More details would be welcomed.

Re: Thanks. We agree with the reviewer that "new" should be used with caution. We have removed relevant terms in the revised manuscript. Then in the "Materials and methods" section, we have added more details in I/SVOC emission estimation.

Changes in manuscript:
2. Materials and methods:
2.1 I/SVOC emission inventory

I/SVOCs commonly exist in both gas- and particle-phase in the atmosphere. Previous studies usually used POA scaling factors to estimate the I/SVOC emissions, which may lead to large uncertainties in the estimation of gas-phase organic compound-dominated sources, like oil refinery, chemical production, and industrial solvent-use. Herein, we compiled both gas-phase I/SVOCs (I/SVOCs-G) and particle-phase I/SVOCs (I/SVOCs-P) emission inventories and incorporate them into the model. Detailed process of the inventories is as follows.

(1) Source classification: To refine the I/SVOC emissions from different sources, we divided the sources into five major categories and then further grouped them into 21 sub-categories. The major categories include industrial process sources, industrial solvent-use sources, mobile sources, residential sources, and agricultural sources. As shown in Table S1, the industrial process sources include the sectors such as oil refinery, chemical production, and pulp and paper production; Industrial solvent-use sources include textile, leather tanning, timber processing, and various industrial volatile chemical products use; Mobile sources include gasoline and diesel vehicle emissions, fuel evaporation, diesel machinery, marine vessel, and aircraft; Residential sources include coal combustion, residential solvent-use, and cooking emissions; Agricultural source is specifically referred to biomass burning in household stoves, and open burning was not included in this study.

(2) Emission estimation: I/SVOCs-G emissions for each specific source were estimated by the ratios of total I/SVOC components to anthropogenic VOC (AVOC) components (I/SVOCs-to-VOCs). Similarly, I/SVOCs-P emissions were estimated by the ratios of total particle-phase I/SVOC components to POA (I/SVOCs-to-POA). The I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each source were determined according to their fractions of total I/SVOC species in VOC and POA emissions. Then we grouped different I/SVOC species into lumped I/SVOC bins based on their C^* to determine the volatility distributions of each source. The I/SVOCs-G emissions were distributed into four lumped aliphatic IVOC bins across the volatility basis set from $C^*=10^3$ to 10^6 $\mu g \cdot m^{-3}$, two aromatic IVOC bins with the $C^*=10^5$ and 10^6 $\mu g \cdot m^{-3}$, and four lumped SVOC bins with C^* from 10^{-1} and 10^2 $\mu g \cdot m^{-3}$. The I/SVOCs-P emissions were distributed into five bins spanning C^*

from 10^{-1} and 10^3 µg·m^{-3}. Source profiles of I/SVOC species for different sources were referenced from the results in previous studies. Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each specific source and their references. For industrial process, industrial solvent-use, and residential solvent-use sources, only I/SVOCs-G emissions were considered. Their I/SVOCs-G-to-VOCs ratios and emission profiles were derived from the latest version of SPECIATE 5.1 database (US EPA, 2021). For gasoline and diesel vehicles, the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and emission profiles were referenced from a new mobile-source parameterization recommended by Lu et al. (2020). Those of diesel machinery, marine vessel, and residential coal combustion were determined by recent measurement results in China (Qi et al., 2019; Huang et al., 2018; Cai et al., 2019). The I/SVOCs-G-to-VOCs ratios and profiles of cooking and biomass burning emissions were derived from SPECIATE 5.1 database, while their particle-phase ratios and profiles were referenced from two previous studies (May et al., 2013; Louvaris et al., 2017). Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and their emission profiles of each specific source. The base emissions of AVOCs and POA (See Table S3) were taken from a high-resolution emission inventory for the year of 2017 developed in our previous study (An et al., 2021).

(3) Model input: Before being input into the model, the estimated I/SVOC-G and I/SVOC-P emissions were summed and then redistributed according to their phase equilibrium under their actual atmospheric state. The formula of phase equilibrium is shown in Equation (1).

$$F_p = \frac{C_{OA}}{C_{OA} + C^*} \tag{1}$$

Where, F_p is the fraction of particle-phase I/SVOC emissions for each volatility bin under actual atmospheric state. C_{OA} represents the OA concentration under actual atmospheric state. We assumed it to be 10 µg·m^{-3} in this study. C^* is the effective saturation concentration of each volatility bin. After redistribution, the I/SVOC emissions for each source category were allocated into 4 km × 4 km grids and hourly temporal profiles using the same method as the criteria pollutants.

New references:

Louvaris, E. E., Florou, K., Karnezi, E., Papanastasiou, D. K., Gkatzelis, G. I., and Pandis, S. N.: Volatility of source apportioned wintertime organic aerosol in the city of Athens, Atmos. Environ., 158, 138–147, 2017.

L153-165 : it is mentioned that the gas-phase IS-VOCs are obtained from ratios of IS-VOCs to VOCs and particle phase IS-VOCs are obtained from ratios of IS-VOCs to POA). Then IS-VOCs are distributed into volatility bins. The ratios are detailed in the supplementary material S1 (which should be refered to in the paper, but is not in the current version). How are the ratio defined ? Only a few references are given in Supp S1. Please add a reference for each activity sector. How are the distributions into volatility bins defined ?
By setting up differently the IS-VOCs in the gas phase and the IS-VOCs in the particle phase, how can we ensure that the gas and particle phases are consistent ?

Re: Thanks for the comments. We agree with the reviewer that the methodology of I/SVOC emission inventory compilation needs more elaboration. Therefore, in the revised manuscript, we

provide a more detailed description of the estimation for I/SVOCs-G and I/SVOCs-P emissions and their volatility distributions. After the I/SVOC emission estimation, we then summarize them and redistributed their gas- and particle-phase emissions in each bin according to their phase equilibrium under actual atmospheric state to ensure the consistency of gas- and particle-phase emission before input into the model. We have made more detailed additions and revisions in the revised manuscript.

Changes in manuscript:
2.1 I/SVOC emission inventory

I/SVOCs commonly exist in both gas- and particle-phase in the atmosphere. Previous studies usually used POA scaling factors to estimate the I/SVOC emissions, which may lead to large uncertainties in the estimation of gas-phase organic compound-dominated sources, like oil refinery, chemical production, and industrial solvent-use. Herein, we compiled both gas-phase I/SVOCs (I/SVOCs-G) and particle-phase I/SVOCs (I/SVOCs-P) emission inventories and incorporate them into the model. Detailed process of the inventories is as follows.

(1) Source classification: To refine the I/SVOC emissions from different sources, we divided the sources into five major categories and then further grouped them into 21 sub-categories. The major categories include industrial process sources, industrial solvent-use sources, mobile sources, residential sources, and agricultural sources. As shown in Table S1, the industrial process sources include the sectors such as oil refinery, chemical production, and pulp and paper production; Industrial solvent-use sources include textile, leather tanning, timber processing, and various industrial volatile chemical products use; Mobile sources include gasoline and diesel vehicle emissions, fuel evaporation, diesel machinery, marine vessel, and aircraft; Residential sources include coal combustion, residential solvent-use, and cooking emissions; Agricultural source is specifically referred to biomass burning in household stoves, and open burning was not included in this study.

(2) Emission estimation: I/SVOCs-G emissions for each specific source were estimated by the ratios of total I/SVOC components to anthropogenic VOC (AVOC) components (I/SVOCs-to-VOCs). Similarly, I/SVOCs-P emissions were estimated by the ratios of total particle-phase I/SVOC components to POA (I/SVOCs-to-POA). The I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each source were determined according to their fractions of total I/SVOC species in VOC and POA emissions. Then we grouped different I/SVOC species into lumped I/SVOC bins based on their C* to determine the volatility distributions of each source. The I/SVOCs-G emissions were distributed into four lumped aliphatic IVOC bins across the volatility basis set from $C^*=10^3$ to 10^6 $\mu g \cdot m^{-3}$, two aromatic IVOC bins with the $C^*=10^5$ and 10^6 $\mu g \cdot m^{-3}$, and four lumped SVOC bins with C* from 10^{-1} and 10^2 $\mu g \cdot m^{-3}$. The I/SVOCs-P emissions were distributed into five bins spanning C* from 10^{-1} and 10^3 $\mu g \cdot m^{-3}$. Source profiles of I/SVOC species for different sources were referenced from the results in previous studies. Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each specific source and their references. For industrial process, industrial solvent-use, and residential solvent-use sources, only I/SVOCs-G emissions were considered. Their I/SVOCs-G-to-VOCs ratios and emission profiles were derived from the latest version of SPECIATE 5.1 database (US EPA, 2021). For gasoline and diesel vehicles, the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and emission profiles were referenced from a new mobile-source parameterization recommended by Lu et al. (2020). Those of diesel machinery, marine vessel, and residential coal combustion were determined by recent measurement results in China (Qi et al.,

2019; Huang et al., 2018; Cai et al., 2019). The I/SVOCs-G-to-VOCs ratios and profiles of cooking and biomass burning emissions were derived from SPECIATE 5.1 database, while their particle-phase ratios and profiles were referenced from two previous studies (May et al., 2013; Louvaris et al., 2017). Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and their emission profiles of each specific source. The base emissions of AVOCs and POA (See Table S3) were taken from a high-resolution emission inventory for the year of 2017 developed in our previous study (An et al., 2021).

(3) Model input: Before being input into the model, the estimated I/SVOC-G and I/SVOC-P emissions were summed and then redistributed according to their phase equilibrium under their actual atmospheric state. The formula of phase equilibrium is shown in Equation (1).

$$F_p = \frac{C_{OA}}{C_{OA} + C^*} \tag{1}$$

Where, F_p is the fraction of particle-phase I/SVOC emissions for each volatility bin under actual atmospheric state. C_{OA} represents the OA concentration under actual atmospheric state. We assumed it to be 10 $\mu g \cdot m^{-3}$ in this study. C^* is the effective saturation concentration of each volatility bin. After redistribution, the I/SVOC emissions for each source category were allocated into 4 km × 4 km grids and hourly temporal profiles using the same method as the criteria pollutants.

Changes in Supplementary information:

Table S1. Gas-phase I/SVOCs-to-VOCs ratios for specific sources and emission profiles used in CMAQ simulations. The characters in brackets are the source codes in the SPECIATE 5.1 database.

Source		I/SVOCs-G to VOCs	Volatility (C^* at 298 K, $\mu g \cdot m^{-3}$)										References
			IVOCP6	IVOCP5	IVOCP4	IVOCP3	SVOCP2	SVOCP1	SVOCP0	SVOCN1	IVOCP6ARO	IVOCP5ARO	
			10^6	10^5	10^4	10^3	10^2	10	1	10^{-1}	10^6	10^5	
Industrial process	Oil refinery	0.039	0.759	0.123	0.004	0.110	0.003	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Chemical production	0.282	0.430	0.230	0.025	0.116	0.199	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Pulp and paper	0.140	0.571	0.393	0.028	0.006	0.001	0.001	0.000	0.000	0.000	0.000	SPECIATE 5.1
Industrial solvent-use	Textile	2.473	0.041	0.448	0.182	0.268	0.040	0.002	0.019	0.000	0.000	0.000	SPECIATE 5.1
	Leather tanning	0.231	1.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Timber processing	0.119	0.584	0.416	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Furniture coating	0.021	0.888	0.112	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Solvent-based coating	0.177	0.948	0.044	0.008	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Water-based coating	0.504	0.096	0.893	0.011	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Dry cleaning	0.004	0.885	0.115	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Paint remover	0.072	0.987	0.010	0.004	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
Mobile sources	Gasoline vehicle	0.265	0.206	0.056	0.113	0.098	0.000	0.000	0.000	0.000	0.406	0.121	Lu et al., 2020
	Diesel vehicle	1.358	0.331	0.318	0.244	0.095	0.000	0.000	0.000	0.000	0.004	0.007	Lu et al., 2020
	Fuel evaporation	0.002	0.841	0.159	0.000	0.000	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
	Diesel machinery	0.400	0.282	0.279	0.264	0.102	0.057	0.012	0.003	0.000	0.000	0.000	Qi et al., 2019
	Marine vessel	0.300	0.230	0.375	0.193	0.097	0.029	0.000	0.000	0.000	0.077	0.000	Huang et al., 2018
	Aircraft	0.482	0.761	0.148	0.063	0.028	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1
Residential sources	Coal combustion	0.180	0.439	0.439	0.088	0.035	0.000	0.000	0.000	0.000	0.000	0.000	Cai et al., 2019
	Residential solvent-use	0.240	0.938	0.047	0.003	0.007	0.000	0.003	0.000	0.001	0.000	0.000	SPECIATE 5.1
	Cooking	0.036	0.554	0.374	0.052	0.015	0.003	0.001	0.000	0.000	0.000	0.000	SPECIATE 5.1
Agriculture sources	Biomass burning	0.064	0.337	0.330	0.215	0.118	0.000	0.000	0.000	0.000	0.000	0.000	SPECIATE 5.1

Table S2. Particle-phase I/SVOCs-to-POA ratios for specific sources and emission profiles used in CMAQ simulations.

Source		I/SVOCs-P to POA	Volatility (C* at 298 K, µg·m⁻³)					References
			IVOCP3	SVOCP2	SVOCP1	SVOCP0	SVOCN1	
			10^3	10^2	10	1	10^{-1}	
Mobile sources	Gasoline vehicle	0.901	0.000	0.323	0.406	0.073	0.197	Lu et al., 2020
	Diesel vehicle	0.867	0.000	0.419	0.420	0.099	0.063	Lu et al., 2020
	Diesel machinery	0.420	0.455	0.204	0.123	0.131	0.087	Qi et al., 2019
	Marine vessel	0.469	0.305	0.140	0.185	0.166	0.204	Huang et al., 2018
Residential sources	Cooking	0.830	0.670	0.157	0.003	0.000	0.000	Louvaris et al., 2017
Agriculture sources	Biomass burning	0.150	0.500	0.250	0.125	0.125	0.000	May et al., 2013

Results and discussion

L293-296. The estimation of IS-VOC in the gas phase computed in this study is compared to what was found in other studies. However, there is not much details and it is hard to understand what differs between the studies. A section should be added to detail what was done in previous emission inventory of IS-VOC in China and aboard. This should be probably added in the introduction. I guess that the factors used to estimate IS-VOC are different in this study than in other studies over China, because IS-VOC are estimated from AVOCs. This should be clearly stated in the introduction. Also, note that IS-VOCs have been estimated from AVOCs in other previous studies over US and Europe.

Re: Thanks for the comments. We have supplemented the details of I/SVOC emission estimation in previous studies in the introduction section of the revised manuscript.

Changes in manuscript:
1. Introduction:

   I/SVOC emission inventories have been developed and applied into air quality models over the past decade. Most of them were estimated by applying different scaling factors based on their relationship with POA, volatile organic compounds (VOCs), or some proxies like naphthalene (Pye and Seinfeld, 2010; Shrivastava et al., 2011; Jathar et al., 2017; Wu et al., 2019, 2021; Li et al., 2020, 2022; Ling et al., 2022). Yet in practice, a same scaling factor was applied to most of the sources in previous studies due to the lack of measurements on I/SVOC emission factors. For example, except biomass burning (0.75–1.5), Wu et al. (2019) utilized scaling factors of 8–30 for all of the other emission source categories, which was estimated based on the measurements of on-road mobile source. Li et al. (2020) assumed scaling factors of 1.5 for on-road mobile source, and 0.34–1.5 for the other sources, such as industrial and residential sources, which were much lower than the estimations in Wu et al. (2020). Huang et al. (2021) have tried emission factor method to quantify the I/SVOC emissions, yet the results were 60% lower than the scaling factor method, far from catching the measured amount of SOA. Obviously, roughly estimating I/SVOC emissions using one or two emission profiles as surrogates for all emission sources will create large uncertainties.

New references:

Jathar, S. H., Woody, M., Pye, H. O. T., Baker, K. R., and Robinson, A. L.: Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions, Atmos. Chem. Phys., 17, 4305–4318, 2017.

Li, J., Han, Z., Wu, J., Tao, J., Li, J., Sun, Y., Liang, L., Liang, M., and Wang, Q.: Secondary organic aerosol formation and source contributions over east China in summertime, Environ. Pollut., 306, 119383, 2022.

Ling, Z., Wu, L., Wang, Y., Shao, M., Wang, X., and Huang, W.: Roles of semivolatile and intermediate-volatility organic compounds in secondary organic aerosol formation and its implication: A review, J. Environ. Sci., 114, 259–285, 2022.

Pye, H. O. T., Seinfeld, J. H.: A global perspective on aerosol from low-volatility organic compounds, Atmos. Chem. Phys., 10, 4377–4401, 2010.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmos. Chem. Phys., 11, 6639–6662, 2011.

L352. Please summarize the method of Lu et al. (2020) in a few sentences.

Re: Thanks for the comments. We have supplemented the summarization of the methods of Lu et al. (2020) in the revised manuscript.

Changes in manuscript:

3.1.2 Volatility distributions of I/SVOCs

Note that IVOCs in vehicle exhaust are dominated by aromatics, which have faster OH reaction rates and higher SOA yields compared to aliphatics in the same volatility bin (Zhao et al., 2016b; Drozd et al., 2019). Lu et al. (2020) therefore defined two additional lumped IVOC species with $logC^*$ bins at 5 and 6 to account for the aromatic IVOCs in vehicle exhaust according to the measurements in previous studies (Zhao et al., 2015; Zhao et al., 2016b). Here in this study, we also split the aromatic IVOC emissions from mobile sources and found that aromatic IVOCs accounted for 23% of the total I/SVOC emissions from the mobile source.

L449. Is there an increase of AVSOA in IMPROVE compared to BASE ? It is surprising if there isn't, as organic concentrations are higher in the IMPROVE simulation, leading to higher absorbing mass.

Re: Thanks. The AVSOA concentration in IMPROVE simulation case increased compared to the BASE. The increasing rate was about 30%. We have supplemented some explanation in the revised manuscript.

Changes in manuscript:

3.2.1 Simulation results of OA concentrations

The seasonal average concentration of AVSOA in the BASE case was only 0.25 $\mu g \cdot m^{-3}$. The average AVSOA concentration in the IMPROVE case increased by 30.1% compared with the BASE

case due to higher OA loading. Nonetheless, AVSOA still exhibited very limited contribution to the regional OA concentration, whereas average concentration of BVOC derived SOA (BVSOA, 1.7 $\mu g \cdot m^{-3}$) was much higher.

L451. What was expected?

Re: Sorry for the mistake. We have deleted this statement in the revised manuscript.

Changes in manuscript:

The seasonal average concentration of AVSOA in the BASE case was only 0.25 $\mu g \cdot m^{-3}$. The average AVSOA concentration in the IMPROVE case increased by 30.1% compared with the BASE case due to higher OA loading. Nonetheless, AVSOA still exhibited very limited contribution to the regional OA concentration, whereas average concentration of BVOC derived SOA (BVSOA, 1.7 $\mu g \cdot m^{-3}$) was much higher.

Conclusions
L655. « SOA increased by 1.2 times in IMPROVE simulation ». However, L431 stated that OA are 38% higher in IMPROVE simulation. Why are these numbers different?

Re: OA is composed of POA and SOA. On L655, we only compared SOA in IMPROVE and BASE cases, while on L431, total OA concentrations were compared.

Anonymous Referee #2, 27 May 2022

General Comments:

An and coauthors have assembled an emission inventory of organic compounds spanning likely particle-phase, semivolatile, intermediate volatility, and highly volatile pollutants for a comprehensive list of key sources in China. After a brief summary of the changes incurred by this new inventory relative to an existing Base, the authors apply both inventories to model ambient air concentrations of primary and secondary OA using a state-of-the-art chemical transport model, CMAQ. The methods used in this study appear generally sound (with an exception discussed below) and the analysis is complete. The information provided by the study in terms of source contributions for OA should be valuable, I expect, to policy-makers in China, and to the larger Earth system modeling community. I was particularly impressed at the model performance improvement that the authors were able to document. I have some concerns about details of the implementation, some questions about surprising results, and several minor suggestions, which I have included below.

Major Concerns:

1. *I am missing something fundamental from the presentation in the paper. The authors have documented both I/SVOC-G emissions and I/SVOC-P emissions, but are they being added together in the model? I think it is very likely that there is overlap in this region and that if both are used, then they will be double-counting some emissions, especially the SVOCs, but also the IVOCs. Can the authors be much more specific about how they fit these pieces together? In lines 241-242, the authors mention subtracting the semivolatile portion from the total POA, but there needs to be more detailed description in one place of what is going on here.*

Re: Thanks for the reviewer's comments. The I/SVOCs-G and I/SVOCs-P emissions in this study were estimated independently. The reason for this consideration is that the I/SVOC emissions of most sources include both gas- and particle-phase. Therefore, we determined the I/SVOCs-G based on the fractions of I/SVOC components in VOCs in the gas-phase profiles, while the I/SVOCs-P were determined based on the fractions of I/SVOC components in POA in the particle-phase profiles. The fractions in the gas- and particle-phase profiles were usually obtained from the gas- (eg Tenax tube) and particle-phase (eg filters) sampling system, respectively. We believe there was no double-counting for the I/SVOC emissions. After estimating I/SVOCs-G and I/SVOCs-P emissions, we added them together and redistributed into the model according to their phase equilibrium under the actual atmosphere state. In the revised manuscript, we have supplemented the description of I/SVOC emission estimation. The SOA formed from I/SVOCs-G was estimated using the parameterization within the VBS framework in Lu et al. (2020). POA was treated as semivolatile to account for its gas-particle partitioning and ageing process and segregated to several particle species, which varied in their volatility that quantified with the metric $C^* = 10^{-1}$ to 10^3 μg·m^{-3} (Donahue et al., 2006). I/SVOCs-P emissions from different sources were then speciated and input as semivolatile accordingly. The remaining POA emissions excluding I/SVOCs-P were treated as nonvolatile POC (primary organic carbon) and PNCOM (primary non-carbon organic matter). We have supplemented

the description of POA modeling scheme in the revised manuscript.

Changes in manuscript:

2.1 I/SVOC emission inventory

I/SVOCs commonly exist in both gas- and particle-phase in the atmosphere. Previous studies usually used POA scaling factors to estimate the I/SVOC emissions, which may lead to large uncertainties in the estimation of gas-phase organic compound-dominated sources, like oil refinery, chemical production, and industrial solvent-use. Herein, we compiled both gas-phase I/SVOCs (I/SVOCs-G) and particle-phase I/SVOCs (I/SVOCs-P) emission inventories and incorporate them into the model. Detailed process of the inventories is as follows.

(1) Source classification: To refine the I/SVOC emissions from different sources, we divided the sources into five major categories and then further grouped them into 21 sub-categories. The major categories include industrial process sources, industrial solvent-use sources, mobile sources, residential sources, and agricultural sources. As shown in Table S1, the industrial process sources include the sectors such as oil refinery, chemical production, and pulp and paper production; Industrial solvent-use sources include textile, leather tanning, timber processing, and various industrial volatile chemical products use; Mobile sources include gasoline and diesel vehicle emissions, fuel evaporation, diesel machinery, marine vessel, and aircraft; Residential sources include coal combustion, residential solvent-use, and cooking emissions; Agricultural source is specifically referred to biomass burning in household stoves, and open burning was not included in this study.

(2) Emission estimation: I/SVOCs-G emissions for each specific source were estimated by the ratios of total I/SVOC components to anthropogenic VOC (AVOC) components (I/SVOCs-to-VOCs). Similarly, I/SVOCs-P emissions were estimated by the ratios of total particle-phase I/SVOC components to POA (I/SVOCs-to-POA). The I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each source were determined according to their fractions of total I/SVOC species in VOC and POA emissions. Then we grouped different I/SVOC species into lumped I/SVOC bins based on their C* to determine the volatility distributions of each source. The I/SVOCs-G emissions were distributed into four lumped aliphatic IVOC bins across the volatility basis set from $C^*=10^3$ to 10^6 $\mu g \cdot m^{-3}$, two aromatic IVOC bins with the $C^*=10^5$ and 10^6 $\mu g \cdot m^{-3}$, and four lumped SVOC bins with C^* from 10^{-1} and 10^2 $\mu g \cdot m^{-3}$. The I/SVOCs-P emissions were distributed into five bins spanning C^* from 10^{-1} and 10^3 $\mu g \cdot m^{-3}$. Source profiles of I/SVOC species for different sources were referenced from the results in previous studies. Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios for each specific source and their references. For industrial process, industrial solvent-use, and residential solvent-use sources, only I/SVOCs-G emissions were considered. Their I/SVOCs-G-to-VOCs ratios and emission profiles were derived from the latest version of SPECIATE 5.1 database (US EPA, 2021). For gasoline and diesel vehicles, the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and emission profiles were referenced from a new mobile-source parameterization recommended by Lu et al. (2020). Those of diesel machinery, marine vessel, and residential coal combustion were determined by recent measurement results in China (Qi et al., 2019; Huang et al., 2018; Cai et al., 2019). The I/SVOCs-G-to-VOCs ratios and profiles of cooking and biomass burning emissions were derived from SPECIATE 5.1 database, while their particle-phase ratios and profiles were referenced from two previous studies (May et al., 2013; Louvaris et al., 2017). Table S1 and S2 show the I/SVOCs-G-to-VOCs and I/SVOCs-P-to-POA ratios and their

emission profiles of each specific source. The base emissions of AVOCs and POA (See Table S3) were taken from a high-resolution emission inventory for the year of 2017 developed in our previous study (An et al., 2021).

(3) Model input: Before being input into the model, the estimated I/SVOC-G and I/SVOC-P emissions were summed and then redistributed according to their phase equilibrium under their actual atmospheric state. The formula of phase equilibrium is shown in Equation (1).

$$F_{\mathrm{p}} = \frac{c_{\mathrm{OA}}}{c_{\mathrm{OA}}+C^*} \tag{1}$$

Where, F_{p} is the fraction of particle-phase I/SVOC emissions for each volatility bin under actual atmospheric state. C_{OA} represents the OA concentration under actual atmospheric state. We assumed it to be 10 μg·m^{-3} in this study. C^* is the effective saturation concentration of each volatility bin. After redistribution, the I/SVOC emissions for each source category were allocated into 4 km × 4 km grids and hourly temporal profiles using the same method as the criteria pollutants.

New references:
Louvaris, E. E., Florou, K., Karnezi, E., Papanastasiou, D. K., Gkatzelis, G. I., and Pandis, S. N.: Volatility of source apportioned wintertime organic aerosol in the city of Athens, Atmos. Environ., 158, 138–147, 2017.

2.2 Model configuration

POA was treated as semivolatile to account for its gas-particle partitioning and ageing process and segregated to several particle species, which varied in their volatility that quantified with the metric $C^* = 10^{-1}$ to 10^3 μg·m^{-3} (Donahue et al., 2006). I/SVOCs-P emissions from different sources were then speciated and input as semivolatile accordingly. The remaining POA emissions excluding I/SVOCs-P were treated as nonvolatile POC (primary organic carbon) and PNCOM (primary non-carbon organic matter).

2. It is surprising in Table 2 that there are no I/SVOC-P emissions from the industrial processes, aircraft, and coal combustion emissions. Do the authors consider this realistic, or is this an area where more data are needed?

Re: Thanks. We agree with the reviewer's comments. There should be some I/SVOCs-P emissions from the industrial processes, aircraft, and coal combustion like power plants and boilers. However, we didn't consider the I/SVOCs-P emissions from the above source for the following two reasons. One is that POA emissions from these sources were limited, which will not affect the results in this study. According to our base emissions inventory, their POA emissions accounted for 1.6%, 0.1% and 4.0% of total emissions, respectively. Second, the profiles of I/SVOCs-P components are difficult to obtain, which also indicates that more measurements of I/SVOCs emission sources are needed in the future. We have addressed the absence of these emission sources in the revised manuscript.

Changes in manuscript:
3.1.1 Source-specific I/SVOC emissions

I/SVOCs-P emissions were 82.96 Gg. The largest contributor of I/SVOCs-P emissions came from biomass burning and diesel vehicle, accounting for 53.24% and 11.88% of the total, followed by gasoline vehicle (5.23%), marine vessel (2.66%), diesel machinery (2.54%), and biomass burning (1.75%). Note that the I/SVOCs-P emissions from coal combustion (e.g. power plants, boilers, etc.), other industrial processes, and aircraft were not included in this study. On the one hand, the POA emissions (see Table S3) from these sources were limited, accounting for less than 5%, which could be expected that their I/SVOCs-P emissions were also relatively low. On the other hand, the profiles of I/SVOCs-P components of these sources were still difficult to obtain. More measurements of the I/SVOC emissions from these sources is very necessary in the future.

3. *Can the authors explain some of the interesting trends shown in Fig. 2. Specifically, I find it curious that there is a substantial fraction of I/SVOC-G in the C*=0.1 ug m-3 bin for agricultural sources. Also, the residential source has an odd volatility trend, with very little emissions in the lower IVOC range. Do the authors think this is realistic? I recommend replacing gas-phase and particle-phase in Fig. 2 with 'I/SVOC-G' and 'I/SVOC-P' or something similarly precise. These are not necessarily gas-phase or particle-phase emissions, but are instead emissions based on VOC and filter OC measurements (I assume; it's not really discussed in the description of the emission inventory).*

Re: Thanks. We agree with the reviewer's comments. The I/SVOCs-G and I/SVOCs-P profiles in Fig. 2 still cannot capture the actual emission characters in real-world. This is mainly due to the limited available I/SVOC emission measurements. Regarding the problem that the agricultural source has a substantial fraction of I/SVOCs-G in the $C^*=0.1$ μg·m^{-3}, we find there are some mistakes in the profile mapping after inspection. Therefore, we have made the following changes in the revised manuscript according to the comments. First, we have checked and corrected the profile data in Fig. 2 again. Then we summed the "Gas-phase" and "Particle-phase" emissions in each bin in Fig. 2(b-g) since the fractions of gas- and particle-phase emissions were determined by real atmospheric conditions, which was meaningless to be exhibited in the figures.

Changes in manuscript:
3.1.2 Volatility distributions of I/SVOCs

Figure 2 shows the volatility distribution of I/SVOC emissions from different sources as well as their gas-particle distributions under actual atmospheric state. The I/SVOC emissions generally showed an increasing trend with the increase of volatility. As shown in Figure 2(a), IVOC emissions (logC* bins at 3−6) accounted for 86% of the total I/SVOCs emissions, overwhelmingly dominated by industrial process and mobile sources. SVOCs (logC* bins at 0−2) and low-volatile organic compounds (LVOCs, logC* bins at -1) contributed to 11% and 3% of the total I/SVOCs emissions. In terms of the contributing sectors, mobile sources, industrial process, and solvent-use dominated the total I/SVOC emissions. While the IVOCs were equally contributed by above-listed three sources, residential and mobile sources dominated the SVOCs and LVOCs emissions.

We further investigated the contributions of different volatility bins to each source category. The mobile source was dominated by IVOC emission (88%). Note that IVOCs in vehicle exhaust are dominated by aromatics, which have faster OH reaction rates and higher SOA yields compared

to aliphatics in the same volatility bin (Zhao et al., 2016b; Drozd et al., 2019). Lu et al. (2020) therefore defined two additional lumped IVOC species with logC* bins at 5 and 6 to account for the aromatic IVOCs in vehicle exhaust according to the measurements in previous studies (Zhao et al., 2015; Zhao et al., 2016b). Here in this study, we also split the aromatic IVOC emissions from mobile sources and found that aromatic IVOCs accounted for 23% of the total I/SVOC emissions from the mobile source. The industrial process and solvent-use sources were also dominated by IVOC emissions, accounting for 81% and 97%, respectively. The volatility distribution of residential sources was relatively uniform, with IVOCs, SVOCs and LVOCs accounting for 40%, 30%, and 30%. Agricultural (i.e., biomass burning) sources were more concentrated in IVOCs, accounting for 76%, while SVOCs accounted for 24%. It should be noted that other than mobile sources, the emission profiles of the other sources were mainly derived from SPECIATE 5.1 database (US EPA, 2021) in this study, which may be inconsistent with real-world emissions in China. To further reduce the uncertainty in the I/SVOC emission inventory, measurements of I/SVOC emissions from different local sources are therefore important and urgently needed in the future.

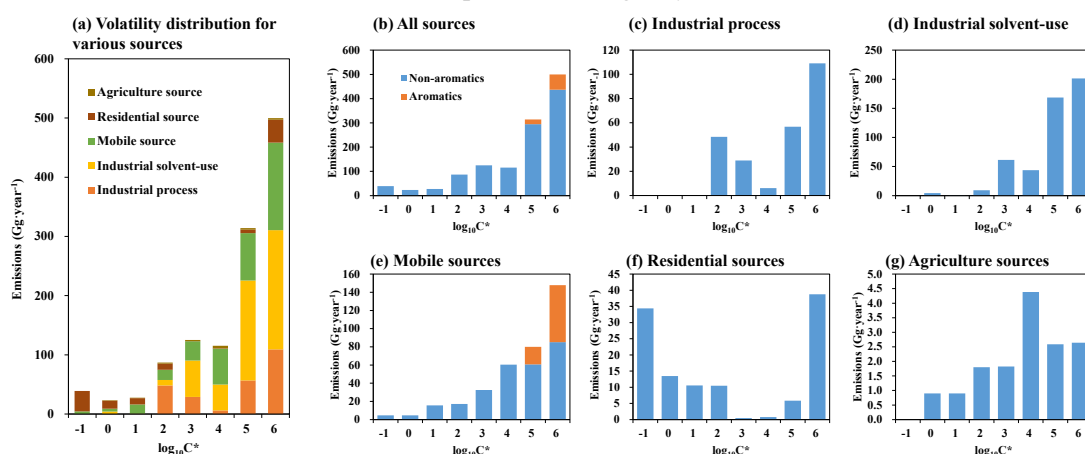



Figure 2. Volatility distributions of I/SVOCs emitted from different sources in the YRD region.

4. I was surprised to not see a multi-panel figure showing the spatial emissions of LVOC, SVOC, IVOC and VOC. Can this be added? This would aid comparison to contemporary studies like Chang et al. (2022; https://www.sciencedirect.com/science/article/pii/ S2590332222001488)

Re: Thanks for the reminding. We have supplemented the figures and associated descriptions of spatial emissions of VOCs, IVOCs, SVOCs, and LVOCs in the revised manuscript.

Changes in manuscript:
3.1.3 Spatial distributions of I/SVOC emissions in YRD region

Figure 3 compares the spatial distributions of AVOC, IVOC, SVOC, and LVOC emissions in the YRD region. The IVOC, SVOC, and LVOC emissions were largely concentrated in city clusters in eastern YRD, and hotspots can also be observed in the northern agglomerations. The distributions of I/S/LVOC emissions were generally consistent with that of the AVOC emissions in the region. Compared to the spatial distributions of I/S/LVOC emissions in Chang et al. (2022), our emissions had similar spatial distributions but at a higher resolution. Emission hotspots in urban areas can be captured more clearly in this study, which will help improve the simulation in urban areas.

Figure 4 shows the spatial distributions of source-specific I/SVOC emissions in the YRD region. There were considerable differences in the spatial distributions of I/SVOC emissions from different sources. The I/SVOC emissions from industrial sources (including industrial process and industrial solvent-use) were mainly concentrated in the eastern urban agglomeration, which was related to the developed industrial activities in the region. The I/SVOC emissions from mobile and residential sources clustered into multiple hotspots in urban areas, while emissions from agricultural sources were mainly distributed in northern YRD, where frequent agricultural activities exist.

We also compare the spatial distributions of I/SVOC emissions with those of POA and BVOCs. We found that POA emissions were more concentrated in urban centers associated with mobile and residential sources (See Figure S1). BVOC emissions in the YRD region were mainly distributed in the southern area, where AVOC and IVOC emissions were relatively low. The difference in the spatial distributions of I/SVOC, AVOC, BVOC, and POA emissions implies that the sources of organic components in different areas of the region are quite different, which will be discussed in the following sections.

New references:

Chang, X., Zhao, B., Zheng, H., Wang, S., Cai, S., Guo, F., Gui, P., Huang, G., Wu, D., Han, L., Xing, J., Man, H., Hu, R., Liang, C., Xu, Q., Qiu, X., Ding, D., Liu, K., Han, R., Robinson, A. L., and Donahue, N. M.: Full-volatility emission framework corrects missing and underestimated secondary organic aerosol sources, One Earth, 5, 403–412, 2022.

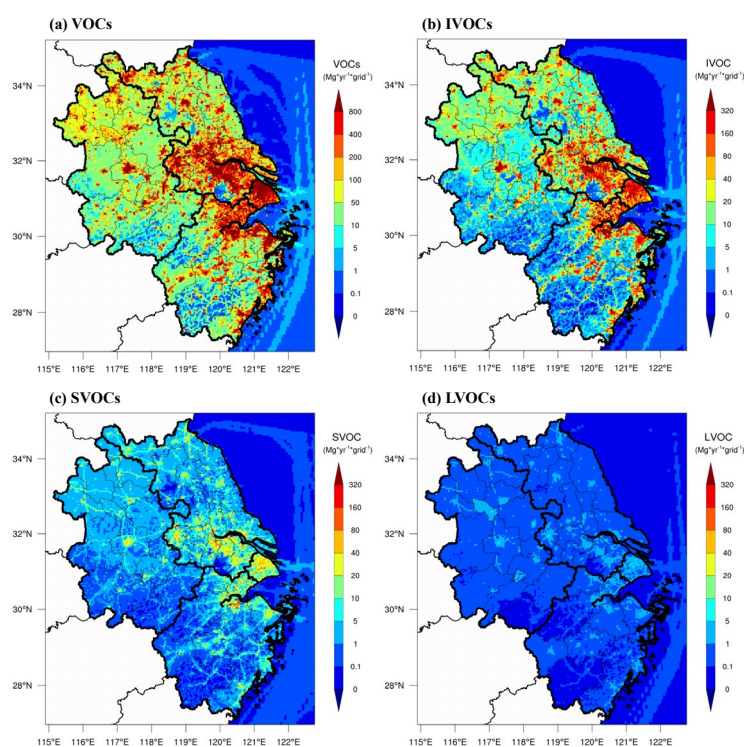



Figure 3. Spatial distributions of VOC, IVOC, SVOC, and LVOC emissions in the YRD region for the year 2017.

5. *In Fig. 6, it is surprising that the POA trend is relatively unchanged between the BASE and IMPROVE cases, when perhaps 60% of the POA is being evaporated (line 329-330). Can the authors shed some light here? Is most of the I/SVOC-P going into the LVOC bins? Judging by Table S2, it doesn't seem like it.*

Re: Thanks for the comments. We re-examined the simulations of POA concentrations for the IMPROVE simulation case after re-distributing the gas- and particle-phase I/SVOC emissions according to their phase equilibrium under the actual atmosphere state. The results indicated POA concentrations in the IMPROVE case decreased by 12%−20% compared with the BASE case. We have made the corrections in the revised manuscript.

Changes in manuscript:

3.2.1 Simulation results of OA concentrations

The seasonal average concentration of POA was 5.5 μg·m^{-3} in the BASE case, with the lowest in summer (3.8 μg m^{-3}) and the highest in winter (6.9 μg m^{-3}). High POA concentrations in winter was mainly induced by the stagnant meteorological conditions such as low wind speed and boundary layer height, and vice versa in summer. For the spatial distributions as presented in Figure 6, POA concentrations in northern YRD were high and mainly concentrated in urban areas, which was consistent with the distributions of POA emissions (Figure S1). The POA concentrations in the IMPROVE simulation decreased by 12%−20% compared with the BASE case. In the IMPROVE simulation, the POA was treated as semi-volatile, where gas–particle partitioning and multigeneration oxidation were considered (Murphy et al., 2017). Entering into the atmosphere, more semi-volatile compounds evaporated into gas-phase and then generated SOA through multigeneration oxidation, which reduced the POA concentrations relatively.

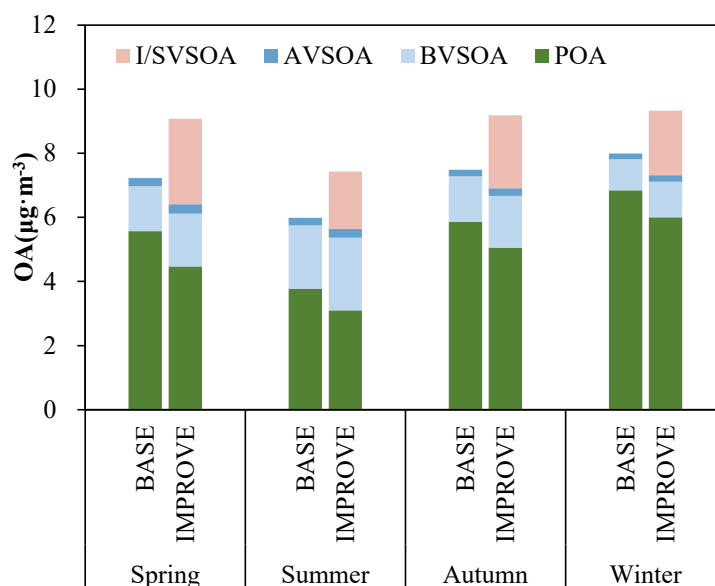



Figure 5. Comparisons of the regional average concentrations of POA and SOA formed from AVOCs, BVOCs, and I/SVOCs in different seasons from the BASE and IMPROVE simulations.

6. *Although the authors have done a commendable job in the writing, there are more than a*

few grammatically incorrect and awkward phrases throughout. I recommend the authors have a technical writer give feedback, or someone from the journal staff.

Re: Thanks. Grammar and writing have been checked by some technical writers in the revised manuscript.

Minor Suggestions/Typos:
1. *Line 53: Jathara --> Jathar.*

Re: Thanks. It has been revised.

2. *Line 53-54: Recommend rephrasing to: "It is challenging to constrain the abundance of OA precursors and to identify key sources."*

Re: Thanks. We have rephrased the sentence as the reviewer recommended.

3. *Line 57: Please add references for studies using the three source apportionment techniques. Also add a reference for the AMS to the next sentence.*

Re: Please see the following changes.

Changes in manuscript:
1. Introduction

Great efforts have been made in the identification of OA sources through source apportionment of the measured OA components, such as positive matrix factorization (PMF) (Zhang et al., 2011), chemical mass balance (CMB) model (Zheng et al., 2002) or multilinear engine (ME-2) (Canonaco et al., 2013). The Aerodyne high-resolution time-of-flight aerosol mass spectrometer (AMS), has been proven to be a powerful tool in quantification and chemical characterization of different OA components in real-time (Canagaratna et al., 2007).

New references:

Zhang, Q., Jimenez, J. L., Canagaratna, M. R., Ulbrich, I. M., Ng, N. L., Worsnop, D. R., and Sun, Y.: Understanding atmospheric organic aerosols via factor analysis of aerosol mass spectrometry: a review, Anal. Bioanal. Chem., 401, 3045−3067, 2011.

Zheng, M., Cass, G. R., Schauer, J. J., and Edgerton, E. S.: Source Apportionment of $PM_{2.5}$ in the Southeastern United States Using Solvent-Extractable Organic Compounds as Tracers, Environ. Sci. Technol., 36, 2361−2371, 2002.

4. *Line 74: model --> modeling*

Re: Thanks. It has been revised.

5. *Line 75: which advantages in --> which has advantages for*

Re: Thanks. It has been revised.

6. *Line 77-78: Please be careful here. Some models do include multi-generational oxidation of vapors. Are you referring to a specific model here? A specific CMAQ version? Note that CMAQ has included aging via oligomerization of anthropogenic and biogenic vapors for some time.*

Re: Thanks for the reminding. We have deleted this statement in the revised manuscript.

7. *Line 87: Recommend rephrasing 'due to the missing of' to 'due to the underestimation of'*

Re: Thanks. It has been revised.

8. *Line 102-104: Many of these profiles are not in SPECIATE5.1, and SPECIATE5.1 generally does not support lumped species by volatility. Recommend removing this sentence, or change 'most' to 'some'.*

Re: Thanks. We have removed this statement in the revised manuscript.

9. *Line 106: Is 30% really dominating? Maybe replace 'dominates' with 'contributes significantly to'?*

Re: Thanks. It has been revised.

10. *Line 116-127: Recommend moving to Methods section*

Re: Thanks for the comments. According to another reviewer's comments, we have rewritten this section to highlight the novelty of this study in I/SVOC emission estimation through a survey of established methodologies in previous studies. The specific modifications are as follows:

Changes in manuscript:
1. Introduction:
     I/SVOC emission inventories have been developed and applied into air quality models over the past decade. Most of them were estimated by applying different scaling factors based on their

relationship with POA, volatile organic compounds (VOCs), or some proxies like naphthalene (Pye and Seinfeld, 2010; Shrivastava et al., 2011; Jathar et al., 2017; Wu et al., 2019, 2021; Li et al., 2020, 2022; Ling et al., 2022). Yet in practice, a same scaling factor was applied to most of the sources in previous studies due to the lack of measurements on I/SVOC emission factors. For example, except biomass burning (0.75–1.5), Wu et al. (2019) utilized scaling factors of 8–30 for all of the other emission source categories, which was estimated based on the measurements of on-road mobile source. Li et al. (2020) assumed scaling factors of 1.5 for on-road mobile source, and 0.34–1.5 for the other sources, such as industrial and residential sources, which were much lower than the estimations in Wu et al. (2020). Huang et al. (2021) have tried emission factor method to quantify the I/SVOC emissions, yet the results were 60% lower than the scaling factor method, far from catching the measured amount of SOA. Obviously, roughly estimating I/SVOC emissions using one or two emission profiles as surrogates for all emission sources will create large uncertainties.

New references:

Jathar, S. H., Woody, M., Pye, H. O. T., Baker, K. R., and Robinson, A. L.: Chemical transport model simulations of organic aerosol in southern California: model evaluation and gasoline and diesel source contributions, Atmos. Chem. Phys., 17, 4305–4318, 2017.

Li, J., Han, Z., Wu, J., Tao, J., Li, J., Sun, Y., Liang, L., Liang, M., and Wang, Q.: Secondary organic aerosol formation and source contributions over east China in summertime, Environ. Pollut., 306, 119383, 2022.

Ling, Z., Wu, L., Wang, Y., Shao, M., Wang, X., and Huang, W.: Roles of semivolatile and intermediate-volatility organic compounds in secondary organic aerosol formation and its implication: A review, J. Environ. Sci., 114, 259–285, 2022.

Pye, H. O. T., Seinfeld, J. H.: A global perspective on aerosol from low-volatility organic compounds, Atmos. Chem. Phys., 10, 4377–4401, 2010.

Shrivastava, M., Fast, J., Easter, R., Gustafson, W. I., Zaveri, R. A., Jimenez, J. L., Saide, P., and Hodzic, A.: Modeling organic aerosols in a megacity: comparison of simple and complex representations of the volatility basis set approach, Atmos. Chem. Phys., 11, 6639–6662, 2011.

11. Line 199: What was used for biogenic emissions? MEGAN?

Re: Yes. Sorry forgot to explain in the text. We have supplemented the statement of biogenic emission estimation in the revised manuscript.

Changes in manuscript:
2.2 Model configuration

Biogenic volatile organic compounds (BVOCs) emissions were estimated based on MEGAN (the Model of Emissions of Gases and Aerosols from Nature) version 2.10 driving by inputs of the leaf area index (LAI) from MODIS product, plant functional types (PFT) base on remote sensing data, inline coupled emission factors and meteorology simulated by the WRF model. Detail configurations of MEGAN can be obtained from our previous study (Liu et al., 2018).

12. Line 206-207: The AERO7 CMAQ aerosol scheme does not include IVOCs with C* between 104 and 106 ug m-3. It also doesn't include aromatic IVOCs. Did the authors add these?

Re: Yes. We included a new-developed of VBS scheme recommended by Lu et al. (2020) in this study. The scheme included the OH oxidation of IVOCs with C* between 10^4 and 10^6 $\mu g \cdot m^{-3}$ and aromatic IVOCs. The description of this scheme is shown in the fifth paragraph in Section "2.2 Model configuration".

13. Line 228-230: Is the multigenerational oxidation scheme equivalent to that used in Lu et al and Murphy et al, or did the authors derive their own?

Re: The scheme was equivalent to that used in Lu et al. and Murphy et al. We have mentioned in the manuscript.

14. Table 1: Recommend reformatting to get rid of the repeated phrase "VOC and I/SVOC emissions in the region". Maybe change the column 'Emission settings' to something like 'Sources with added I/SVOC emissions'. Then the first row would be 'none', the second row 'all', and each latter row would be something like 'all except XX'. I don't think you need the Notes column.

Re: Thanks for the reviewer's recommendation. We have reformatted the table in the revised manuscript.

Changes in manuscript:

2.3 Model simulations

Table 1. Settings of simulation cases.

Name	Sources with added I/SVOC emissions
BASE	none
IMPROVE	all
CASE1	all except industrial process
CASE2	all except industrial solvent-use
CASE3	all except mobile sources
CASE4	all except residential sources
CASE5	all except biomass burning
CASE6	all except biogenic sources
CASE7	without VOCs and I/SVOC emissions
CASE8	all except gasoline vehicle
CASE9	all except diesel vehicle
CASE10	all except diesel machinery
CASE11	all except marine vessel
CASE12	all except cooking

15. Line 296-300: I don't understand these sentences. Please rephrase.

Re: Thanks for the reminding. We also find these sentences doesn't seem to make much sense. We have removed these statements in the revised manuscript.

16. Line 358-359: Please be careful here when referring to species as 'gas-phase'. You have chosen to plot these data as particle-phase or gas-phase, which I don't think is the best way to go, since in reality, they are just scaled off the particle-phase and gas-phase emissions, respectively. They could be in either phase when emitted.

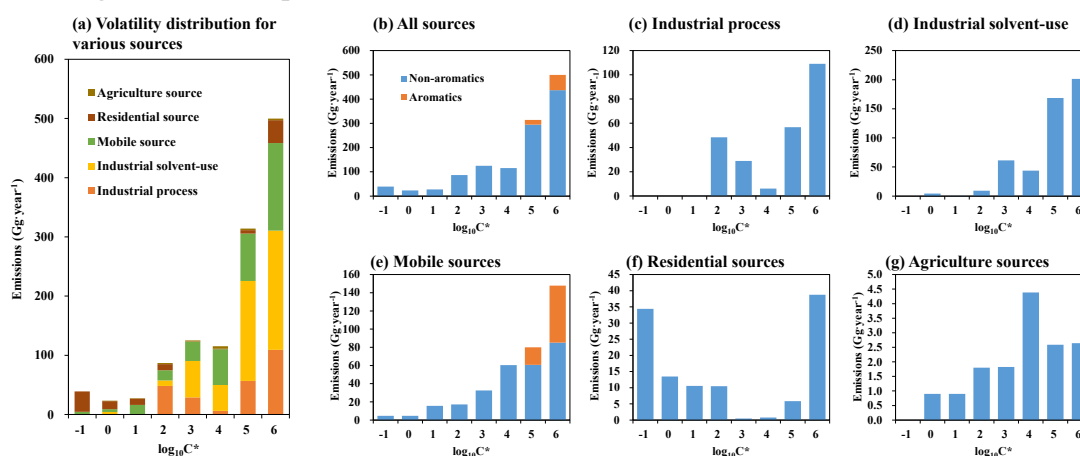
Re: Thanks for the reminding. We understand the reviewer's suggestion that the gas- and particle-phase of the I/SVOCs components are dynamically changing and should not be simply divided. Therefore, we have merged the gas- and particle-phase data of I/SVOCs in each volatility bin in Figure 2 in the revised manuscript.

Changes in manuscript:

Figure 2. Volatility distributions of I/SVOCs emitted from different sources in the YRD region.

17. There are two tables labelled Table S6.

Re: Thanks. It has been revised.

18. 5: Color axes are difficult to read. Please enlarge them.

Re: Thanks. We have enlarged the color axes of the figures.

Changes in manuscript:

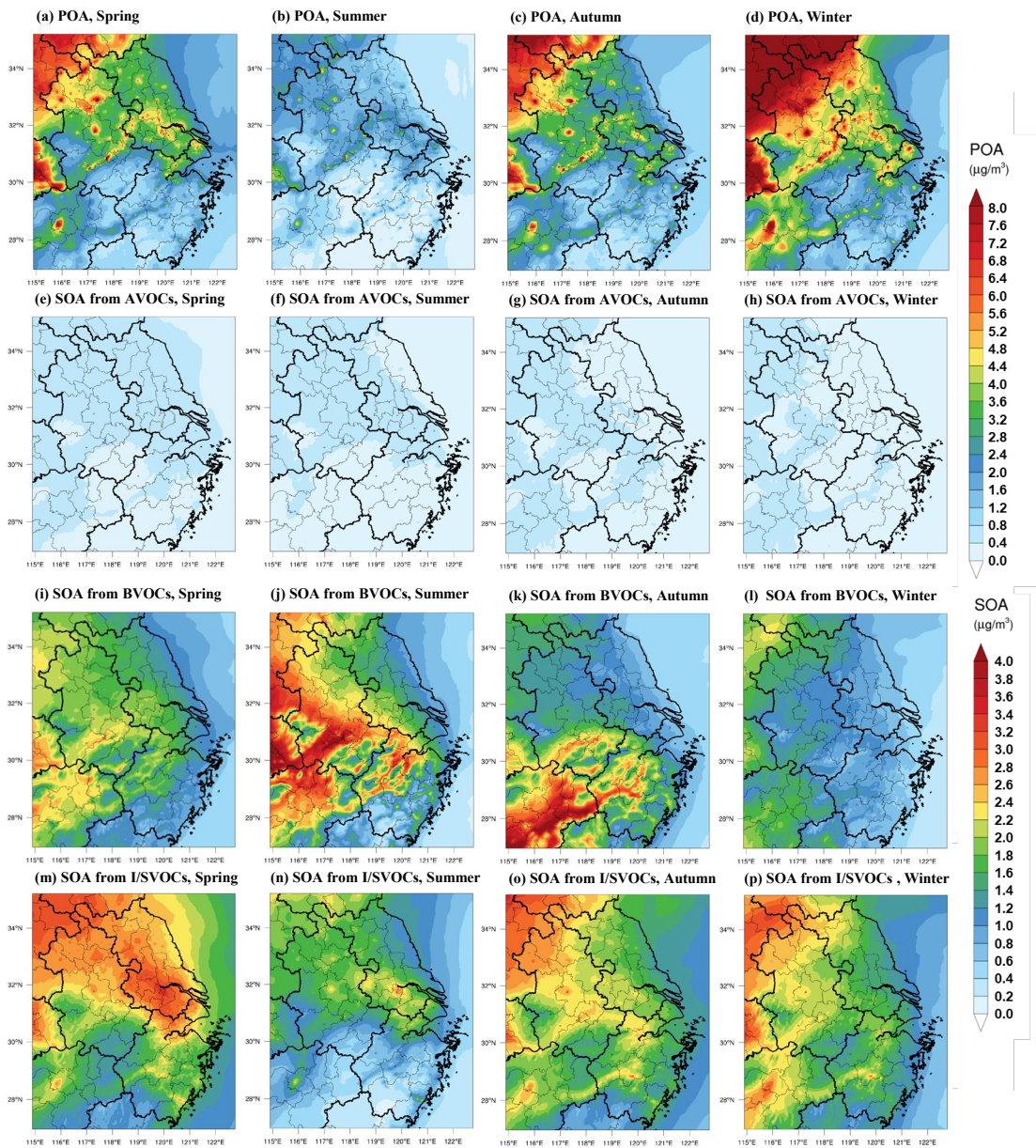



Figure 6. Spatial distributions of modeled POA and SOA formed from AVOCs, BVOCs, and I/SVOCs in different seasons in the IMPROVE simulation.

19. Section 3.3: I recommend adding a few sentences comparing to the YRD-specific source contributions reported by Chang et al. (2022) since the goals of these studies are highly related.

Re: Thanks for the comments. We have supplemented some statements on the comparison with the results reported by Chang et al. (2022) in the revised manuscript.

Changes in manuscript:
3.3.1 POA and SOA sources in the region

    Our results were generally similar with those of Chang et al. (2022) for YRD region. We both

found the domestic combustion mainly engaged in cooking emissions had a major contribution to OA. Next was volatile chemical products (VCPs), especially the use of solvents, paints, and adhesives in industrial sector, also made a high contribution. Note that industrial process also took up a high fraction in our OA simulation, while it was lower in Chang et al. (2022)'s study. The difference in I/SVOC emission estimates was the main reason for this divergence. Mobile sources in both studies had similar contributions, which accounted for about 10% to total OA. Comparatively, our source classification was more specific, which will help identify more specific OA sources to design more refined regional control countermeasures.

20. Line 610: supe-regional --> super-regional

Re: Thanks. It has been revised.

Anonymous Referee #3, 26 Nov 2022

Major comments:

7. Lines 165-193 and lines 246-262: I am disinclined to use the label 'I/SVOC-P' since IVOCs (assuming 'I' stands for IVOCs in the label) shouldn't really be in the particle phase. The ratios the authors are using from Lu et a. (ACP, 2020) and likely the other sources are to determine SVOCs only. This issue was raised by referee #2

Re: Thanks for the reviewer's comments. According to this comment and that raised by referee #2, we have changed the labels of 'I/SVOC-G' and 'I/SVOC-P' to be 'gas-phase' and 'particle-phase'. The labels of 'I/SVOCs-G-to-VOCs' and 'I/SVOCs-P-to-POA' were changed to be 'G-ratio' and 'P-ratio' in the entire text.

8. I would recommend the authors use a different name for the 'IMPROVE' simulation since it doesn't convey how it is different from the 'BASE' simulation.

Re: Thanks for the comments. We have changed the name of 'IMPROVE' to be 'I/SVOC-E' in the whole text.

9. Section 3.2.2: This is an important section of the paper, but it seems like the authors overgeneralize the model-measurement comparison and give the model too much credit; I found this to be a common theme in the paper. For instance, while the model holds diurnal trends across seasons, it isn't clear that the observations do the same, particularly in the winter months. It is easy to claim that the 'model works' but it comes at the cost of ignoring useful insight that the comparison sheds.

Re: Thanks. We agree with the reviewer's comments and make some revisions in the revised manuscript as follows.

Changes in manuscript:

The I/SVOC-E simulation also demonstrated improvements in reproducing the temporal variations of SOA, especially during the daytime (Figure 7e−7h). Compared with the BASE simulation, evident increases in SOA concentrations during daytime can be observed in I/SVOC-E simulation (Figure 7e−7h), which agrees better with the observation. However, the model is still hard to capture the diurnal patterns of SOA observed in most seasons, except for the summer, when both the concentrations and diurnal variations of SOA are well reproduced, which indicates that SOA in summer is mainly subject to photochemical oxidation of I/SVOC emissions, while SOA formation will be largely affected by other factors in other seasons, especially during the nighttime in cool seasons. For example, an increasing body of experimental and observational evidence suggest that heterogeneous and multiphase reactions also played important roles in SOA formation especially during pollution episodes (Guo et al., 2020; Kim et al., 2022). Recent studies also found that nocturnal NO_3 oxidation was also an important route for SOA formation, which would drive

the enhancement of SOA during the nighttime (Yu et al., 2019; Decker et al., 2021). Yet mechanism and parameterizations of these processes remain unclear, making the involvement of these processes in the model difficult. In addition, I/SVOC emissions from outside of the YRD region might be underestimated due to the lack of detailed base emission inventory, resulting in the corresponding underestimation of the transported SOA, which were prominent especially in autumn, winter and spring in Shanghai. High-resolution I/SVOCs emissions inventory is urgently needed to be developed at a larger regional scale.

10. Too much precision (3/4 significant figures) in the numbers presented in the text?

Re: Thanks. We have reduced the retained significant figures in the revised manuscript.

11. Figure 8: What are the reasons for why the spatial distribution of SOA changes between seasons for the same source? I understand that this could be the case for biogenic VOCs but I am confused why one would expect this for mobile or industrial sources, emissions from which should be less sensitive to seasons.

Re: Thanks for the comments. The main reason for the spatial difference of each season is the different dominant wind direction. In the Yangtze River Delta region, northwest wind prevails in spring, southeast wind prevails in summer, and northeast wind prevails in autumn and winter. However, in general, the SOA spatial distribution of the same source in different seasons is not very different, which has been explained in the original text.

12. While the emissions, chemistry, and SOA potential are much better understood for I/SVOC emissions from mobile sources, that isn't necessarily the case for non-tailpipe sources of I/SVOCs (e.g., solvent use). This needs to be discussed in the final section of the paper.

Re: Thanks for the reviewer's comments. We have made some revisions for this section in the revised manuscript as follows.

Changes in manuscript:

With the addition of source specific I/SVOC emissions, we successfully quantified the contribution of each source to POA and SOA concentrations in YRD. For POA, cooking emission is the predominant source, which concentrates in urban area of YRD in accordance with the population distribution. For SOA, for the first time, we demonstrate that I/SVOCs from industrial sources are dominant contributor, followed by those from mobile sources. In summer, the contributions of biogenic emission to total SOA are also non-negligible, especially for the cities in southern YRD. Spatial and seasonal variations in the source contributions suggest that control strategies for OA pollution should vary by cities and seasons. For urban area, cooking emissions has been emerging as an important POA source, not to mention their impacts on SOA formation are not yet certain. Our results suggest the control measures on the cooking emissions should be

strengthened in the future for the further reduction of POA. Another important source of SOA in urban area is mobile source, especially gasoline and diesel vehicles. Reduction in I/SVOC emissions from vehicles are effective measures in the mitigation of urban air pollution, which is also technically feasible as has been demonstrated in Qi et al. (2021). Continuous improvement in emission standards is one way to promote the reduction of motor vehicle related SOA. Our study further reveals that non-tailpipe sources of I/SVOCs (e.g., solvent use, petrochemical, etc.) are major contributors to SOA formation in the YRD region, consistent with Chang et al. (2022)'s model results in the national scale. However, current understanding of SOA formation potentials from these sources are still far from enough. For example, the localized I/SVOC emission factors and source profiles of these sources are still missing. Their chemical behavior and SOA yields may be different from the emissions from mobile sources which have been widely studied, which urges in-depth studies on these sources as well as the corresponding control measures.

13. For referee #2, comment #2, consider the work of Morino and coworkers: https://doi.org/10.1021/acs.est.8b01285 and https://doi.org/10.1016/j.atmosenv.2022.119319.

Re: Thanks for the reviewer's suggestion. The work of Morino and coworkers is interesting. The work of Morino and coworkers is interesting, which to some extent complements the contribution of stationary source emissions from the industrial and energy sectors to organic aerosols, and improves the simulation of OA in winter. At present, their results cannot be included in the model system in this study, but we discussed them in Section 3.2.2, see the revised manuscript.

Changes in manuscript:
A recent study furtherly found that there were considerable emissions of condensable organic aerosols from stationary sources in the industrial and energy sectors, which would effectively improve the contributions of the industrial sector to OA simulation especially in winter, should also be considered in the future (Morino, et al., 2018; Morino, et al., 2022).

New references:
Morino, Y., Chatani, S., Tanabe, K., Fujitani, Y., Morikawa, T., Takahashi, K., Sato, K., and Sugata, S.: Contributions of condensable particulate matter to atmospheric organic aerosol over Japan, Environ. Sci. Technol., 52, 8456−8466, 2018.
Morino, Y., Chatani, S., Fujitani, Y., Tanabe, K., Murphy, B. N., Jathar, S. H., Takahashi, K., Sato, K., Kumagai, K., and Saito, S.: Emissions of condensable organic aerosols from stationary combustion sources over Japan, Atmos. Environ., 289, 119319, 2022.

14. For referee #2, comment #13, consider adding details about the multigenerational oxidation scheme to the methods section.

Re: Thanks. We have supplemented some details about the multigenerational oxidation scheme in the revised manuscript.

Changes in manuscript:

Multi-generation oxidation was considered by implementing further oxidation of the vapors from the initial oxidation, which redistributes the mass across the volatility bins of $C^* = 10^{-2}$ to 10^2 $\mu g \cdot m^{-3}$, and thus fragmentation and functionalization were included. It is worth noting that only one-step oxidation of the vapor products was considered, using the default aging scheme for the oxidation products of POA in the CMAQ (Murphy et al., 2017). Additionally, SOA formation from SVOCs were treated similarly, and more details can be found in Murphy et al. (2017).

Minor comments:
21. Line 72: Given the hard ionization in the AMS, there are limits to how much source information can be extracted from AMS data.

Re: Thanks for the reminding. We have considered the reviewer's comments in the revised manuscript.

Changes in manuscript:

However, given the hard ionization in the AMS, there are limits to how much source information can be extracted from AMS data. Further deconvolution on the contributions of different sources to OA production is challenging.

22. Line 82: I don't think that's right. Material in volatility bins is moved as opposed to moving the bins themselves.

Re: Thanks. We have rephrased the sentence as the reviewer recommended.

23. Line 83: It should be made clear that VBS is a framework to represent OA, i.e., the implementation of a VBS does not improve model performance. As the authors already know, it's how the VBS is configured and parameterized is what makes it useful and powerful.

Re: Thanks. We agree with the reviewer's comments. Each study has improved the configuration and parameters of VBS in the model. We have re-written this sentence in the revised manuscript as follows.

Changes in manuscript:

Previous studies have successively configured the VBS scheme from one-dimensional (1-D) to 1.5-/2-dimensions (1.5-/2-D), which can better describe the evolution of OA in the 2-D space of oxidation and volatility in the model, and coupled the simplified emission inventory of SOA precursors estimated from POA to improve the model performance on SOA simulation (Tsimpidi et al., 2010; Koo et al., 2014; woody et al., 2016; Zhao et al., 2016a; Yang et al., 2019).

24. Line 110: 'catching'?

Re: Thanks. It has been revised to be 'reproducing'.

25. Line 146: By definition, IVOCs are unlikely to partition to the particle phase at typical environmental conditions.

Re: Thanks. We have deleted this sentence in the revised manuscript.

26. Line 162-163: What is an example of residential solvent use? What was the reason for not including biomass burning?

Re: Thanks for the reminding. We have deleted this statement in the revised manuscript.

27. Line 226: missing 'and'?

Re: Thanks. We have added 'and' in the revised manuscript.

28. Line 311-313: Rephrase SO2 sentence.

Re: Thanks. We have rephrased this sentence in the revised manuscript as follows.

Changes in manuscript:

The simulated SO_2 was slightly overestimated, which might be caused by the overestimation of SO_2 emissions due to the fact that China's SO_2 emission reduction was far beyond the expectation.

29. Line 313: 'In contrast'?

Re: Thanks. It has been revised.

30. Section 3.1.1: What would be useful is for the authors to discuss why the IVOC-to-VOC ratios are different between sources.

Re: Thanks for the comments. We have modified the statement in 10. Section 3.1.1 as follows. More detailed discussion may require subsequent experimental studies.

Changes in manuscript:

The chemical materials and production process of these industries were quite different, which

would make their G-ratios quite different in the profiles.

31. Line 376-379: Lu et al. (ACP, 2018) claim that IVOCs from mobile sources are dominated by cyclic and branched alkanes so I take objection with this statement about aromatics being important (I do find it odd that Lu et al. (ACP, 2020) allot half of the IVOC range emissions to aromatics). More clarification is needed.

Re: Thanks for the comment. It is really hard to say that the aromatics are dominant in the IVOCs emissions from vehicles, so we have modified this sentence in the revised manuscript as follows.

Changes in manuscript:
Note that IVOC emissions from vehicles included a certain fraction of aromatics, which have faster OH reaction rates and higher SOA yields compared to aliphatics in the same volatility bin (Zhao et al., 2016b; Drozd et al., 2019)

32. Table S1: Are the IVOCP4-6 the same as the IVOCP4-6-ALK in Lu et al. (ACP, 2020)?

Re: Sorry for the mistake. The IVOCP4-6 fractions are also referenced from SPECIATE 5.1 (although the fractions in the database should be derived from Lu et al. and coworkers). We have revised the references in Table S1.

33. Lines 448-455: The IVOC model comparison with observations is less than impressive. The model is higher in the summer (9 versus 6.8) and the model is lower in the winter (12.4 versus 18.2). Discussing if this is an emissions or chemistry (or both?) issue would be helpful.

Re: Thanks for the comment. We have added some discussion in the revised manuscript as follows.

Changes in manuscript:
The modeled IVOCs was higher in summer while lower in winter, not to mention the diurnal patterns and spatial distributions also remained unknown. This may be due to the difference in monthly profiles of I/SVOC emissions, which has not been considered in this study. Another important reason should be the chemical mechanism of IVOCs to SOA evolution still needs to be improved. Continuous long-term measurements of I/SVOC at multiple locations are strongly recommended in the future to help to improve the SOA model performance.

34. Line 471: Effect of stronger photochemistry in the summer?

Re: Thanks for the reviewer's reminding. We have modified this sentence in the revised manuscript.

35. Figure S3: Why are there so few OC measurements to compare against? Or have these been averaged for the entire season? Is the model able to capture intra-season variability at each of the sites?

Re: Due to limited access to data, we used the average OC concentration of the whole season for comparison. We used the observation data from Shanghai Supersite to compare the intra-season variations in this study. In the future, we will try to make more fully comparisons on the daily or hourly variations of other sites with more sufficient data.

36. Figure 7: It needs to be made clear how the modeled species representing OA as well as the different AMS factors are aggregated to perform comparisons for 'POA' and 'SOA'. My sense is that there are uncertainties in this aggregation and it is not self-evident that the AMS gets you clean POA and SOA data to compare model predictions against.

Re: Thanks. We agree with the reviewer that uncertainty exist during the deconvolution of the factors by AMS-PMF analysis. To support factor separations and interpretations, in our current study as well as in two of our companion studies (Huang et al., 2021; Zeng et al., 2022), multiple high time-resolution mass spectrometric techniques were applied and multiple source apportionment methods, i.e., AMS-PMF, Molecular-Marker-PMF, Integrated-PMF were compared to improve the separation and accuracy of the factors. For example, the collocating measurements of primary and secondary organic tracers from cooking (i.e., oleic acid, azelaic acid) sources enable us to further separate an oxygenated cooking organic aerosol factor (OCOA) from the original "lumped" COA factor, yielding a narrowed range for the contributions from COA (Huang et al., 2021). Similarly, an oxygenated hydrocarbon-like organic aerosol (OHOA) was also resolved based on the measurements of vehicular related organic tracers (i.e., alkanes and hopanes). The oxygenated factors were thus aggregated into the SOA, which originally might be counted as POA in traditional AMS-PMF analysis. Our comprehensive measurements on OA components provide the up-to-date most adequate deconvolution of POA and SOA factors from the perspective of field measurements. To make it clear to the readers, we have revised in the manuscript as follows.

Changes in manuscript:

To further validate the model performance, we compared the simulation results of POA and SOA with those resolved through PMF analysis of organic mass spectral data acquired by an AMS at the SAES supersite. Note that uncertainty exist when directly compare the modeled OA factors with those resolved by AMS-PMF analysis since a clear split of POA and SOA from a measurement point of view can hardly be achieved. To minimize the uncertainty associated with the PMF analysis, comprehensive molecular identification of OA components was conducted and multiple source apportionment model results were compared following the method in Huang et al. (2021a) to improve the accuracy of the factor separation.

Response to the Editor,

Scientific comments:

l. 15: What do you mean by 'absence of precursors'? Do you mean 'the fact that precursors of intermediate-volatility and semi-volatile organic compounds (I/SVOCs) are not included in models...'?

Re: Thanks for the comments. This statement is really inappropriate. We have modified this sentence to "The fact that precursors of intermediate-volatility and semi-volatile organic compounds (I/SVOCs) are not included in models has a significant impact on the performance of SOA simulation."

l. 74: Please rewrite this sentence. 'Large gaps' should be clarified. I guess you refer to the underprediction of SOA concentrations by models.

Re: Thanks. We have rewritten this sentence to "the models generally underpredicted the measured SOA concentration in the atmosphere."

l. 79: Are indeed the volatility bins shifted? To my understanding, in the VBS, the volatility bins are fixed but compounds are sorted into the different bins depending on their volatility. Please clarify.

Re: Thanks for the comments. We agree that the statement "*volatility bins shift*" is incorrect. In fact, what we want to state is the proportion of compounds in different volatility bins are shifting. We have revised this sentence to "by shifting the proportion of these compounds in different volatility bins".

l. 80ff: "Previous studies have successively configured the VBS scheme from one dimensional (1-D) to 1.5-/2-dimensions (1.5-/2-D), which can better describe the evolution of OA in the 2-D space of oxidation and volatility in the model, and coupled the simplified emission inventory of SOA precursors estimated from POA to improve the model performance on SOA simulation."

The sentence is overall too long and confusing. Please split it into two.
1) What is the parameter of the second dimension in the 2-dimensional VBS? The first parameter is volatility, but 'Oxidation' is a process; how can this be accounted for in the scheme. Do you mean 'oxidation state'?
2) The second part of the sentence ('...and coupled...') is not clear either. What is a simplified emission inventory? Why was it simplified and compared to what? Are you implying that POA are SOA precursors?

Re: Thanks. We agree with the comments. We have made some correction for the first part of the

sentence. For the second part, we originally wanted to state that the emission inventories of precursors developed in the previous studies were roughly estimated using the POA scaling factor methods. Considering that this meaning will be uniformly introduced in the next paragraph, we decide to delete the second part of this sentence without affecting the overall purpose of this paragraph. In the revised manuscript, this sentence has been rewritten to "Previous studies have successively configured the VBS scheme from one-dimensional (1-D) to 1.5-/2-dimensions (1.5-/2-D), which can better describe the evolution of OA in the 2-D space of volatility and oxidation state in the model."

l. 89: Do you mean 'limitation' rather than 'constraint'?

Re: Thanks. It has been revised.

l. 133/4: What is a 'full volatility organic emission inventory'? It might be clearer to reword it as 'emission inventory that takes into account ....'

Re: Thanks. We have revised this sentence to "Chang et al. (2022) developed an emission framework that achieves a full volatility coverage in both the gas and particle phases of organic compounds for China".

l. 136: Can you quantify 'gaps'? Underprediction by 'x' percent?

Re: Thanks for the comments. In fact, Chang et al's work has largely improved the model performance of SOA simulation. It is not appropriate to emphasize the underprediction. We decided to rewrite this sentence from the perspective of improving the spatial and source resolution. In the revised manuscript, we have revised this sentence to "However, detailed source contributions of SOA in city scale still needs to be refined. Studies on high-resolution I/SVOC emission inventory for more specific sources are highly needed."

l. 252: Doesn't the sentence 'It is worth noting... ' contradict the preceding one? In the sentence starting in line 249, you mention 'multi-generation oxidation' whereas in the second sentence you state only one generation was considered. Please clarify.

Re: Sorry for the unclear expression. The meaning of "one-step oxidation" here is different from that of "one-generation oxidation". The former refers to the parametric treatment, while the latter refers to actual reaction. "one-step oxidation" means that there is only one reaction when parameterizing, but this parameter is obtained according to the smog chamber experiments, including second, third and even fourth generation oxidations. However, only one step from organic vapors to product is considered in parameterization in the model. To avoid confusion, we have rewritten this sentence to "Multi-generation oxidation was considered by implementing further

oxidation of the vapors from the initial oxidation, which redistributes the mass across the volatility bins of C* = 10^{-2} to 10^{2} μg·m^{-3}, and thus fragmentation and functionalization were included. The further oxidation of the vapor products used the default aging scheme for the oxidation products of POA in the CMAQ".

l. 259: '...and input as semivolatile accordingly' – is not clear. Do you mean 'considered as semivolatile species accordingly'?

Re: Yes. We have rewritten this sentence to "Particle-phase emissions from different sources were then speciated and input considered as semivolatile species accordingly."

l. 271: The semivolatile emissions that were particle phase'.... – could you simply say 'Emissions of semivolatile POA..' – if not, please clarify.

Re: Thanks. We have rewritten this sentence to "Emissions of semivolatile POA were treated with variable gas–particle partitioning and multigenerational aging in this simulation case."

l. 450ff: "The modeled IVOCs was higher in summer while lower in winter, not to mention the diurnal patterns and spatial distributions also remained unknown. This may be due to the difference in monthly profiles of I/SVOC emissions, which has not been considered in this study."
1) As also mentioned by the last reviewer, some more discussion would be useful here. Even though you do not have the exact monthly emission profiles, can you be a bit more explicit on they qualitatively may vary with season or month and how this could play out for the IVOC profiles?
The explanation that the chemical mechanism might be responsible for not being able to reproducing the trends is not convincing.
2) "not to mention the diurnal patterns and spatial distributions also remained unknown" is not clear. Please reword.

Re: Thanks for the comments. 1) We have supplemented the statement of seasonal difference of IVOC emissions to discuss the difference of modeled and observed IVOC concentrations. Then we deleted the explanation about the chemical mechanism. 2) This statement does not matter. We have deleted it. The whole sentence has been revised to "The modeled IVOCs was higher in summer while lower in winter. This may be attributed to the unreasonable estimate of monthly profiles of I/SVOC emissions. In this study, I/SVOC emissions in winter were only 5% higher than those in summer, consistent with the trends simulated by the model, but far from reaching the large difference (~2.7 times) between the observed concentrations in winter and summer."

l. 495: What do you mean by 'better meteorological conditions'?

Re: Sorry for the mistake. We mean "more favorable diffusion conditions" here. In the revised manuscript, it has been rewritten to "The low concentration in summer was likely due to more favorable diffusion conditions than the other seasons."

l. 522 -524: "Note that uncertainty exist when directly compare the modeled OA factors with those resolved by AMS-PMF analysis since a clear split of POA and SOA from a measurement point of view can hardly be achieved."
Please clarify this sentence. What do you mean by 'modeled OA factors'? POA and SOA or the various SVOC, I/SVOC etc categories? I understand that the PMF of AMS results gives different factors – but can't some of them be ascribed to POA and SOA?

Re: Thanks. "modeled OA factors", we meant to say that the POA and SOA concentrations modeled by the CMAQ. And yes, as the reviewer said, AMS-PMF did provide detailed OA factors, such as less oxygenated organic aerosol, more oxygenated organic aerosol, etc. We ascribed the PFM resolved OA factors into POA (hydrocarbon like organic aerosol and cooking organic aerosol) and SOA (including all oxygenated organic aerosol factors). We have rewritten this sentence to "Note that uncertainty exist when directly compare the POA and SOA derived from the model with those resolved by AMS-PMF analysis since a clear split of POA and SOA from a measurement point of view can hardly be achieved."

l. 554: "For example, an increasing body of experimental and observational evidence suggest that heterogeneous and multiphase reactions also played important roles in SOA formation especially during pollution episodes" reads as if you imply that heterogeneous and multiphase reactions are 'other factors', i.e. different from the photochemical oxidation in the summer. However, also heterogeneous and multiphase reactions in the summer can be oxidation reactions, initiated by photochemistry. Please clarify.

Re: Thanks. It's really unclear here. What we want to say is the heterogeneous and multiphase reactions have not been included in the model of this study but have important contributions to SOA formation especially during pollution episodes. In the revised manuscript, we have rewritten this sentence to "For example, heterogeneous and multiphase reactions have not been included in the model of this study but played important roles in SOA formation especially during pollution episodes in cool seasons."

l. 562: What do you mean by 'condensable organic aerosols'?

Re: "condensable organic aerosols" refers to the organic compounds in condensable particulate matter. To make the statement clearer, we have rewritten this statement to "A recent study furtherly found that there were considerable emissions of condensable particulate matter (CPM) from stationary sources in the industrial and energy sectors, which would effectively improve the contributions of the industrial sector to OA simulation especially in winter, should also be

considered in the future".

Technical comments:
l. 102: replace 'a same' by 'the same'

Re: Thanks. It has been revised.

l. 219: The link to MOZART does not work. Please update.

Re: Sorry, we found that the link was invalid since March 18, 2022. Therefore, in the revised manuscript, we added a reference to explain.

New references:
Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J. F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., Laepple, T., Orlando, J., Tie, X., Tyndall, G., Wiedinmyer, C., Baughcum, S. L., and Kloster, S.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geosci. Model Dev., 3, 43–67, 2010.

l. 254: replace 'were treated' by 'was treated'

Re: Thanks. It has been revised.

l. 257: 'that quantified with the metric' can be simply replaced by 'with'

Re: Thanks. It has been revised.

l. 467: replace 'was' by 'were'

Re: Thanks. It has been revised.

l. 549: 'the model is still hard to capture the diurnal patterns' is not correct English. Replace by 'The model cannot fully capture the diurnal patterns...' or something similar.

Re: Thanks. It has been revised according to your comment.

Additional changes:
In the author list, Dr. Jingyu An is studying in Shanghai Key Laboratory of Atmospheric

Particle Pollution and Prevention, Department of Environmental Science and Engineering, Fudan University at the same time, so we added another affiliation in the revised manuscript.