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. Brasseur, 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.

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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.

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.

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 $\mu g\cdot m^{-3}$. 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 $\mu g\cdot m^{-3}$ 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.

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 μg·m^{-3}, two aromatic IVOC bins with the C*=10^5 and 10^6 μg·m^{-3}, and four lumped SVOC bins with C* from 10^{-1} and 10^2 μg·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).

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

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.

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, μg·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, $\mu g \cdot m^{-3}$)					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 μg·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 μg·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 μg·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 μg·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.