Dear Editor,

Thank you for handling our submission and giving us so careful comments. Your comments are very valuable to improve our manuscript. We have fully addressed all your comments. Please find below our point-by-point response.

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