

We thank the reviewers for the effort to review the manuscript and to provide constructive comments and good suggestions to improve our manuscript. Our replies to the comments and our actions taken to revise the paper (in blue) are given below (the original comments are copied here).

The modifications corresponding to the comments are labeled **in red color and highlighted**.

The language and grammar in the revised manuscript have been edited carefully and polished native English speakers by according to the reviewers' comments. (labeled in red color)

Referees' comments:

Referee #1

The microphysical processes of organic aerosol have not yet been well represented, which lead to large uncertainties in current simulation studies. In this article, authors used a new global-regional nested aerosol model combined a particle microphysics module and a volatility basis-set organic aerosol module to simulate microphysical processes of organic aerosol. The model can reproduce the organic aerosol components and the particle number size distribution in Beijing, and spatial distributions of organic carbon and number concentrations of particles condensation larger than 10 nm. They further explored the model's sensitivity to the size distribution of primary emission and volatility distribution of primary organic aerosol. I am glad to see the amount of work presented in the manuscript. This reviewer doesn't find apparent flaw in the method and data the manuscript shows. I think the manuscript can be accepted after the following concerns are addressed.

Reply: Thanks to the reviewer for providing good suggestions to improve our work.

General Comments:

1. The writing is a bit difficult to understanding in many places, which leaves itself open to misinterpretations or confusion, and so the paper could really use a thorough edit from a native English speaker.

Reply: The writing has been edited carefully and polished by native English speakers according to the reviewers' comments. The major revisions are showed in red color in the revised manuscript.

2. To provide a reliable foundation for further analysis, a comprehensive model evaluation including aerosol optical depth, $PM_{2.5}$ is recommended.

Reply: Due to space limitation and continuity of the article, the comparison of annual mean global aerosol optical depth between simulation and MODIS data and $PM_{2.5}$ evaluation in Beijing and surrounding cities are added in the supplement material. The description of $PM_{2.5}$ evaluation in Beijing and surrounding cities is also added in the revised manuscript. (seen in Line 447-449)

Special Comments:

1. Line 85, “also” may be deleted.
2. Line 92-93, “the complication of processes and the mechanisms not well understood” may be replaced with “the unclear complication of processes and the mechanisms”.
3. Line 96, “find” should be “found”.
4. Line 158, “indicate” should be “indicated”.

Reply: The above modifications have been made.

5. Line 266, “primary OA (POA)” should be “POA”.

Reply: "POA" is the first appearance and so “primary OA (POA)” is used here.

6. Line 306, “When necessary, SP-LV is redistributed to size-bins ...”, please clarify the specific situations.

Reply: The situations to redistribute SP-LV to size-bins include: (1) calculating the particle size in order to simulate the condensation growth and coagulation of secondary particles; (2) the coagulation scavenging of secondary particles by primary particles. The related descriptions were added in the revised manuscript (seen in Line

320-321).

7. Please provide the full name for “LV-OA” and “POC” at the first appearance.

Reply: The full name for “LV-OA” were provided in Line 328 and “POC” in Line 330.

8. Line 378, “More details on the observation can be found in the published paper (Du et al., 2017)” may be “More details on the observation can be found in Du et al.(2017)”.

Reply: Revised (seen in Line 401-401).

9. Please also provide the correlation coefficients between the observed and simulated BC and simulated POA and observed HOA.

Reply: The correlation coefficients between the observed and simulated BC and simulated POA and observed HOA are showed in Fig.1 (seen in Fig.1) and presented in the revised manuscript (seen in Line 425-426 and 431-432).

10. For figure 4 and 7, the shaded circles are difficult to observe. And the concentrations of secondary organic aerosol and CN10 are recommended to display.

Reply: The observed values of SOA and CN10 are labeled with shaded colors in black circles. The shaded circles have been made clear in Figure 4 and 7. The concentrations of SOA and CN10 are too dense to be clearly displayed, so the exact values are not displayed in Figure 4 and 7.

11. Line 515-516, “The higher concentrations of ASOA than BSOA are also demonstrated by other studies”, please some references there.

Reply: The references are added (seen in Line 548).

12. Please give some potential reasons for the differences between spatial

distributions of SV-SOA and LV-SOA.

Reply: The differences between spatial distributions of SV-SOA and LV-SOA are mainly caused by their different formation mechanisms. SV-SOA is mainly from the products of VOCs whereas LV-SOA is from the further oxidation of SV-SOGs. The multi-generation aging processes can make the LV-SOA formed downwind the source regions. Globally, high SV-SOA and LV-SOA concentrations are mainly located in the continental source regions. However, the concentration of LV-SOA is higher than that of SV-SOA in downwind regions. Even over source areas with low emission intensity, such as North America and Europe, LV-SOA also has a higher concentration than does SV-SOA. In the VBS scheme, the organic compounds could undergo the multi-generation aging processes during transport and produce a higher concentration of LV-SOA which mostly remains in particle phase. Consequently, LV-SOA distribution is more homogeneous than SV-SOA does and has a wider spread over the ocean. The reasons for the differences between spatial distributions of SV-SOA and LV-SOA are added in the revised manuscript. (seen in Line 577-582)

13. Line 563, the authors did not provide “Table 3” and “observed values in Fig.6a”. Please modify.

Reply: “Table 3” were revised to “Table S1” and “Fig.6a” were revised to “Fig.6a” (seen Line 600 and 601).

14. Figure 7d shows that the high value CN10 is mainly primary over Northeast China where the concentration of secondary organic aerosol is relative high shown in Figure 6. Please explain this phenomenon.

Reply: Though SOA concentration is relative high in North China Plain, they are coated on the primary particles (BC and POC particles) due to the high concentration of primary particles. In our model, primary and secondary particles are distinguished by their physical origin rather than chemical composition (seen in Line 218-221). Even though the concentration of secondary coatings is high, the primary particles with secondary coatings are defined as "primary particles". The large primary emission leads to the high concentration of primary particles (BC and POC; served as the core of "primary particles"), which can scavenge the secondary particles by coagulation and reduce the growth rate of secondary particles by competing for

condensable gases. Therefore, CN10 is dominated by "primary particles" over North China Plain. The corresponding explanation are added in Line 632-634.

15. Line 631, "indicate" should be "indicated".

16. Line 835, "top panel" and "bottom panel" should be "left panel" and "right panel".

Reply: Revised (seen in line 669 and 877).