Responses to the comments of Reviewer #4:

We are truly grateful to yours' positive comments and thoughtful suggestions. Those comments are all valuable and very helpful for revising and improving our paper, as well as the important guiding significance to our researches. Based on these comments and suggestions, we have studied comments carefully and have made correction which we hope meet with approval. All changes made to the text are marked in blue color. Below you will find our point-by-point responses to the reviewers' comments/questions:

Specific Comments:

1. *L144 duo to->due to*

Response:

Done. (L147)

2. *L291 carton-> carbon*

Response:

Done. (L294)

3. *L305 What do you mean distributing the increments using the mass concentration background error STD? Please clarify this.*

Response:

We really appreciated your question. The assimilation process will directly generate analysis increments of 20 control variables, however, these control variables are not completely consistent with model variables within MOSAIC. For those consistent with model variables, their increments can be directly used to adjust model variables, while for those lumped control variables, their increments correspond to 2 or 3 model variables, for instance, the control variable SSN1 correspond to 3 model

variables, i.e. *so4_a01*, *no3_a01*, and *nh4_a01*, which are sulfate, nitrate, ammonium mass concentrations at the first size bin, respectively, thus, distributing the increment of SSN1 over three model variables *so4_a01*, *no3_a01*, and *nh4_a01* is necessary. How to distribute? A simple way is to determine the distribution ratio. When estimating background error covariance using the NMC method, we can employ differences between 48 h and 24 h forecasts valid at the same time (i.e. 0000 UTC) for every model variable within a period of one month (November 2018) to set up a sample and figure out the background error standard deviation (STD) in mass concentration. For example, the computed STDs of *so4_a01*, *no3_a01*, and *nh4_a01*are c1, c2, and c3, respectively, thus, the corresponding distribution ratios are calculated as c1/(c1+c2+c3), c2/(c1+c2+c3), c3/(c1+c2+c3).

4. *L540 You said the vertical correlation of every variable is similar, however, you subsequently said vertical correlations differ among aerosol variables. Please clarify it. Besides, since the AOT observation has no vertical information, how do you assume the vertical information of the AOT observations?*

Response:

We really appreciated your question. We said the vertical correlation of every variable is similar, meaning that vertical correlation plots for every variable look similar. Because the vertical correlation describes the auto-correlation between two layers at different heights, the vertical correlation is a symmetric matrix and the maximum 1 is on the diagonal, which is common to all variables. Therefore, the vertical correlation of every variable is similar. However, vertical correlations among aerosol variables are not the same. Given a correlation more than 0.8, some variables have a larger domain while some have a less domain, which indicates that vertical correlations differ among aerosol variables.

AOT is an atmospheric column measurement, it has no vertical information. When assimilating AOT observations, it does not need to assume the vertical information of the AOT observations.

5. *Fig.7 Can you explain why the assimilation has little effects on the significant underestimates of the AOTs? Such as the observed AOTs are around 1-1.5, whereas the simulated ones are around 0.*

Response:

Thank you so much for your question. In general, the assimilation has significant effects on AOT simulation, but has little effects on the some significant underestimates of the AOTs. This phenomenon is probably due to uncertainties in aerosol emissions as well as meteorological boundary conditions. Emission data is another important factor that influences the aerosol simulation. Simultaneous assimilation of aerosol data to updating aerosol emission and initial field may reduce this phenomenon in the future.

We would like to express our great appreciation to you for the valuable and pertinent comment on our manuscript, which is crucial to improve the quality of our work. We hope that these revisions are satisfactory and that the revised version will be acceptable for publication in Geoscientific Model Development. Thank you very much for your work concerning my paper.

Wish you all the best!

Yours sincerely,

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11/24/2021
