Responses to the comments of Reviewer #2:

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 green color. Below you will find our point-by-point responses to the reviewers' comments/questions:

Major Comments:

1. *Using a constant observational error covariance of 0.06 seems not very convincing. For AOD of 1.8, the error is only 3.3%. Is this realistic? The observational error plays an important role in the DA analysis. Some justification for using this value is needed.*

Response:

We really appreciate your question. The observation error plays an important role in assimilation process, however, no relevant theoretical basis on its construction has been found so far. The observation error depends on measurement error and representation error (Elbern and Schmidt, 2001; Schwartz et al., 2012; Jiang et al., 2013), and is difficult to accurately estimate so that how to determine it is also a matter of assimilation practice. In several studies, the observation error is given by a tuning parameters. Based on the 3DVAR principle, the function of the observation error can be easily analyzed, namely, the observation error determines the weight of observation across the analysis. Given a background field, the smaller observation error produces the greater increments in terms of absolute value to make the analysis closer to observations and away from the background field and vice versa. No matter how large the observation error is, as long as the observation operator is correct, the generated analysis theoretically will fall between the background field and observations, demonstrating a positive assimilation effect, even though not the best. Consequently, it is inclined to construct the simple observation error to run the assimilation system in practice. It is apparent that using a constant observation error only to test the developed system is rational.

Even though the observation error can be roughly determined based on experience, it is necessary to select a rational value. According to Yumimoto et al. (2016), the observation error was estimated to be the retrieval uncertainty attached to the Himawari-8 AOT data plus a standard deviation calculated as the representative error in the regridding. The retrieval uncertainty ranged from 0.0001 to 1.04 with average of 0.013 and has larger values in the land relative to over the ocean. Thus it can be seen that using a constant observation error of 0.06 is rational in this study, which is also obtained after several tests. As you mentioned, as for AOD of 1.8, the value seems somewhat irrational, but these high AOD data account for a small proportion during the study period. It should be pointed out that the observation error varies with data values, which also needs some further researches in the future.

2. *More detailed information in numerical experiment design is needed. Is AOD DA performed every hour whenever AOD data are available? Does the forecast last for 24 h only? For each 24-h DA cycle, are the meteorological data in the first guess from FNL or from data at the end of the previous cycle? Similarly, for each forecast starting at 0300 UTC, while aerosols are taken from the analysis after a 24-h DA cycle for the Analysis run and from the previous 24-h forecast for the Control run, are meteorological conditions taken from FNL?*

Response:

We really appreciate your question. AOD DA is not performed every hour during the period of 0300 UTC to 0800 UTC when the Himawari-8 AOD observations are available for China. AOD observations at 0300 UTC every day from 23 to 29 November 2018 was only assimilated to provide the analysis (L460-461), and the forecast last for 24 h, which means that the assimilation frequency is 24 h. Comparing to its high temporal resolution (an hourly product), the 24-h assimilation frequency seems to be an underutilization of AOD observations. However, the AOD retrievals are found with much noise, which will have a significantly negative impact on assimilation. For example, surface $PM_{2.5}$ concentration and original (not thinned) Himawari-8 AOD observations at 0300 UTC and 0600 UTC are plotted in Fig. 1 and Fig. 2, respectively. Overall, surface $PM_{2.5}$ mass concentrations change little even with a small decrease at some areas from 0300 to 0600 UTC (Fig. 1b, Fig. 2b) while there is a remarkably increase in AODs during the same period (Fig. 1a, Fig. 2a). In terms of $PM_{2.5}$, the noticeably increase in AOD observations should not be considered as normal changes of aerosol but much noise. As a result, more frequent assimilation of AOD observations like this will certainly result in a dramatic overestimation of $PM_{2.5}$ mass concentrations. In terms of evaluation with $PM_{2.5}$ mass concentration observations, AOD observations at 0300 UTC without no temporal collocation were only assimilated in this study to test the developed assimilation system. As known, DA serves only as a mathematical approach on how to introduce observations into the model, and then improves model initial and forecast fields. Assimilation results are largely determined by observational data, as for how to deal with those with much noise and improve the quality, more researches are needed in the future.

Additionally, for each 24-h DA cycle, the meteorological data in the first guess are from FNL, and the meteorological conditions in both the Analysis run and Control run are taken from FNL, meaning that the Analysis run and Control run utilized the same meteorological conditions. It should be noted that meteorological states were not assimilated in this study because the developed DA system has no capacity of assimilating meteorological data, which aims at aerosol DA.

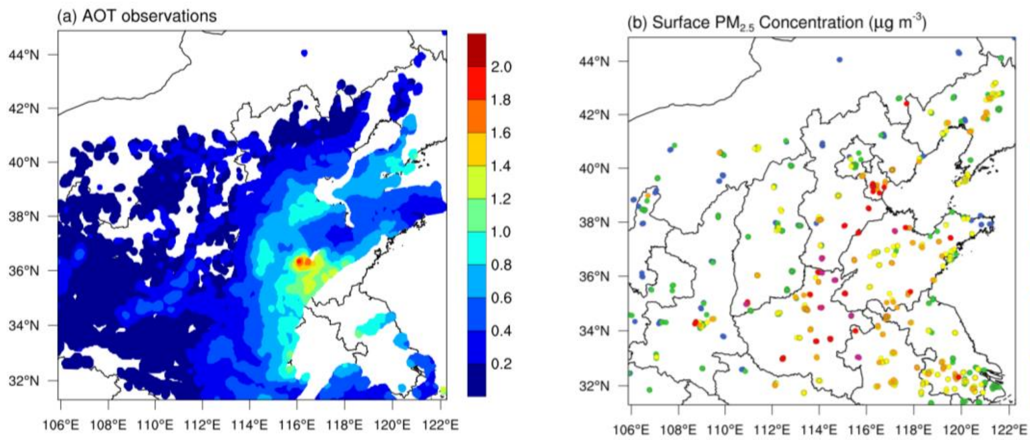

Figure 1. Observations of the original (not thinned) Himawari-8 AOTs (a) and surface $PM_{2.5}$ mass concentration (b) in D02 at 0300 UTC on 25 November 2018.

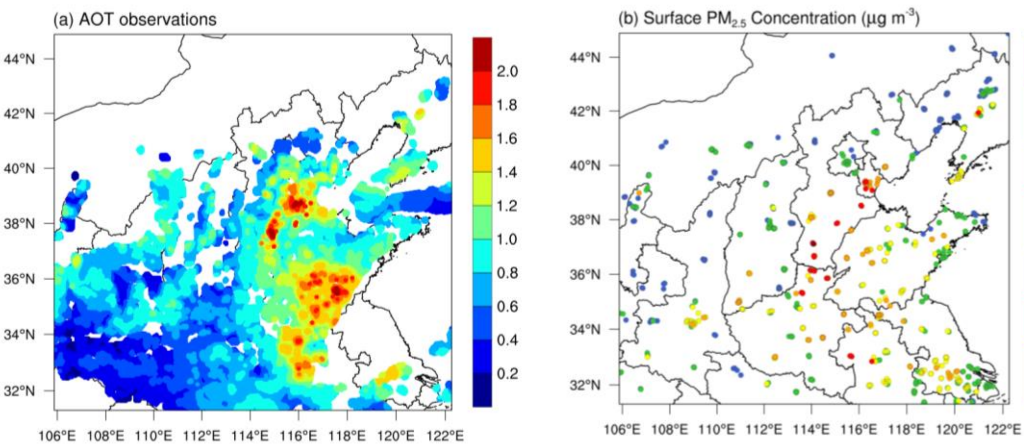

Figure 2. Same as Fig.1, but at 0600 UTC on 25 November 2018.

3．The development of assimilating optical properties was built on the framework of Li et al. (2013). The authors should discuss major differences between the two analysis systems and major differences in the conclusions of the two studies.

Response:

We really appreciate your question. The DA system presented in this manuscript is an upgrade of that developed by Li et al. (2013). Li et al. (2013) developed a 3DVAR aerosol DA system to work with the sectional scheme MOSAIC within WRF-Chem for the first time. However, it can only assimilate aerosol mass concentrations, including total mass such as $PM_{2.5}$ and PM_{10} and composition mass, without the ability of assimilating aerosol optical properties. In order to develop the DA system for aerosol optical properties, the basic framework of Li et al. (2013) including the minimization process as well as the **B**-matrix computation was employed, but new aerosol state variables are designed based on the MOSAIC scheme. There are a total of 20 state variables in this DA system while there are 5 variables in Li et al., (2013). More importantly, an optical module consisting of the nonlinear forward operator achieved by simplifying the Optical Module inside the WRF-Chem model and its tangent linear (TL) as well as adjoint (AD) codes has been added in order to directly assimilate optical properties. In the study of Li et al. (2013), $PM_{2.5}$ mass assimilation has a significant improvement for $PM_{2.5}$ initial conditions and its 24-h subsequent forecasts, whereas, this study mainly focus on the validation of the new development with AOD observations and shows that AOD assimilation improves 24-h $PM_{2.5}$ forecasts and model AOD initial simulations.

4. The improvement of aerosol forecasts only lasts for 24 hours in this study. Although Li et al. (2013) also showed a similar result, this seems a little bit short in terms of forecast length. Some studies have shown the benefit of assimilating AOD data in longer aerosol forecasts (48 h), such as Benedetti, et al. 2019 and Choi et al. 2020. Could it be due to, for example, no assimilation of meteorological data, the quality of AOD data, the assimilation method, the study location, etc.? The authors should compare their results with others' or make some comments about this issue (24 h versus 48 h).

Response:

We really appreciate your suggestion. In short, the benefit of assimilating AOD data can last longer than 48 h in the studies conducted by Benedetti et al. (2019) and Choi et al. (2020), which is in terms of AOD simulations, however, the improvement lasting for 24 h in this study is in terms of $PM_{2.5}$ forecasts. It is obvious that the results can not be comparable. In our study, AOD assimilation significantly improves AOD initializations and simulations, but the improvement for the forecast length is not evaluated. Both Benedetti et al. (2019) and Choi et al. (2019) assimilated MODIS AOD to improve the dust analysis and forecasts. In the study of Choi et al. (2019), only MODIS AOD was employed to evaluate the assimilation benefits, whereas, independent AOD data from two established ground-based networks as well as PM_{10} data from the China Environmental Protection Agency were used in the evaluation in the study of Benedetti et al. (2019). In spite of the better improvement for AOD simulations, the AOD assimilation can only make small adjustments to PM_{10} but is unable to improve the quality of forecast fundamentally.

Major Comments:

5．Line 65. "... monitoring, for instance, China has..." should be ""... monitoring. For instance, China has..."

Response:

We really appreciated and followed your valuable suggestion. This sentence has been revised as "For instance, China National Environmental Monitoring Centre (CNEMC, http://www.cnemc.cn/en/) has established a nationwide monitoring network consisting of more than 1500 stations since 2013 to provide near-time data of pollutants, including $PM_{2.5}$, PM_{10}, SO_2, NO_2, CO, and O_3."(L66-67)

6. Line 74. "... detailed aerosol profiles (Kaufman et al., 2002), ..." Kaufman et al., 2002 used AOT and aerosol index for their study. Both are vertically integrated data and thus do not provide vertical profile information.

Response:

We really appreciated and followed your valuable suggestion. This sentence has been revised as "Remote sensing optical properties can cover a much larger domain (Kaufman et al., 2002) and provide detailed aerosol profiles (Young and Vaughan, 2009)" (L75-76), at the same time, this piece of reference below has been added:

Young, S. A. and Vaughan, M. A.: The retrieval of profiles of particulate extinction from Cloud-Aerosol Lidar Infrared Pathfinder Satellite Observations (CALIPSO) data: Algorithm description, J. Atmos. Ocean. Tech., 26, 1105–1119, https://doi.org/10.1175/2008JTECHA1221.1, 2009.

7. Line 98. What does the "control variable scheme" mean? DA methods usually need control variables. Do you mean "...PM10, which is used as a control variable?"

Response:

We really appreciate your question. The control variable scheme means how many control variables, one or more, are employed in DA analysis. The early aerosol DA usually employed a control variable. For example, PM_{10} (mass concentration) rather than its compositions is directly employed as the control variable so that observation is the control variable self.

8. Lines 120-122. I believe that ECMWF uses a 4DVAR method to assimilate AOD and it is an online approach. Check out Benedetti et al. 2019 paper listed above.

Response:

We really appreciate your suggestion. ECMWF has incorporated atmospheric composition variables into its 4DVAR meteorological assimilation analysis system.

The aerosol assimilation uses total aerosol mass rather than composition mass as a control variable, and it can only assimilate satellite-derived AODs and work with the global model. The sentence has been revised as "Although the four-dimensional variational (4DVAR) technique has been extensively used in operations (Gauthier at al., 2007; Benedetti et al., 2019), and has also been employed to assimilate atmospheric chemical compositions such as O_3, SO_2, and CO based on the simple offline chemical transport model (CTM) (Eibern and Schmidt, 1999; Elbern and Schmidt, 2001), it is greatly challenging to develop a 4DVAR DA system coupled with the sophisticated aerosol model such as MOSAIC because of the high computational cost and complex adjoint model" in the revised manuscript. (L121-127)

9. Lines 236-237. "...observation errors associated with AOD retrievals are determined by measuring instruments..." It is probably more than just the instrument itself, but also the retrieval algorithm and surface emissivity, to name a few.

Response:

Thank you so much for your correction. This sentence has been revised as "In general, observation errors associated with AOT retrievals are determined by measurement and representation errors (Elbern and Schmidt, 2001; Schwartz et al., 2012; Jiang et al., 2013)" in the revised manuscript. (L240-241)

10. Line 261. Define BEGS.

Response:

We are so sorry for the misspelling. It should be written as BFGS. The L-BFGS algorithm is a limited memory quasi-Newton method for large scale unconstrained optimization, which was developed by four mathematician Broyden, Fletcher, Goldfarb, and Shanno, BFGS is their initials. The L-BFGS code has been developed at the Optimization Center, a joint venture of Argonne National Laboratory and Northwestern University (http://users.iems.northwestern.edu/~nocedal/lbfgs.html). (L264)

11. Lines 440 and 442. The data reduction used in this study is not a thinning procedure but a superobbing procedure.

Response:

We really appreciate your question. We thinned AOD observations in the spatial resolution of the model, which is also employed by other researchers (Yumimoto et al., 2016; Dai et al., 2019; Ha et al., 2020). This approach certainly leads to a great data reduction, however, it can smooth out some noise in retrieved data to improve the quality, which is also of great significance for assimilation. At least, the assimilation practice has demonstrated that assimilating thinned AOD observations is better than original observations. More researches on how to thin data with a high spatial resolution are needed in the future.

12. Line 457. Add "AOT" in front of assimilation.

Response:

Done. (L466)

13. Line 569. "… with negative increments marked in blue." Improve the color shading in Figure 6c. Make warm and cold colors for positive and negative values, respectively. The current plot mixes red and blue colors for positive values, while it uses blue shading for negative values. This is confusing. A similar problem is seen in Figure 9c.

Response:

Done. The color shadings in both Figure 6c and Figure 9c have been improved in the revised manuscript so that warm and cold colors are for positive and negative values, respectively.

14．Line 594. "… BIAS increase…" This statement sounds like that the assimilation of AOD data makes the result worse, but it is not true. Need to rewrite this. The same for line 663.

Response:

We followed the suggestion. This statement has been rewritten as "BIAS is reduced by about 77 percent" (L603). The statement in line 663 has also been written as "reducing BIAS by 4.97 ug m^{-3}" (L688).

15．Try to use words consistently throughout the paper, such as "cost function" versus "objective function", "AOD" versus "AOT", "Control" versus "control" experiment, and "Assimilation" versus "assimilation" experiment.

Response:

Done. We used the words "cost function", "AOT", "Control", and "Assimilation" consistently throughout the paper in the revised form.

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,

Daichun Wang and Wei You

11/23/2021
