Responses to the comments of Reviewer #3:

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 carefully and have made correction which we hope meet with approval. All changes made to the text are marked in yellow color. Below you will find our point-by-point responses to the reviewers' comments/ questions:

General Comments:

1. *The model resolution, meteorological conditions, and emission data could be other important sources of uncertainty in the air pollution modeling and in fact some of them can be identified in the diagram you show in Fig. 2. However, they are not discussed in the manuscript. Would you be able to quantify these uncertainties in relation to the impact of aerosol field initialization (DA) based on the design of model experiment? For instance, you may consider conducting additional experiment which assimilate meteorological states and aerosol to explore their relative impacts on the subsequent forecast.*

Response:

We really appreciate your valuable suggestion. Discussing various sources of uncertainty in the air pollution modeling is of significance, however, this manuscript presented a new development of aerosol optical properties data assimilation (independent developed), which is coupled with the MOSAIC scheme for the first time and different from the GSI tool, so a validation of the developed assimilation system using Himiwari-8 AOT observations was focused in the study. Quantifying these uncertainties may need well-designed model experiments, which would be carried out in the following researches.

We are sorry to say that the developed assimilation system has no capacity of assimilating meteorological data, namely, it only aims at improving aerosol initial conditions. Nevertheless, it can assimilate a wide range of aerosol observations, including total aerosol ($PM_{2.5}$, PM_{10}) or component mass concentration, optical properties such as AOD, extinction and backscatter profiles, and attenuated backscatter profile, which would advance aerosol data assimilation. Moreover, we will develop meteorological and aerosol coupling DA methods in the future.

2. *The under-utilization of Himawari-8 AOT product (hourly data) in the context of assimilation frequency (24 h) seems to be obvious. I imagine a strategy with more frequent assimilations could be a unique point to make in this research as the geostationary satellite product used here has such a high temporal resolution. Nevertheless, the relevant discussion is not covered in the manuscript. I would suggest adding more content to address this comment.*

Response:

We really appreciate your suggestion. Himawari-8 level 3 AOT_Merged, an improved hourly product, which is derived from level 2 AOT retrievals at a 10 min interval, was employed to conduct assimilation experiments. A daily assimilation frequency seems to be an underutilization of Himawari-8 observations in comparison to its high temporal frequency. Since AOT observations are retrieved at the visible and infrared bands, observations between 0300 and 0800 UTC in the daytime are available for China. In fact, AOT observations are noticeably noisy, which will have a greatly negative impact on assimilation results. Moreover, observations at afternoon are much noisier than those in the morning. For example, surface $PM_{2.5}$ concentration and original (not thinned) Himawari-8 AOT observations at 0300 UTC and 0600 UTC on 25 November 2018 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 AOTs during the same period (Fig. 1a, Fig. 2a). In terms of $PM_{2.5}$, the noticeably increase in AOT observations should not be considered as normal changes of aerosol but much noise. As a result, more frequent assimilation of AOT 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, AOT observations at 0300 UTC without no temporal collocation were only assimilated in this study to test the developed assimilation system. As known, data assimilation serves only as a mathematical approach on how to introduce observations into the model, and then improves model initializations and forecasts. Assimilation results are largely determined by observational data, as for how to deal with those with high noise and improve the quality, more researches are needed in the future.

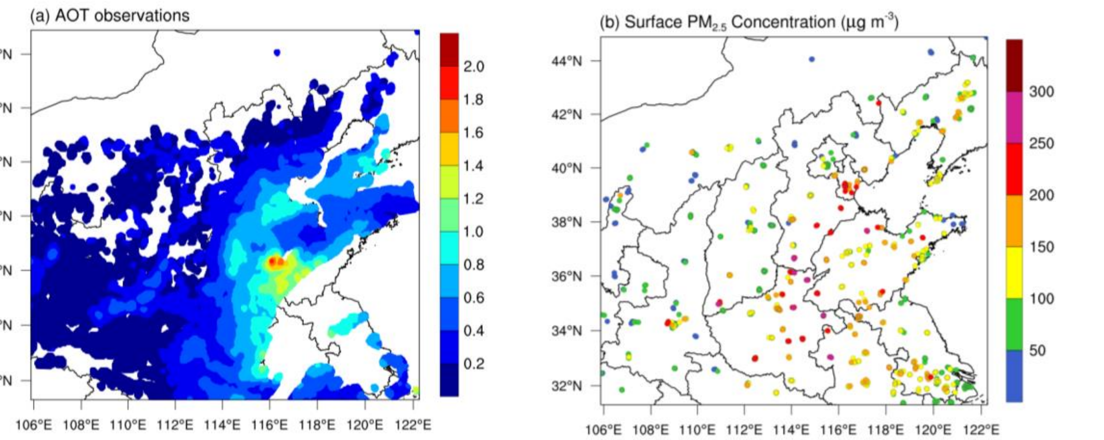

Figure 1. Observations of the original (not thinned) Himawari-8 AOTs (a) and surface $PM_{2.5}$ mass concentration (b) in D02 at 0300 UTC on 25 November 2018.

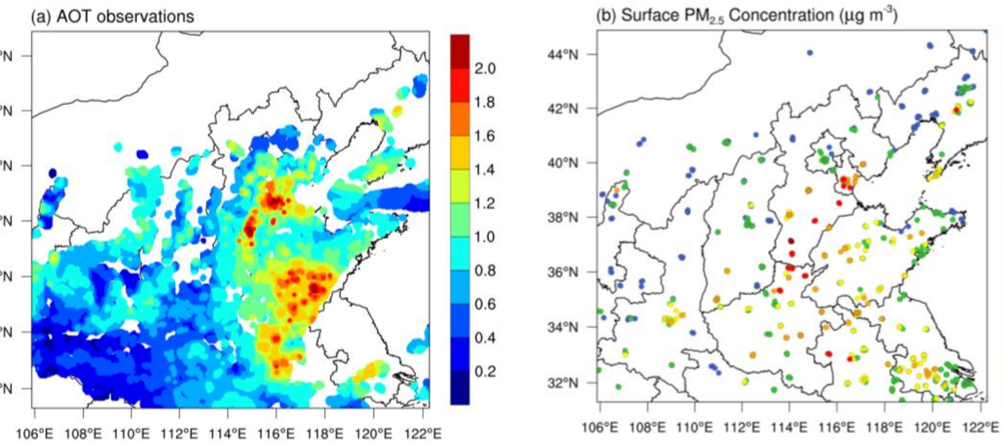

Figure 2. Same as Fig.1, but at 0600 UTC on 25 November 2018.

3. *Despite the vertical profiles of background error STDs and auto-correlations are given, the analyzed increments of each aerosol state variables are not seen anywhere in the document. Since the AOD is obtained through the integration of aerosol properties in the atmospheric column, it would be useful to show analyzed results in terms of their vertical distributions and further discuss how would that contribute to the uncertainty of simulation.*

Response:

We really appreciated and followed the suggestion. The assimilation process directly produces the analysis increments of 20 aerosol state variables, so it is natural to give the analyzed increments of each aerosol state variable. The analyzed $PM_{2.5}$ increments were computed based on those of each variable and given in Fig. 9 in light of comparing with $PM_{2.5}$ observations (no aerosol state variable observations are available at present). Actually, the increment of each variable contributes greatly to the total $PM_{2.5}$ increment and differs significantly according to its background error STD. In general, the variable with a larger background error STD has a larger increment and vice versa. Of all state variables, SSN2 has the greatest background error STD, its increment in case of November 25, 2018 is shown in Fig. 3 here, which is similar to that of $PM_{2.5}$.

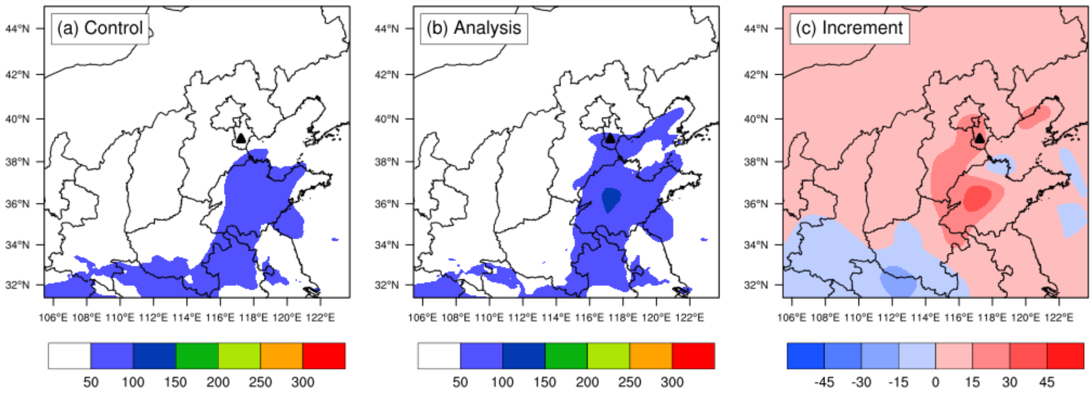

Figure 3. Spatial distribution of SSN2 in the background field (a) and analysis (b) as well as the increment (c) in D02 at 0300 UTC on 25 November 2018, these quantities are in unit of ug m^{-3}.

As you mentioned, it would be useful to show vertical distributions of the analyzed increments. Similarly, we has added the vertical distribution of $PM_{2.5}$ analyzed increment, which is shown in Fig. 10 in the revised manuscript (here is shown in Fig. 4), helping to demonstrate the impacts of AOD assimilation on aerosol vertical distributions. And the following information has also been added in the revised manuscript (L670-681). “Since AOD is an atmospheric column measurement, it naturally includes the information of aerosol vertical distributions. Consequently, AOT assimilation can improve aerosol vertical distributions as well. A vertical cross-section of $PM_{2.5}$ at 0300 UTC on 25 November 2018 is shown in Fig. 10, this cross-section is through Tianjin (marked by the black triangle in Fig. 9). Similar to surface $PM_{2.5}$, suspended $PM_{2.5}$ mass concentrations in the upper air are also enlarged with a wide range from the ground to about 1 km by significantly positive increments generated by assimilation (Fig. 10c). In spite of no observational $PM_{2.5}$ profiles to compare, the vertical distribution in analyses is believed to be closer to the real in terms of the ground $PM_{2.5}$ level (Fig. 10b). It should be noted that the vertical increments are determined by the background error vertical correlation. In a summary, AOD assimilation is certainly helpful to improve the three-dimensional structures of $PM_{2.5}$.”

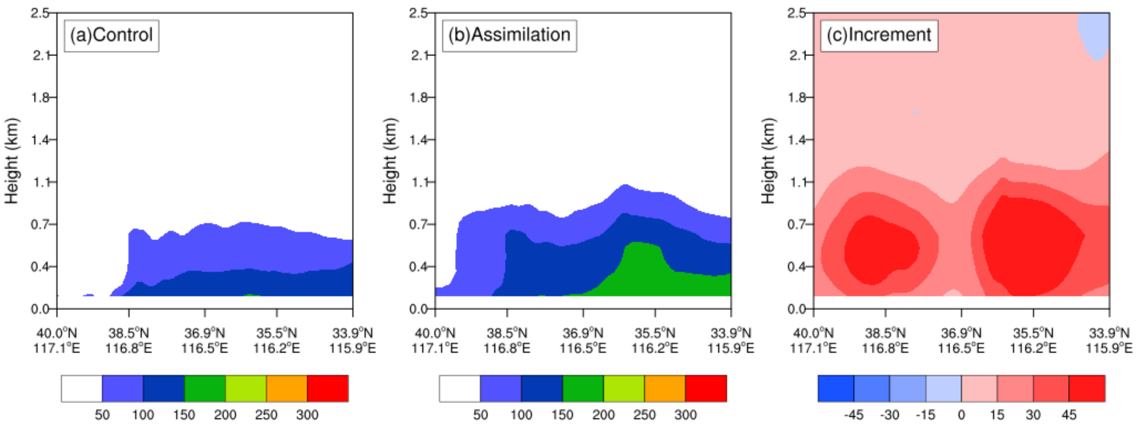

Figure 4. Vertical cross-section of $PM_{2.5}$ in the background field (a) and analysis (b) as well as the increment (c) in D02 at 0300 UTC on 25 November 2018.

Specific Comments:

4. *L32: It is mentioned here that the developed DA system is able to assimilate lidar-based aerosol profiles. However, I did not find any relevant description with respect to the treatment in the followed sections. Would you clarify this?*

Response:

We really appreciate your question. Developing a new aerosol data assimilation system, especially for variational method to assimilate unconventional observation data (such as aerosol optical data sources), is a challenging work. Based on the 3DVAR principle, the observation operator determines what type of observations can be assimilated, that is, you need to design and construct the operator according to the observations which will be assimilated. In fact, various aerosol optical properties can be simultaneously calculated through the previous same steps, for example, the process from the size parameter, complex refractive, and aerosol number to optical properties such as extinction and backscatter coefficients, go further, AOD and attenuated backscatter can be computed using extinction and backscatter. In the data assimilation system, these optical quantities have individually corresponding observational data interface. What type of observations are inputted, the assimilation system run corresponding program codes, and this design is easily implemented in practical coding. For example, if extinction and backscatter profiles are to be assimilated, then the terms in the cost function and its gradient associated with the following AOD and attenuated backscatter are no longer computed. It is worth mentioning that only AOD observations are employed to test the developed assimilation system in this study, so any relevant descriptions of lidar-based extinction or backscatter profiles assimilation are not given. We will combine assimilate more data sources including surface PM data, satellite derived AOD, attenuated backscatter et al in the near future.

5. *L237-240: Have you conducted any experiment to test how sensitive this constant error is?*

Response:

We really appreciate your question. We have not conducted any experiment to test how sensitive the observation error is. The development and validation of the assimilation system are focused in this study. The observation error plays an important role in the assimilation process, however, it is very difficult to accurately determine it and usually determined based on experience (or tuning parameters).

6. *L260: Can you give an example of the minimization process, such as reduction of cost function in function of iteration numbers?*

Response:

We really appreciate you question. The minimization process is to find the minimum solution to the cost function, which usually employs the descent algorithm, such as the L-BFGS algorithm here which is a limited memory quasi-Newton method for large scale unconstrained optimization and available at http://users.iems.northwestern.edu/~nocedal/lbfgs.html. In general, the minimization process is a process of iteratively updating control variables. At first, the cost function and its gradient are computed with an initial value of control variables, and the function and gradient values along with control variable values are put into the descent algorithm to obtain a new value of control variables. Then come to the next step, new values of the function and its gradient as well as control variables are altogether put into the descent algorithm again to update the value of control variables, go on like this. The process ends until the convergence condition (the gradient is equal to 0 in theory) is meet or iteration number for example 50 is reached. In the minimization process, the cost function keep reducing, and the reduction is fast in the beginning while it becomes slowly lately. Further more, the reduction depends on the case and is hard to describe in function of iteration numbers. In our study, the max number of iterations is set to 50. The number of iterations varies with experimental cases.

7. *L288-289: Please include references to supplement statement here*

Response:

Done. The following reference has been added: (L293)

Barnard, J. C., Fast, J. D., Paredes-Miranda, G., Arnott, W. P., and Laskin, A.: Technical Note: Evaluation of the WRF-Chem "Aerosol Chemical to Aerosol Optical Properties" Module using data from the MILAGRO campaign, Atmos. Chem. Phys., 10, 7325–7340, https://doi.org/10.5194/acp-10-7325-2010, 2010.

8. *L291: Should be black car"b"on and organic car"b"on*

Response:

Done. (L294-295)

9. *L369: Would this introduce any inconsistency between nonlinear model and TL? Also, I am curious how did you deal with if statements in the code if there's any.*

Response:

We really appreciate your question. The Optical Module within WRF-Chem is a developed routine package, it can compute a large number of aerosol optical quantities, such as aerosol scatter phase functions. However, these codes have nothing with the development of the assimilation system. Thus, when transplanting the Optical Module to establish the observation operator, these irrelevant codes should be removed to reduce the difficulty in tangent linear (TL) and adjont (AD) coding. Also, above-mentioned process can improve computing efficient.

The conditional statements remain unchanged when establishing the TL or AD codes of if statements. TL or AD codes of the assignment statements are needed to add into if statements. TL statements are arranged in the same order as assignment statements, but AD statements are arranged in a reverse order.

10. L389: Since this manuscript documents the development of a DA package, it is of necessity to show the result of TL/AD test. For example, it is common to show the plot of gradient check with respect to various orders of perturbation.

Response:

We really appreciated your suggestion. TL/AD test is necessary for establishing TL and AD codes, which only serves as the validation of the codes after all it is a huge work to finish the TL/AD codes and easy to make mistakes, so it seems unnecessary to give the result of TL/AD test in the manuscript. The following table (Tab. 1) shows the gradient with respect to perturbations in both directions. It is noted that initial perturbations are set to 20 and -20, respectively, and the gradient (radio) of AOD with respect to control variables was calculated by halving the perturbation every time. Eventually, the gradient approaches 1 in both directions.

Table 1. TL/AD test results

number	positive perturbation	ratio (gradient)	negative perturbation	ratio (gradient)
1	20.00000	1.02831070096536	-20.00000	0.995594423135122

2	10.00000	1.02728481026492	-10.00000	0.997059224601074
3	5.000000	1.02644276988709	-5.000000	0.997750286836985
4	2.500000	1.02579561769594	-2.500000	0.998080650773033
5	1.250000	1.02542213463021	-1.250000	0.998239359890258
6	0.6250000	1.02522400926412	-0.6250000	0.998316741235688
7	0.3125000	1.02512225357477	-0.3125000	0.998354903969795
8	0.1562500	1.02507072260859	-0.1562500	0.998373850019414
9	7.8125000E-02	1.02504479642776	-7.8125000E-02	0.998383288869707
10	3.9062500E-02	1.02503179348556	-3.9062500E-02	0.998387999717800
11	1.9531250E-02	1.02502528213119	-1.9531250E-02	0.998390352987688
12	9.7656250E-03	1.02502202388487	-9.7656250E-03	0.998391529132607
13	4.8828125E-03	1.02502039438236	-4.8828125E-03	0.998392116963912
14	2.4414062E-03	1.02501957932535	-2.4414062E-03	0.998392411082556
15	1.2207031E-03	1.02501917199313	-1.2207031E-03	0.998392557990852

11. L418: Please cite this reanalysis product and provide the link of the data source.

Response:

Done. We have added the link of the data source (L424-425).

12. L422: The assimilation cycle time (24 hours) seems to be coarse in relation to data availability. Please discuss how it is designed and clarify if there's any limitation on the data coverage or quality, etc.

Response:

We really appreciate your question. As discussed above, Himawari-8 level 3 observations between 0300 and 0800 UTC in the daytime are available for China. AOT observations are noticeably noisy, which will have a greatly negative impact on assimilation results. In terms of $PM_{2.5}$, directly assimilating AOT with noises will result in a dramatic overestimation of $PM_{2.5}$ mass concentrations. The 24 h assimilation frequency was designed only to test the developed system.

13. L424-426: The statement here is contradictory to the design of assimilation cycles. Please explain.

Response:

We really appreciate your question. As explained above, more frequent assimilation of AOT observations with much noise will cause the significant overestimation of $PM_{2.5}$ mass concentrations. Nevertheless, In terms of evaluating with AOT observations, more frequent assimilation may have better effects.

14. L441: I am not sure this is the best treatment as it could further smooth out the observed data. Please address.

Response:

We really appreciate your question. We aggregated AOT 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). How to treat the dataset with a high spatial resolution before assimilation may need further researches. We aggregated AOT observations by averaging them in one grid cell so that the resolution of them matches that of the model, smoothing out the observed data, however, this approach can filter out much noise to improve the quality.

15. L443 and L463: Fig. 3b is mentioned earlier than Fig. 3a. I would suggest swapping them for the fluency of reading.

Response:

We followed the suggestion. Fig.3b and Fig. 3a have been swapped in the revised manuscript (L451, L472).

16. L492: It looks like the similar DA procedure is also carried out over the D01 but at least with different treatment in data thinning. Have you done any experiment *without assimilation in D01? If true, what was the impact of additional DA in D01.*

Response:

We really appreciated your question. A two-level nested domain configuration was employed to run simulation experiments. The outer domain D01 is at a horizontal resolution of 27km, and the inner domain D02 is at a resolution of 9km. The AOT observations are thinned using D01 grid and D02 grid, respectively. The same assimilation procedure was carried out over D01 and D02, separately, but with data of different resolutions, to improve individual aerosol initial conditions. In the control experiment, both D01 and D02 simulations were performed without assimilation. The D02 simulations were only evaluated with various observations and the evaluation was shown in this study because the AOT observations are mainly distributed in D02. Of course, we can evaluate the impacts of D01 assimilation on D01 simulations as well, nevertheless, it seems a repeated work in terms of testing the development.

17. L532: Is it possible to estimate the correlation length with the observational data or alternatively the analysis after assimilation?

Response:

We really appreciated your question. It is a good idea that using the analysis after assimilation to estimate the correlation length. We will conduct the test in the future.

18. L577-579: Sentences such as these in the manuscript could be trimmed to shorten the length.

Response:

We followed your valuable suggestion. The relevant sentences have been revised as "The higher scores of the metrics CORR, RMSE, and BIAS would demonstrate the better assimilation performance and vice versa" (L586-587).

19. L587: Please elaborate more on this. Would the uncertainty mostly be on the *magnitude or something else?*

Response:

We really appreciated your suggestion. AOD simulation was performed at a wavelength of 500 nm, the same as Himawari-8 AOT retrievals, whereas MODIS AOD is retrieved at 550 nm. It is obvious that the difference in the wavelength (500nm and 550nm) would affect the evaluation when evaluating the AOD simulation with MODIS AOD, however, the evaluation is convincing because the wavelength difference is minor.

20. L606: The red triangles in Fig. 1b are hardly distinguished from one another as they are basically overlapped with each other. Please try to make them more visible. Add another zoomed-in map may help achieve that.

Response:

We have added a zoomed-in map as Fig. 1c for AERONET sites in Beijing area in the revised version, which is also given as Fig. 5 below:

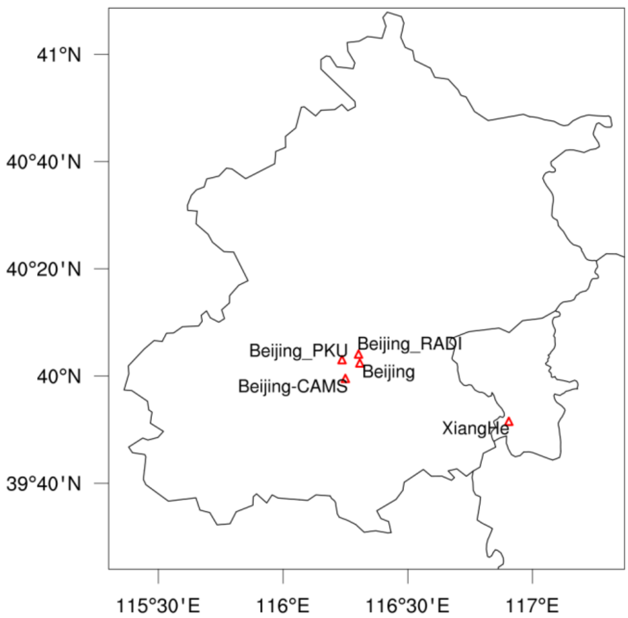

Figure 5. A zoomed-in map for AERONET sites in Beijing area, including Beijing, Beijing-CAMS, Beijing_PKU, Beijing_RADI, XiangHe.

21. L610: What is the temporal resolution of AERONET observations? From the time *series plot of Fig. 8, it looks like the data is mostly only available around 00 UTC of each day.*

Response:

We really appreciate your question. The temporal resolution of AERONET observations is several minutes, and the data in the daytime is only available because sun photometer measurements of the direct solar radiation is used to retrieve AOD.

22. L615-616: Any explanation why model has worse skill at XuZhou-CUMT? It seems the event on Nov. 25 is more severe than Nov. 26 at this site and not captured as well.

Response:

We are so sorry to give a rational explanation, the worse model skill at XuZhou-CUMT is probably due to emissions, which is needed to further study.

23. L617-618: Any guess on this? Have you looked at the meteorological conditions on these days? Could it be associated with the intensity of wind speed?

Response:

We are so sorry that we have not looked at the meteorological conditions on these days, and studied the impacts of them on assimilation. The intensity of wind speed has actually an important impact on assimilation, so combined assimilation of meteorological and aerosol states should be performed in the future.

24. L622: It would be easier for reader to understand if the data distribution map of Nov. 26 is also provided. Along the same line, I would suggest adding information of available data amount in Fig. 8 to address this.

Response:

We really appreciate your suggestion. The AOD data amount has a significant impact on assimilation, for example, no available AOD data shown in Fig. 6a can be assimilated in Beijing area due to cloud contamination where a more severe pollution happened on 26 November 2018 shown in Fig. 6b so that no assimilation benefits are generated to improve aerosol forecasts in Beijing area, meaning the control experiment and assimilation experiment on 26 November 2018 have the same performance (shown in Fig. 8a, 8b, 8c, 8d, 8e in the manuscript). The available data amount is variable from 23 to 29 November 2018. What is more, the amount of data is same, the assimilation effect may differ greatly due to different pollution cases.

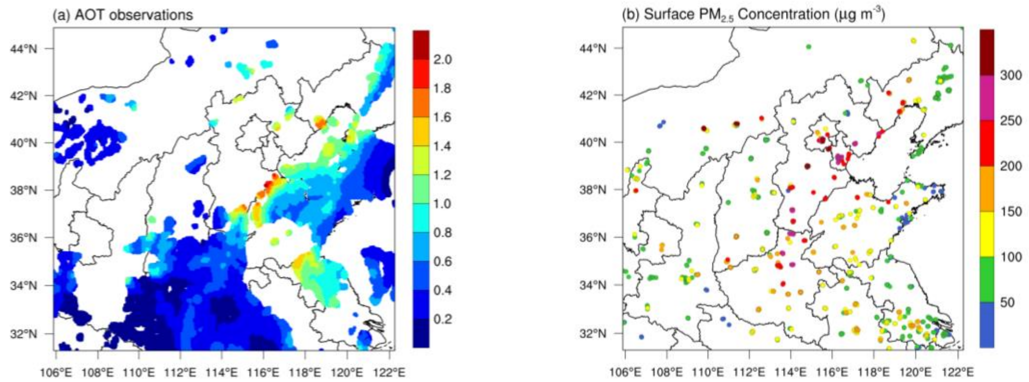

Figure 6. Observations of the thinned Himawari-8 AOTs (a) and surface $PM_{2.5}$ mass concentration (b) in D02 at 0300 UTC on 26 November 2018.

25. L643: You may remove "between analyses and the background field" since increment has been defined in the earlier paragraph.

Response:

Done. The words “between analyses and the background field” has been removed in the revised manuscript (L656-657).

26. L644-645: The of color bar scales in Fig. 3a and Fig. 9 are not consistent, which makes it hard to compare them visually. Please consider modify them.

Response:

Done. We have modified the color bar scales in Fig. 9.

27. L645: Need to mark where Tianjin is in the map, otherwise one may not know which location you talked about.

Response:

Done. We have marked Tianjin with a small black triangle in the map (L659).

28. L644: Panels in Fig. 9 are not sufficient to conclude the underestimation in control experiment as no observation is provided.

Response:

We really appreciated your suggestion. Fig. 9a shows surface $PM_{2.5}$ mass concentrations in the background field at 0300 UTC on 25 November 2018, whereas corresponding observations are provided in Fig. 3b.

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

11/24/2021