Responses to the Community comment

We would like to thank to the reviewer for giving constructive criticisms, which are very helpful in improving the quality of the manuscript. We have made major revision based on the critical comments and suggestions of the referees. The referee's comments are reproduced (*black*) along with our replies (blue) and changes made to the text (red) in the revised manuscript. All the authors have read the revised manuscript and agreed with submission in its revised form.

Anonymous Referee #1

Comment NO.1: The manuscript, titled "Improving estimation of a record breaking East Asian dust storm emission with lagged aerosol Ångström Exponent observations", by Yueming Cheng et al. is a data assimilation (DA) case study for improving dust storm predictions in China using WRF-Chem. The authors chose a dust storm in Mar 2021 as a case study and successfully showed that using AERONET observations for DA could improve hindcasting ability of dust in WRF-Chem. Authors further showed that employing the Angstrom Exponent (AE) benefits more than employing the aerosol optical thickness (AOT) in improving DA results. The methodology is generally scientifically valid and clear in presentation, although there are occasional grammar issues or unclear descriptions that could use a little more editing. The main issue I thought was that there are insufficient science discussions to the results, such as why AE benefits more than AOT, or how could the WRF-Chem dust model be improved. The manuscript so far feels a little more like a technical report on improving hindcasts rather than a scientific development on our understanding of dust modelling. I have a few major comments below for science, and I have some other specific comments on technical questions or presentation. I suggest major revision for the current revision. Response: Done. The occasional grammar issues or unclear descriptions have been rewritten.

Dust emission is a significantly uncertain process in dust simulation. In AFWA module, The dust emission factor is an important parameter needed tuning. Due to manual tuning is computationally expensive, it is beneficial to replace the tuning process with an automatic parameter estimation system to improve dust emission simulation. In this study, data assimilation, which feeds the observation information into numerical model, can be a valuable tool to automatic adjust dust emission parameters for the optimization of the estimates of dust emissions. Aerosol optical thickness (AOT) represents the total amount of atmospheric column, however, lacks the aerosol size information. Ångström Exponent (AE) which measures the wavelength-dependence of AOT and is significantly sensitive to size of aerosol particle, may have a positive impact on data assimilation (Tsikerdekis et al., 2022, 2023).

Due to WRF-Chem model has uncertainties not only on dust emission but also on dust deposition (Huang et al., 2020) and dust optical properties (Di Biagio et al., 2019) in simulation, two assimilation experiments with perturbation of dust emission and size distribution are conducted. One assimilation experiment named AOT DA-SZD only assimilates AERONET AOT observations, and the other assimilation experiment named AOT+AE DA-SZD assimilates both AERONET AOT

and AE observations. The comparison between AOT DA-SZD and AOT+AE DA-SZD experiments shows the effects of the additional AE information on dust emission optimization. Our results demonstrate that the additional assimilation of AE observations with consideration of the dust emission size distribution uncertainty are helpful to the optimization of dust emission through better adjustment of dust size distribution. AOT assimilation can only optimize the dust emission flux depending on the covariance between time-lagged AOT observations and the simulated total dust emission, while the additional inclusion of AE assimilation can optimize the size distribution of dust emission and the associated total flux depending on the covariance between time-lagged AE observations and the simulated dust emission in each bin. The sufficient science discussions to the results are given in Sect. 3.

References:

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmos. Chem. Phys., 19, 15503–15531, https://doi.org/10.5194/acp-19-15503-2019, 2019.

Huang, Y., Kok, J. F., Kandler, K., Lindqvist, H., Nousiainen, T., Sakai, T., Adebiyi, A., and Jokinen, O.: Climate Models and Remote Sensing Retrievals Neglect Substantial Desert Dust Asphericity, Geophysical Research Letters, 47, e2019GL086592, https://doi.org/10.1029/2019GL086592, 2020.

Tsikerdekis, A., Schutgens, N. A. J., Fu, G., and Hasekamp, O. P.: Estimating aerosol emission from SPEXone on the NASA PACE mission using an ensemble Kalman smoother: observing system simulation experiments (OSSEs), Geosci. Model Dev., 15, 3253–3279, https://doi.org/10.5194/gmd-15-3253-2022, 2022.

Tsikerdekis, A., Hasekamp, O. P., Schutgens, N. A. J., and Zhong, Q.: Assimilation of POLDER observations to estimate aerosol emissions, Atmos. Chem. Phys., 23, 9495–9524, https://doi.org/10.5194/acp-23-9495-2023, 2023.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 2 Line 44-47, Page 3 Line 64-65, Page 7 Line 194-208, and Sect. 3.

Comment NO.2: I think there needs to be some science discussions on what caused the biases in WRF Chem dust emissions in China. The paper currently leaves readers with puzzles regarding why WRF-Chem underestimates dust so much. Is it problems in simulated dust emissions, lifetime (dust deposition), or optical properties? If the bias comes from emission, is it a problem in wind speed, soil moisture, vegetation, or other met fields? If it's deposition or optics, is there anything to do with size distribution, dust particle shape, or dust refractive index? Does the dust underestimation also occur over the rest of the world in WRF-Chem? How does changing the a priori emissions (using other dust_opt or other emission schemes) alter the FR underestimations? Modellers would like to know how could our process-based dust understanding benefit from the

insights from this DA study.

Response: Done. Due to the predicted meteorological fields such as wind and soil moisture are constrained by NCEP Final (FNL) analysis, dust emission parameterization is a significantly uncertain process in the dust emission simulation. As Su and Fung. (2015) pointed out the underestimation of the dust emission over the Gobi desert by the AFWA scheme, in this study, to reduce the underestimation of dust emission factor is calibrated and selected as 21 based on the AERONET-observed AOT and AE. After the bias calibration of dust emission, due to WRF-Chem model has uncertainties not only on dust emission but also on dust deposition (Huang et al., 2020) and dust optical properties (Di Biagio et al., 2019) in simulation, two assimilation experiments with perturbation of dust emission and their size distribution are conducted. One assimilation experiment named AOT DA-SZD only assimilates AERONET AOT observations, and the other assimilation experiment named AOT DA-SZD and AOT+AE DA-SZD experiments shows the effects of the additional AE information on dust emission optimization.

References:

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmos. Chem. Phys., 19, 15503–15531, https://doi.org/10.5194/acp-19-15503-2019, 2019.

Huang, Y., Kok, J. F., Kandler, K., Lindqvist, H., Nousiainen, T., Sakai, T., Adebiyi, A., and Jokinen, O.: Climate Models and Remote Sensing Retrievals Neglect Substantial Desert Dust Asphericity, Geophysical Research Letters, 47, e2019GL086592, https://doi.org/10.1029/2019GL086592, 2020.

Su, L. and Fung, J. C. H.: Sensitivities of WRF-Chem to dust emission schemes and land surface properties in simulating dust cycles during springtime over East Asia, JGR Atmospheres, 120, https://doi.org/10.1002/2015JD023446, 2015.

Changes in Manuscript: Please refer the description in the revised manuscript, from Page 5 Line 144 to Page 6 Line 146 and Page 7 Line 194-208.

Comment NO.3: It looks like authors attributed all the differences/biases between AERONETmeasured and WRF-simulated AOT to dust. Could the biases be attributed to other natural and anthropogenic emissions? Although you only assimilated three days where dust was dominant, there must be some strong anthropogenic and natural emissions that got captured by AERONET, especially over Beijing, a heavily polluted metropolitan area. You only used AE values < 0.4 for evaluation to focus on dust, but it seems you didn't do the same when doing the DA. From my point of view, it could be better to use the coarse-mode AERONET AOT from the spectral deconvolution algorithm (SDA) to do the DA since all fine-mode aerosols are truncated and only dust/sea-salt

remains.

Response: Done. Since the East Asian dust storms are triggered by an exceptionally strong Mongolian cyclone and accompanied by strong northwesterly wind (Gui et al., 2021), human pollutants are difficult to accumulate in the downwind areas during this period. To minimize the influences of anthropogenic emissions, only the AERONET AOT and AE dominated by dust are assimilated to optimize the dust emissions, which are chosen with AE at 440-870 nm less than 0.4 (Huneeus et al., 2011). Therefore, the observables generally match the state variables.

Thank you for your advice. The coarse-mode AERONET AOT from the spectral deconvolution algorithm (SDA) is useful for data assimilation. However, due to the dust particle from 0.2 to 2 μ m in diameter simulated by WRF-Chem is partly included in the fine-mode fraction of SDA retrievals (O'Neill et al., 2001, 2023), it is difficult to construct new observation operator and this important work will be completed in the future.

References:

Gui, K., Yao, W., Che, H., An, L., Zheng, Y., Li, L., Zhao, H., Zhang, L., Zhong, J., Wang, Y., and Zhang, X.: Two mega sand and dust storm events over northern China in March 2021: transport processes, historical ranking and meteorological drivers, https://doi.org/10.5194/acp-2021-933, 1 December 2021.

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C. S.: Global dust model intercomparison in AeroCom phase I, Atmospheric Chemistry and Physics, 11, 7781–7816, https://doi.org/10.5194/acp-11-7781-2011, 2011.

O'Neill, N. T., Eck, T. F., Holben, B. N., Smirnov, A., Dubovik, O., and Royer, A.: Bimodal size distribution influences on the variation of Angstrom derivatives in spectral and optical depth space, J. Geophys. Res., 106, 9787–9806, https://doi.org/10.1029/2000JD900245, 2001.

O'Neill, N. T., Ranjbar, K., Ivănescu, L., Eck, T. F., Reid, J. S., Giles, D. M., Pérez-Ramírez, D., and Chaubey, J. P.: Relationship between the sub-micron fraction (SMF) and fine-mode fraction (FMF) in the context of AERONET retrievals, Atmos. Meas. Tech., 16, 1103–1120, https://doi.org/10.5194/amt-16-1103-2023, 2023.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 4 Line 101-102 and Page 20 Line 403-404.

Comment NO.4: Even if all biases in AOT/AE were assumed to be due to dust, currently the authors attribute all differences/biases between observations and simulations to emissions and only correct emissions. This presumes there are no biases in dust settling/deposition and dust optical properties. But if so, assimilating AOTs should correct most of the error, and AE would not be needed. A science discussion is needed on why the AE information could further reduce bias. Authors could not just conclude that the more you use for DA the better the hindcast results. My thought is that if AE is

additionally needed for DA, this either means there are problems in AERONET AOT, or (more likely) WRF-Chem has problems not only on dust emissions but also on dust optics. Studies also have pointed out issues in both settling velocity (e.g., Huang et al., 2020) and optics (e.g., Di Biagio et al., 2019) in models. If so, it does not make so much sense to attribute all AOT/AE biases to emissions to compensate other errors in WRF-Chem. Maybe optics should also be inverted, not just emissions.

Response: Done. To investigate the influences of AERONET AOT and AE assimilation on the dust emission optimization, three assimilation experiments are conducted from 12:00 UTC on 11 March 2021 to 00:00 UTC on 24 March 2021. Due to WRF-Chem model has uncertainties not only on dust emission but also on dust deposition (Huang et al., 2020) and dust optical properties (Di Biagio et al., 2019) in simulation, two assimilation experiments with perturbation of dust emission and size distribution are conducted. One assimilation experiment named AOT DA-SZD only assimilates AERONET AOT observations, and the other assimilation experiment named AOT+AE DA-SZD assimilates both AERONET AOT and AE observations. 20 ensemble members are generated by perturbing the dust emission in each bin, and the perturbation factor of each bin has a mean of 1 and a spread of 0.6 followed the lognormal distribution. Correlated noise is used across the dust size bins in the perturbation, and the noise correlation decreases with increased difference of the diameter among the bins (Di Tomaso et al., 2017). The ensemble prediction dynamically estimates the covariance between the dust emission in each bin and the aerosol optical properties. The comparison between AOT DA-SZD and AOT+AE DA-SZD experiments shows the effects of the additional AE information on dust emission optimization. The effects of dust emission size distribution perturbation are investigated by one additional assimilation experiment named AOT+AE DA, which is conducted as same as the AOT+AE DA-SZD experiment except the 20 ensemble members are generated by perturbing the dust emission in each bin with same perturbation factor.

References:

Di Biagio, C., Formenti, P., Balkanski, Y., Caponi, L., Cazaunau, M., Pangui, E., Journet, E., Nowak, S., Andreae, M. O., Kandler, K., Saeed, T., Piketh, S., Seibert, D., Williams, E., and Doussin, J.-F.: Complex refractive indices and single-scattering albedo of global dust aerosols in the shortwave spectrum and relationship to size and iron content, Atmos. Chem. Phys., 19, 15503–15531, https://doi.org/10.5194/acp-19-15503-2019, 2019.

Di Tomaso, E., Schutgens, N. A. J., Jorba, O., & Pérez García-Pando, C. Assimilation of MODIS Dark Target and Deep Blue observations in the dust aerosol component of NMMB-MONARCH version 1.0. Geoscientific Model Development, 10(3), 1107–1129. https://doi.org/10.5194/gmd-10-1107-2017, 2017.

Huang, Y., Kok, J. F., Kandler, K., Lindqvist, H., Nousiainen, T., Sakai, T., Adebiyi, A., and Jokinen, O.: Climate Models and Remote Sensing Retrievals Neglect Substantial Desert Dust Asphericity, Geophysical Research Letters, 47, e2019GL086592,

https://doi.org/10.1029/2019GL086592, 2020.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 194-208.

Comment NO.5: *Line 78: It's a little difficult to grasp how many AERONET stations you used for assimilation. Please state in text. I suggest plotting out the locations of the AERONET stations, with values of AOT and AE, either in Fig. 1 or in SI.*

Response: Done. As shown in Fig. 1(a), there are 5 AERONET sites with available observations from 14 March to 23 March 2021 for data assimilation, including 4 sites named as Beijing-CAMS (39.93°N, 116.32°E), Beijing (39.98°N, 116.38°E), Beijing_PKU (39.99°N, 116.31°E), and Beijing_RADI (40.00°N, 116.38°E) in the downwind area and a site near the dust source region named as Dalanzadgad (43.58°N, 104.42°E). The assimilated AOT and AE values at the AERONET sites are also given in Fig. 1(b,d). For Dalanzadgad site, the AOT values from 14 March to 17 March 2021 are significantly higher than those from 18 March to 23 March, while AE values show the opposite features.

Changes in Manuscript: The AERONET stations used for assimilation are stated in Page 4 Line 105-111 and the locations of the AERONET stations are plotted in Fig. 1.

Comment NO.6: *Lines 87-88: Authors wrote two observation errors: "observation error is a sum of representation error and observation error", which is confusing. Maybe use another word like instrument error for the latter one?*

Response: Done. We have replaced the latter "observation error" with "instrument error".

Changes in Manuscript: Please refer the modification in the revised manuscript, Page 3 Line 92.

Comment NO.7: Line 89: I glanced through Schmid 1999 but didn't see such characterization of representation error. Schmid 99 was not a WRF-Chem modelling study either. Why should representation error be 0.055τ ? Should it be more related to WRF-Chem grid resolution? Also, please define τ .

Response: Done. The representation error has been recalculated.

Due to the representation error is related to the WRF-Chem grid resolution, the representation error in AERONET AOT and AE is calculated depending on the AOT and AE temporal variability of AERONET and WRF-Chem with 45 km horizontal resolution (Schutgens et al., 2010). By averaging results at all AERONET sites in March 2021, the relative AOT temporal variations of AERONET and WRF-Chem in 1 h interval are 0.11τ and 0.1τ , while the AE temporal variations of AERONET and WRF-Chem in 1 h interval are 0.05 and 0.02, respectively. Therefore, the representation errors in AERONET AOT (τ) and AE in the 1 h interval are $\epsilon_r = 0.01\tau$ and $\epsilon_r =$ 0.03, respectively. The instrument error in AOT is defined as 0.015 and the instrument error of AE is estimated by propagating the instrument error in AOT at 440 and 870 nm (Schutgens et al., 2010). References:

Schutgens, N. A. J., Miyoshi, T., Takemura, T., & Nakajima, T. Applying an ensemble Kalman filter to the assimilation of AERONET observations in a global aerosol transport model. Atmos.

Chem. Phys., 16, 2010.

Changes in Manuscript: Please refer the description in the revised manuscript, from Page 3 Line 92 to Page 4 Line 100.

Comment NO.8: *Line 92: I think it is needed to state why authors chose to assimilate AERONET instead of SONET or CALIOP. It looks random to me.*

Response: AErosol RObotic NETwork (AERONET), which both include ground-based AOT and AE observations, is chosen as the assimilated observations to investigate the sensitivity of dust emission to size information in this study. The Sun-Sky Radiometer Observation Network (SONET) and CALIPSO observations are used for independent validation.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 3 Line 72-73 and Line 76-77.

Comment NO.9: *Line 93: Please also plot out the locations of the SONET sites and their values of AOT and AE, in main text or supplement.*

Response: Done. As shown in Fig. 1(a), there are 5 SONET sites with available observations from 14 March to 23 March 2021, including: Yanqihu (40.40°N, 116.67°E), Beijing (40.00°N, 116.37°E), Jiaozuo (35.18°N, 113.20°E), Songshan (34.53°N, 113.09°E), and Zhengzhou (34.70°N, 113.66°E). The AOT and AE values at the SONET sites are also given in Fig. 1 (c,e). Similar with Dalanzadgad site, Jiaozuo, Songshan, and Zhengzhou sites experience a stronger dust process from 14 March to 17 March 2021 with higher AOTs and lower AEs.

Changes in Manuscript: The locations of the SONET sites and their values of AOT and AE are plotted in Fig. 1.

Comment NO.10: *Line 117: Please define the acronym MOSAIC. I am not sure how important this modification is if you don't concern dust chemistry, since MOSAIC never appeared in the text again. How are the spatiotemporal distributions of metal ions changed through this modification?*

Response: Done. We have defined the acronym MOSAIC. The aerosol module used in this study is GOCART and the chemical composition of dust is unchanged during Mie calculation.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 6 Line 150.

Comment NO.11: Line 140: Again, it looks like the whole difference between AERONET AOTs and simulated AOTs are attributed toward dust emission biases. Can't there be biases from other natural and anthropogenic emissions?

Response: Since the East Asian dust storms are triggered by an exceptionally strong Mongolian cyclone and accompanied by strong northwesterly wind (Gui et al., 2021), human pollutants are difficult to accumulate in the downwind areas during this period. To minimize the influences of anthropogenic emissions, only the AERONET AOT and AE dominated by dust are assimilated to optimize the dust emissions, which are chosen with AE at 440-870 nm less than 0.4 (Huneeus et al., 2011). Therefore, the whole difference between AERONET AOTs and simulated AOTs are attributed toward dust emission biases. References:

7

Gui, K., Yao, W., Che, H., An, L., Zheng, Y., Li, L., Zhao, H., Zhang, L., Zhong, J., Wang, Y., and Zhang, X.: Two mega sand and dust storm events over northern China in March 2021: transport processes, historical ranking and meteorological drivers, https://doi.org/10.5194/acp-2021-933, 1 December 2021.

Huneeus, N., Schulz, M., Balkanski, Y., Griesfeller, J., Prospero, J., Kinne, S., Bauer, S., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Fillmore, D., Ghan, S., Ginoux, P., Grini, A., Horowitz, L., Koch, D., Krol, M. C., Landing, W., Liu, X., Mahowald, N., Miller, R., Morcrette, J.-J., Myhre, G., Penner, J., Perlwitz, J., Stier, P., Takemura, T., and Zender, C. S.: Global dust model intercomparison in AeroCom phase I, Atmospheric Chemistry and Physics, 11, 7781–7816, https://doi.org/10.5194/acp-11-7781-2011, 2011.

Comment NO.12: *Line 145: I am not sure if the error covariance includes forward model error (that is, error in the WRF-Chem H operator). Please clarify.*

Response: The analysis error covariance includes forward model error. WRF-Chem is served as observation operator and the dust emissions in each size bin are perturbed independently for AOT DA-SZD and AOT+AE DA-SZD experiments. Perturbation in the dust size distribution can lead to variations in the deposition process and optical calculations, further resulting in modifications to the meteorological field due to aerosol-radiation interactions. Therefore, forward model error is included in the error covariance.

Comment NO.13: Line 155: A localization length of 600 km sounds a little too big to me for assimilating AERONET data. It is almost a meso-synoptic length scale and is much bigger than your WRF-Chem horizontal grid resolution.

Response: The localization length of 600 km, which obtained through tuning, is selected as a reasonable parameter for the dust emission inversion used in Dai et al. (2019). Asian dust can be transported over long distances and affect areas far downwind. The localization length determines the assimilated observations in the horizontal space. Localization length larger than 600 km causes more observations be assimilated for the analyses at a grid point, while localization length less than 600 km makes it difficult for the time-lagged observations in downwind areas to be utilized. References:

Dai, Cheng, Goto, Schutgens, Kikuchi, Yoshida, et al. Inverting the East Asian Dust Emission Fluxes Using the Ensemble Kalman Smoother and Himawari-8 AODs: A Case Study with WRF-Chem v3.5.1. Atmosphere, 10(9), 543. https://doi.org/10.3390/atmos10090543, 2019.

Comment NO.13: Line 155: "Grid centroid" instead of "centre grid"?

Response: Accept. We have replaced "centre grid" with "grid centroid".

Changes in Manuscript: Please refer the modification in the revised manuscript, Page 7 Line 191. **Comment NO.14:** Figure 1: It's interesting that using AE measurements in Beijing could lead to changes in the posterior AE (AOT+AE DA) over Taklimakan, or even in India, in comparison to the a priori AE (FR). How does DA generate emission changes in Taklimakan and India, if you were using AERONET sites in Beijing? **Response:** In the original manuscript, all the AERONET sites in the domain are used for data assimilation, hence, the posterior AEs in Taklimakan and India are both changed in comparison to the a priori AEs. In the revised manuscript, only 5 AERONET sites in Fig. 1 are used for data assimilation, hence, the regions (e.g., India) beyond the 600 km observation localization length are generally unchanged. If we only using AERONET sites in Beijing, DA is difficult to generate obviously emission changes in Taklimakan and India because of the 600 km localization length.

Comment NO.15: Line 203-206: It is not clear what this means. There were no observations because AERONET sites were down, like because of the dust storm? Please rephrase. Does this mean if you use the observations on 14-15 March, dust emissions would be even higher? Please clarify.

Response: We have deleted the sentence. It doesn't mean if we use the observations on 14-15 March, dust emissions would be even higher. Based on the assumption that the model background error covariance is correct and reasonable, the optimization of dust emission is generally invariant with the assimilated observations at different times.

Comment NO.16: *Line 208: I suggest adding map plots on the prior error of the WRF FR emissions, as well as the posterior errors of inverted emissions for the AOT DA and the AOT+AE DA cases. It helps visualize how adding AOT and AE for DA reduces the posterior errors of DA emissions.*

Response: Accept. The ratios between posterior error of dust emission and the prior one in each dust bin for the three assimilation experiments during 14-17 March 2021 and 18-23 March 2021 are given in Fig. 4 and Fig. 6.

Changes in Manuscript: Please refer the detailed description in Sect. 4.1 in the revised manuscript. **Comment NO.17:** *Figure 4: When you say "aggregate", is this plot averaged across or summed across the domain? Please clarify in text.*

Response: The plot is summed across the domain.

Changes in Manuscript: Please refer the caption of Fig. 2 in the revised manuscript.

Comment NO.18: *Line 213: I suggest either saying posterior and prior dust emissions, or a posteriori and a priori dust emissions.*

Response: Done.

Changes in Manuscript: Please refer the modification in the revised manuscript.

Comment NO.19: *Lines 217-219: So, did you use more AERONET sites than listed here for DA above? Or are these all the sites used for DA? I am still confused, please clarify.*

Response: In the revised manuscript, the AERONET sites used for data assimilation are same as those for self-validation.

Changes in Manuscript: Please refer the detailed description of assimilated AERONET observations in the revised manuscript, Page 4 Line 105-111.

Comment NO.20: *Line 236: Here authors should suggest scientific reasons for why using AE would benefit DA so much, while AOT DA would not.* **Response: Done**.

9

Our results demonstrate that the additional assimilation of AE observations with consideration of the dust emission size distribution uncertainty are helpful to the optimization of dust emission through better adjustment of dust size distribution. AOT assimilation can only optimize the dust emission flux depending on the covariance between time-lagged AOT observations and the simulated total dust emission, while the additional inclusion of AE assimilation can optimize the size distribution of dust emission and the associated total flux depending on the covariance between time-lagged AE observations and the simulated dust emission in each bin.

Changes in Manuscript: Please refer the detailed description in Sect. 4 in the revised manuscript. **Comment NO.21:** *Line 248: What's the reason of selecting these two SONET sites but not the other two? It looks a little random here. I suggest plotting the comparisons for Songshan and Jiaozuo in main text or supplement too.*

Response: Done.

Changes in Manuscript: The comparison of the simulated AOTs and AEs with the observed ones over all the AERONET and SONET sites are given in Fig. 9 and Fig. 10. Please refer the detailed description in Sect. 4.2 in the revised manuscript.

Comment NO.22: *Line 255: From here on, I start to find the message for the next few subsections a bit repetitive, stating that the AOT+AE DA run is better than the FR run and the AOT DA run. The manuscript could use a little rewriting to make the discussion and message more succinct.* **Response:** Done.

Changes in Manuscript: Please refer the detailed description in Sect. 4 in the revised manuscript. **Comment NO.23:** Figure 6: It seems to readers there are insufficient SONET data points for the time series plot. I suggest authors also include SONET data points for AE > 0.4 since you used it for DA, in any color other than grey.

Response: As shown in Fig. 9 and Fig. 10, we extended the experiment period from 14-17 March 2021 to 14-23 March 2021 to get enough SONET data points. Due to only the AERONET observations with AE<0.4 are used for data assimilation, it is not necessary to include SONET data points for AE > 0.4.

Comment NO.24: Figure 7-9: These are nice plots. Though, readers find the message across Figs. 7-9 a little repetitive. I would suggest showing the extinction coefficient (second rows) and skip the AOT plots (first rows), and maybe combine second rows of all three figures together. The first rows could be put in supplement.

Response: Done.

Changes in Manuscript: Please refer the extinction coefficient plots in Fig. 12 in the revised manuscript and the AOT plots in Fig. S8 in supplement.