Response to the Comments of Referees

We would like to thank to the reviewers for giving constructive criticisms, which are very helpful in improving the quality of the manuscript. We have made minor 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: *Thanks for spending a lot of effort revising the manuscript following the comments and suggestions. I think most of my comments and suggestions are addressed. I just wanted to echo what the 2nd reviewer has said, that the DA gave a relatively homogeneous dust emission change across the domain (Fig. 3a and 3b-d). This makes sense because the AERONET sites are far away from the two deserts. I think partly due to this reason, in the introduction and conclusion sections the authors kind of shifted the emphasis from improving spatiotemporal variability of dust to improving dust size distribution. My only concern here is that there is little evaluation on the posterior dust size distribution by comparing simulations against observations, which makes this paper's ending not very strong. At current stage I think authors could argue DA changes size distribution, but it is less obvious that DA improves it. Of course the posterior error decreases, but it does not necessarily mean improvements in the simulations.*

I think there are little in-situ size distribution data from field campaigns, so there is not much to do about it. But, I would just suggest that authors make a simple plot on the prior and posterior dust volume size distributions from WRF (e.g., Ryder et al., 2018, Figs. 5-7; Li et al. 2022, Fig. 6), for grids containing the AERONET site and the two deserts. This will be helpful if future in-situ measurements come up from Chinese field campaigns. This also helps readers more directly visualize how DA changes the prior WRF dust size distribution to the posterior. (Fig. S3 is also very helpful by breaking things down into bins.)

Optional: Authors could also do a simple comparison against AERONET inversion data for some references of size distribution (e.g., https://aeronet.gsfc.nasa.gov/cgi-bin/data_display_inv_v3?site=Dalanzadgad&nachal=0&year=2021&month=3&day=16& aero_water=0&level=2&if_day=0&if_err=0&place_code=10&DATA_TYPE=76&year_or_m onth=0). Doing a small cross-validation for the AERONET size distribution and the WRF size distribution will make the paper's position stronger. I think this suggestion is more optional since AERONET inversion data also has uncertainties based on inversion of remote sensing data.

References:

Ryder, C. L., Marenco, F., Brooke, J. K., Estelles, V., Cotton, R., Formenti, P., McQuaid, J. B., Price, H. C., Liu, D., Ausset, P., Rosenberg, P. D., Taylor, J. W., Choularton, T., Bower, K., Coe, H., Gallagher, M., Crosier, J., Lloyd, G., Highwood, E. J., and Murray, B. J.: Coarse-mode mineral dust size distributions, composition and optical properties from AER-D aircraft measurements over

the tropical eastern Atlantic, Atmos. Chem. Phys., 18, 17225–17257, https://doi.org/10.5194/acp-18-17225-2018, 2018.

Li, L., Mahowald, N. M., Kok, J. F., Liu, X., Wu, M., Leung, D. M., Hamilton, D. S., Emmons, L. K., Huang, Y., Sexton, N., Meng, J., and Wan, J.: Importance of di2erent parameterization changes for the updated dust cycle modeling in the Community Atmosphere Model (version 6.1), Geosci. Model Dev., 15, 8181–8219, https://doi.org/10.5194/gmd-15-8181-2022, 2022.

Response: Done. The normalized atmospheric volume distribution in Gobi desert, Taklimakan desert, and downwind area including AERONET sites for the four experiments during 14-17 March 2021 and 18-23 March 2021 is given in Fig. S6.

Changes in Manuscript: Please refer the caption of Fig. S6 in the Supplements.

Comment NO.2: *Figure 5: I am interested in Fig. 5 in terms of why there are increases over the Taklimakan and some regions of the Gobi but decreases over the majority of the Gobi. I think a science explanation is needed in the main text, but I could not find it.*

Response: Done. Compared with the FR experiment, the dust emission in the AOT+AE DA-SZD experiment is increased over the Taklimakan desert and some regions of the Gobi desert but decreased over the majority of the Gobi desert. This is because the dust emission periods vary across different grids, leading to the opposite trends among different dust source regions.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 11 Line 279-282.

Comment NO.3: *Lines 247-250: I think there needs a science explanation on why or how adding AE changes dust emissions more so in bin 1 and bin 3 than other.*

Response: Done. The significant decrease in bin 1 is due to bin 1 having the highest dust extinction efficiency, and the significant increase in bin 3 is due to bin 3 having the largest proportion in the fine-mode dust emission.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 10 Line 256-257.

Comment NO.4: *Figs. 7-8: Please add site labels to figure caption so people don't need to scroll back to Fig. 1 to look for what the colors mean.*

Response: Done. The site labels are added to figure caption.

Changes in Manuscript: Please refer the caption of Figs. 7-8 in the revised manuscript.

Comment NO.5: *Fig. 3a-d: Please indicate the unit of dust emissions on the colorbars or in the figure caption. Same for Fig. 5.*

Response: Done. The unit of dust emissions is given in caption of Fig. 3 and Fig. 5.

Changes in Manuscript: Please refer the caption of Fig. 3 and Fig. 5 in the revised manuscript.

Comment NO.6: *In Fig. 1a: Please indicate what the color scale means (although we somehow can guess it).*

Response: Done. The meaning of color scale is given in the figure caption.

Changes in Manuscript: Please refer the caption of Fig. 1 in the revised manuscript.

Anonymous Referee #2

Comment NO.1: *The study investigates the effect of assimilating Aerosol Optical Depth (AOD) at 550nm and Angstrom Exponent (AE) between 440 to 870nm simultaneously for a dust event over East Asia in 2022. The authors attempt to explain the added value of assimilating AOD+AE compared to only AOD by conducting assimilation experiments with WRF-Chem where the emissions of each bin are perturbed independently. In addition, a distinctive experiment where the emissions of each bin are perturbed with the same lognormal distribution is conducted to highlight the limitation of that set up. The evaluation is done with both assimilated (AERONET) and independent (SONET and CALIOP) observations, which further solidifies the validity of the results. Overall, the figures are well presented and discussed. As in prior studies, the use of additional optical properties (except AOD) is highlighted, hence I believe the topic is relevant to ACP scope. Since this is a resubmission, I had a brief look at the initial version and confirmed that a lot has been improved (period expanded, statistics, text). I recommend the publication of the manuscript after addressing the following issues:*

The drawback of the current version is the language and the grammatical mistakes on parts of the manuscript, which sometimes get in the way of understanding parts of the study (see specific comments and technical corrections). I am sure that this can be much improved by the author if they do a thorough re-check of all the text.

Response: We thank the referee for this very positive assessment of our manuscript. The thorough re-check of all the text has been done.

Comment NO.2: *In addition, defining a model uncertainty based on either prior studies or on some kind of evaluation analysis is essential. At the moment a specific standard deviation (0.6) is set for the perturbation lognormal distribution. If this is set higher than the true model uncertainty the assimilation will falsely trust the observations more and if this is set lower than the true model uncertainty the assimilation will falsely trust the model more. I would highly recommend being more specific on that matter.*

Response: Refer to Dai et al. (2019), the standard deviation of 0.6 corresponds to the uncertainty of the dust emissions for 14 global models (Huneeus et al., 2011).

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.

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.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 206-207.

Comment NO.3: *L17-19: This sentence needs to be rephrased.*

Response: Accept. The sentence has been modified as "Due to the lack of observation during dust storms and the accuracy of satellite-retrieved AE depending on the instrument and retrieval algorithm, it is possible to estimate the dust storm emission using the time-lagged ground-based AE observations."

Changes in Manuscript: Please refer the description in the revised manuscript, Page 1 Line 17-19.

Comment NO.4: *L17: What do you mean by "lagged" ground observations? Lagged in the and space, as the dust was emitted a few days earlier in the source region before it was transported over the observation site? Could you be more specific and rephrase?*

Response: The "lagged" means the observation at a later time contains the information about the dust emitted a few days earlier in the source region. We have replaced "lagged" with "time-lagged".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 1 Line 17-19.

Comment NO.5: *L24: This sentence needs to be rephrased. In the current form the reader cannot determine where these improvements are referring to. Be more specific on the experiments you are comparing, the reference you are using, which variables you are evaluating and the metrics you calculate. Smaller sentences always help.*

Response: Done. The sentence has been rewritten as "Validation by independent observations from Skynet Observation NETwork (SONET) shows that assimilating additional AE information reduces the root mean square error (RMSE) of simulated AOT and AE by approximately 17% and 61% respectively, as shown by the comparison between the AOT DA-SZD and AOT+AE DA-SZD experiments."

Changes in Manuscript: Please refer the description in the revised manuscript, Page 1 Line 23-26.

Comment NO.6: *L25-26: "can be improved" or "it is improved"?*

Response: Agree, "can be improved" has been replaced by "are improved".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 1 Line 26-27.

Comment NO.7: *L45-47: Consider looking at the AEROCOM I and AEROCOM III studies. The simulated dust size range and model resolution are major factors for the diversity we observe in the simulated total emission flux.*

Response: Done. The sentence has been modified as "However, due to differences in the parameterizations of dust source fluxes, dust particle sizes, and model resolutions, simulated East Asian dust emissions vary by more than an order of magnitude among different models (Textor et al., 2006; Uno et al., 2006; Gliß et al., 2021; Kok et al., 2021), indicating that dust emission is a highly uncertain process in dust simulation."

References:

Textor, C., Schulz, M., Guibert, S., Kinne, S., Balkanski, Y., Bauer, S., Berntsen, T., Berglen, T., Boucher, O., Chin, M., Dentener, F., Diehl, T., Easter, R., Feichter, H., Fillmore, D., Ghan, S.,

Ginoux, P., Gong, S., Grini, A., Hendricks, J., Horowitz, L., Huang, P., Isaksen, I., Iversen, T., Kloster, S., Koch, D., Kirkeva, A., Kristjansson, J. E., Krol, M., Lauer, A., Lamarque, J. F., Liu, X., Montanaro, V., Myhre, G., Penner, J., Pitari, G., Reddy, S., Seland, Ø., Stier, P., Takemura, T., and Tie, X.: Analysis and quantification of the diversities of aerosol life cycles within AeroCom, Atmos. Chem. Phys., 37, 2006.

Gliß, J., Mortier, A., Schulz, M., Andrews, E., Balkanski, Y., Bauer, S. E., Benedictow, A. M. K., Bian, H., Checa-Garcia, R., Chin, M., Ginoux, P., Griesfeller, J. J., Heckel, A., Kipling, Z., Kirkevåg, A., Kokkola, H., Laj, P., Le Sager, P., Lund, M. T., Lund Myhre, C., Matsui, H., Myhre, G., Neubauer, D., Van Noije, T., North, P., Olivié, D. J. L., Rémy, S., Sogacheva, L., Takemura, T., Tsigaridis, K., and Tsyro, S. G.: AeroCom phase III multi-model evaluation of the aerosol life cycle and optical properties using ground- and space-based remote sensing as well as surface in situ observations, Atmos. Chem. Phys., 21, 87–128, https://doi.org/10.5194/acp-21-87-2021, 2021.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 2 Line 46-49.

Comment NO.8: *L64-66: Also consider the series of paper by Chen (2018 ACP, 2019 ACP).*

Response: Done. The estimated emission may be misrepresented by not including observations related to size (Chen et al., 2018, 2019; Tsikerdekis et al., 2021).

References:

Chen, C., Dubovik, O., Henze, D. K., Lapyonak, T., Chin, M., Ducos, F., Litvinov, P., Huang, X., and Li, L.: Retrieval of desert dust and carbonaceous aerosol emissions over Africa from POLDER/PARASOL products generated by the GRASP algorithm, Atmos. Chem. Phys., 18, 12551–12580, https://doi.org/10.5194/acp-18-12551-2018, 2018.

Chen, C., Dubovik, O., Henze, D. K., Chin, M., Lapyonok, T., Schuster, G. L., Ducos, F., Fuertes, D., Litvinov, P., Li, L., Lopatin, A., Hu, Q., and Torres, B.: Constraining global aerosol emissions using POLDER/PARASOL satellite remote sensing observations, Atmos. Chem. Phys., 19, 14585–14606, https://doi.org/10.5194/acp-19-14585-2019, 2019.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 3 Line 67-68.

Comment NO.9: *L98-100: Could you provide the instrument error for AE?*

Response: Done. The instrument error in AOT (ϵ_{AOTi}) is defined as 0.015 and the instrument error of AE (ϵ_{AEi}) is estimated by propagating the instrument error in AOT at 440 and 870 nm as $\epsilon_{AEi} = \sqrt{((\epsilon_{AOTi}/\tau_{870})^2 + (\epsilon_{AOTi}/\tau_{440})^2)/(ln(870/440)^2)}$ (Schutgens et al., 2010).

Changes in Manuscript: Please refer the description in the revised manuscript, Page 4 Line 100-102.

Comment NO.10: *L199-201: It is not clear if you are using the same perturbation factor across your whole domain or if you are defining your perturbation per grid. Could you please mention it here? Also please consider revising this sentence, it is very hard to read.*

Response: We use the same perturbation factor across the whole domain. This sentence has been modified as "20 ensemble members are generated by perturbing the emission fluxes in each of the five dust bins. The same perturbation factor is used across the whole domain. The random

perturbation factor is drawn from a lognormal distribution with a mean of 1 and a standard deviation of 0.6."

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 204-206.

Comment NO.11: *L201: The lognormal distribution spread is set to 0.6. By spread you mean standard deviation? Also, since this value determines the emission/optical property uncertainty of your ensemble, is it based on a specific analysis you conducted or based on a prior study?*

Response: Yes, spread means the standard deviation. Refer to Dai et al. (2019), the standard deviation of 0.6 corresponds to the uncertainty of the dust emissions for 14 global models (Huneeus et al., 2011).

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.

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.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 206-207.

Comment NO.12: *L209: As a general comment here: There is model spin-up as well as data assimilation spin-up (see Tsikerdekis et al., 2022). Are you referring to model spin-up here? Note that emissions may be corrected even in the first cycle (fully in the 6th cycle on your system), but it may take several days to see the effect on the aerosol optical properties, assuming of course you have a continues flow of assimilated observations.*

Response: This is not referring to model spin-up and we have modified the statement.

The initial condition at 12:00 UTC on 11 March 2021 is prepared by an 11-day simulation executed by WRF-Chem without any aerosol data assimilation as a spin-up. The results from 12:00 UTC on 11 March 2021 to 23:59 UTC on 13 March 2021 are excluded in the analysis.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 198-200 and Page 8 Line 214-215.

Comment NO.13: *L209-210: Is the baseline experiment an ensemble run or a single run? The difference between the two may be minuscule since you lognormal distribution has a mean of 1, but still it is important to check the differences per bin. If it is an ensemble run, which perturbation set-up is using, the DA-SZD or the regular SZD?*

Response: The baseline experiment is a single run. We have checked that the differences per bin are limited.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 8 Line 215.

Comment NO.14: *L84: "in global" to"globally"*

Response: Done. "in global" has been replaced by "globally".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 3 Line 86.

Comment NO.15: *L88: "of the ones at" to "of the AOTs at"*

Response: Done. "of the ones at" has been replaced by "of the AOTs at".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 3 Line 90.

Comment NO.16: *L150: "as same as" to "the in"*

Response: Done. "as same as" has been replaced by "the in".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 6 Line 153.

Comment NO.17: *L196: "has uncertainties" to "uncertainties"*

Response: Done. "has uncertainties" has been replaced by "uncertainties".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 200.

Comment NO.18: *L197: "in simulation" can be deleted.*

Response: Done. "in simulation" has been deleted.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 201.

Comment NO.19: *L198: "only assimilates" to "assimilated only"*

Response: Done. "only assimilates" has been replaced by "assimilated only".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 7 Line 202.

Comment NO.20: *L206: "as same as" to "as"*

Response: Done. "as same as" has been replaced by "as".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 8 Line 213.

Comment NO.21: *L207: "except the" to "except that the"*

Response: Done. "except the" has been replaced by "except that the".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 8 Line 213.

Comment NO.22: *L207: "with same" to "with the same"*

Response: Done. "with same" has been replaced by "with the same".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 8 Line 214.

Comment NO.23: *L209: "as the spin-up" to "as spin-up"*

Response: Done. "as the spin-up" has been deleted.

Changes in Manuscript: Please refer the description in the revised manuscript, Page 8 Line 215.

Comment NO.24: *L274: "with additional AE" to "with the addition of AE"*

Response: Done. "with additional AE" has been replaced by "with the addition of AE".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 12 Line 286.

Comment NO.25: *L301: "due to the uncertainty" to "since the uncertainty"*

Response: Done. "due to the uncertainty" has been replaced by "since the uncertainty".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 314.

Comment NO.26: *L302: "Those" to "These results"*

Response: Done. "Those" has been replaced by "These results".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 315.

Comment NO.27: *L303: "to the optimization" to "for the optimization"*

Response: Done. "to the optimization" has been replaced by "for the optimization".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 316.

Comment NO.28: *L303: "better" to "the"*

Response: Done. "better" has been replaced by "the".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 316.

Comment NO.29: *L304: "The similar" to "Similar"*

Response: Done. "The similar" has been replaced by "Similar".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 317.

Comment NO.30: *L304: "with assimilated" to "with the assimilated"*

Response: Done. "with assimilated" has been replaced by "with the assimilated".

Changes in Manuscript: Please refer the description in the revised manuscript, Page 14 Line 317.