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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Improving the Sectional MOSAIC Aerosol models of WRF-Chem with the revised Gridpoint Statistical Interpolation System and multi-wavelength aerosol optical measurements: DAO-K experiment 2019 at Kashi, near the Taklamakan Desert, northwestern China" *by* Wenyuan Chang et al.

Anonymous Referee #1

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The manuscript presents the development of assimilation of aerosol observations into WRF-Chem using the GSI system using approaches that are different to those used in previous studies. These developments are then tested for a case of assimilating ground-based observations of particle mass concentration, scattering and absorption coefficients, and AOD, performing sensitivity simulations on assimilating datasets in-

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Discussion paper

dependently and jointly. This is done for a single site located in Kashi, representative of dust conditions. This study it's within the scope of ACP and represents good contributions to the field as it develops a tool that could be used by the community and highlights shortcomings in the techniques a and how could they be improved. I think the paper needs a bit more work before it's ready for publication based on the comments below.

My main comments are the following.

- While the WRF-Chem optical properties module assumes Mie theory which is based on particles being spherical, the testing of the tool is focused on dust which are mostly non-spherical particles. This is briefly mentioned in the article, but I would like to see more on the subject, including looking into literature that has explored this topic and discussion on what discrepancies obtained in this study could be explained by this issue. See more on by line comments

- I believe that what the authors defined as Adjoint operators are really the tangent linear models, i.e., the derivative of the observables with respect to the inputs (aerosol mass). The adjoint operates on perturbations on the observables and outputs the expected perturbations on inputs. Please verify with the literature and correct accordingly.

- Assess representation of some intensive properties such as size (e.g., angstrom exponent, ratio of pm2.5 to pm10), single-scattering albedo, and mass scattering efficiency to try to understand mismatches when doing assimilation. A little bit is done but it would be very helpful to expand this topic and use the nomenclature used in the literature. See more on by line comments

- Absorption seems completely biased even after assimilation, this points to issues probably related to underestimation of imaginary refractive index of dust. Look for literature on this depending on the deserts, I believe Chinese deserts tend to have darker (i.e., more absorbing) sands.

Comments by line:

103-108. For completion, consider citing and discussing the study by Kumar et al (2019) that also uses GSI with CMAQ but does not use the CRTM as Tang study. This study also provides an alternative way of computing the BEC matrix (other than the NMC method) which you discuss in section 2.3

545-552. Are all of these observations in the same location? If not how far apart are they? How many PM2.5/PM10 sites are used? Also, what's the inlet cutoff size used for the scattering and absorption measurements? This is important to related mass and optical properties properly

554-561. Could you add justification for the PM2.5/PM10 observation errors stated in Table 1? There is no explanation how the errors were picked. Also, why do you only use representative error for PM2.5/PM0 but not for the other observations?

569. Can you add a bit more info on the vertical resolution? For instance, thickness of the 1st level and number of levels within 1km.

594-596. Can you clarify if you did 2 simulations every cycle with and without aerosol interacting with radiation, or it was a single simulation with two calls to the radiation code within the same simulation?

632-638. You are also missing some processes of potential importance such as secondary organic aerosol formation and heterogeneous sulfate formation influencing low-dust days.

645-647. I think a better fit to PM2.5 could be achieved if you relaxed the interbin correlation. It looks like PM10 is fitting pretty well but it's going a bit over the observation, so this is restricting increases in PM2.5 due to the correlation. Since bin 4 is 2.5-10um, in theory, if no interbin correlation was present, PM10 and PM2.5 should be able to fit independently. For this study it would make sense to relax the interbin correlation due to the known issues in dust size distributions (see next comment)

647-652. Literature on dust modeling states that parameterizations tend to overpredict the fine dust and underpredict the coarse dust (see Kok et al., 2011, Adebiyi and Kok 2020). So the joint assimilation of PM2.5 and PM10 could be somewhat correcting for that, which is a additional possible explanation to the behavior explained in these sentences.

672-685. Another reason for the discrepancy is related to the size distribution. Are you assimilating multi-wavelength AOD here, right? If so, I would expect some modifications to the size distribution. It looks you are effectively modifying size distr. as the ratio of PM2.5 to PM10 ratio is reduced from 0.31 in the background to 0.11 in the DA_AOD simulation but it might be going a bit too far as the observed ratio is 0.28. You can also check angstrom exponent. You can also explore the point you make at the end related to the dust mass extinction efficiency, you have observations to compute this at the surface. Additionally, there is also potential for your vertical distribution to be off and be generating these issues. You can diagnose this by comparing the ratio of surface extinction vs AOD. It seems the model is overpredicting this ratio, which could mean too much aerosol close to the surface.

Related to this point. You are actually already computing mass scattering efficiency (2nd column in Table 3). The background already underpredicts it, and the assimilation makes it worse as you are increasing the coarser fraction. You could explore if there is an underprediction of the dust refractive index. You could look into values provided in the literature for the region studied and compare to what WRF-Chem uses.

678-683. I think there is no need for this very long description of the Ma paper as these results are not that relevant to the area study as RH is likely low in the desert and dust aerosols tend to be hydrophobic

690-692. AOD to PM10 ratios depends on many variables. Since you are blaming discrepancies to issues in mass scattering/absorption efficiency it makes more sense to do direct comparissons to this variable as you have in-situ measurements of scattering

and absorption

693-696. You can assess issues with size distribution by using the angstrom exponent.

Table 2 and 3. Is there any reason behind using the lower wavelength (440-450nm) for these comparisons? Since the focus of this work is on dust, it would be preferable to compute optical properties for longer wavelengths where coarse aerosols contribute more to the scattering

702-706. There is extensive literature on how optical properties of dust particles deviate from Mie theory (e.g., Dubovik et al, 2006, Nousiainen et al 2015). It would be good for the authors to reference this work and attempt to explain what could be the implications of using Mie theory, and if those can explain any of the discrepancies found when assimilating multiple datasets in this study

Figure 12. It would help to see an additional panel with these profiles being normalized, so we can more easily assess by how much the assimilation of the different datasets is changing the vertical distribution.

730 You know it overestimated PM10, not sure about aerosol number concentration (you would need a different observation for assessing that)

734. Use single-scattering albedo for this

738-746. This is a misconception, aerosol light extinction and AOD does not depend on sun light intensity (for instance, you can sample both at night time with different methods). What's going to change with sunlight are the radiative effects. There are likely other reasons to explain this diurnal behavior. Look into the diurnal evolution of your BEC, and also into diurnal evolution of dust reaching the city. Similar misconnection is mentioned in lines 770-771.

960-963. This is probably due to underprediction of dust imaginary refractive index

Minor Edits

Fig 5 caption. It reads like a) and b) represent PM10 and winds, respectively, but I think that's not the case. Please revise

623. Did you mean "underestimates" instead of "lowered"?

780. Do you mean "particles that absorb radiation" rather than "aborting particles"? Also, I would like black carbon in that list as well.

781-791. I believe primary dust in WRF-Chem is also considered to be a bit absorbing (has a imaginary refractive index above 0). As mentioned in a previous comment, this number might be too low for dust in this region.

802-804. I disagree with this statement. If the model has biases that the assimilation is not able to correct (for instance, inaccurate real and imaginary refractive indexes) then assimilating multiple observation could also create unrealistic modifications to the model.

806-828. I wouldn't put DA_Esca_Eabs as an improvement over DA_Esca, they show pretty much the same results. This means that the absorption observations are not really generating any differences in the results. Also, DA_PMx_AOD matches better the assimilated variables (which off course is expected) and the better agreement with scattering you happened to underpredict it with PM assimilation, and overpredict it with AOD assimilation, so assimilating both yields you something in between.

832-834. As mentioned earlier, it would be better to check this using normalized profiles. The background profiles already had aerosols up to 4km, so is likely that the assimilation is just scaling this profile upwards rather than adding a larger fraction of the mass in these layers

Section 4.1. I don't think this section is very relevant, the aerosols are so dominated by dust and your BEC is constructed in a way dust aerosols will be the ones largely modified. So just briefly mentioning that the composition of these other aerosols doesn't change would do.

Section 4.2. Might want to discuss in this section how the large underprediction of dust absorption would impact these results.

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