Response to Reviewer 1

Comments: The authors assimilate MODIS observations of AOD at 550 nm into a coupled meteorological and atmospheric chemical transport model. They chose a wildfire event in North America for their study period, and they analyse the achieved improvements in PM2.5, OC, and EC forecasts, as well as the changes in meteorological parameters caused by the changes in aerosol concentrations. The paper is clear and concise, the methodology is state-of-the-art, and the results are quite interesting. I have only one point that could help to clarify the description of the methodology and results.

Response: Thank you for the positive comments.

Comments: It is quite remarkable that the assimilation of AOD-550 improves not only the total mass concentration, but also OC and EC concentrations. I wonder how exactly this is achieved; the discussion in the manuscript does not really help me in understanding this. It would be very helpful to know how the aerosol concentration field is corrected by the assimilation. Do you allow each chemical species to be corrected independently (essentially by distributing the innovations to the various chemical components in model space according to the background error covariances)? Or does your assimilation only correct (i.e. scale) the total aerosol mass PM2.5, thus applying the same scaling factor to each chemical species? The latter approach has been pursued, e.g., by [A] (cited below), the former was tested, e.g., by [B], and the conclusion was that observations of optical parameters do contain sufficient information to retrieve PM2.5, but not for retrieving the chemical composition. If your assimilation algorithm only corrects PM2.5, while constraining the relative proportions of the different species to those predicted by the model, then the improvements achieved for OC and EC by assimilating AOD-550 are, most likely, the results of OC and EC being strongly correlated to PM2.5 in your case. On the other hand, if you allow EC and OC to be corrected independently by the assimilation algorithm, then the good result is quite surprising. Surely, an AOD observation at a single wavelength does not contain sufficient information to allow you to retrieve the chemical composition of the aerosols!?

Response: Our AOD data assimilation method was detailed in Liu et al. (2011), which is a 3DVAR approach and allows independent correction of individual aerosol species (so similar to Kahnert 2009). In Liu et al. 2011, we already cited Benedetti and Fisher (2007) and Kahnert (2008).

We added a sentence in the revised manuscript (in the last paragraph of Introduction section): “As pointed out in L11, the aerosol data assimilation system used here directly analyzes 3D mass concentration of individual aerosol species and allows them to adjust independently with additional constraint from the background error covariance for individual species. Similar method was also adopted by Kahnert (2009) for aerosol inverse modeling.”

Kahnert, M. (2008), Variational data analysis of aerosol species in a regional CTM: Background error covariance constraint and aerosol optical observation operators, Tellus, Ser. B, 60, 753–770, doi:10.1111/j. 1600-0889.2008.00377.

We agree that AOD at a single wavelength has no sufficient information on the aerosol composition. The background error covariances, which are event and period specific statistics from the NMC method, played important role for mapping observed optical information into aerosol mass concentration space. While this does not always work very well, but it was indeed able to produce some good results with event-specific background error statistics such as for this case and others (e.g., dust dominated event as in Liu et al. 2011 and our recent unpublished work on Saharan Air Layer simulation over Atlantic ocean).

Comments:

In summary, the paper can be published as is. I would leave it up to the authors if they want to take the opportunity to revise their paper before publication in GMD.

References:

[A] Benedetti, A. and Fisher, M. 2007. Background error statistics for aerosols. Q. J. R. Meteorol. Soc. 133, 391-405.

[B] Kahnert, M. 2009. On the observability of chemical and physical aerosol properties by optical observations: Inverse modelling with variational data assimilation. Tellus 61B, 747-755.

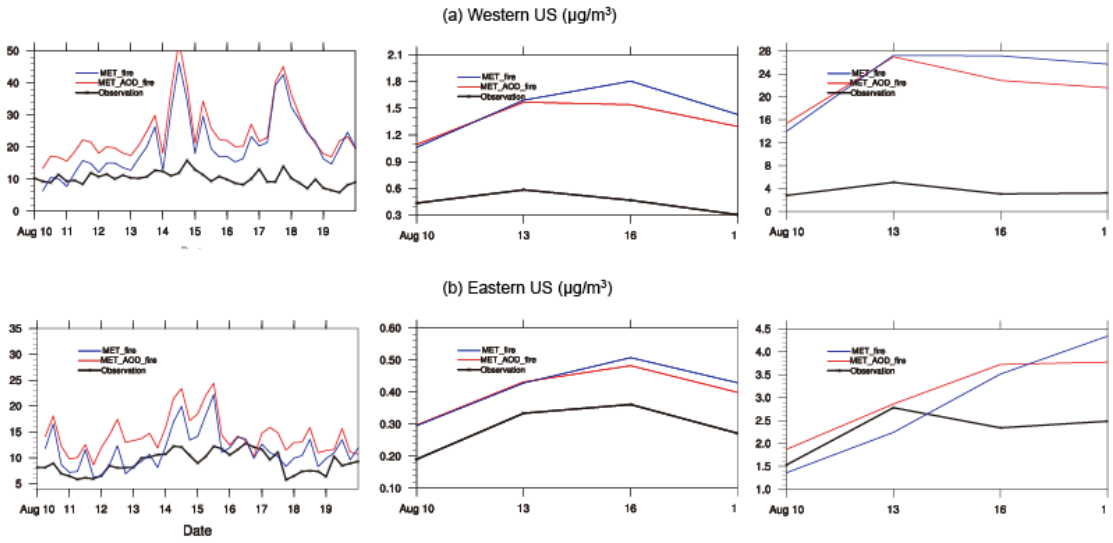
Response: We added Kahnert, M. 2009 in the reference.

Response to Reviewer 2

General Comments: *This reviewer does not find this paper very interesting at this point, although it has large potential to be a very interesting study. Unfortunately, the authors start with a bad model forecast (almost no AOD in there, probably because no fire emissions were used). That makes it very easy to show improvement. Large changes in AOD make it quite expected to see differences in the meteorological forecasts. Does this represent an improvement? Is there any evaluation of this? Even without the evaluation this could still be a very interesting paper with a few more diagnostics on the differences. I recommend publishing with major revisions. I do not expect a major rewrite (even though my sentences above may sound like this and I am somewhat disappointed by what is in the paper). The following are my most important comments at this point:*

Response: We appreciate the reviewer's detailed comments.

For your first point regarding fire emission, we have a short paragraph in the conclusion section. In fact, we have conducted experiments with the inclusion of GOES WF_ABBA fire emissions: MET_FIRE (without AOD DA, but with fire emissions and meteorology data assimilation) and MET_AOD_FIRE (with AOD DA based on MET_FIRE). However, the results showed large overestimation of AOD and surface aerosol concentrations, as shown below for surface PM2.5 (left), OC (middle), and BC (right) for MET_FIRE (blue curves) and MET_AOD_FIRE (red curves), which led to smaller AOD data assimilation impact when compared to the impact from largely overestimated fire emission. A more thorough study about the fire emission impact could be done in the future but beyond the scope of this study.

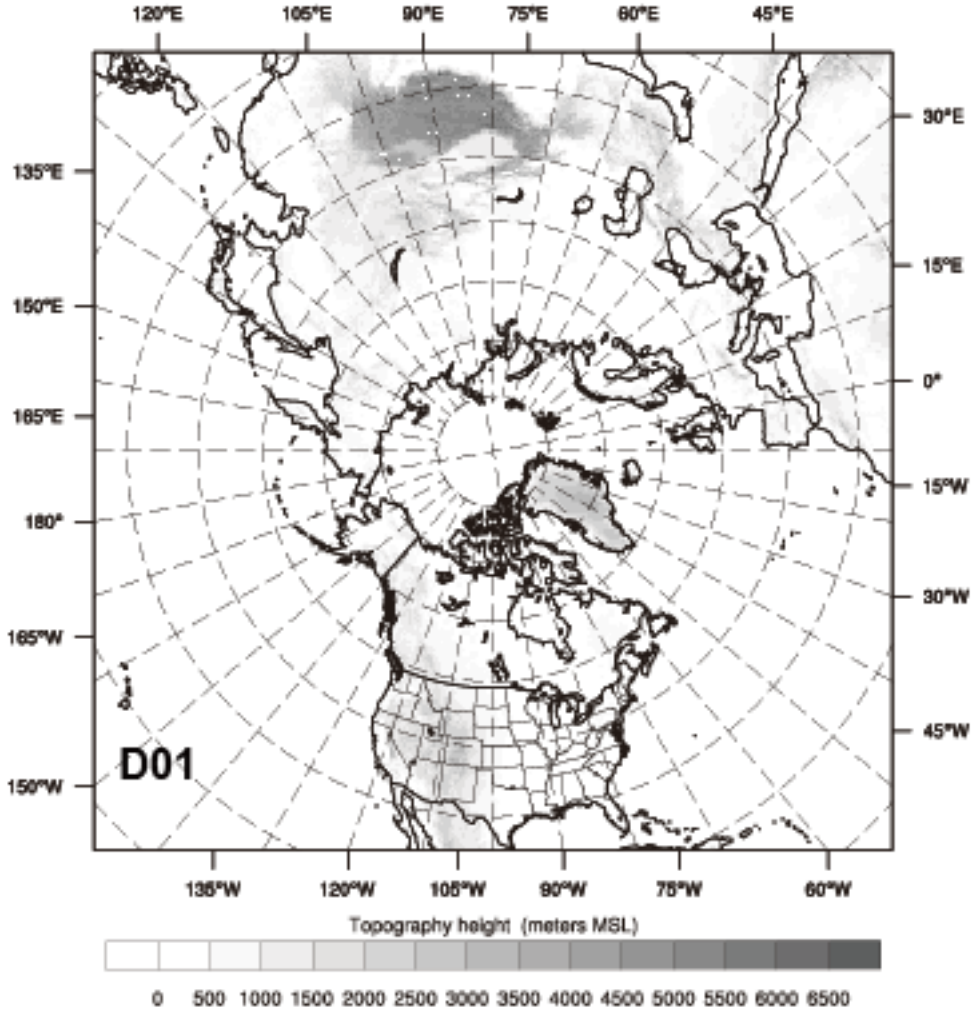


Main point of this paper is not to prove the improvement of weather forecasts with the inclusion of dynamic aerosol fields, but to demonstrate the WRF/Chem's ability to simulate aerosol direct and semi-direct effects with aerosol data assimilation. For demonstrating the improvement of weather forecasts, we think it is important to also

include indirect effect that beyonds the scope of this study. We included some more diagnostics plots as suggested in your specific comments.

1. ***Comments:*** *Pg. 3854, line 10 "covered most of the : : :" What does that mean? Either a figure or an explanation is needed*

   Response: The model domain is shown below. As our analysis only focused on a sub-area of the whole model domain. We think it is unnecessary to include this figure in the paper. We revised the sentence like this: "The model domain (operationally used at the US Air Force Weather Agency) with 20-km horizontal grid spacing covered a large portion (20-degree north) of the Northern Hemisphere with the polar projection (not shown), although our analysis will focus on North American regions where a wild fire occurred (Fig. 1)."



2. ***Comments:*** *Line 13/14: The use of "were implemented" makes it sound like you did this work. Is this the case? If not, does a reference exist?*

   Response: Thanks for pointing out this. We have added the following reference there to clarify the text.

   Fast, J.D., Gustafson, W.I., Easter, R.C., Zaveri, R.A., Barnard, J.C., Chapman, E.G., Grell, G.A., Peckham, S.E., Evolution of ozone, particulates, and aerosol direct radiative forcing in the vicinity of Houston using a fully coupled meteorology-chemistry-aerosol model. J Geophys Res-Atmos 111. D21305. Doi 10.1029/2005jd006721. 2006.

3. ***Comments:*** *Line 15: Why did you choose this shortwave scheme? Was RRTMG not available for your version? That would allow you to also look at LW impacts.*

   Response: We selected Goddard scheme is simply because it is the sponsor's operational configuration. It might be interesting to compare two schemes in the future work.

4. ***Comments:*** *Line 18: Fast et al. 2006 must be cited in addition to Barnard et al. Fast et al were the first to look at the direct effect and to implement the interaction with radiation.*

   Fast JD, WI Gustafson Jr., RC Easter, RA Zaveri, JC Barnard, EG Chapman, and GA Grell. 2006. Evolution of ozone, particulates, and aerosol direct forcing in an urban area using a new fully-coupled meteorology, chemistry, and aerosol model. J. Geophys. Res., 111:D21305, doi:10.1029/2005JD006721.

   Response: We added this reference in the revised manuscript.

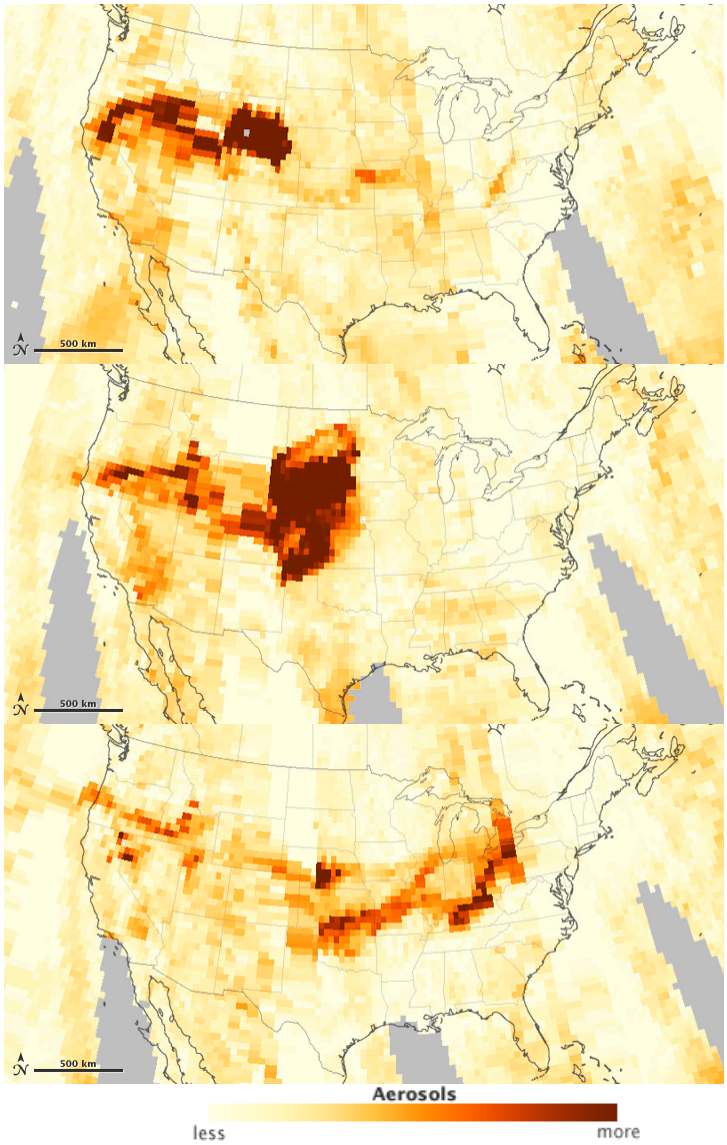
5. ***Comments:*** *Line 27: "parameterized within the GOCART model"? This sentence is not clear. It sounds like you used a different model to calculate the fluxes? Do you mean to say "Within WRF-Chem::: .. are parameterized using the GOCART dust and sea-salt modules" ?*

   Response: You are right. We have clarified in the text, "Within WRF-Chem, emissions of dust and sea-salt are parameterized using the GOCART dust and sea-salt modules".

6. ***Comments:*** *Pg. 3855, line 6/7: Wouldn't it be better to have this figure in the paper instead of putting in this obscure web address? It would also be useful to see a comparison figure from your simulations. What does your integrated PM2.5 look like if plotted in similar fashion? What is the data source for the MODIS data? What level of quality control? Are they real-time or near real-time?*

   Response: The figures (attached below for your reference) in that website are displayed only in a qualitative way, but not clear what are exactly shown (e.g.,

quantity and unit). We think it is not very useful to show this kind of qualitative figures (without the full knowledge of the figures) in our quantitative analyses. The Figure 1 already shows mean AOD difference between two simulations over fire event days. The qualitative pattern is very similar to that in that website. The total 550-nm aerosol optical depth (AOD) retrievals from MODIS were obtained from ftp://ladsweb.nascom.nasa.gov/allData/51/MOD06_L2, which are 10kmx10km Level-2 data from the collection 5.1. We used only the data flagged as the best quality. They are not real-time. We added related information in the revised version.



7. ***Comments:*** *Line 12: Only 6 hours for spin up? AOD over the western US is probably only available at afternoon local time (maybe 18Z and 00Z?). Why do you use a domain that covers almost all of the northern hemisphere? What happened over the US for times when you do not have AOD data available? Do*

you use the same AOD data every 6 hours? I assume you are using a time window?

Response: The fire event started from Aug. 14 and our WRF/Chem simulations start from 00 UTC 1 August, thus for the fire event simulation, the spin up time is more than 14 days. The 6-hour spin-up is referring to each of the DA process, as we did four times DA at 00, 06, 12 and 18 UTC each day. We rephrased the sentence as "Each experiment started WRF/Chem simulation with a 6-h cycling interval from 00 UTC 1 August in order to spin up aerosol fields before the fire event.".

The larger domain is the sponsor's operational domain and we tried to conduct some scientific study within the limitation of the project delivery. We think the model simulation with a larger domain should be beneficial instead of harmful.

AOD data were assimilated only at times when they were available around 18 UTC over US. For other times (00, 06, 12 UTC), aerosol fields were simply carried over from previous cycle's forecast although MET fields were still updated. We clarified this in the revised text: "For MET_AOD, GSI 3DVAR updated both meteorological and GOCART aerosol variables (only at 18 UTC when AOD data were available over US) every 6 h". We also added a sentence regarding assimilation time window: "The assimilation time window was ±1.5-h centered at analysis times (00, 06, 12, and 18 UTC)."

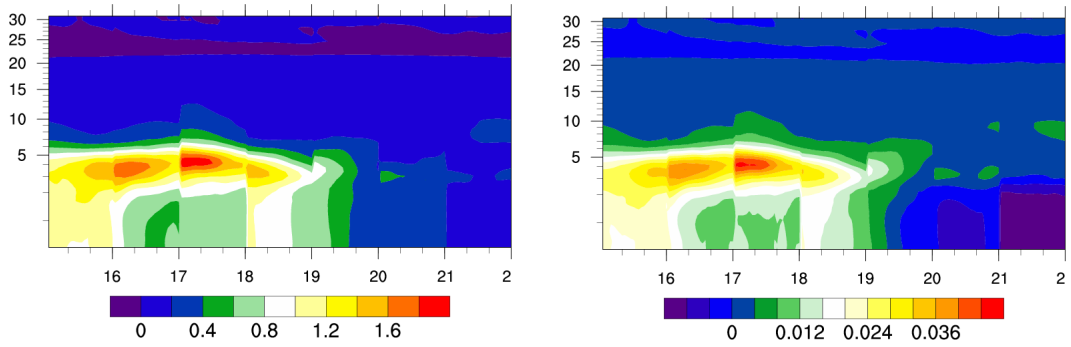
8. ***Comments:*** Section 3, pg 3856: Do you have any sulfate comparison available? Why is OC so much more affected than BC?

Response: Sulfate is the secondary PM component and formed from anthropogenic emissions, such as So2 and NOx. It is not relevant to this natural fire event. Thus, We limited its modification by AOD data assimilation. Modification for individual species is largely controlled by the background error statistics. OC has larger error than BC from our background error statistics.

9. ***Comments:*** Section 4: pg 3857: what is "accumulated day-2 forecasts"? What do the vertical profiles of bc, oc, and sulf look like compared to Fig. 4? What are the impacts on parameterized versus resolved precipitation? Would a horizontal difference plots for the precipitation fields look different? FDR is a large area. Precipitation differences in areas where AOD is largest may be different than in areas where AOD is low.

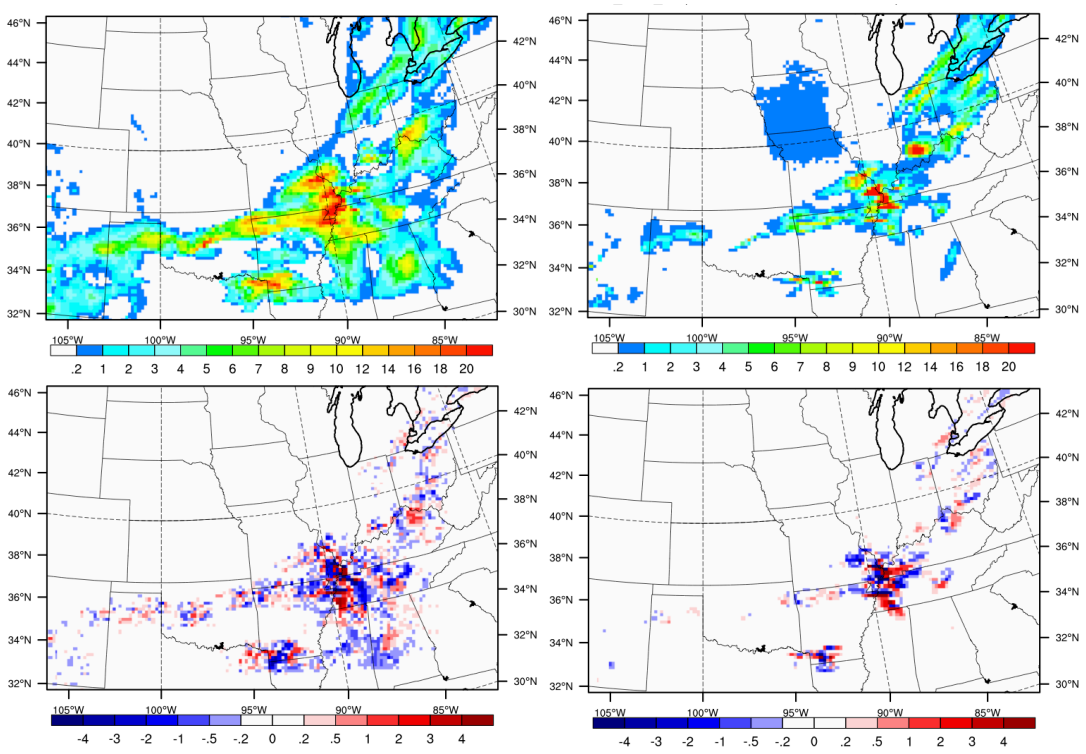
Response: We used the wrong word. Now we changed it to "hourly output of day-2 forecasts".

Similar to Figure 4(a), OC and BC difference plots (MET_AOD minus MET) are provided below. It can be seen that the peak difference for OC and BC is basically in the same height and time. We revised the text as: "The largest AOD (also OC and BC, not shown) increase due to AOD DA occurred at around 4-5 km altitude …".



The same figure as Fig.4 but for OC (left) and BC (right), unit: μg/m3

Figures below show the accumulated 12-h forecast of RAINC (upper-left) and RAINNC (upper-right) initialized from 00UTC Aug. 17, 2012 for the experiment MET_AOD. The corresponding difference fields (MET_AOD minus MET) are displayed in (lower-left) and (lower-right). The difference fields eventually followed rainfall pattern and look like parameterized (RAINC) and resolved (RAINC) portion of rainfall are equally affected. Grid points with positive and negative difference are mixed up and scattered, which makes interpretation difficult. This paper intended to primarily demonstrate data assimilation impact on air quality prediction and potential effects on weather forecasts, but not attempted for a physical process study. We intentionally make the manuscript short and prefer to leave figures here instead of including them in the paper.



10. ***Comments:*** Section 5, pg 3859, line 13: Sloppy writing. “Enhanced aerosol (varied from : : :)” What are you talking about?

    Response: we changed it to “increased AOD”.

11. ***Comments:*** Line 22: “However, in our trials, : : : : : :” This sentence does not belong here. Is this comment supposed to be related to the paper by Grell et al ? I do not think that WF_ABBA data were available for Alaska. Are you saying the Grell et al paper was only seeing a noticeable difference since the fire emissions were overestimated? Even if that were so, you don’t show anything in your paper to prove that suspicion.

    Response: Here we talked about additional experiments we did for this California fire event, not Grell’s Alaska fire event. We now clarified in the revised text: “ :::::: (Prins et al., 1998) fire emissions in the simulation of this fire event over California”.