

Geosci. Model Dev. Discuss., referee comment RC1  
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## Comment on gmd-2021-172

Anonymous Referee #1

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Referee comment on "Development of aerosol optical properties for improving the MESSy photolysis module in the GEM-MACH v2.4 air quality model and application for calculating photolysis rates in a biomass burning plume" by Mahtab Majdzadeh et al., Geosci. Model Dev. Discuss., <https://doi.org/10.5194/gmd-2021-172-RC1>, 2021

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The manuscript shows the development and evaluation of aerosol effects on photolysis rates on the GEM-MACH air quality model, and how the inclusion of aerosol feedbacks in meteorology play a role in photolysis rates as well. The manuscript is very well written, is within the scope of GMD, and represents an excellent contribution to the field. 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.

- The manuscript is very thorough in terms of their methods and results, but I believe is missing more discussion of the results. I highlighted many missed opportunities to do this in the comments by line below
- It would be nice to expand this analysis all the way to the trace gases (O<sub>3</sub> and NO<sub>2</sub>) assessing the impact of the changes into air quality. This is done for the fire case study, but it would also be nice to see it over cities during high AOD events.
- The aerosol and optical properties treatment shown in this manuscript is very detailed, more detailed than what's in many other air quality models. Thus, I think there is a missed opportunity to evaluate aerosol optical properties other than AOD. For instance, the authors do a detailed treatment of BC mixing state, so evaluating absorption (e.g., AERONET provides SSA retrievals) would be of interest. This model has a detailed treatment of aerosols size, so evaluating Angstrom exponent (a proxy for aerosol size) would also be of interest.

- The fire analyzed shows model peak PM<sub>2.5</sub> concentration ~50 ug/m<sup>3</sup> which is a relatively weak smoke plume. I think a more extreme fire (i.e., PM<sub>2.5</sub> concentrations > 1000 ug/m<sup>3</sup>) should be analyzed in which case I think the effects of the photolysis rates updates will play a more important role. The NSF WE-CAN and NOAA-NASA FIREX-AQ field campaigns measured some of these extreme fires, including a full suite of trace gas measurements and photolysis rates which would allow for a more thorough evaluation of the updated photolysis module. I would encourage the authors to take advantage of those datasets

- While the title suggests the work is mostly about aerosol feedbacks on photolysis rates, a good portion of the study describes the new treatment of aerosol optical properties including considerable evaluation of it. Thus, I suggest to somehow reflect this into the title for consistency

Comments by line:

265. It would if the authors could list the refractive indexes used for all species at dry conditions at mid-visible wavelength for comparison to other studies.

310. It would be desirable if the authors list the aerosol densities used for the mass to volume conversion

345-346. I would also expect a dependence on the refractive index of the shell. What is assumed here?

355. In this treatment you are still computing the optical properties independently for each aerosol type and then adding up, so it's not fully representing an internal mixture. My understanding of computing aerosol properties for internal mixture is that a volume-weighted average refractive index is first computed and then this goes to Mie theory. I understand that the use of table look ups might limit the application of this approach, but at least there should be some sensitivity analysis to show how the current approach compares to fully internal mixture and fully external mixture treatments for a range of mixtures.

Table 3. What does "AOD Calculations" mean in Table 3? How are aerosol feedbacks included when there are no AOD calculations (S2 and W2)? Are the aerosol optical properties computations consistent between the photolysis and aerosol feedback modules when columns 3 and 4 are set to "yes"?

Section 3.1. There is no discussion of these results. The reader is left wondering what

generates such a large range of values in the base model

497-499. Any insights on why would the model underpredict the maximum values? Perhaps a bit of analysis could provide paths for future improvements. For instance, one thing I would look for is if RH at the location of the aerosols was high for these days. Is the model representing RH/water vapor for these days? You could look into nearby sounding, AERONET also has a precipitable water product. Also what type of aerosol types were prevalent those days. Was surface aerosol elevated on those days as well (could look into PM2.5 observations)?

515. More discussion is needed here. Is this larger observed variability linked to the underprediction of the peaks mentioned earlier?

530. Additional analysis could be added. What is the hotspot shown by MERRA2 in Alberta during winter not shown by GEM-MACH?

550. There some "hotspots" in the JNO<sub>2</sub> differences (Fig 10f), specially in some regions of the US. It would be nice if you could provide an interpretation. Are these due to aerosols only? Are clouds playing a role?

554-559. These results are a bit puzzling, it would be good to gain better understanding on what's going on. On Fig 10 you show negligible changes of including or not AOD on photolysis rates (f). So a +/-30% in AOD is likely not responsible for the +/-40% change in J values in Fig 11. I would add a panel showing the changes in cloud properties to see if that explains the the J value differences. Differences of dry aerosol column would also help explain the AOD changes.

595 The MODIS level 3 data is pretty coarse, so a higher resolution datasets would be better. MODIS level 3 is an aggregation of MODIS level 2, which is ~10km but it will have the tendency of screen dense smoke plume out as clouds (you can check this on NASA worldview). Products that consider a smoke detection algorithm are generally recommended for these comparisons. These include NOAA enterprise VIIRS AOD product and the MODIS MAIAC retrievals.

599-605. MERRA has coarser resolution than GEM-MACH simulations so resolution is likely not an issue. MERRA tends to use a relatively flat diurnal cycle of emissions which can sometimes overpredict emissions outside of the peak fire hours. So one issue could be that that plume corresponds to the same fire earlier in the day, but that is not really there. Using better AOD retrievals could help corroborate this

621. Might be worth mentioning that (based on the spatial plot) the injected plume seems to be transported more towards the northeast, which is probably why it doesn't show up in the cross-section away from the fire

634-635. Looking at Fig. 16 I'm seeing much larger deviations than stated in the text, please double-check. Also, looks like differences are enhanced away from the surface (but still in the PBL) where it can still affect trace gases after chemistry and transport, which I also think is worth discussing.

637-640. Since BC is changing and you don't expect significant BC changes due to photolysis changes, then it is worth mentioning here that this is likely related to the aerosol feedbacks

#### Minor Edits

Throughout the text there were places where it looks like spaces between words were missing, not sure if this is actually happening or it was some artifact of the text editor or pdf printer. Please double check

48. on "whereas an "externally mixed" population of particles consists of only one chemical species" I think you meant something like "whereas an "externally mixed" population of particles consists of each particle being of only one chemical species"

55. Instead of "Black carbon can exist in different mixing states:" I would say "Black carbon is generally modeled using different assumptions on its mixing state:" as these correspond to modeling approximations

109. I think you should remove the parenthesis from "(revised)"

203: there is a "(" that's not closed

Fig 10. Using blue for the lake boundaries and as a shade for the differences make it confusing. I would recommend changing the lake boundary color to one not used in the difference color-scale.