Geoscientific Model Development Discussions

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Reply on RC1

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Author 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-AC1, 2021

We thank the reviewers for their time and effort to provide us with their feedback and to make valuable improvements to our manuscript.

Our responses to reviewer 1:

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 (O3 and NO2) 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.**

Authors response: We have looked at the impact on photolysis rates and trace gases over urban areas in our domain and the impacts are minor for the period of simulation considered.

- **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.**

Authors response: This is an excellent suggestion and is inline with the outline of our next manuscript of further investigating the effects of the improved photolysis module with a nested configuration of GEM-MACH 10-km domain to a 2.5 km Athabasca Oil Sands domain. In addition, in the new study, we will perform a more detailed comparisons of model output with observations (e.g. black carbon mass concentrations and particle absorption coefficients) under the Oil Sands Monitoring Program, 2018 aircraft campaign.

- **The fire analyzed shows model peak PM2.5 concentration ~50 ug/m3 which is a relatively weak smoke plume. I think a more extreme fire (i.e., PM2.5 concentrations > 1000 ug/m3) 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**

Authors response: This is a good recommendation. In this work, our funding was through the Joint Oil Sands Monitoring program and this required us to focus on the period of Oil Sands intensive measurements; other fires are indeed available in other datasets, and we will pursue this in future work

- **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.**

Authors response: Thanks for your comment. We changed the title to address your suggestion: "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".

Comments by line:

- **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.**

Authors response: We added the refractive indices of all aerosol types in the new Table 2 (can be found in the supplement file).

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

Authors response: We added the densities of all aerosol types in the new Table 2 (can be found in the supplement file).

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

Authors response: The core-shell parameterizations used in our study (Bond et al., 2006) is for a range of realistic particle sizes containing black carbon (core) surrounded by a concentric shell of weakly absorbing material. Bond and Bergstrom (2006), pointed out that scattering and absorption depend on the physical form of the particles and that the light absorption of carbonaceous particles depends strongly on the size of the bonded structure. According to Bond and Bergstrom (2006), since the absorptive properties of light-absorbing carbon are not very variable, a narrow range of refractive indices for strongly-absorbing carbon particles was recommended, of which the highest is 1.95 - 0.79i.

Bond et al. (2006), provided amplification factors on the basis of core and shell size, which are not very sensitive to assumed refractive index of light absorbing carbon (LAC). According to Bond et al. (2006) the volume-weighted mixing approximations produce unjustified overestimates of absorption. In order to examine the sensitivity of the

absorption amplification to different refractive indices, they performed some sensitivity tests and calculated the error to the fitted amplification factors with three different refractive indices (base: 1.85 + 0.71i, Low m: 1.55 + 0.55i, High m: 2.0 + 1i, and Less Absorbing: 1.96 + 0.66i). They found that the predicted amplifications are not very sensitive to the assumed refractive index of LAC (within ~15% for those three refractive indices).

We listed the refractive indices of the (shell) aerosols in the new Table 2 (in the supplement file).

- **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.**

Authors response: We used the terms "internal particle components" and "volume-averaged homogeneous mixture of ammonium sulfate, ammonium nitrate, primary and secondary organics" to describe the soluble surrounding species in the core-shell treatment. The most common assumptions for aerosol mixing states in the literature are the external mixture and internal mixture. The term internal mixture can also be applied to the core-shell mixing state. In the 'external mixture', extinction, scattering and absorption efficiencies are calculated separately for each chemical species, and the total optical efficiencies are calculated by adding the efficiencies of the components in each size bin (Péré et al., 2009; Lesins et al., 2002). In most studies, the refractive index of an 'internally mixed' particle is calculated by volume-weighting of all the refractive indices of pure species (Péré et al., 2009; Jacobson, 2001).

The internally mixed assumption using the volume-weighted refractive indices has its limitations, e.g. Bond and Bergstrom (2006), refers to this method as the most common, but one of the poorest ones. The calculations of the scattering and absorption with the assumption of a single value for the refractive index of the internally mixed aerosols would strongly depend on the underlying model assumptions (Bond et al., 2006). According to Jacobson (2000) the internally well-mixed method for elemental carbon is physically unreal, and the core treatment for elemental carbon is more representative compared to the externally mixed treatment.

In the case of internal mixture in our study, we used the computationally-efficient method of calculating the optical properties of each individual aerosol type using the offline Mie code and created a lookup table for aerosol optical properties to be used by the model. The resulting Mie lookup table and the volume average of the mixed particles was used to calculate the scattering and absorption coefficients and asymmetry factor of mixed particles. On the other hand, the term 'external mixture' would not be applicable in our case, in that an external mixture was not maintained in the model code.

Table S1 (in the supplement file) shows an example of different treatments of the soluble species surrounding the black carbon core: ammonium sulfate, organics and water. We calculated the optical properties of these species using two different methods: 1) volume averaging the refractive indices and calculating the extinction, scattering and absorption efficiency of the mixture, 2) using the optical properties of pure components and volume-weighting the pure optical properties, the latter being the approach we adopted in the model. We used an on-line Mie calculator (https://omlc.org/calc/mie_calc.html) and the

optical properties of ammonium sulfate, organics and water at 0.5 μm (Jacobson, 2005) for particle diameter of 0.2 μm to carry out these calculations. We calculated a maximum difference of -2.7% for four different combinations of aerosol concentrations. The narrow range of the real part of the refractive indices in the soluble species in the fine mode particle mixture yields a low level of uncertainty in the two different particle mixture treatments. This level of uncertainty is much less than the error in the vertical profile of $PM_{2.5}$ mass concentration and humidity calculated by GEM-MACH model.

- **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"?**

Authors response: Having no AOD means that in the photolysis module we set the total AOD, scattering and absorption optical depth at each grid point to zero, meaning that in the simulations without AOD there is no direct aerosol optical effect on the photolysis rate calculations. The aerosol feedbacks on meteorology code was developed in a previous study for the GEM radiative transfer scheme. GEM uses the particle mass size distribution predicted internally by MACH at each time step, but assumes a constant chemical speciation profile for gas species (typical of an aged polluted air mass) when calculating optical properties. This is largely due to the computation cost of Mie calculations in the GEM model. Our next step is to modify the GEM radiation code to include the particle chemical composition on-line and our hybrid particle mixing state assumptions.

We have included the following text in the revised paper:

Note that in the "no-feedback" simulations, aerosol optical (and cloud condensation nucleation) properties in GEM come from default climatological properties used in the GEM weather forecast model (Makar et al, 2015a, b). That is, our "no-feedback" simulation is not a "no aerosol" simulation – rather, the "no-feedback" simulation makes use of spatially invariant, "typical" optical properties of our weather forecast model. Having "no AOD" in this case means that model aerosol AOD had not been calculated since it is not used in the feedback code. Further, the aerosol feedback on meteorology code developed in a previous study for the GEM radiative transfer scheme (Makar et al, 2015a, b). In the feedback mode, GEM-MACH uses a separate Mie calculation generating a lookup-table online, using the particle mass size distribution predicted internally by MACH at each time step, but with a single typical complex refractive index. In contrast, our photolysis rate calculations and the AODs we calculate in the current work are decoupled from this feedback portion of the model. Our next step in this work is to further modify the GEM radiation code to include the particle chemical composition on-line, and our hybrid particle mixing state assumptions, as an alternative to the existing AOD calculation within the feedback portion of the code.

- **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**

Authors response: We added the following text to the manuscript following figure 5:

The base MESSY module uses a climatology for aerosol number density with one fixed vertical profile for grid cells over-land and another fixed vertical profile over water (see Table 1). The uniformity of the fixed profiles does not account for the real atmospheric variability resulting in the larger differences between model and observed aerosol optical depths for the base MESSY version.

- **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)?

Authors response: Thank you for your recommendation.

We compared the AOD time series of Fort MacKay and Toronto with humidity and $PM_{2.5}$ concentrations for both seasons. The high values of AOD correspond to the increase in the concentration of $PM_{2.5}$, and the difference in the maximum values are linked to the underestimation of $PM_{2.5}$ by GEM-MACH that needs improvement, and will be investigated in the upcoming project.

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

Authors response: Yes, there is more variability in the observed time series than model and this is shown in time series in Figure 6 and in the standard deviation vertical bars in Figure 8.

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

Authors response: Thank you for your recommendation. We added the following text to the manuscript:

The location of the hotspot in central Alberta in winter season in the MERRA product is coincident with the location of coal-fired power plants and oil refinery emissions of SO_2. This could be the result of different SO_2 emissions used in MERRA and GEM-MACH models or an over-prediction error when sulfate aerosol is retrieved over a snow covered surface.

- **550. There some "hotspots" in the JNO2 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?**

Authors response: Figures 10(e) and (f) are the % difference in J(O1D) and J(NO_2), respectively, for the direct effect of aerosols on photolysis rates, and the locations of greatest impact are different for J(O1D) and J(NO_2). The difference between JNO2 and JO1D hotspots in south-east US likely reflect the wavelength dependence of the scatter of solar radiation and the high humidity and more aerosol water in that area.

We added the following text to the manuscript:

The hotspots of greatest difference in Fig. 10(f) reflect the direct effects on aerosol with no weather feedback in these simulations.

- **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 photolisys 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.**

Authors response: The moist marine air masses have the greatest JNO_2 change likely due to changes in low clouds. The vertical profiles in Figure S1 in the supplement file show the percentage difference in (a) JNO_2, (b) number mixing ratio of clouds (droplets) and (c) specific humidity over an area with high values of JNO_2 difference. Note that 0.988 hybrid level corresponds to ~820 hPa or ~1750 meters above seal level. These vertical profiles support the importance of low clouds. The areas impacted are mostly over the ocean or along ocean boundaries (northeast US/Canada, Northwest Territories, north Pacific). Over land, in convective air masses, there is less impact on JNO_2 averaged over a region. These changes in humidity impact low clouds which, in turn, impact the radiative transfer and photolysis rates.

We added the following text along with the new Fig. 11 to the manuscript (the new Fig. 11 can be found in the supplement file):

As we can see in Fig. 11, the moist marine air masses have the greatest impact on J-values change likely due to changes in low clouds. The areas impacted are mostly over the ocean or along ocean boundaries (northeast US/Canada, Northwest Territories, north Pacific). Over land, in convective air masses, there is less impact on J-values averaged over a region. These changes in humidity impact low clouds which, in turn, impact the radiative transfer and photolysis rates.

- **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.**

Authors response: We thank the reviewer for this information. We replaced MODIS and MERRA-2 plots in figure 14 with MAIAC and VIIRS plots (the new Fig. 14 can be found in the supplement file).

- **599-605. MERRA has coarser resolution that 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**

Authors response: We replaced MODIS and MERRA-2 (hourly) plots in Figure 14 with MAIAC and VIIRS plots, with the following text appearing in the revised manuscript (the new Fig. 14 can be found in the supplement file):

Measurement data from MAIAC ("Multi-Angle Implementation of Atmospheric Correction (Lyapustin et al., 2018)"), VIIRS ("Visible Infrared Imaging Radiometer Suite (Hillger et al., 2013)"), the MERRA-2 re-analysis (Modern-Era Retrospective analysis for Research and Applications - Version 2, Global Modeling and Assimilation Office, 2015) and AERONET (Aerosol Robotic Network) ground-based measurements, were used in this study to to evaluate the modifications in the photolysis module in GEM-MACH.

MAIAC is an advanced algorithm which produces global AOD from MODIS (Moderate Resolution Imaging Spectroradiometer) Terra and Aqua satellite data. NASA's Terra and Aqua satellite are in polar orbits. The MODIS instruments have a global coverage of one to two days in 36 spectral bands between 0.405 to 14.385 μm. For the purpose of this study, we used MAIAC gridded daily average AOD data at 550 nm, with 1 km spatial

resolution (obtained from: https://e4ftl01.cr.usgs.gov/MOTA/MCD19A2.006/). VIIRS is one of the Earth observing instruments aboard the Suomi National Polar-Orbiting Partnership (Suomi NPP) satellite. Level 2 VIIRS AOD data at 550 nm with a spatial resolution of 6 km at nadir were used in this study (obtained from https://www.avl.class.noaa.gov/saa/products/welcome).

Figure 14(a) shows the daily AOD at 550 nm from the MAIAC, Fig. 14(b) is the daily AOD at 550 nm from the VIIRS, and Fig. 14(c) is GEM-MACH AOD at 580 nm over La Loche area at 23:00 UTC on June 25, 2018. Both MAIAC and VIIRS plots (Fig. 14(b) and 14 (c)) show two areas of maxima; one is directly over the forest fire plume, similar to the hotspot on GEM-MACH plot (Fig. 14(c)), and a weaker hotspot over the north-east of the major forest fire plume, which is more intensified compared to the GEM-MACH secondary hotspot. The aging of the major fire plume downwind is evident in all three plots. The maximum GEM-MACH AOD (0.625) is underestimated compared to the MAIAC (maximum of 3) and VIIRS (maximum of 1.7). One possible explanation for this underestimation could be the potential deficiencies in the CFFEPS forest fire emissions or aerosol processes in this simulation. Curci et al (2015) suggest that AOD underpredictions may be a common problem for current air-quality models.

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

Authors response: The following sentences have been added to the text:

There are multiple injected fire plumes and they move off in different directions as a result of different plumes reaching different heights in the atmosphere as well as variation over time. The model captures the branch moving toward Fort MacKay but not the branches that move farther north. Only the branch traveling to Fort MacKay is shown in the vertical cross section. While this aged plume still has character originating from the forest fire, it does not originate from all the fire emissions near Lac La Loche.

- **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.**

Authors response: We have changed the text to reflect the corrected numbers and included them as a range corresponding to the values at the surface up to the mid-boundary layer:

Figure 16(a) shows the reduction of O_3 in the fresh fire plume below the PBL due to the decrease in $J(O^1D)$, reaching a difference close to 1% to 6% from the surface up to the mid-boundary layer. The OH radical concentration difference (Fig. 16(b)) responds to the O_3 change and decreases by 10% to 20% from the surface up to the boundary level, due to the AOD feedback on the photolysis rates.

As shown, differences can affect chemistry not just at the surface but also in the mid-boundary layer. Even larger differences are observed at the upper boundary of the mixed layer. These are more likely due to changes in mixing height when the feedback on meteorology version of the model is turned on. We have less confidence in the reproducibility of these changes near the upper boundary layer because of their more irregular nature.

- **637-640. Since BC is changing and you don't expect significant BC changes due to photoliysys changes, then is worth to mention here that this is likely**

related to the aerosol feedbacks

Authors response: Yes, the changes in black carbon concentration (±10%) in Figure 16d at the surface up to the mid-boundary layer are mostly due to aerosol feedbacks on meteorology. We have added this sentence to the discussion of Figure 16.

The changes in black carbon concentration (±5%) in Fig. 16(d) at the surface up to the mid-boundary layer are mostly due to aerosol feedbacks on meteorology.

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**

Authors response: We made the correction when creating the pdf file.

- **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"**

Authors response: Yes, thank you. We made the correction as follows:

When all the species are homogeneously mixed within an aerosol particle, the population is known to be "internally mixed", whereas an "externally mixed" population of particles consists of each particle being of only one chemical species.

- **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 aproximations**

Authors response: We made the improvement as follows:

Black carbon is generally modeled using different assumptions on its mixing state:

- **I think you should remove the parenthesis from "(revised)"**

Authors response: We made the minor change.

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

Authors response: Thank you for the correction.

- **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.**

Authors response: We have changed the lake boundary colour to black.

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Please also note the supplement to this comment:
https://gmd.copernicus.org/preprints/gmd-2021-172/gmd-2021-172-AC1-supplement.pdf