Geoscientific Model Development Discussions

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Reply on RC2

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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-AC2, 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 2:

General comments

This paper presents updates made in the aerosol feedbacks and photolysis rates implemented in the GEM-MACH model. The document is well explained, easy to read and the research is in the scope of GMD. These improvements in the code are very clever and represent a state-of-the-art model that is a great contribution to the field. The results are good; however, I would recommend adding more explanations and discussion to the figures and explaining why the model shows some biases in certain cases. Please, read my comments below where I note them.

Specific comments

- **Is there any particular reason the simulation periods were chosen to be January and June 2018? I believe a more useful study would have been conducting simulations during July-August 2019, since NASA's FIREX-AQ field campaign compiled data that would have been useful for assessing the model, specially the photolysis developments.**

Authors response: This is a good recommendation. We were funded by the Joint Oil Sands Monitoring program and this required us to focus on the period of Oil Sands intensive measurements. Some of us were involved in a separate publication examining the effects of feedbacks just prior to the FIREX-AQ period (Makar et al, 2021)

- **Section 2.1: You say that you created an initial lookup table using the refractive indices of 6 aerosols, however in the introduction, you mention that GEM-MACH considers 8 aerosols. What are these 6 aerosols you considered for**

creating the lookup table? What happens with the other 2 components that you do not have data for?

Authors response: The initial version of the new lookup table (before the interpolation of size parameters into GEM MACH's) contains the optical properties of seven aerosol types (originally didn't mention pure water in the text): sea-salt, black carbon, dust, ammonium sulfate, ammonium nitrate, pure water and organic carbon.

The final version of the lookup table contains the optical properties of 7 aerosol types: black carbon, sea-salt, dust, primary organics, secondary organics, ammonium sulfate, ammonium nitrate. For primary organics, we used the optical properties of secondary organics and pure water droplets and the hygroscopic growth factor of primary organics (the equations used in these calculations can be found in the supplement file):

The new algorithm uses the eight dry chemical components of GEM-MACH aerosol (sulfate, nitrate, ammonium, primary organic matter, secondary organic matter, black carbon, dust and sea-salt). We used the predicted mass concentration of nitrate in GEM-MACH, and the molecular weight of ammonium nitrate to calculate the mass concentration of ammonium nitrate. The remaining mass of ammonium and the mass concentration of sulfate were used to calculate the mass concentration of ammonium sulfate.

We addressed your comment and added pure water to the list of aerosols in the manuscript as follows:

In order to update the aerosol effects in the MESSy photolysis module in GEM-MACH, we calculated a new lookup table using the Mie scattering code within the VECTOR model (McLinden et al., 2002) for extinction efficiency, single scatter albedo and asymmetry factor for seven aerosol types, which within the lookup table are treated as pure-composition particles of sea-salt, black carbon, dust, ammonium sulfate, ammonium nitrate, organic carbon, and pure water.

- **Section 2.4: You used CFFEPS biomass burning emissions. Does this inventory include fire emissions outside Canada? If not, did you account for fires in the US?**

Authors response: CFFEPS include emissions in North America, inside the FireWork domain (including US):

see https://weather.gc.ca/firework/firework_anim_e.html?type=em&utc=00.

Details on the CFFEPS version used in these simulations, and other CFFEPS references and documentation, may be found in Makar et al (2021), referenced in the revised paper. We also used a projected 2017 US anthropogenic emissions inventory obtained from the U.S. Environmental Protection Agency (EPA) 2011 Air Emissions Modelling Version 6.3 Platform (https://www.epa.gov/air-emissions-modeling/2011-version-63-platform; Eyth et al., 2013) and the 2008 Mexican inventory obtained from the EPA's 2011 Air Emission Modelling Version 6.2 Platform (https://www.epa.gov/air-emissions-modeling/2011-version-62-platform).

- **Section 3.1: Figure 4 shows many AERONET sites that fall within the domain of the simulations. Why do you use just focus in 4 of them? I think an explanation is needed somewhere in this section or the section before. Also, I think more discussion should be added to this figure since it is just mentioned in section 3.2.1.**

Authors response: Thanks for your comment. Our project was funded by the Joint Oil Sands Monitoring program, with the focus on the Canadian domain, including the Oil Sands area. We chose four Canadian AERONET sites, covering urban and industrial sites with reasonable number of observation data for the period of our simulations.

- **Figure 6 shows that some high AOD values from AERONET in Toronto are not well represented by the model. Could this be because of long-range transport of smoke plumes that are not well simulated by the model? I think you can add more discussion about these particular days.**

Authors response: The pollutant rates in urban areas are expected to have strong spatial variations due to different emission sources. The model resolution of 10 km should also be taken into consideration; the Toronto AERONET station is located 16 km away from Pearson International airport and only 4 km from Downsview airport. There was no evidence of the impact of forest fire on Toronto using ECCC's FireWork model output. The other possible reason could be linked to the underestimation of $PM_{2.5}$ by GEM-MACH that needs improvement, and will be investigated in the upcoming project. That is, this seems to be a local effect more linked to Toronto urban model performance than long-range transport.

- **Figure 9: I am surprised that GEM-MACH underestimates the AODs in the whole domain for both seasons since the comparison with the AERONET stations is good. What can be happening? Perhaps you could include a (c) panel showing the AOD from MODIS.**

Authors response: As shown in the manuscript, the monthly average plots show four major forest fires in the domain, and the AOD pattern follows the MERRA-2 AOD. Having said that, there are lots of factors involved in the uncertainties in the monthly average values, including the monthly map biased by a few large forest fires.

We replaced the MODIS plots with MAIAC and VIIRS plots in the revised version of the manuscript, but we note that our simulated AOD is biased low with respect to MODIS, MERRA-2, MAIAC and VIIRS AOD, while the bias is less noticeable for AERONET comparisons. The difference may also be due to systematic differences between AERONET observations (derived from surface-based observations) and satellite AOD: Fu et al (2018) shows comparisons between MODIS and AERONET AOD observations in China, with the slope of the MODIS to AERONET best fit line varying between 1.25 and 1.02 – that is, MODIS AOD observations are biased high relative to AERONET, suggesting that at least some of our positive biases have been seen in comparisons between these two AOD observation methodologies. Past comparisons between several different methods of calculating AOD within models and satellite-derived AOD suggest a general negative bias with respect to satellite AOD values (Curci et al., 2015).

- **Figure 14: MODIS L3 data is too coarse (I think 1 degree resolution). For your case I think using AOD from MODIS MAIAC (3 km) would be more appropriate to compare against your simulations.**

Authors response: We replaced MODIS and MERRA-2 (hourly) plots in Figure 14 with MAIAC and VIIRS plots and changed the text as follows (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.

In line comments

- **Line 202: Consider rewording the sentence that begins with “Using the hygroscopic…”. It is long and difficult to understand.**

Authors response: We changed the text as follows:

Using the hygroscopic growth factor of each aerosol type, we calculated the dry size parameter (Section 2.2, equation1) of the initial lookup table, which was then interpolated into GEM-MACH dry size parameter. The final lookup table which was implemented into GEM-MACH is a function of GEM-MACH dry size parameters and wavelengths.

- **Line 203: the parentheses in “(Section 2.2” never closes.**

Authors response: Thank you for the correction.

- **Line 220: it seems there is an extra “(“.**

Authors response: Thank you for the correction.

- **Lines 489-499: You can mix this paragraph with the following one since you are describing and discussing Fig. 6. E.g.: “were used to plot these time series. The sample size for Toronto…”. Also, I suggest sticking to the nomenclature shown in Table 3 for each simulation instead of saying “GEM-MACH AOD” since**

this can cause misunderstandings.

Authors response: Thank you for your suggestion. We changed the text as follows:

For example, the maximum AERONET AOD for the month of June in Fort McKay and Toronto were measured ☐0.27 and ☐0.42 respectively, whereas the GEM-MACH AOD (simulations S1 and W1) were ☐0.19 and ☐0.29 for those sites.

- **Lines 507-508: "There is a higher correlation for Toronto compared to FortMcKay for both seasons." But why?**

Authors response: In an urban area, the vehicle emissions are a large source of pollution and this source has a consistent diurnal profile which is captured well in modelled emissions. While in the oil sands region, the industries emit with less diurnal change and the pollution predicted at a location is dependent on the model predicting the wind direction quite well. Industrial pollution plumes can also be quite narrow.

- **Lines 602-604: I do not think the resolution of GEM-MACH is the problem here. MERRA-2 and the MODIS data you used are much coarser than 10km. Also, the second maxima shown in MERRA-2 does not appear well in the MODIS data. I see some gray shades in that place that would mean that is a grid point not available from MODIS. I think maybe CFFEPS is underestimating the fire emissions, and that could also explain the discrepancies shown in Fig. 9.**

Authors response: We replaced MODIS and MERRA-2 (hourly) plots in Figure 14 with MAIAC and VIIRS plots.

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Please also note the supplement to this comment:
https://gmd.copernicus.org/preprints/gmd-2021-172/gmd-2021-172-AC2-supplement.pdf