Atmospheric Chemistry and Physics Discussions

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Comment on acp-2021-409

Anonymous Referee #2

Referee comment on "Quantifying the structural uncertainty of the aerosol mixing state representation in a modal model" by Zhonghua Zheng et al., Atmos. Chem. Phys. Discuss., https://doi.org/10.5194/acp-2021-409-RC3, 2021

General Comments:

Zheng and coauthors have undertaken a novel comparison of predicted aerosol mixing state between their highly-detailed particle-resolved model and an existing Earth system model (CESM2). The authors are able to leverage the impressive numerical formulation of PartMC-MOSAIC and the machine learning approaches they have developed to provide some context as to how well the widely used modal method is able to capture aerosol mixing state. The paper is especially well-written, the content is well-organized, figures are well-constructed. In short, the manuscript was a pleasure to read. I encourage ACP to publish this paper, considering it is one of many steps toward a more sophisticated future landscape of aerosol modeling approaches. However, I do have a couple of topics I would appreciate the authors commenting on in the manuscript to make sure they are resolved or documented before moving on from this study.

Major Concerns:

- The relevance of the selected ML input parameters for predicting aerosol mixing state is clear, especially for an exercise where one is feeding the inputs from actual measurements to model. However, in this study, it is the CESM2 fields that are being used to specify the parameters, including the concentrations of each aerosol species. Since the aerosol formulation in the CESM2 has an impact on the speciated aerosol concentrations (e.g. the authors mention the example of BC transferring to the mixed mode and depositing faster), how can the authors be confident that the ML fields in Fig. 3 are actually realistic if they are based on CESM2 model data that they are arguing is at least partially corrupted? Forgive the unproven hypothetical here, but in the extreme, couldn't it be possible that the MAM4 model gives erroneous PM species concentrations that compensate with an erroneous framework to yield an accurate mixing ratio field? The authors provide helpful discussion in section 5.3 to address this, so that extreme example is unlikely. But would they consider adding some results that show the variability in their ML fields with reasonable variation in the underlying inputs, perhaps based for input ranges informed by the biases of the CESM2 speciated PM predictions?

- Additionally, would the authors please explain why other parameters relevant for sources are not appropriate for the ML input? I'm thinking of data like land use, human population, and wind speed that could help inform dust, carbon, and sea spray mixing state. If it's been documented, as the authors say, that the mixing state parameter goes down close to sources, why not use that information in this exercise? Maybe this would require running the ML training cases for longer than 24 hrs to get a signal for aging (i.e. distance from sources) on mixing state?
- My main concern with this study is that the authors have chosen to focus on surface data from the global model, and yet are interested in impacts on radiative forcing and cloud-aerosol interactions. Certainly, to know the true scope of the potential problem in misrepresentation of aerosol mixing state, the full atmospheric column needs to be taken into account.
- Do the authors have any idea how PartMC-MOSAIC ML performs with scavenging in place? The ML input parameters don't include hydrometeors or cloud cover as a marker for aerosol removal. It seems like the impact of a major process like cloud scavenging will be extremely important to understand from at least a few points of view: 1) impact on ML representation of PartMC-MOSAIC and 2) divergence between ML and MAM4 in the context of clouds/fog.

Minor Suggestions/ Typos:

- Abstract: I recommend adding a few words or a sentence to the end of the abstract that explicitly connects back to the main motivation, which I assume to be a better quantification of radiative forcing and aerosol-cloud interactions. How significant is the 70% modal model bias for those aims?
- Page 2, lines 38-43: A related issue is the transfer of particles from smaller modes to larger ones during growth, even if this is among "pure" modes. Also, removal of particles due to scavenging by cloud activation is difficult to reconcile.
- Page 4, line 88: Please add an explicit reference for the MAM4 long-range transport bias.
- Fig. 1: Legend obscures data in right panel.
- Section 3.3: I'm sure there are many applications for grouping surrogate species in different ways, but I think an important one worth mentioning is heterogeneous reactions on surfaces of (e.g.) primary dust or sea spray particles. There is probably much overlap with the hygroscopicity mixing state index. If the authors agree, perhaps a comment could be added to that effect.