

In this study, the authors implemented a three-category representation of aerosol mixing-state (1L2B) into an air quality and weather forecast model (GEM-MACH), which can better resolve differences in hygroscopicity due to aerosol composition and the amount of absorption enhancement of BC due to non-absorbing coatings. A two-category representation of aerosol mixing-state (HYGRO) was also implemented in the model. They found that the more-detailed representation of the aerosol hygroscopicity in both 1L2B and HYGRO decreases wet deposition, which increases aerosol concentrations, particularly of less-hygroscopic species. They further show that these increased aerosol concentrations increase cloud droplet number concentrations and cloud reflectivity in the model, decreasing surface temperatures. The manuscript is well written, and results are clearly presented. I have a few comments for the authors to consider.

General Comments:

1. It is a bit unclear to me how the aerosol species in each category are treated during physical and chemical processes. I would suggest the authors to make it clear and explain it a bit more in section 2. Some details can go to appendix or supplement.

I wonder how many aerosol species are in the three categories. Does each category all have 8 aerosol species? In each size bin, it has  $8 \times 3 = 24$  aerosol species. In total, the model has  $8 \times 3 \times 12 = 288$  aerosol species being transported/advected? Please give some more details.

The authors assume that aerosol in the lo- $\kappa$  mixing-state categories does not participate in aqueous chemistry and is not removed by cloud-to-rain conversion and subsequent wet deposition. Can aerosols in the lo- $\kappa$  mixing-state categories be activated as cloud droplet? Or the authors mean they can be activated but would not be allowed to be removed by in-cloud wet scavenging processes? How does the model treat the aerosols for droplet and rain evaporation processes? Does the model have some kind of “cloud borne” tracers to track these aerosol species?

I agree with reviewer #1 and would also suggest that the authors to explain in the manuscript how chemical reactions could also modify aerosol mixing state. According to section 2.1, emitted dust would go to lo- $\kappa$ \_lo-BC category. After gas-aerosol partitioning, sulfate, nitrate, and chloride can form on the particle surface making it more hygroscopic. All aerosol species in this category will be moved to hi- $\kappa$  when reaching the threshold?

2. There are also uncertainties in the hygroscopicity values used for aerosol species. I would love to hear the authors' comments on the impact of the hygroscopicity uncertainties on the three-category aerosol mixing state treatment.

3. Regarding the analysis, I wonder if the authors have checked the vertical profiles of the

aerosol species and use some aircraft campaigns during the simulation period, like NASA ATom-1? The comparisons of aerosol species with IMPROVE and CSN tend to give similar results. To me, it would be more useful to compare concentrations of large aerosol particles as well (e.g., against EPA CASTNET measurements). Since the authors have looked at cloud properties like cloud droplet number concentrations, I wonder if the authors have further looked at the changes in the radiative fluxes or aerosol forcing.

#### Specific Comments:

For Figures 1-7, I don't see a strong need to organize the panels in this way. SRIM panel plots are much larger than the other panel plots, and there is much white space there. To me, three panel plots in a row looks more organized. There are no marks/lines showing latitudes and longitudes. Please add them. Although the authors stated that most variables shown are identical in the HYGRO and 1L2B simulations, it may still be useful to show the spatial distributions of differences/relative differences between 1L2B and HYGRO. Do 1L2B and HYGRO have identical spatial distributions of the variables shown.

Lines 43-58, consider adding a summary table for the various representations of aerosol mixing-state?

Lines 101-102, I'm quite confused here. Please try to rephrase/clarify it. What do you mean "our BC-free category is also hydrophobic"? If the authors mean hi- $\kappa$  category, why it is also hydrophobic. Or the authors mean lo- $\kappa$ \_lo-BC category? "we have a single category for all hydrophilic particles" seems also to be hi- $\kappa$  category?

Lines 142-143, "Therefore, there is never any absorption enhancement for particles in the hi-BC mixing-state category". The authors assume that there is no absorption enhancement for BC cores that comprise more than 40% of the particle by mass. The authors also stated previously that a threshold of 0.3 between lo-BC and hi-BC is used. I wonder what about the BC mass fraction between 0.3 and 0.4.

Lines 160-165, it is also not very clear to me. What do you mean "dust is defined to be the residual after the other components are accounted for"? Is the dust emission calculated online or prescribed?