An Improved Representation of Aerosol Mixing State for Air-Quality-Weather Interactions
Stevens et al., 2022

Summary

This study is about a development of a model representation of the aerosol mixing state in a chemical transport model called GEM-MACH. The authors developed a three-category representation (1L2B) to resolve an aerosol population into three categories, i.e. (1) high hygroscopicity, (2) low hygroscopicity with high BC content, and (3) low hygroscopicity with low BC content. The authors further compared the 1L2B representation to the other two representations called HYGRO (2-category, high hygroscopicity, low hygroscopicity) and SRIM (the original one in GEM-MACH which assume complete internally-mixed aerosols) by evaluating against the observation of a case of biomass-burning over North America. Besides, the study investigated the impact of aerosol mixing state representation on meteorology with and without feedback. The study matches the scope of *ACP*. I recommend acceptance of this manuscript for publication upon satisfactorily clarifications of the following issues.

Questions/clarifications

1. (Line 25) Apart from condensation and coagulation, chemical reactions could also modify aerosol mixing state (a.k.a. chemical aging of aerosol). I suggest the authors to clarify this.

2. (Line 69-75) In the aerosol representation of 1L2B, the fixed bin boundary of hygroscopicity (kappa) and BC mass fraction of 0.1 and 0.3 proposed by Ching et al., 2016 was derived based on urban environments. It is possible that in the scenarios of biomass burning (test case in this study), such fixed boundary values may not be the most optimal in resolving aerosol mixing state. Should the author provide some sensitivities studies on the bin boundary positions, it would enhance the scientific values of this manuscript and applicability of the model representation to various kinds of environments.

3. (Section 2.1) Apart from the atmospheric physical and chemical processes, the sensitivity study of aerosol mixing state also depends on mixing state of the aerosols at the instant of their

emissions. It is expected that the authors should state clearly or tabulate the assumed chemical composition of the particles from biomass burning (i.e. smoke particles from forest fire) as well as other aerosol types when they are emitted. This would definitely affect the evaluation of the simulations.

4. (Line 303) According to the latest particle-resolved study published by *Yao et al., 2022 ACP*, based on 1800 environmental scenarios, simplified aerosol representation (assuming internally-mixed aerosol particles) could potentially lead to a wide range of underestimation in aerosol scattering with a maximum of -32% among the environment scenarios they studied. Therefore, I think it is better for the authors to clarify in the text that error in scattering coefficient of an aerosol population may not be negligible, instead it could depend on the interaction between the aerosol particles and the environment, such as relative humidity and chemistry processes involved.

5. (Line 311) Please explain why "higher black carbon mass fraction in HYGRO … SRIM case would be expected to reduce the AAOD", it is not clear to me why this is the case, please explain further.

6. (A general question and section 3.1.5) It is well known that relative humidity affects aerosol scattering through the size of the particles (aerosol swelling) and aerosol water content. These are related to the aerosol hygroscopicity which is determined by the aerosol mixing state. I would like to see some discussion about the role of relative humidity in the case of biomass burning (in this study), for example when the authors discuss aerosol-radiation interactions in 3.1.5 (p.16). For example, over the model domain, there is variation of relative humidity, how does the difference between 1L2B, HYGRO and SRIM relate to the relative humidity?

7. (Model infrastructure) It is expected to see in the methodology section about the construction of aerosol mixing state representation schemes of HYGRO and 1L2B. For example, how the aerosol particles are transferred from one bin to another. Please provide some descriptions in this aspect.

8. (Model computational aspect) It is expected to see some information of the computational cost and accuracy comparisons among the three aerosol mixing state representations. The accuracy could be calculated with respect to observations or SRIM.