

Review on “Potential limitations of using a modal aerosol approach for sulfate geoengineering applications in climate models” by Visioni et al.

This study investigates the potential issues of using a modal aerosol scheme for simulating geoengineering sulfate aerosols. Simulation results from the CESM1 Geoengineering Large Ensemble (GLENS) project are analyzed and impacts on aerosol concentrations and radiative fluxes are quantified. This is a useful and important contribution which points out the importance of carefully designing aerosol configuration (e.g., mixing state, mode standard deviation, size range), particularly for representing the unconventional cases of geoengineering stratospheric aerosols.

My main comment is that the issues are not due to the modal aerosol scheme itself (as pointed out in the title of this paper), but to the negligence in the scheme design. Modal schemes assume internal mixing between aerosol species within a single aerosol mode, and this is the case of bin schemes which assume internal mixing between aerosol species within a single aerosol bin. Different aerosol modes should be created for tropospheric coarse dust/sea salt and for stratospheric coarse sulfate as they have vastly different properties. We cannot simply lump these in the same aerosol mode. We note that the aerosol schemes used in global climate models often have to be simplified (minimalized) due to the consideration of computational efficiency. For the comparison, the bin scheme (CARMA in CESM) uses a group of bins for pure sulfate and another group of bins for mixed aerosols (POM, BC, dust, sea salt, sulfate internally mixed in a bin). The modal scheme (MAM4 in CESM) has done similarly to design the primary carbon mode specifically for carbonaceous aerosols (POM/BC). Currently efforts are underway (to develop MAM5) by adding a new mode for stratospheric sulfate separated from the tropospheric coarse mode for dust/sea salt.

Based on the above comments, major revision of the manuscript is required before the paper can be accepted by ACP, including the title, abstract, and conclusions in the text to make it clear that the issues are not due to the modal scheme itself and call for more careful design of mode (bin) structures.

We thank the reviewer for their supportive comments, and for making a very important point regarding our manuscript. We have modified all necessary parts as the reviewer suggested.

The title has been changed to: “*Limitations of assuming internal mixing between different aerosol species: a case study with sulfate geoengineering simulations*”

The abstract has been changed to (changes in bold, line and page referring to the revised version)

(lines 1-5, p1) “*Simulating the complex aerosol microphysical processes in a comprehensive Earth System Model can be very computationally intensive and therefore many models utilize a modal approach, where aerosol size distributions are represented by observations-derived lognormal functions, and internal mixing between different aerosol species is often assumed*”

(line 6, p1) “*In this work we show specific conditions under which the current approximations used in some modal approaches might yield some incorrect answers*”

In the Conclusions, some remarks have been added:

(line 392, p19) “*MAM3 only separates the species in three modes depending on their size, by treating all aerosol species as the same (internal mixing assumption).*”

(line 423-424, p. 21) “A similar approach has already been used to include an additional primary carbon mode in MAM4 (Liu et al., 2016) in order to account for processes that affect the microphysical properties of primary carbonaceous aerosols in the atmosphere.”

Minor comments:

Line 50-54, please refer to Riemer et al. for more accurate definition of aerosol mixing state: Riemer, N., Ault, A. P., West, M., Craig, R. L., & Curtis, J. H. (2019), Aerosol mixing state: Measurements, modeling, and impacts. *Reviews of Geophysics*, 57, 187–249. <https://doi.org/10.1029/2018RG000615>.

Added.

Line 101. “observed” change. I am sure the “observed” means the “identified”, “noticed” or “found” in your study, not change from observations (in the field campaign or laboratory). To avoid confusion, please use another word. This same is true for “observed” and “observation” in many places in the following text.

We have modified “observed” in “identified” as suggested by the reviewer. We have identified all cases in the manuscript where the term “observed” or “observation” was used improperly, and modified them all accordingly.

Line 131-137. Some statements are not clear: “only one type of particles for each mode is considered”; “Therefore, each particle species shares the same radius and number concentration per each mode,”. Because of internal mixing of aerosol species in an aerosol mode, composition of aerosols within one mode is the same, as well as size. Refer to the above Riemer et al. paper for the definition of aerosol mixing state.

*We have simplified the phrase in “Compared to the more computationally-demanding MAM version with 7 aerosol modes (MAM7), **in MAM3 all aerosol species are considered internally mixed within each of the three modes, thus sharing composition and size distribution.** The mass for the single species has to be conserved, both globally and locally, and thus can only change in each gridbox if particles are moved from one gridbox to the other, either because of air mass movement, or because of gravitational settling or other tropospheric removal processes.”*

Line 187. “which can then oxidize and form sulfate particles of sulfuric acid in the smaller (Aitken) mode.” The sentence is awkward. Might change to “which can be oxidized to form sulfuric acid and then sulfate particles by condensation in the smaller (Aitken) mode.”

We thank the reviewer for the suggestion. The phrase has been changed accordingly.

Line 234. “In MAM3, all aerosol species are assumed to be internally mixed.” should be “In MAM3, all aerosol species *within an aerosol mode* are assumed to be internally mixed.”

Added, thank you!

Line 252-253 and Figure 1. The increases of dust and sea salt in UTLS regions in GLENS is likely

due to the renaming (transfer) of the accumulation mode dust and sea salt (along with stratospheric sulfate) to the coarse mode. Because of the small standard deviation of coarse mode, dust and sea salt are accumulated and increased there.

If the observed change was due to a transfer from one mode to the other, however, there would have to be a similar reduction in the accumulation mode in those species. Figure 3, on the other hand, shows that accumulation mode dust and sea salt are slightly higher above 300 hPa in GLENS compared to the Baseline case, and little change is observable below that. This lead us to exclude a significant contribution from a renaming of accumulation mode particles into coarse mode.

Line 282. “known as immersion or heterogeneous freezing”. Use “heterogeneous nucleation”. Immersion is just one of the mechanisms of heterogeneous nucleation of ice.

Changed, thank you for the suggestion.

Line 364. “positive in the shortwave (implying a cooling)”. In Figure 7, it is shown to be negative.

Sorry, that should have said “negative in the shortwave”. We have corrected it.