Response to Comments of Reviewer #2

Title: Size-resolved process understanding of stratospheric sulfate aerosol following the Pinatubo eruption

General comments:

I think the article is well written and a nice advancement to stratospheric aerosol modeling but may be better suited in its current form for a journal such as Geoscientific Model Development. While the differences in aerosol loading due to model configuration are made clear, it is left to the reader to interpret how this improves understanding of atmospheric chemistry or physics. The authors provide in-depth discussion on the relative importance of coagulation, nucleation and renaming/growth in the model, but have little discussion on the physical processes this may help resolve. Similarly, few comparisons are made with measurements for AOD, particle size or radiative flux, with no discussion given to possible sources of disagreement, or what implications these results may have for observations (e.g. the assumptions going into the HIRS results used here). Personally, I think addressing any of these points would help expand the applicability of the paper to a more general audience.

Thanks to the referee for the helpful comments and constructive suggestions. We have revised the manuscript carefully and the point-to-point responses are listed below.

As shown by the title of the manuscript, this work aims to provide in-depth discussion on the relative importance of coagulation, nucleation and renaming/growth in stratospheric sulfate formation.

With regards to comparisons with observational data, we have already compared our results against stratospheric sulfate burden from HIRS observations (Figure 4 of revised manuscript) and AOD against AVHRR observations (Figure 8 of revised manuscript). Other sources of observational data are available, such as TOA radiative flux with ERBS and aerosol size comparisons with WOPC and SAGE. However, Brown et al. (2024), another study regarding the simulation of Pinatubo in E3SMv2 (the same model as we used in this work), has mostly already covered these comparisons. Their PA experiment is extremely similar to the MAM4SC experiment in this study, while their SPA experiment is extremely similar to MAM5SC excluding the addition of an independent stratospheric coarse mode. Neither the addition of the new mode or the use of a more complex chemistry scheme is meant to significantly alter model output of TOA radiative flux, and so we do not think it is necessary to repeat such comparisons in this work, and instead refer the reader to Brown et al. (2024).

Since a new stratospheric coarse mode is added in this work, we have included a new Figure 6 to compare simulated volume-size distribution against the observations from WOPC following your suggestions. It can be seen that MAM5FC did better capture the coarse mode volume (or mass) of sulfate aerosol in 1992 and 1993.

We have also added a global mean AOD anomaly comparison between model simulations and satellite-derived AOD datasets, AVHRR and GloSSAC (Figure 9 in the revised manuscript), which provided global coverage during the Mt. Pinatubo eruption. AVHRR is more sensitive to rapid AOD increases caused by eruptions but becomes less accurate for AOD values below 0.01, while GloSSAC is accurate at lower AOD values but becomes saturated above 0.15. These observations help quantify the bounds of AOD changes from Mt. Pinatubo. The MAM5FC and MAM5SC simulations showed reasonable AOD peaks and decay patterns, while MAM4FC and MAM4SC tended to underestimate AOD strength and overestimate the decay rate.

Major concerns/questions:

1. Line 50-65: This geoengineering section seems a bit out of place to me. I'm sure this work has implications for geoengineering studies, but no indication of exactly what those may be is provided. I recommend clarifying the link to this work or removing this paragraph. Perhaps the geoengineering discussion and the link to this work would be better placed in the discussion/conclusion?

   Response:

   Geo-engineering is commonly cited as a motivation into the simulation of volcanic aerosols/stratospheric sulfates, e.g. Mills et al. (2017) introduction and Tilmes et al. (2022).

2. Line 415: "MAM4 also generally has stronger nucleation and coagulation processes than MAM5." From Figure 6 it isn't clear whether MAM4FC has greater coagulation tendencies for physical reasons or if it is just due to the increased NUCL. This is discussed in the conclusion of the paper (Line 503), but I think should be mentioned here. I would suggest rewording to something like: "MAM4 also generally has stronger nucleation than MAM5, and due to these higher concentrations, increased coagulation processes as well" Or, if there are other reasons for increased COAG then this should also be discussed.

   Response:

   Thank you for the suggestion. A short explanation has been added to the sentence: "MAM4 generally has stronger nucleation leading to higher Aitken mode concentrations, and therefore also has stronger coagulation processes than MAM5."

3. Line 490: "large differences in both the temporal variations and the spatial distributions of sulfate concentrations" It is difficult to tell from Figures 7/8, but in Figures 9/10 there doesn't appear to be much change in spatial distribution. Both MAM4 and MAM5 show large increase in the tropics and later transport to the NH. Some expansion on the spatial differences the authors are referring to would be welcome.

   Response:

   This is primarily referring to Figure 3, where SC experiments had sulfate concentrations at a noticeably higher altitude than FC. This is mentioned further on in the paragraph: "Vertically, sulfate distributions were generally at lower altitudes in FC compared to SC…"

4. Line 521-522: If the use of full chemistry and MAM5 helped improve agreement with AVHRR, why is the TOA flux more comparable to Brown (2024) and Mills (2017) results than the MAM4 version? Is this related to the geometric standard deviation that was used?

   Response:

   The fundamental changes made to the aerosol module are similar across MAM5 in this work, the altered MAM4 scheme in Brown et al. (2024), and the altered scheme in Mills et al. (2017): they all allow sulfate to rename into coarse mode and adjusting some of the coarse mode parameters (e.g., reduced the geometric standard deviation of the coarse mode in their altered MAM4 to that as used in the stratospheric coarse mode in MAM5 of this work) to better fit observational data. On the contrary, MAM4 in this work does not allow sulfate to rename into coarse mode (i.e., continued increase in sulfate mass leads to an increase in the accumulation mode number concentration, rather than a transition from accumulation mode mass to coarse mode mass). As a result, the TOA flux from MAM5 agrees closer to Brown (2024) and Mills (2017) than that from MAM4, and this is the expected result.

References:

Brown, H. Y., Wagman, B., Bull, D., Peterson, K., Hillman, B., Liu, X., Ke, Z., and Lin, L.: Validating a microphysical prognostic stratospheric aerosol implementation in E3SMv2 using observations after the Mount Pinatubo eruption, Geosci. Model Dev., 17, 5087-5121, 10.5194/gmd-17-5087-2024, 2024.

Mills, M. J., Richter, J. H., Tilmes, S., Kravitz, B., MacMartin, D. G., Glanville, A. A., Tribbia, J. J., Lamarque, J. F., Vitt, F., and Schmidt, A.: Radiative and chemical response to interactive stratospheric sulfate aerosols in fully coupled CESM1 (WACCM), Journal of Geophysical Research: Atmospheres, 122, 13,061-013,078, 2017.

Tilmes, S., Visioni, D., Jones, A., Haywood, J., Séférian, R., Nabat, P., Boucher, O., Bednarz, E. M., and Niemeier, U.: Stratospheric ozone response to sulfate aerosol and solar dimming climate interventions based on the G6 Geoengineering Model Intercomparison Project (GeoMIP) simulations, 2022.