Response to Comments of Reviewer #1

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

General comments:

The paper is mostly well written with a specific purpose but there are difficulties. The first difficulty is the limited comparisons with observations, just two sets of observations and two figures. Both figures raise questions about the comparison, but little discussion is given to the disagreements. A lot of papers have been published describing the post Pinatubo aerosol from a variety of instruments. Why aren't additional comparisons made with a much wider set of data? The model has pretty fine resolution, so it should not be limited to comparisons to measurements with global coverage. The authors spend a lot of energy comparing the various size distribution modes from the different model configurations, but make no attempt to compare any of these size distributions to observed size distributions. The authors offer no explanation for their limited comparison with observations.

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.

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 (see below) in the revised manuscript 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.

Furthermore, we have added a global mean AOD anomaly comparison between model simulations and two satellite-derived AOD datasets, AVHRR and GloSSAC (Figure 9 in the revised manuscript, see below). These two observations provided global coverage during the Mt. Pinatubo eruption. Due to limitations in the onboard instruments, AVHRR is more sensitive to the rapid AOD increase caused by volcanic eruptions but becomes less accurate when AOD values fall below 0.01 (Russell et al., 1996; Quaglia et al., 2023). Conversely, GloSSAC measurements become saturated when AOD exceeds 0.15, but are accurate when AOD values are relatively small (Thomason et al., 2018). This justifies using AVHRR and GloSSAC to quantify the upper and lower bounds of the AOD changes caused by Mt. Pinatubo. Consequently, Figure 9 in the revised manuscript has been added to evaluate the performance of different aerosol-chemistry schemes. We assessed the time evolution and e-folding time of the simulated global AOD anomaly against satellite observations (AVHRR and GloSSAC). The MAM5FC and MAM5SC simulations generally produced reasonable AOD peaks and decay patterns, while MAM4FC and MAM4SC tended to underestimate AOD strength and overestimate the decay rate.

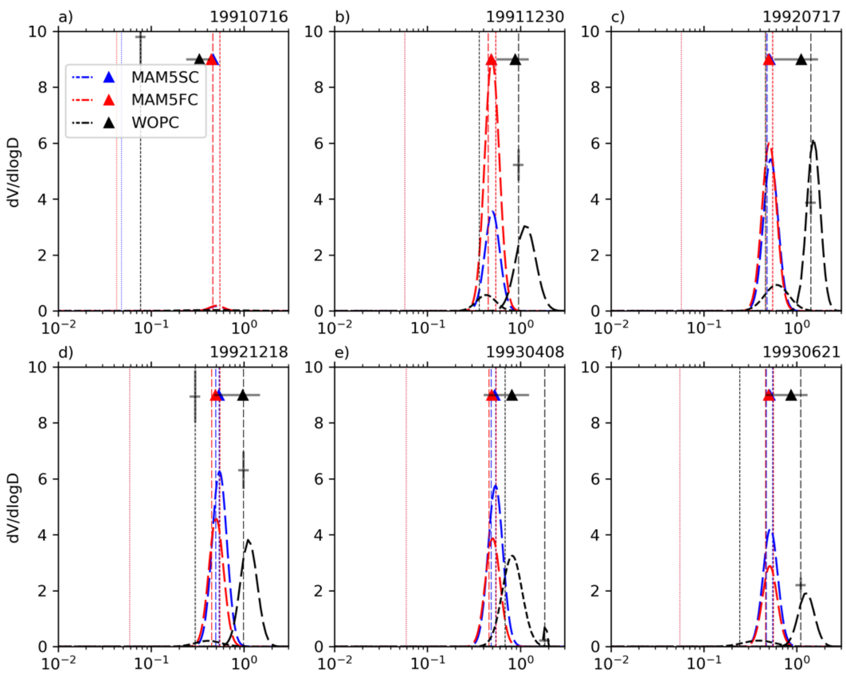

Figure 6: Comparisons of stratospheric aerosol size distributions from MAM5SC and MAM5FC with observations from WOPC for 1991–1993. WOPC launches are samples taken from the 18 km measurements and matched to the nearest model height and grid cell over Laramie, Wyoming (41.3° N, 105° W).

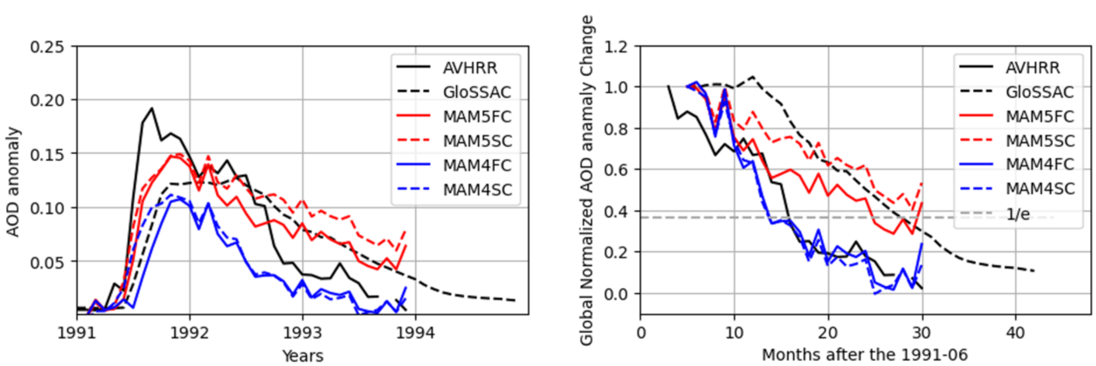

Figure 9: Comparison of AOD anomaly between simulations and observations. The left panel shows the time evolution of monthly AOD anomaly values from simulations and AVHRR and GloSSAC observations, while the right panel shows the time evolution of normalized AOD anomaly values.

Major concerns/questions:

1. 119-120 In terms of size order aren't the modes: Aitken, accumulation, coarse, rather than accumulation first? If so then they should be listed in that order.

   Response:

   Thank you for pointing this out. We have revised the text to be ordered by size.

2. Fig. 1 What happens to particles in MAM4 when they exceed Dg_hi (0.48 µm), which is not that large for sulfate particles following Pinatubo? See e.g. Deshler et al., GRL, 1992.
   Response:
   In MAM4 renaming is not turned on, therefore the geometric mean diameter is not allowed to exceed Dg_hi. The model will instead increase the aerosol number to maintain conservation of mass while also maintaining a maximum geometric mean diameter.

3. 178-179 The parenthetical clause is so long the reader has lost the thread as to what limits the aerosol formation rates.
   Response:
   We have restructured this sentence to make it more readable.
   "Most importantly, OH radicals (as well as other oxidants) are prognostically calculated rather than using prescribed values. This allows the model to represent the localized depletion of OH radicals due to the injection of large amount of SO_2, bottlenecking aerosol formation rates."

4. Fig. 2 and its discussion. What is the explanation for the aerosol in the Southern Hemisphere, which appears at most longitudes almost simultaneous with the Pinatubo eruption, particularly in MAM5FC? The presence of this aerosol clearly above background should be acknowledged and if possible explained.
   Response:
   The issue with aerosol in the Southern Hemisphere is caused by the color bar, because the background values for sulfate aerosol are just slightly larger than the upper limit of the white color. To avoid confusion, we have replotted it with a different color bar (see below).

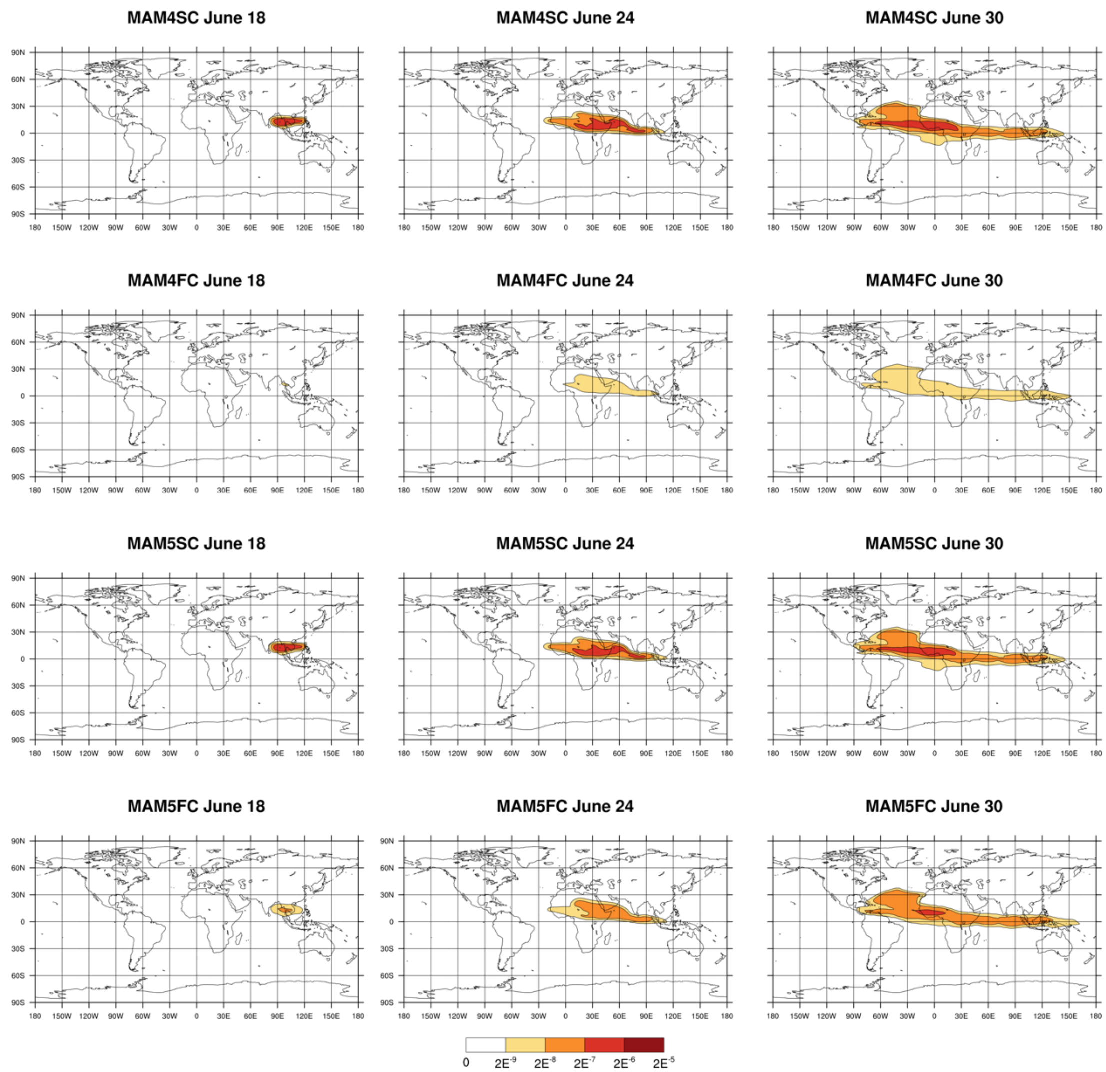

Figure 2: Simulated distributions of sulfate aerosol concentrations (kg/kg) at 53 hPa for days 3, 9 and 15 after the eruption of Pinatubo, respectively, for experiments MAM4SC (first row), MAM4FC (second row), MAM5SC (third row), and MAM5FC (bottom row).

5. 284-286 Why does the smaller geometric standard deviation in MAM5 lead to more persistence? Is it because the particles are smaller in MAM5 compared to MAM4 and therefore less sedimentation? In any case there should be a sentence to describe the physics involved.

   Response:

   The removal rate of aerosols from the stratosphere in E3SM depends heavily on aerosol size, with larger aerosols having a significantly shorter lifespan. This is because the Stokes' settling velocity is roughly proportional to the square of aerosol diameter. With all other factors being identical, when the geometric standard deviation is smaller (i.e. smaller particles are less small, and larger particles are less large), a smaller portion of the aerosol population is large enough to be removed more quickly. We have added a brief explanation and a reference to the Seinfeld and Pandis textbook (Seinfeld and Pandis, 2016).

6. Fig. 4 and its discussion. The discussion mostly consists of describing the figure providing specific dates and sulfate burden peaks for the various modes. While perhaps these details are interesting they are all available in the figure for the interested reader. More interesting for the reader would be more discussion of the model / measurement discrepancies. Why do all models except MAM4FC over estimate the peak observed sulfur burden by 20%? Are the HIRS data reliable at the peak or are they suffering a saturation problem? Why does HIRS fall off so much faster than the MAM5 models in 1993? Additional interesting detail would be the range of median sizes involved in the large particle mode.

   Response:

   There are several possible reasons for the disagreement. The first is that HIRS results from Baran and Foot (1994) are not particularly accurate outside of the tropical areas due to errors introduced from the background signal (this is acknowledged in their results and discussion section), and as time passes sulfate aerosol is transported towards the poles.

   In general, MAM5FC is able to accurately reflect the observed burden prior to 1993. With respect to the slower fall off in MAM5 models relative to HIRS in 1993, further improvement can be done in our future study. As can be seen in the newly added Figure 6 above, the stratospheric coarse mode burden in MAM5FC consists of particles somewhat smaller than observations, leading to a longer lifespan.

7. Because MAM4 doesn't have a coarse mode and MAM5 has a very narrow accumulation mode, both Figs 4 and 5 show the same thing, no contribution in the accumulation mode (or very little) from MAM5 and no contribution in the coarse mode from MAM4. Why then show the accumulation mode at all? Combine the MAM4 accumulation mode and MAM5 coarse mode into one figure. MAM5 accumulation mode could be included as a dotted line in the coarse mode plot. Then the two models can be more easily compared in terms of sulfate burden carried in the large particle mode.

   Response:

   The stratospheric coarse mode is unique to MAM5 which does not exist in MAM4, so we would prefer to keep the plots separate in order to emphasize this. However, we have changed the line styles in the figures according to your suggestions here and in your question #16 (we consistently use blue color for MAM4, red for MAM5, solid line for FC, and dashed line for SC). See revised Figure 5 below as an example.

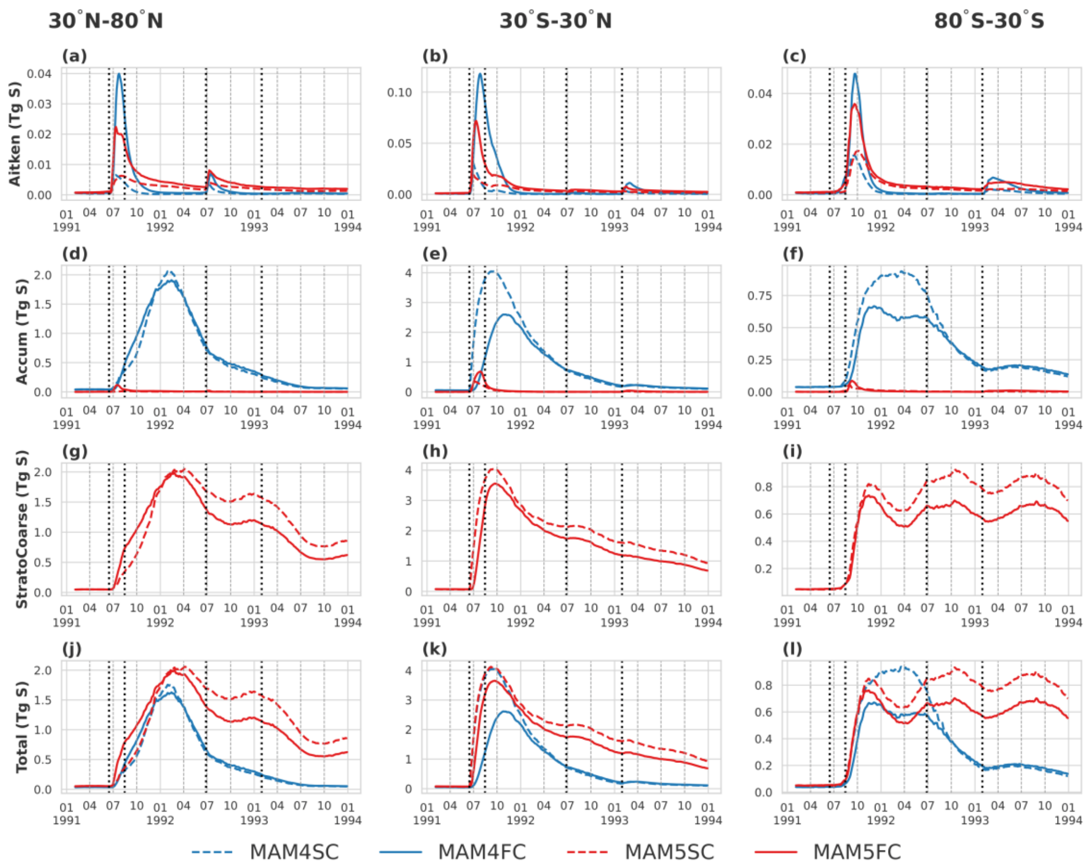

Figure 5: Simulated stratospheric sulfate burden (in Tg S) divided by mode and latitudinal region. The vertical lines represent the Pinatubo, Cerro Hudson, Spurr and Lascar eruptions respectively.

8. Fig. 5 Same comments as Fig. 4 but in addition the ordinates should all be the same scale (0-4 Tg S), so the relative contributions from the different latitude zones can be seen directly. Without that the reader immediately wonders about the Southern Hemisphere signal which seems to persist at high levels. The label on the ordinate is wrong. It should be Sulfur burden (Tg S). Label the rows in some other way or describe them in the figure caption. Again combine accumulation and coarse mode into one plot then there will only be three rows. The latitudes of the eruptions should be listed in the figure caption.

   Response:

   Thank you for the suggestion. While using the same scale does have some merits for comparison of magnitude, our original intention is to focus on the aerosol microphysical processes (for example, how long they last in each different mode, etc.). If we set the scale to be identical across the figures, some features would not be very readable, such as Aitken mode with very small burdens.

9. 331-332 What is the reason for this oscillation, when the sulfur burden is declining everywhere else? A somewhat similar oscillation, but offset, is observed in the Northern Hemisphere. Why does the sulfur burden persist to 1994 in the Southern Hemisphere while it decays everywhere else?

   Response:

As mentioned in our responses to your question #6, further improvement can be done in our future study. As can be seen in the newly added Figure 6 above, the stratospheric coarse mode burden in MAM5FC consists of particles somewhat smaller than observations, leading to a longer lifespan. If we adjust the size parameters to have larger sulfate particles, we would see larger sedimention along the path of transport and hence smaller burden and oscillation in high latitudes.

10. 341-343 This point would be a lot clearer for the reader if the ordinate scale on all plots was the same. But this statement, "Starting from 1993, the stratospheric burden south of 30 S in MAM5 begins to make up at least half of the total between 80 S and 80 N (about 0.8 Tg S out of the total of 1.7 Tg S)", doesn't make sense. Between 80 S and 80 N includes 30 S – 30 N, so the total sulfur burden in early 1993 for MAM5FC is ~4.1 Tg S.
Response:
We prefer to keep the current scales for the reasons described in our responses to your question #8.
Thank you for pointing out the error in numbers. We have corrected it as "(0.8 Tg S out of the total of 4.1 Tg S)".

11. 349-350 Suggest rephrasing to. "The tendencies are the integrals over three-dimensions of: all longitudes, …"
Response:
Text has been rephrased as suggested:
"The tendencies are integrated over the three dimensions of: all longitudes, the above-mentioned latitude range, and vertically above the tropopause."

12. Fig. 6 is problematic. There are too many rows making the figure so small that most readers have to blow it up to see it. This could be fixed by breaking it into two figures: the first containing the first 4 rows, the next containing the last 3. Another suggestion is to combine the right and left panels into a single plot with separate ordinates on the right and left. Since the left shows a rate and right shows an accumulation the lines will generally not overlap but rather complement each other. Again the ordinate labels are incorrect. They should be tendency (kg/s) for the left plots and their integrals (or cumulative) (Tg S) for the right. Include the name of the row as a label in each plot. The labels RNMxx are too tied to the inner workings of the model, "renaming". But physically what is happening? The particles are growing to the next largest size distribution mode. If labels were added to the plots to identify them RNMaa could become Aitken->Accumulation mode and RNMasc Accumulation->Coarse mode.
Response:
We actually present the units at the top of the figure for both columns, since each column shares the same unit.
Renaming is not the only way for aerosols to transit between modes in E3SM. Please see definitions of all processes in our response to your question #13 below.

13. Fig 6 e, f) shows the growth from Aitken to accumulation mode presumably by several processes including growth by condensation and coagulation, correct? If that is the case then why are the ordinate scales on COAG so much larger than for RNMaa? Both are showing

mass loss rate and total mass lost. It seems it should be the other way around with COAG less than RNMaa. Why is this one process coagulation singled out for a special plot?

Response:

In the model, condensation, coagulation and renaming are each different aerosol processes. Condensation/evaporation represents the gas-aerosol mass exchange, e.g. transition from H_2SO_4 gas to sulfate aerosol. Coagulation is the process of multiple smaller particles colliding into each other to form larger particles. Renaming is not a "real" physical process and is instead an internal calculation within the model where particles that have grown sufficiently large are moved from one aerosol mode to another. Coagulation and renaming are two separate ways for aerosols to transit from the Aitken mode to accumulation mode; coagulation is not a subset of renaming or vice versa.

Because COAG and RNM are separate mechanisms, we think it is better to keep them separate in figures and discussions. A description of what renaming and condensation represent are in the model overview in Part 2.

14. Fig 6 i – n) Condensation? Why are these processes now called condensation? Condensing from what? Weren't these earlier called renaming, which is also not that helpful or descriptive. Isn't this particle growth from one mode to another? Ordinate problems again. What is on the left and right ordinates? Is it again rate (kg/s) and cumulative mass (Tg S)? The reader doesn't know and the figure caption does not help.

Response:

See our responses to your question #13. We have now clarified the terminology in figure captions.

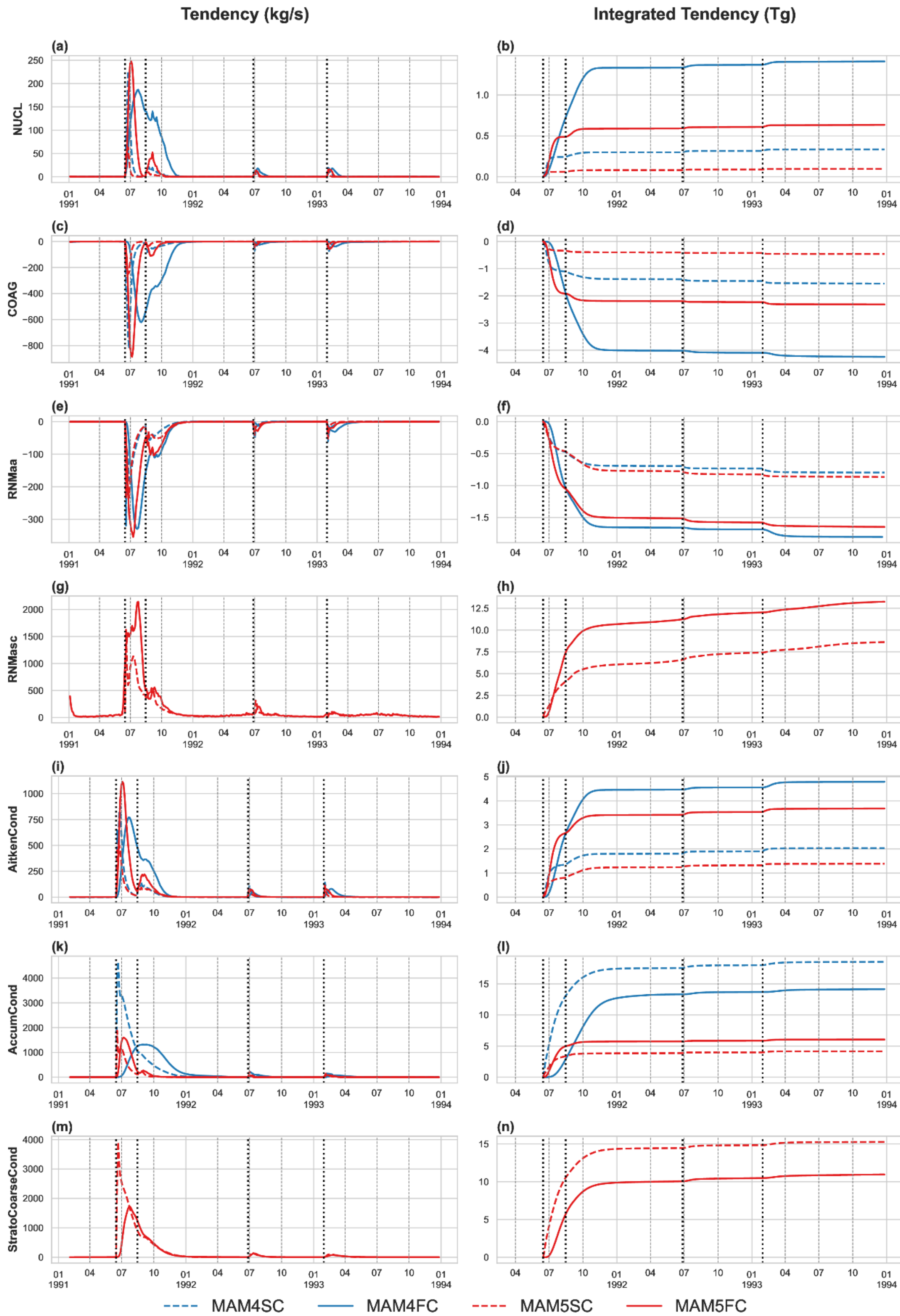

Figure 7: The relevant tendencies for each mode in each experiment in the stratosphere between 80°S and 80°N. See Figure 1(a) for description of processes. Positive values represent gained mass from the associated microphysical process from the perspective of the aerosol mode in question. NUCL represents the aerosol mass gain for Aitken mode due to nucleation (i.e.,

aerosol formation), COAG represents Aitken mode mass loss due to coagulation into the accumulation mode, RNMaa represents Aitken mode mass loss due to renaming into the accumulation mode, RNMasc represents the gain in mass for stratospheric coarse mode in MAM5 due to renaming from the accumulation mode. The left column plots represent the tendency values over time, while the right column represents the cumulative mass change due to the associated microphysical process (i.e. tendency integrated over time). The bottom three rows represent the condensation tendencies for each mode. The vertical lines represent the Pinatubo, Cerro Hudson, Spurr and Lascar eruptions respectively.

15. 351-415 Again the figure discussion consists primarily of describing the figure, pointing out maximum values and dates when they occur. Relatively little is describing what can be learned from the figure which is the importance of the figure. Here and elsewhere, if these dates and amounts are particularly important organize them into a table. Then they could really be compared. It is not clear how listing them in the text helps the reader.
    Response:
    Following the reviewer's suggestion, we have simplified this paragraph, trying to remove some of the numbers and to emphasize the most important information of the figure.

16. Figs. 4-8, 11 The plotting for these figures could be made much more intuitive, so the reader doesn't constantly have to refer to the legend to remember which is which. It would be quite easy to do. Use one color for MAM4 and one for MAM5, then one line style (e.g. solid) for FC and another (dashed or dotted) for SC. Then each figure can be immediately understood without referring to the legend but once.
    Response:
    Thank you for the suggestion. We have changed the line styles in these figures to consistently use blue color for MAM4, red for MAM5, solid line for FC, and dashed line for SC).

17. Figs 7 and 8 suffer from the same problem as Fig. 6. There are too many panels and they are too small. What are Figs. 7 and 8 adding to what we learned from Fig. 6? Are all these rows necessary? Which ones are the most informative?
    Response:
    Figure 7 (previously Figure 6) indeed has too many panels to show processes for all the modes. These are important information for process understanding. We could split the figure into two parts but we would like to keep it for now and discuss it later with the journal editorial team.

18. 420-430 In fact the discussion of Figs. 7 and 8 acknowledges that not much new is added. "The same patterns as above apply between 30 S and 30 N. Above 30 N and below 30 S the same signal from the Pinatubo is still present, though slightly delayed due to the time that it took for the aerosol to transport poleward. Signals from other eruptions (Spurr and Lascar) are also present" Then a few interesting differences are discussed. Just show the interesting panels and combine Figs 7 and 8 into one figure with the few interesting panels.
    Response:
    We had discussions regarding condensation across different latitudinal regions in the text. However, following your suggestions, we have moved the original Figures 7 and 8 to the supplementary.

19. 503 condensation? Same questions as above. What does this mean?
    Response:
    See our responses to your question #13.

20. 503-505 What is the reason for quoting these numbers? How will the reader use such information rather than the already stated comparison about the difference rates? Too detailed.
Response:
We have cut out the specific numbers and left the qualitative comparisons.

21. 506 Here COAG is separated from RNMaa, but aren't both processes doing the same thing? There is only the transition from Aitken to accumulation. Is it important how it happens? If so why isn't that mentioned earlier?
Response:
See our responses to your question #13.

22. 509 deposition? Does this mean sedimentation out of the stratosphere? Generally deposition refers to losing aerosol due to contact with a surface.
Response:
"deposition" has been changed to "sedimentation".

References:

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