Response to the handling editor:

Thank you very much for taking the time to update the questions and comments to the latest version of the manuscript. Please find our responses below:

1. L251ff: What is the difference between R1 and R2? Why is R1 included? **Response:** 

The listed reactions are all of the reactions within the default model's chemistry processor package. R1 and R2 are listed in Table 3 of Emmons et al. (2010) as part of the MOZART chemistry scheme which "simple chemistry" is based on. Within the model, these two reactions have different reaction rates in the absence and presence of a reaction chaperone M, respectively. To clarify, we have added the following sentence before the list or reactions: "R1 and R2 have different reaction rates, with the latter requiring an additional chaperone molecule."

2. L254ff: It seems intermediate steps are left out in this chemistry? Or why don't the equations balance, e.g. R4, R6. R7?

#### **Response:**

Intermediate steps are left out to simplify calculations and reduce computational costs. Some byproducts (e.g., O<sub>2</sub>, H<sub>2</sub>O) are not produced in sufficient quantities to significantly influence their concentration in the atmosphere (e.g., O<sub>2</sub> production in R4) or affect their participation in SO<sub>2</sub> production or other reactions, so these byproducts are simply omitted from the reactions. This is why some of the equations do not balance.

3. Fig 5 caption: please include a warning of different scales on vertical axes **Response:** 

Thank you for your comment. A note has been added to the end of the Fig. 5 caption: "Note the differing vertical axes in different plots."

4. L481: "In order" The authors use this throw away phrase too much. It is almost never necessary.

# **Response:**

Thank you for the suggestion. Several instances of the phrase have been removed. Now it is "To examine the differences..." in Section 2.3 and "To better understand..." in Section 3.3.

5. Figs 7,8, and 9, please include warnings of different scales on vertical axes **Response:** 

Thank you for your comment. Warnings have been added to the end of the captions of Figs. 7,8 and 9: "Note the differing vertical axes in different plots."

6. Fig 13: It would be worth pointing out again here why the SC models estimate more cooling in both MAM4/5 than the FC models.

#### **Response:**

We have revised the sentence to explain the difference in cooling between SC and FC in MAM4/5:

"The time variations of the global mean top-of-model net radiative flux shows that, during October-November of 1991, MAM4 experiments produce weaker peak values of around -1.5 W/m² while MAM5-PSA produces peak values of roughly -3.0 W/m² (Fig. 13). The weaker cooling in FC corresponds to the smaller stratospheric sulfate burden as seen in Fig. 4, while SC's stronger cooling also corresponds to its larger stratospheric sulfate burden."

7. L208: this text and the bullet list that follows refers to "processes", but the list includes both physical processes and the technical/computational process of renaming. This writing style can be confusing to readers who may not be familiar with the material. Please reorganize slightly (i.e., remove renaming from the list of physical processes) and be more explicit about the difference between the models' representation of physical processes versus technical steps. Response:

Thank you for pointing this out. To clarify this, we have removed renaming from the list of physical processes. We've also added the following sentences to avoid confusion for the readers:

"Each of these processes have their physical rules within MAM, and represent an actual physical process with the exception of renaming, which is a technical step in MAM."

"We note that renaming is a computational step in MAM, and does not correspond to a physical process".

8. L232: symbols used in the figure should be defined in the figure caption.

# **Response:**

We have restored the definition of Dg, Dg\_hi and Dg\_lo in the Fig. 1 caption:

"Dg, Dg\_hi, and Dg\_lo represent the initial values of the geometric dry mean diameter and its upper and lower limits, respectively."

9. L282: Is this justification for the smaller SO2 emission for Pinatubo compared to observations something that is established, or more speculated? There is also evidence that the stratospheric injection by Pinatubo is actually much smaller than the total observed emission, see Ukhov et al. (2023).

# **Response:**

We used emissions of 10 Tg following Mills et al. (2016). Ukhov et al. (2023) specifically mentions this assumption used in Mills et al. (2016) and agrees with it.

We have added a brief justification:

"Pinatubo eruptions are assumed to have occurred on June 15 1991, with 10 Tg of SO<sub>2</sub> evenly emitted between 18 and 20 km altitude. This corresponds to the mass detected in the stratosphere by the TIROS Operational Vertical Sounder and Total Ozone Mapping Spectrometer 7-9 days after the beginning of the eruption (Guo et al., 2004)."

10. L330: please point out here and in figure caption the figure displays tropical sulfate concentrations

# **Response:**

Thank you for the suggestion. We have pointed this out at the start of the paragraph and the caption.

- "Figure 3 depicts the vertical profiles of tropical sulfate concentrations"
- "Figure 3: Vertical profiles of tropical sulfate concentrations in the four experiments."
- 11. L366: "A smaller geometric standard deviation for stratospheric coarse mode in MAM5-PSA means that there are fewer super-coarse aerosols within the population. " Although counterintuitive (adding a larger mode leads to smaller size distribution), this mechanism does seem like a possibility. But, there is no evidence shown to support this claim. Also, later (1393) it is pointed out that in MAM5, certain growth processes are not operational in the stratospheric course mode. At line 399, both processes are referenced as reasons for the sulfate having a longer lifespan in MAM5-PSA than MAM4. Again, both reasons seem possible, but is their evidence to support the idea that they are important?

# **Response:**

Thank you for the comment. As of right now, no observational data of the size distribution of Pinatubo's sulfate aerosols exists on a global scale, with only local measurements as seen in Fig. 6. A reduction in the geometric standard deviation of dust aerosols leading to a longer lifespan in CESM and E3SM is also reported by Wu et al. (2020).

Similarly, observational data is also lacking for process understanding of stratospheric sulfates. Our goal for this study is to use our existing knowledge of aerosol processes to deduce what may occur within the stratosphere.

We have added a sentence to point this out below Fig. 1:

"The reduction in the geometric standard deviation (from 1.6 in the accumulation mode to 1.2 in the stratospheric coarse mode) is a method previously used in Wu et al. (2020) to lengthen the lifespan of aerosols."

12. L376: Yes, Baran and Foote have subtracted a baseline value away from the sulfate mass loadings they compute for Pinatubo. This does not mean their values are stratospheric burdens, and they do not use that language. This is because the tropospheric burden after the Pinatubo eruption can and likely is elevated compared to its background state.

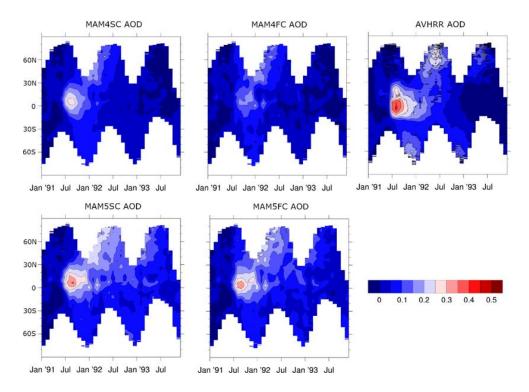
# **Response:**

Thank you for pointing out that the calculated sulfate burden by Baran and Foot (1994) also includes the troposphere. We have clarified this in the first paragraph of section 3.2:

"The derived sulfate burden from HIRS is calculated by taking the differences in the signal between the post-eruption period and an aerosol-free background, which means that it also includes tropospheric sulfate; however, tropospheric sulfate from the Pinatubo eruption has a lifespan of only several days and contributes only negligibly to the total burden past the first few days (Ukhov et al., 2023)."

13. Fig 10: Please update panel titles in figure to remove word "normalized" **Response:** 

Thank you for the reminder; we have revised the panel titles as suggested.



14. Fig 11 and related text: it is important to be clear that AVHRR is a total AOD anomaly, while GloSSAC reports a stratospheric AOD. Similar to the HIRS data, AVHRR's AOD likely includes some tropospheric anomaly component in the first months after the eruption.

#### **Response:**

We have clarified this within the discussion of Fig. 11.

"It is also worth noting that GloSSAC data includes only stratospheric AOD, and is missing tropospheric AOD anomalies that are included in AVHRR and the model results. Similar to the sulfate burden as discussed in section 3.2, the tropospheric AOD from the Pinatubo eruption is not significant compared to the stratospheric values."

15. L690: What results are you citing here, observations, models, etc? If models, what kind of models are they? The range of model results in that study appears to be 8-23 months, but that is for the decay of sulfate burden, not AOD. The two can be different as the size distribution changes with time.

# **Response:**

We are citing a multi-model study by Quaglia et al. (2023). To avoid confusion and mismatching comparisons we have removed this sentence.

16. L699ff: please be clear through this section whether you are comparing SW fluxes or SW+LW total fluxes. The discussion starts with reference to solar (i.e., SW) fluxes, but the comparison to results from Hansen et al. (2005) refers to their total (SW+LW) flux anomaly values, with a peak of about -3.0 W/m<sup>2</sup>.

#### **Response:**

Thank you for pointing this out. We have separated the discussion of solar fluxes and total (SW+LW) fluxes into two separate paragraphs to avoid confusion. The first paragraph contains only discussion of solar fluxes, while the second paragraph discusses the total flux.

We also made it clearer in the text by adding "(shortwave plus longwave)" flux anomaly at the beginning of the second paragraph.

#### **References:**

Baran, A. and Foot, J.: New application of the operational sounder HIRS in determining a climatology of sulphuric acid aerosol from the Pinatubo eruption, Journal of Geophysical Research: Atmospheres, 99, 25673-25679, 1994.

Emmons, L. K., Walters, S., Hess, P. G., Lamarque, J.-F., Pfister, G. G., Fillmore, D., Granier, C., Guenther, A., Kinnison, D., and Laepple, T.: Description and evaluation of the Model for Ozone and Related chemical Tracers, version 4 (MOZART-4), Geoscientific Model Development, 3, 43-67, 2010.

Guo, S., Bluth, G. J., Rose, W. I., Watson, I. M., and Prata, A.: Re-evaluation of SO2 release of the 15 June 1991 Pinatubo eruption using ultraviolet and infrared satellite sensors, Geochemistry, Geophysics, Geosystems, 5, 2004.

Mills, M. J., Schmidt, A., Easter, R., Solomon, S., Kinnison, D. E., Ghan, S. J., Neely III, R. R., Marsh, D. R., Conley, A., and Bardeen, C. G.: Global volcanic aerosol properties derived from emissions, 1990–2014, using CESM1 (WACCM), Journal of Geophysical Research: Atmospheres, 121, 2332-2348, 2016.

Quaglia, I., Timmreck, C., Niemeier, U., Visioni, D., Pitari, G., Brodowsky, C., Brühl, C., Dhomse, S. S., Franke, H., and Laakso, A.: Interactive stratospheric aerosol models' response to different amounts and altitudes of SO 2 injection during the 1991 Pinatubo eruption, Atmospheric Chemistry and Physics, 23, 921-948, 2023.

Ukhov, A., Stenchikov, G., Osipov, S., Krotkov, N., Gorkavyi, N., Li, C., Dubovik, O., and Lopatin, A.: Inverse modeling of the initial stage of the 1991 Pinatubo volcanic cloud accounting for radiative feedback of volcanic ash, Journal of Geophysical Research: Atmospheres, 128, e2022JD038446, 2023.

Wu, M., Liu, X., Yu, H., Wang, H., Shi, Y., Yang, K., Darmenov, A., Wu, C., Wang, Z., and Luo, T.: Understanding processes that control dust spatial distributions with global climate models and satellite observations, Atmospheric Chemistry and Physics, 20, 13835-13855, 2020.