

Replies to Reviewer 1

This new version of the manuscript is much improved compared to the earlier version. The focus is now on how well the model reproduces the available observations. I think this now is a good benchmark paper of what a state-of-the-art climate model can simulate under volcanic conditions. Some remarks and suggestions I included in the annotated manuscript

We thank the reviewer for their comments in the annotated manuscript they provided (numbered). We have corrected the text to address the comments and listed our changes to the text below (shown in Courier New font).

- 1) Page 3 line 22: Changed "by its particle size distribution" → "by the aerosol particle size distribution"
- 2) Page 5 line 1: Changed cite to citep for Bluth et al. (1992) reference
Also corrected typo "Spectrometeter" to "Spectrometer"
- 3) Page 5 line 14: Changed "than Northern Hemispheric mid-latitudes" to "than in Northern Hemisphere mid-latitudes".
- 4) Page 5 line 29: Added missing comma between "However" and "most".
- 5) Page 6 line 26: Changed "during initial few months but showed" to "during the first few months and showed"
- 6) Page 6 line 27: Changed "during later phase" to "during the later phase".
- 7) Page 8 line 18: Changed

We apply a fixed surface boundary condition of 275 \mu pptv .

To instead say:

For OCS, which has tropospheric lifetime of about two years (Montzka2007), we do not include an emissions source, but instead apply a fixed surface boundary condition of 275 \mu pptv .

page10 line 22-25: Changed

We therefore now calculate $p_{\text{H}_2\text{SO}_4}$ online in the model following (Kulmala1990). and calculate the condensation rate consistently with the difference between the vapour pressure and the gas phase partial pressure.

To instead say

We therefore now calculate $p_{\text{H}_2\text{SO}_4}$ online in the model following (Kulmala1990) and the condensation rates are calculated consistently using the difference between the vapour pressure and the gas phase partial pressure.

page11 line 1-2: (*Strange: makes the model dependent on the timestep. Why not a fixed time in seconds?*) We have added to the end of that sentence “, which corresponds to an e-folding timescale of 17 minutes”.

page12 line 13: deleted comma after “volcanically perturbed conditions”

page12 line 28: “coarse soluble” changed to “coarse soluble mode”.

page13 line 24: “B_Control10” changed to “D_NoPrimary10”

page14 line 24: Changed “data Arfeuille et al. (2013)” to instead say “data (Arfeuille et al., 2013)” and reference updated from ACPD to ACP version.

page15 line 3-4: “measurements of the stratospheric aerosol” changed to “measurements of stratospheric aerosol”

page15 line 16: “For example background aerosol” changed to “For example, background aerosol”

page16 line 09: “However, strength of the BD” changed to “However, the strength of the BD”

page17 line 14: Changed “runb” to “B_Control10” (missing back-slash in .tex)

page18 line 2-3: “1.7 Tg S, suggesting 8.3 of the emitted 10 Tg S” changed to “1.7 Tg S, indicating 8.3 of the emitted 10 Tg S”

page18 line 19: Changed “the timeseries of stratospheric aerosol sulphur burden” to “a timeseries of the stratospheric aerosol sulphur burden”

page22 line 1: Changed “Possible causes for these higher biases are” to “Possible causes for these biases are”

page23 line 6--7: *Speculation. You could check this. What would explain the initial formation of large particles? The higher concentrated volcanic plume?*

We have reworded this sentence from:

“This faster decay in the initial period is likely reflecting the shift in size distribution as larger particles are removed earlier in the period, causing slower sedimentation rates afterwards.”

To instead say

“The faster decay in the early phase may be due to the shift in size distribution to larger particles which occurred at this time. Faster sedimentation would remove larger particles during this initial period, leading to slower sedimentation rates later.”

page23 line 21--22: Changed “Similar agreement is seen at 25km however, during 1993 SAGE II measurements...” to “Similarly good agreement is also found at 25km. However, during 1993, SAGE II measurements...”

page24 line 4: Changed "the 1020 km extinction" to "the 1020 nm extinction"

page26 line 24: "to 30-40% lesser than those derived from" to "to 30-40% smaller than those derived from"

page28 line 17--19: *As mentioned earlier in the manuscript, the evaporation rate is already very fast (50% per condensation step). So, this can hardly be true.*

Changed the text:

"This may be indicating that the simple approach to particle evaporation in the model is too slow."

to instead say:

"This suggests that the simple approach to particle evaporation may need improving."

page30 line 11: Changed "agreement agreement" to "agreement".

page30 line 20-23: *Mostly irrelevant here. Maybe in discussion?*

We have moved the text below into the conclusions section where the discrepancies between the model and the OPC measurements are discussed:

"It is worth noting that in radiatively coupled simulations, we expect increased tropical upwelling would dilute the lower part of the plume, decreasing particle concentrations in the lowermost stratosphere."

page 33 line 11: changed "being" to "remaining" as suggested.

page 34 line 1-5: *Maybe good to repeat why you think that the difference between the runs A and B is so small. After all, you emit twice as much SO₂....*

We have added the following sentence to that paragraph

"Although twice as much SO₂ is injected in \runa than \runb, the nucleation rates in the two Runs are similar for July 1991. This could possibly be indicative of a depletion of oxidants which is limiting SO₂ oxidation, although an alternative explanation could be that there is much more surface area in the 20 Tg injection run to act as a condensation sink for sulphuric acid vapour."

We have also re-ordered the existing sentences in that paragraph to read better.

In the preceding paragraph we also changed "...note however that nucleation can be seen in the SH mid-latitudes..." to "...note however that nucleation can be seen in the middle stratosphere at SH mid-latitudes..."

page 34 line 17: Changed "during early phase" to "during the early phase".

page 35 line 14: Changed "growing to larger" to "growing to radii larger"

page 36 line 9-10: Re-worded that sentence to instead read:

“However, our control simulation, with a 20 Tg emission of SO_2 produces much too high a burden of aerosol sulphur compared to the HIRS measurements, whereas a 10 Tg injection is in good agreement. The 10 Tg run also compares better to the magnitude of the enhanced AOD distribution seen in SAGE-II and AVHRR, and captures well the transport to the Southern Hemisphere.”

page 36: line 28-29 Clarified the following sentence:

“In volcanically quiescent conditions, the model finds nucleation only occurs in polar spring, with particles at Laramie in the lower stratosphere originating from the tropical upper troposphere”

...by rewording it to

“The model finds that, in the volcanically quiescent stratosphere, nucleation occurs only during polar spring, with stratospheric particles at Laramie mostly originating from the tropical upper troposphere.”

Page 37, line 26: Reworded “However, alternative explanation could be too young age-of-air (too rapid STE) could be affecting” to instead say

“However, an alternative explanation could be that too young age-of-air and too rapid stratosphere-troposphere exchange is affecting the simulated stratospheric aerosol evolution.”

Replies to Reviewer 2

Review of the revised version of Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UKCA composition-climate model by Dhomse et al

The paper has substantially improved in comparison to the original version. The discussions are more elaborated and the figures are clearer. The additional sensitivity study with an initial injection of 10 Tg SO₂ is a very valuable addition. It is good to know that the authors could detect and solve a model bias.

We thank the reviewer for their comments (numbered and shown in italics) and reply to each showing any changes to the manuscript text in Courier New font.

1) *In general, I would like to recommend publication in ACP now. However, to broaden the perspective I would like to see a somehow critical assessment of the UKCA model results with other Pinatubo simulations. It would be good to mention in the final discussion and conclusion section at the end that other models capture observed quantities after the Pinatubo eruption with a twofold higher sulphur emission.*

We agree that the article could be improved by broadening the perspective of our results in relation to other Pinatubo studies. We have therefore added the following 2 paragraphs to the beginning of the Discussion section:

"In section \ref{ssec:globalburdenefoldingtimescale} we found that injecting 20 Tg \chem{SO_2} into the tropical stratosphere substantially overestimates the stratospheric aerosol sulphur burden, with a 10 Tg \chem{SO_2} injection in much better agreement with observations. Most previous modelling studies of the Pinatubo eruption have also tended to inject 20 Tg of \chem{SO_2}, and we show here that the high bias in our model is also found in other studies. \citet{Oman2006} and \citet{English2013} found peak stratospheric sulphuric acid aerosol burdens of 27 and 24 Tg respectively, translating to 36 and 32 Tg aerosol mass assuming 75\% weight sulphuric acid, similar to our 37 Tg peak value. \citet{Niemeier2009} injected 17 Tg of \chem{SO_2}, and their 30 Tg peak stratospheric aerosol burden also agrees with our simulation, accounting proportionally for the reduced sulphur source. We note also that \citet{Niemeier2009} and \citet{English2013} have presented the HIRS stratospheric aerosol burden timeseries from \citet{Baran1994} assuming the mass burden is for sulphuric acid, without accounting for the fraction of water content implicit in those values.

For our 10 Tg Pinatubo simulation, we found generally good agreement with observed AOD (section \ref{ssec:AODcomparison}), extinction (section \ref{ssec:extinctioncomparison}), and SAD (section \ref{ssec:SADcomparison}). Our 20 Tg simulation gives consistently too high aerosol optical depth in the tropics, mid-latitudes and polar regions, whereas in most of the previous studies mentioned above, reasonable agreement is found in peak AOD, despite the high bias in stratospheric aerosol burden. We note however that there is a considerable diversity in the injection height-range, latitudinal spread and duration of the volcanic source used in these different model experiments."

- 2) *A short paragraph about the necessity of a global aerosol model inter comparison for the Pinatubo episode as planned in the SSIRC model inter comparison study and lead by one of the coauthors would in my opinions be a very valuable addition.*

We agree that our findings further motivate the need for a model intercomparison for the Pinatubo episode have added the following sentence at the end of the conclusions

"The findings highlight the need for a co-ordinated set of experiments to intercompare and evaluate current global stratospheric aerosol models against the wide set of observations available through the Pinatubo-perturbed period."

Minor comments

- 3) *Page 17, line 23 -27 The role of three times higher stratospheric aerosol background load in the UKCA model as one of the possible reasons for the high volcanic aerosol load remains unclear*

We agree that the background stratospheric aerosol sulphur loading in these UKCA simulations is higher than in other studies and have pointed that out already in that particular excerpt of text (lines 23-27). However, as we have clarified in the new paragraph added to the Discussion (see reply to point 1 above), we find that although the peak stratospheric aerosol burden in our 20 Tg Pinatubo simulation is higher than that derived from the HIRS measurements, it is consistent with burden values found in previous Pinatubo model studies. So although we agree that our peak Pinatubo stratospheric aerosol sulphur loading is higher than the HIRS measurements, it is not higher than found in other model studies. We have therefore not commented further on the different stratospheric sulphur burden in background conditions.

- 4) *Page 24 line 10 "Before the eruption (May 1991), the 10 model captures the observed SAD very well with a hemispherical symmetric distribution in the lower stratosphere in the range 0.5 to 2 $\mu\text{m}^2\text{cm}^{-3}$ "*

I would not write that the model captures the observations well, because we see in the satellite data an almost clean UTLS region while in the model the SAD is throughout the UTLS $> 1 \mu\text{m}^2 \text{cm}^{-3}$. This has certainly consequences for the particle growth.

We agree that we may have over-stated how well the model SAD compared to the satellite-derived datasets in background conditions. We have revised the text from

"Before the eruption (May~1991), the model captures the observed SAD very well with a~hemispherically symmetric distribution in the lower stratosphere in the range 0.5 to 2\, \unit{\mu\text{m}^2}\, \text{cm}^{\{-3\}}"

To instead say

"Before the eruption (May~1991), the model captures the global SAD distribution reasonably well compared to the SAGE-derived datasets, although model values are higher in the upper troposphere and lower stratosphere region."

5) *Figure / color bar missing*

Figures 4, 8 Please put the colorbar below the four panels

Done.