Author response to the Referees' comments on "Size-Resolved Stratospheric Aerosol Distributions after Pinatubo Derived from a Coupled Aerosol-Chemistry-Climate Model"

We thank both Referees for their thorough evaluation of the manuscript, which helped us to improve it and to formulate a clearer message. Following the reviewer's comments, we found several bugs in our observational data post-processing and corrected the corresponding parts in the text. We've also improved our representation of the sections 3.3 (temperature) and 3.5 (ozone) and performed a new experiment with the nudged dynamics to have an additional insight into our ozone results.

Following requests of the Executive editor, we included the name and version of our model to the new title: "Stratospheric aerosol evolution after Pinatubo simulated with a coupled size-resolved aerosol-chemistry climate model SOCOL-AERv1.0". We also uploaded the model code (https://doi.org/10.5281/zenodo.1245196) and results of our simulations (https://doi.org/10.5281/zenodo.1245202).

Referees' comments are repeated below in blue and our response follows in black.

Response to comments by Referee 1 (M. Toohey)

This paper describes a series of experiments of stratospheric aerosol following the 1991 Pinatubo eruption with the SOCOL-AER model. The model results are compared extensively with available observations, and the paper serves therefore as a valuable model validation exercise, showing that in general the model reproduces many of the observed aerosol properties rather well. Some differences between model and observations are noted, and sensitivity experiments are used to gauge the importance of a few uncertainties related to the eruption itself and structural uncertainties in model parameterizations.

The results are generally quite impressive, however, for a paper in GMD, there is very little description of the model included here. There is reference given to Sheng et al. 2015, and probably one can find more description there, but some more details should be included in this paper. For instance, in the Results it is mentioned that ECHAM5 is the core GCM of SOCOL-AER–this should be included in the model description section. Also, the model description should include some text relevant to each of the processes that are altered in the sensitivity studies, e.g., the standard sedimentation and coagulation parameterizations used in SOCOL-AER should be described.

We agree that description of the model and some methods, also mentioned later, were a bit vague. We now added more information about the model and methods.

More details on the coupling between aerosol and radiation would be quite useful, e.g., exactly what optical properties are required by the radiation code, and what assumptions and simplifications go into the Mie theory calculations (refractive indices, etc.).

SOCOL-AER uses 6 band shortwave (Cagnazzo et al., 2007) and 16 band longwave (Mlawer et al., 1997) radiation schemes. The required extinction coefficients, single scattering albedos, and asymmetry factors for each of the 22 wavelength bands are calculated for given weight percent and temperature using refraction indices from Biermann et al. (2000). Added to the text.

In the conclusions, it is stated that the "main modelling deficiency found" is the 1-2 K larger lower stratospheric warming compared to reanalyses. This distinction is rather subjective, and I'm surprised there wasn't also mention of the fact the model results show differences compared to observations in aerosol number density of orders of magnitude in the 25-30 km range.

Conclusions are corrected. We now mention the problems in reproducing sizes above 25 km as well as the ozone issues (see below).

P1, I16: "Anthropogenic ... sulfur emissions" sounds like tropospheric aerosols from surface sulfur emissions, but probably you are referring to geoengineering through stratospheric sulfur injection – this sentence could be improved to make the message clearer.

Rephrased as "prerequisite for improved understanding of solar geoengineering effects from sulfur injections to the stratosphere"

P1, l21: Important to be clear that -3 W/mˆ2 is the peak, or maximum radiative forcing.

Referred as "up to"

P2, I11: Distinction between models using prescribed and prognostic aerosols is not really an "approximation", a better word could be found here.

We have removed this confusing part and added later "can be **mainly** discretized into two groups".

P2, l15: Models using prescribed volcanic forcing are not strictly dependent on either observations or prognostic aerosol models–simple reconstruction methods have been used for eruptions before the satellite era (e.g., Sato et al, 1993, Gao et al., 2008, Ammann et al., 2003, Toohey and Sigl, 2017).

We have mentioned this in the text now with a reference to "e.g. Toohey and Sigl (2017)"

P2, I16: What "climate feedbacks" are specifically meant here? It's clear that prescribing aerosols does not allow for feedbacks from atmospheric dynamics onto the aerosol transport and distribution, but the relevance of this on climate seems likely to be small – "climate feedbacks" usually refer to those feedbacks between components of the climate system like atmosphere, ocean, cryosphere, etc.

Here we referred to the feedbacks between aerosols and stratospheric processes that are described later in the penultimate paragraph of the introduction and what the whole paper is about. Namely, stratospheric aerosol lifetime is sensitive to the QBO phase, background temperature important for microphysics, Brewer-Dobson Circulation strength, availability of hydroxyl radicals, etc., which is all modified when a large eruption occurs. Of course, effects of these separate feedbacks on final AOD, (i.e. forcing for atmosphere, ocean, cryosphere) are small compared to the overall effect, but they are certainly not negligible and can be enhanced under certain circumstances and larger eruptions. For example, recent Tambora study by Marshall et al. (2018) demonstrated that MAECHAM5-HAM model, which didn't include interactive chemistry and used prescribed OH radical concentration, provided much shorter aerosol lifetime and therefore shorter AOD increase than other models, which then would also result in additional changes between atmosphere, ocean, and cryosphere.

But to be clearer for readers, we modified the sentence as "*They have only limited ability to reproduce the climate response to volcanic eruptions, as the aerosols are prescribed and therefore the feedbacks between aerosols and the stratosphere are completely missed resulting in biased aerosol radiative forcing depending on concrete circumstances.*"

P2, I19ff: "size-bin resolving" doesn't sound right to me, "size-resolving" is clear.

Corrected

P2, l28: remove "problem"

Done

P2, l31: "and is therefore often..."

Corrected

P3, l4: The model results hint at *differences* in how the models treat aerosol processes –how these differences relate to *uncertainties* in processes is another question.

Rephrased as "*This hints at large differences in how models treat important microphysical and transport processes and significantly increases the uncertainty of the overall aerosol layer understanding.*"

P3, l5: VolMIP is currently ongoing, please replace "A recent" with "An ongoing" or something like this. I'd recommend also removing the "However" in line 6, this seems to shine a light of disappointment on the development of the VolMIP activity!

Accepted both suggestions.

P3, l19: describing

Corrected

P4, l11: prolongs

Corrected

P4, l17: the quasi-biennial oscillation

Done

P4, l28: Are SSTs and SIC climatological values or transient? From what data are they based?

Clarified as "*Monthly mean sea surface temperatures (SSTs) and sea ice coverage (SIC) are prescribed on a transient basis by the Hadley Centre Sea Ice and SST data (Rayner et al., 2003).*"

P4, l30: Guo et al., 2004 estimate 18 or 19 Tg SO2 injection by Pinatubo. The 14 Tg SO2 injection used in this study is within the 1-sigma uncertainty of the estimates from Guo et al. (2004), but some explanation for using a value less than the central estimate should be included here. Similarly, the vertical distribution of the injection is different than that estimated from the satellite observations, which suggest a peak at ~25 km. Some words should be included here to describe why a different vertical distribution was used ("optimized according to Sheng et al, 2015a" doesn't really help the reader).

We have rephrased the whole paragraph making it clearer why we use the estimate of Sheng et al., (2015). In short, they used the same microphysical module to run 300 experiments spanning the observational uncertainties and derived an optimised estimate fitting this model.

P5, l5ff: experiment names like NO_QBO, NO_RAD would be more intuitive.

Corrected.

P5, l10: "We consider two ... experiments concerning the coagulation efficiency" – but only the COAG experiment is described hereafter. The UPWIND experiment seems not directly related to coagulation efficiency.

Corrected. Initially we had two experiments testing coagulation, but decided to stay with one. So, this was an artefact of previous draft versions.

P5, l23: The high latitude of Cerro Hudson may play a role, but also likely the much smaller SO2 injection amount (compared to Pinatubo) and the lower injection height. It would be important to list the injection height used in the simulations here, and mention this as potentially important to its impact.

We have added that "*75% of mass injected between 16 and 18 km*". However, the main factor for relatively low effect of Hudson on the aerosol layer is rather its high-latitude location than a lower plume height, since the tropopause is also lower at 45°N. Of course, it is not the location itself but the associated details of the stratospheric circulation.

P5, l26: teragram defined previously

Corrected

P6, l4: The recent paper from Thomason et al. (2017) is a much better reference for the SAGE_3lambda data. Fig 1: the uncertainty spread in the HIRS data are relatively small, what uncertainties are included in this estimate?

New reference included. We found a bug in our representation of the HIRS uncertainties. The intention was to use ±10% as was already mentioned in the text, we updated the figure now. Problem with HIRS burden data is that it is very old and even doesn't exist in a numerical format. Even less is known about its "real" uncertainty under different aerosol loading. We've added some additional discussion about this to conclusions.

P8, l3: Some description of how the model was sampled is needed here: at the latitude of Laramie I assume, but also the longitude, or zonal mean? Was the model sampled on the days of the balloon flights, or are monthly means used?

Model was sampled as a mean of all gridpoints fitting into a region of ~±5 degrees latitude and longitude around Laramie. We used monthly means for the model. For OPC we also tried to use months with at least two soundings to have a representative of a monthly mean. We have added this additional information to the text.

P8, l4: what types of uncertainties are included in the OPC error bars?

Measurement uncertainty of aerosol surface area and volume as reported in Deshler et al. (2003).

P8, l6: improves

Corrected

P9, l5-6: this statement is arguable for the August comparison

Even though only about a half of SO_2 is converted to H_2SO_4 up to this date, it can also be called a stage of the volcanic cloud evolution.

P9, l7: one order of magnitude seems an optimistic generalization: for May at 28 km the difference looks closer to 3 orders of magnitude.

Corrected to be more precise

P9, l28ff: I disagree with this summary, the COAG experiment clearly shows a different behavior than the other experiments (e.g., Fig 3) and there are strong differences between the model results and the OPC data, suggesting the model has too many, too small particles, especially at heights above 22 km.

The intention of this summary was to compare Fig. 2a and Fig3, i.e behaviour of the model with respect to OPC and to SAGE II. We have rephrased the whole paragraph to make it clearer and also added a note about problems above 25 km.

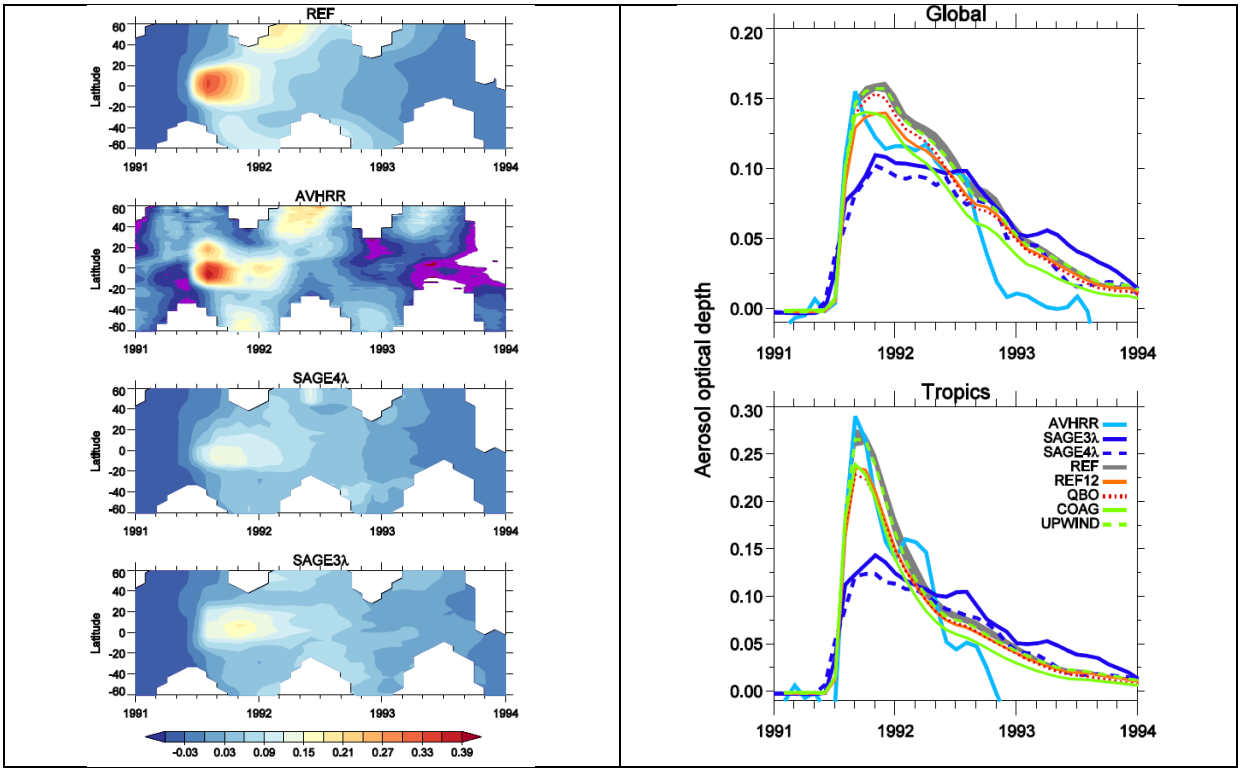
Fig 4 caption: this reads as if all the panels show AOD over oceans, but I assume this is only for AVHRR.

For the model we also used values over oceans, but not for SAGE since these data are zonal means. We corrected the text accordingly.

Fig 5: I was surprised at first to see that the global mean AOD for the SAGE data sets is much larger than in plots from other sources (see, e.g., Fig. 1 of Toohey et al., 2016). But this seems to be related to the inclusion of upper tropospheric volcanic aerosol in the data shown here, since the extinction is integrated over a much larger vertical extent here rather than only above the tropopause. This fact might be emphasized more in the discussion here, (perhaps the plot y-label should be clearer as "AOD anomaly"?) and also the details of the integration more clearly stated: were all altitudes used (i.e., right down to the 5km lower limit of the SAGE data sets)? Also, details of how the "background values" were determined for each data set should be explained.

Fig 5: the spike in the AVHRR global mean AOD data in NH summer of 1993 looks suspicious: it doesn't show up in Fig. 3 of Mills et al., (2016) and no obvious source for the spike can be discerned from the zonal mean values in Fig 4.

Integration was performed over all altitudes down to 5 km, because the contribution of tropospheric aerosols is then supposed to be removed when we subtracted background values. For the background values, similarly to Mills et al. (2016), we used 1995 annual average values from observations and calculations. However, your comments pushed us to carefully recheck the plotting scripts and we've identified two bugs. First of all, we excluded the tropospheric extinctions by performing integration only till the tropopause (defined from the model). This led to slightly lower values similar to those in Toohey et al., 2016. Second, calculation of the zonal means in Fig. 5 was performed over non-masked regions, which were defined as -999., and there was a check not for this exact value but for all negative values which then led to an exclusion of low negative values that were representatives of regional low AOD that appeared when we subtracted the background. New zonal means are also very similar to those presented by Mills et al. (2016). We have corrected the AOD section correspondingly. Below you can see the updated figures. Main message remained the same, namely, the model is closer to AVHRR till mid-1992 and to SAGE-4l later.



P11, l5: Importantly, this procedure doesn't remove (upper) tropospheric aerosols from the Pinatubo eruption itself!

Corrected as "background tropospheric aerosols"

P12, l5: "perfectly" is a strong word, and doesn't quite fit here, e.g., there does seem to be discrepancy in the meridional position of the initial tropical AOD peak.

Corrected as "*nicely*"

P12, l25: some words needed here on how the annual cycle and QBO cycle were subtracted – over what period was the annual cycle determined? How was the QBO defined?

Fig 6: Why is the QBO experiment not shown here? This experiment might shine light on how much of the temperature anomalies shown in Fig 6 are related to the aerosol, and how much to the QBO nudging, and would seem therefore quite important to include in the discussion.

We decided to change our representation of the temperature signal in Fig6. Now we subtract only the annual cycle averaged over 1986-2013 for reanalyses and over 1991-1995 of the noRADnoQBO experiment for all other model experiments (specified in the text now). We additionally plotted results from the noRAD experiment that mainly represent the temperature anomaly in the absence of Pinatubo, which is mostly due to QBO. So that anything between noRAD and other lines can be attributed to volcanic effects. One can also see that, once most of the aerosol mass is removed from the stratosphere in mid-1993, noRad result goes mostly in line with other experiments and reanalyses. Effects of all individual experiments remained the same in this representation. All corresponding text is corrected.

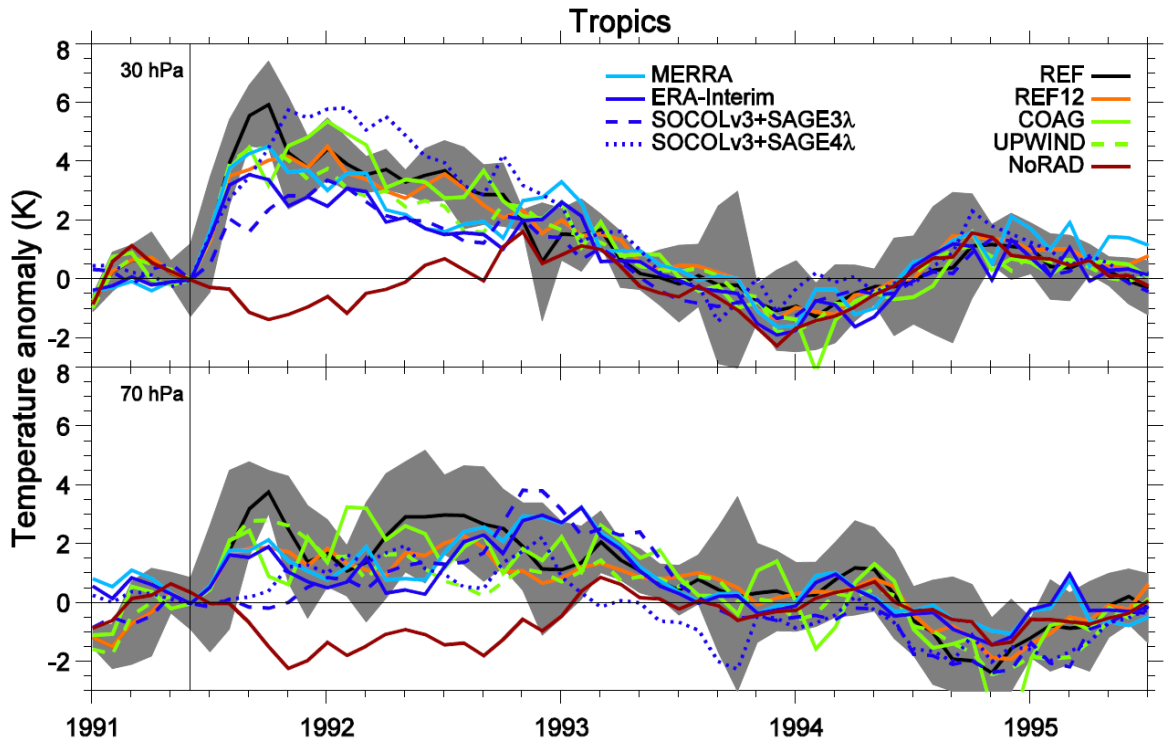
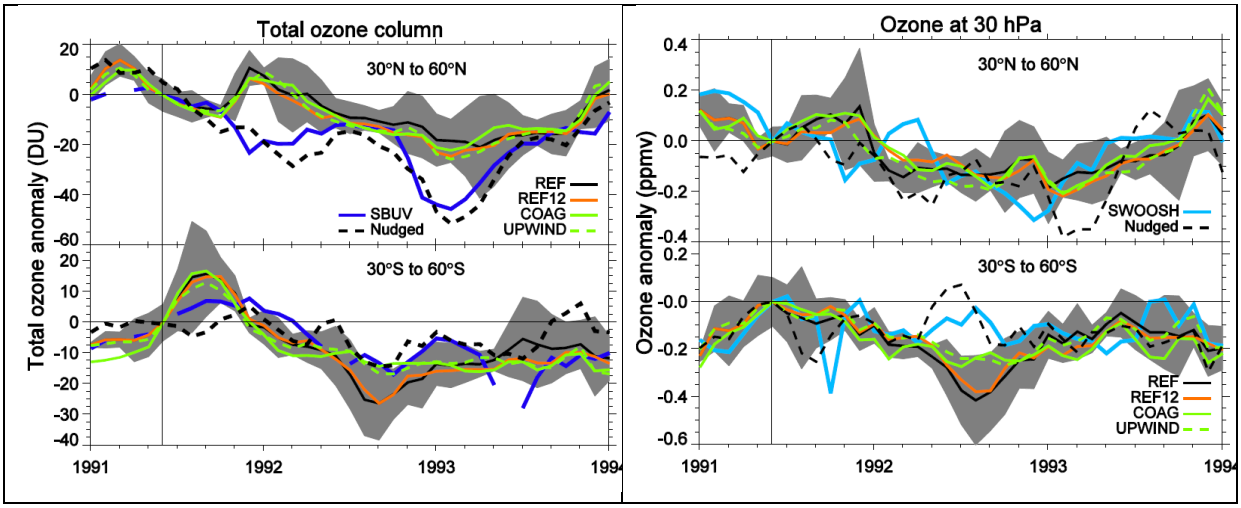


Fig 8: There is a strong QBO signal in tropical ozone–how much of the anomalies shown in this figure are simply a result of the QBO nudging, and how much are due to the volcanic aerosol? Including the QBO simulations in this plot would seem valuable to answer this question.

P15, l9: Actually, most studies have shown that post-volcanic changes in ozone maximize in the midlatitudes (e.g., Randel et al., 1995, Solomon et al., 1998).

We have also changed our representation of the ozone results in order to show midlatitudes and to address the modelled ozone signal problem discussed by Aquila et al. (2012) and Dhomse et al. (2015), i.e. hemispheric asymmetry of the midlatitude ozone responses. New Fig. 8 and additional Fig.9 now show both hemispheres midlatitude ozone changes as a total column and a mixing ratio at 30 hPa, respectively. Ozone anomalies are obtained by subtracting monthly means for 1991-1995. From new figures it can be seen that SOCOL-AER does produce the hemispheric asymmetry right after the eruption in 1991 but overestimates the ozone increase in the southern hemisphere and also has problems in catching other details of the ozone changes. In the northern hemisphere, SOCOL-AER produces an increase in ozone in late 1991 that is not seen in observations (both SWOOSH and SBUV). Interestingly, for the total column the model underestimates ozone decrease in the NH and overestimates it in the SH, while at 30 hPa it is the other way around.

We performed an additional experiment with the nudged dynamics using Era-Interim reanalysis as in Ball et al. (2016). The nudged experiment has a much better agreement with SBUV and SWOOSH reproducing most of the observed features. Given that our tropical temperature signal is relatively well captured (especially in the REF12 experiment), this suggests that further we should look closer on our representation of the dynamical response to volcanic eruptions and the interhemispheric transport. The ozone section and conclusions are corrected.



P15, l3: This was shown much earlier by Tie and Brasseur, 1995.

Corrected

P15, l5: citation for volcanic chlorine contribution would be good.

We have cited a recent study by Klobas et al. (2017), who covers this topic in detail.

P16, l17: roles

Corrected

P16, l19: I think “maintaining the tropical stratospheric aerosol reservoir” is not quite the right message here, these processes are important for more clearly definable and important properties, like the global burden evolution and global AOD.

We have added “*significantly affecting volcanic aerosols lifetime*” to this sentence

Pg 17, l19: This sentence could well cite the recent paper from Timmreck et al. (2018).

Done

Response to comments by Referee 2 (Anonymous)

Here authors use SOCOL-AER model to simulate stratospheric aerosol properties following Mt Pinatubo eruption in June 1991. Authors also use observational data sets to evaluate simulated model response. As expected model sensitivity simulations highlight importance of QBO phase in controlling stratospheric aerosol lifetime and well as spatial distribution. They also find that inclusion of van de Waals forces improves effective radii comparison against observations but it deteriorates aerosol lifetime comparison. Overall, this is well written manuscript and I will like to recommend it for a publication, if authors can address some of the minor comments listed below

Minor Comments:

1. With 14 Tg and 10 Tg SO2 injections, SOCOL simulates higher AOD and relatively more warming in the tropical lower stratosphere, so it would be better if you could (only if it is possible) to add an additional simulation with 10 or 8 Tg SO2 injection? I agree that you want to follow previous studies, but it will be good idea to show if AOD/ lower stratospheric warming comparison improves if you reduce the SO2 amount.

We use 14 Tg and 12 Tg SO_2 and these two estimates already cover mostly all cases, i.e. either one or another agrees well with observations. Another experiment line would complicate figures that are already quite busy with lines.

2. Page 2 Line 21-31: I think you should rewrite this paragraph as both sectional and modal models have their own strengths and weaknesses. And almost all the modelling studies point out that it's not only aerosol microphysics but also input parameters (e.g. SO2 injection amount, plume height) play key role in determining evolution of stratospheric aerosol evolution following any major volcanic eruption, hence having sectional scheme does not guarantee that modelled aerosol evolution would be accurate.

We have added that "*Both modal and size-resolving schemes have their benefits and problems. Modal aerosol schemes prescribe some basic parameters characterizing size distribution (e.g., size distribution function) and therefore have low computational demand. Size-resolving schemes simulate an evolution of the size distribution and can better describe gravitational sedimentation, which crucially affects the stratospheric aerosol lifetime*"

And clarified later that *"…fine resolution of aerosol sizes is not a universal solution and performance of any model, even with highly resolved aerosol sizes, depends on representation of relevant chemical, microphysical and radiative processes, large-scale transport and gravitational sedimentation, as well as their interactions.*"

Technical corrections:

1. Title : What about " Simulating evolution of stratospheric aerosol after Pinatubo eruption using coupled aerosol-chemistry climate model (SOCOL).

We thank the reviewer for the suggestion and derived this: "Stratospheric aerosol evolution after Pinatubo simulated with a coupled size-resolved aerosol-chemistry climate model SOCOL-AERv1.0"

2. Abstract : line 4: 40 size bins with radii spanning from

Corrected

3. Also next statement "Radiative forcing is computed .. " should not be in abstract

We replaced this by "*The aerosol module is coupled to the radiative modules and includes comprehensive…*"

4. Line 7: We performed series of simulations (delete "'a")

Corrected

Page 2 : Line 7: considered as main forcing constraint

Corrected

Line 8: and better understanding about the evolution of stratospheric aerosol layer is crucial

Here the message was that it is important to have a reliable information about the aerosol layer if one wants to study related atmospheric feedbacks.

Page 3: Line 19 : describing Table 1: Experiment QBO should be "noQBO" and nextone should be "noQBOnoRAD"

Corrected throughout the manuscript

Page 6 Line 4: composites is presented in Revell et al., 2017 Page 15 :

We now refer to a recent paper by Thomason et al. (2018).

Line 9: wrong. Ozone response in more pronounced at mid-latitudes Do you get large hemispheric differences in ozone losses (e.g Poberaj et al, 2011, JAS< Aquila et al., 2013, JAS, Dhomse et al, 2015, GRL)

We now address this question of hemispheric asymmetry, please see our response above (with figures). In short, the model does produce this hemispheric asymmetry in 1991, but overestimates ozone increase in the southern hemisphere. The model also has problems compared to observations, more pronounced in the northern hemisphere, which can be corrected if the nudged dynamics used, as we show in an additionally performed experiment.

Page 16: Line 17 : spelling “explore the role of QBO”

Corrected

References:

Ball, W.T., Haigh, J.D., Rozanov, E.V., Kuchar, A., Sukhodolov, T., Tummon, F., Shapiro, A.V., Schmutz, W., 2016a. High solar cycle spectral variations inconsistent with stratospheric ozone observations, Nat. Geoscience, doi:http://dx.doi.org/10.1038/ngeo2640M3.

Biermann, U. M., B. P. Luo, and T. Peter (2000), Absorption spectra and optical constants of binary and ternary solutions of H2SO4, HNO3, and H2O in the mid infrared at atmospheric temperatures, J. Phys. Chem. A, 104(4), 783–793, doi:10.1021/jp992349i.

Cagnazzo, C., E. Manzini, M. A. Giorgetta, P. M. D. F. Forster, and J. J. Morcrette (2007), Impact of an improved shortwave radiation scheme in the MAECHAM5 general circulation model, Atmos. Chem. Phys., 7(10), 2503–2515, doi:10.5194/acp-7-2503-2007.

Deshler, T., Hervig, M. E., Hofmann, D. J., Rosen, J. M., and Liley, J. B.: Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41°N), using balloon-borne instruments, Journal of Geophysical Research: Atmospheres, 108, n/a–n/a, doi:10.1029/2002JD002514, http://onlinelibrary.wiley.com/doi/10.1029/2002JD002514/abstract, 2003.

Klobas, J. E., D. M. Wilmouth, D. K. Weisenstein, J. G. Anderson, and R. J. Salawitch (2017), Ozone depletion following future volcanic eruptions, Geophys. Res. Lett., 44, 7490–7499, doi: 10.1002/2017GL073972.

Marshall, L., Schmidt, A., Toohey, M., Carslaw, K. S., Mann, G. W., Sigl, M., Khodri, M., Timmreck, C., Zanchettin, D., Ball, W. T., Bekki, S., Brooke, J. S. A., Dhomse, S., Johnson, C., Lamarque, J.-F., LeGrande, A. N., Mills, M. J., Niemeier, U., Pope, J. O., Poulain, V., Robock, A., Rozanov, E., Stenke, A., Sukhodolov, T., Tilmes, S., Tsigaridis, K., and Tummon, F.: Multi-model comparison of the volcanic sulfate deposition from the 1815 eruption of Mt. Tambora, Atmospheric Chemistry and Physics, 18, 2307–2328, doi:10.5194/acp-18- 10 2307-2018, https://www.atmos-chem-phys.net/18/2307/2018/, 2018.

Mills, M. J., Schmidt, A., Easter, R., Solomon, S., Kinnison, D. E., Ghan, S. J., Neely, R. R., Marsh, D. R., Conley, A., Bardeen, C. G., and Gettelman, A.: Global volcanic aerosol properties derived from emissions, 1990-2014, using CESM1(WACCM), Journal of Geophysical Research (Atmospheres), 121, 2332–2348, doi:10.1002/2015JD024290, 2016.

Mlawer, E. J., S. J. Taubman, P. D. Brown, M. J. Iacono, and S. A. Clough (1997), Radiative transfer for inhomogeneous atmospheres: RRTM, a validated correlated-k model for the longwave, J. Geophys. Res., 102(D14), 16,663–16,682, doi:10.1029/97JD00237.

Rayner, N. A., D. E. Parker, E. B. Horton, C. K. Folland, L. V. Alexander, D. P. Rowell, E. C. Kent, and A. Kaplan (2003), Global analyses of sea surface temperature, sea ice, and night marine air temperature since the late nineteenth century, J. Geophys. Res., 108(D14), 4407, doi:10.1029/2002JD002670.

Sheng, J.-X., Weisenstein, D. K., Luo, B.-P., Rozanov, E., Arfeuille, F., and Peter, T.: A perturbed parameter model ensemble to investigate Mt. Pinatubo's 1991 initial sulfur mass emission, Atmospheric Chemistry & Physics, 15, 11 501–11 512, doi:10.5194/acp-15-11501-2015, 2015a.

Thomason, L. W., Ernest, N., Millán, L., Rieger, L., Bourassa, A., Vernier, J.-P., Manney, G., Luo, B., Arfeuille, F., and Peter, T.: A global space-based stratospheric aerosol 5 climatology: 1979–2016, Earth System Science Data, 10, 469–492, doi:10.5194/essd-10-469-2018, https://www.earth-syst-sci-data.net/10/469/2018/, 2018.