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 methods were a bit vague. We added more information about the model and methods, although we still leave a detailed description behind a reference to Sheng et al. (2015), as that was a main model description paper. Links to sedimentation and coagulation methods is given in the text.

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 actual aerosol composition 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, l16: "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, l11: 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** separated 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, l16: 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, 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 in the 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, l19ff: "size-bin resolving" doesn't sound right to me, "size-resolving" is clear.

Corrected throughout the text

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 transient sea surface temperatures (SSTs) and sea ice coverage (SIC) are prescribed from 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 explaining 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 the 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. This uncertainty estimate is taken from the paper of Baran and Foot (1994): "Systematic errors through neglect of the aerosol emission at 12.5 μm and uncertainties in properties of the aerosol amount to about 10%."

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 grid points 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. Now, we also show all discrete OPC measurements within chosen months instead of means.

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 added a note about problems above 25 km.

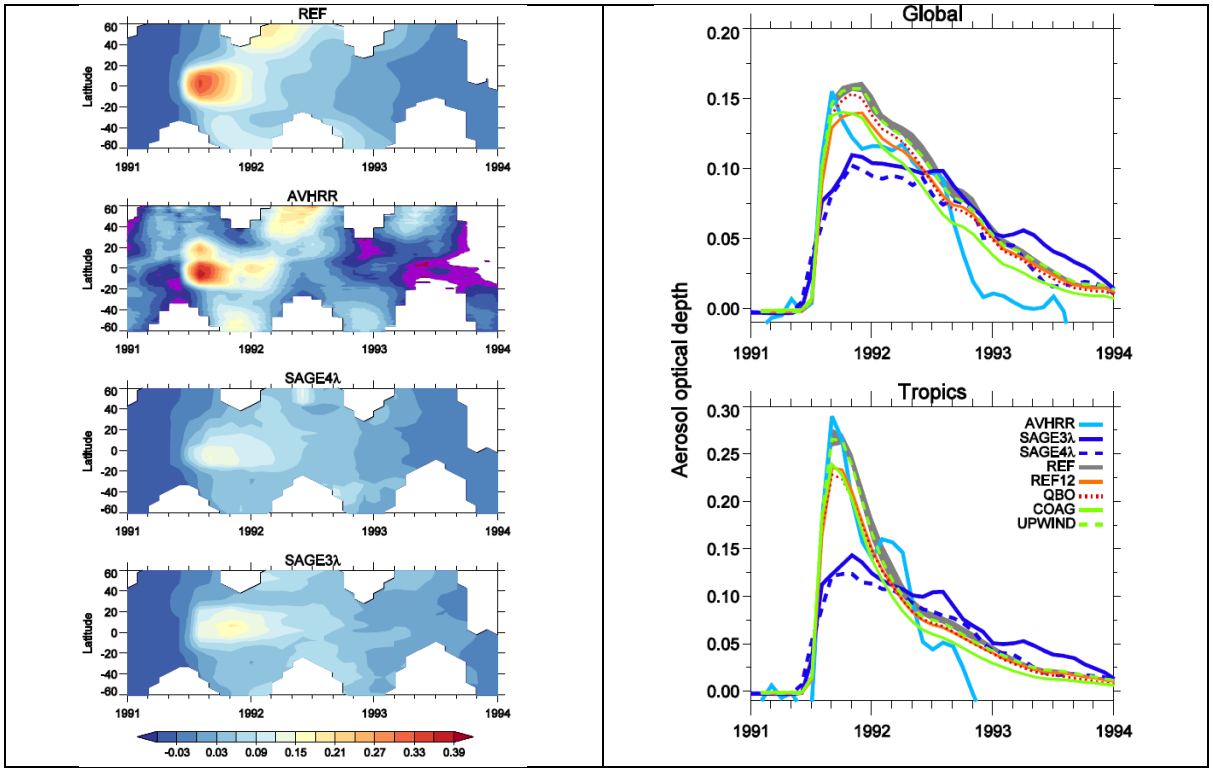
Fig 4 caption: this reads as if all the panels show AOD over oceans, but I assume this is only for AVHRR.

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 close 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!

This part is removed

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.

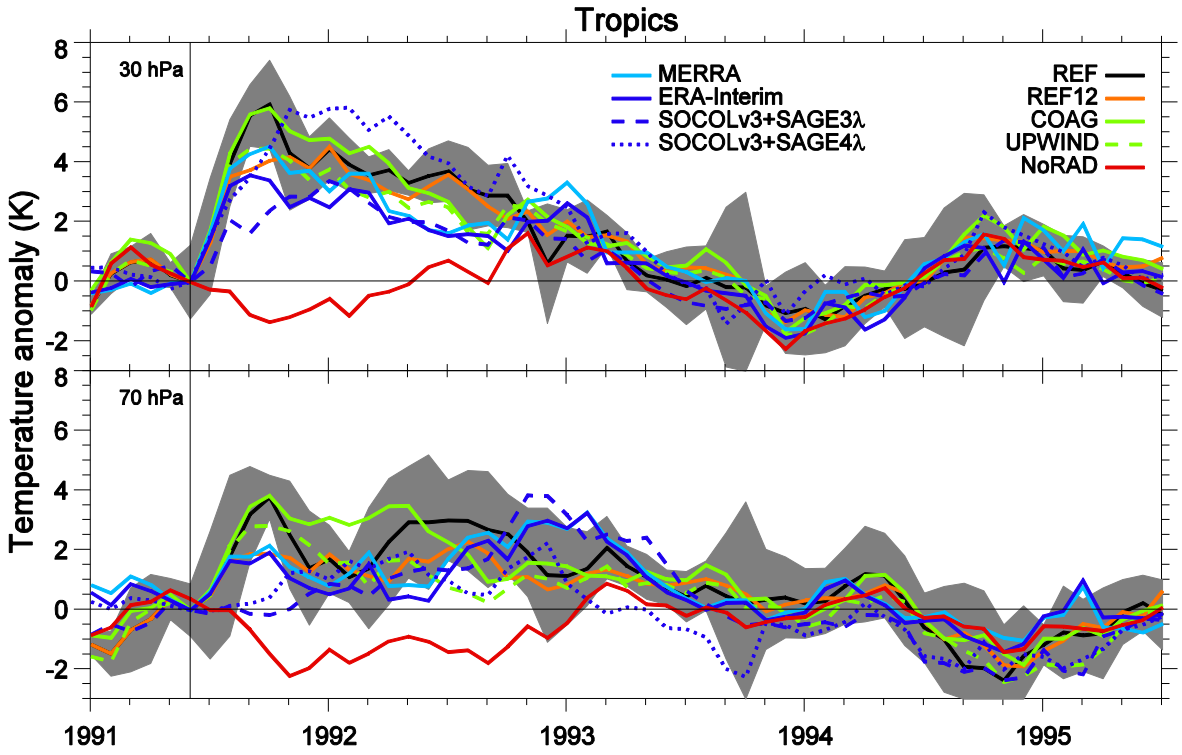
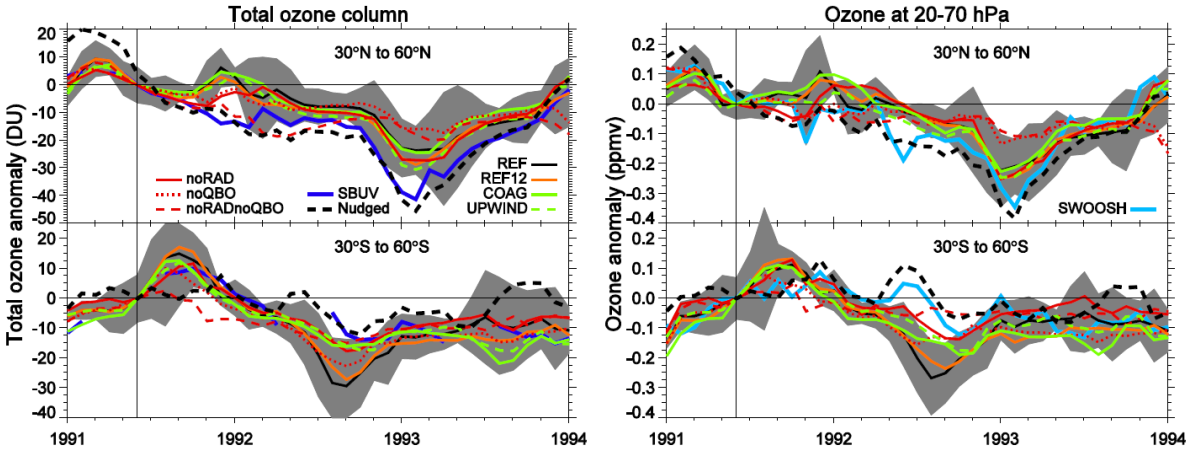


Fig 8: There is a strong QBO signal in tropical ozone–how much of the anomalies shown in this figure are simply a result of the QBO nudging, and how much are due to the volcanic aerosol? Including the QBO simulations in this plot would seem valuable to answer this question.

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 now added noQBO, noRAD, and noQBOnoRAD results to the analysis and changed our representation of the ozone results in order to show middle latitudes 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 middle latitude ozone responses. New Fig. 8 now shows both hemispheres middle latitude ozone changes as a total column and a mixing ratio at 20-70 hPa, respectively. Ozone anomalies are obtained by subtracting monthly means for 1991-1995. From the new figures, it can be seen that SOCOL-AER does produce the hemispheric asymmetry right after the eruption in 1991 but 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).

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 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 to the reviewer #1 above (with figures). In short, the model does produce this hemispheric asymmetry in 1991, but in both hemispheres, while there is an increase only in the southern hemisphere in observations. The model also has other problems compared to observations, 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

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~~Size-Resolved~~ Stratospheric ~~Aerosol Distributions~~ aerosol evolution after Pinatubo ~~Derived from~~ simulated with a ~~Coupled Aerosol-Chemistry-Climate Model~~coupled size-resolved aerosol-chemistry climate model SOCOL-AERv1.0

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Abstract. We evaluate how the coupled aerosol-chemistry-climate model ~~SOCOL-AER~~ SOCOL-AERv1.0 represents the influence of the 1991 eruption of Mt. Pinatubo on stratospheric aerosol ~~loading, aerosol microphysical processes, radiative effects, and atmospheric chemistry~~properties and atmospheric state. The aerosol module is coupled to the radiative and chemical modules and includes comprehensive sulfur chemistry and microphysics, in which the particle size distribution is represented by 40 size bins ~~spanning radii~~ with radii spanning from 0.39 nm to 3.2 μm. ~~Radiative forcing is computed online using aerosol optical properties calculated according to Mie theory.~~ SOCOL-AER simulations are compared with satellite and in situ measurements of aerosol parameters, temperature reanalyses, and ozone observations. In addition to the reference model configuration, we performed ~~a~~ series of sensitivity experiments looking at different processes affecting the aerosol layer. An accurate sedimentation scheme is found to be essential to prevent particles diffusing too rapidly to high and low altitudes. The aerosol radiative feedback and the use of a nudged quasi-biennial oscillation help to keep aerosol in the tropics and significantly affect the evolution of the stratospheric aerosol burden, which improves the agreement with observed aerosol mass distributions. ~~Changes in the aerosol distribution affected by an~~ The inclusion of Van der Waals forces to the particle coagulation scheme suggest improvements in particle effective radius, although other parameters (such as aerosol longevity) deteriorate. Modification of the Pinatubo sulfur emission rate also improves some aerosol parameters, while worsens others compared to observations. Observations themselves are highly uncertain and render it difficult to conclusively judge the necessity of further model reconfiguration. ~~In conclusion~~Model revealed problems in reproducing aerosol sizes above 25 km and also in capturing certain features of the ozone response. Besides this, our results show that SOCOL-AER is capable of predicting the most important global-scale atmospheric ~~and climate~~ effects following volcanic eruptions, which is also a prerequisite for improved understanding of ~~anthropogenic~~ solar geoengineering effects from sulfur ~~emissions~~injections to the stratosphere.

1 Introduction

During the eruption of Mt. Pinatubo in June 1991, a large amount of sulfur dioxide was emitted into the stratosphere, leading to an enhancement of the stratospheric aerosol burden. The aerosol layer perturbed the Earth's radiative balance, resulting in a top-of-the-atmosphere global mean radiative forcing ~~of approximately~~ up to -3 W m^{-2} (Minnis et al., 1993), a global surface cooling of ∼0.4-0.5 K (Dutton and Christy, 1992; Thompson et al., 2009), and a temperature increase of ∼2.5-3.5 K in the tropical lower stratosphere (Labitzke and McCormick, 1992; Randel et al., 2009). During the past decades it was shown that these observed temperature perturbations are connected to many feedbacks in the Earth system such as alteration of the stratospheric circulation with consequences for the troposphere (e.g., Kodera, 1994; Graf et al., 2007), dynamical and chemical effects on stratospheric ozone (Solomon, 1999; Rozanov et al., 2002; Telford et al., 2009), drying of the troposphere causing significant changes in the regional hydrological cycle (Soden et al., 2002), modulation of the global monsoon (Liu et al., 2016), and even modulation of the ocean circulation (e.g., Predybaylo et al., 2017). The distribution and evolution of the stratospheric sulfate can, therefore, be considered as the main forcing constraint for these and many other processes following large volcanic eruptions (Kremser et al., 2016; Timmreck et al., 2016; Swingedouw et al., 2017) and proper information about the aerosol layer is crucial for the characterization and understanding of numerous inherent feedbacks.

Modeling studies help to synthesize our knowledge of how the Mt. Pinatubo and other big eruptions impact the climate system. ~~As a first approximation, global~~ Global three-dimensional general circulation models (GCMs) or chemistry-climate models (CCMs) used for studying the volcanic effects on climate can be ~~discretized~~ mainly separated into two groups: those using prescribed aerosol distributions and those using online aerosol microphysical modules (e.g., Zanchettin et al., 2016). Models of the first type can use aerosol composites derived from satellite and ground-based observations (e.g., Stenchikov et al., 1998), but for studies of the pre-satellite era and the future such models have to rely on estimates provided by models of the second type ~~. Therefore, they~~ or derived by simple reconstruction methods (e.g., Toohey and Sigl, 2017). They have only limited ability to ~~address climate feedbacks~~reproduce the climate response to volcanic eruptions, as the aerosols are prescribed ~~, and also either inherit~~ and therefore the feedbacks between aerosols and the stratosphere are completely missed resulting in biased aerosol radiative forcing depending on concrete circumstances. Such models also inherit either all instrumental uncertainties (see Section 3) or uncertainties ~~inherited~~ from the second type of models and reconstruction models. Models with aerosol microphysics can also be grouped, depending on how they treat the aerosol size distribution: a first class of so-called "modal" and "bulk" (unimodal) schemes and a second class of ~~size-bin resolving~~ size-resolving (also called "sectional") aerosol modules. Currently, there are more than a dozen active global models with aerosol microphysics (see review by Kremser et al., 2016), a smaller part of which employ sectional aerosol schemes.

~~Size-dependent aerosol sedimentation rates crucially affect~~ 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 high computational efficiency. Size-resolving schemes simulate an evolution of the size distribution and can better describe gravitational sedimentation, which crucially affects the stratospheric aerosol lifetime. Arfeuille et al. (2013) argued that bulk schemes are less satisfactory in reproducing volcanic aerosol ~~size~~ distributions, which cast doubts on

the success of such approaches. For 2D models, Weisenstein et al. (2007) have shown that ~~size-bin resolving~~ size-resolving aerosol models are superior to modal approaches in accurately representing the time-dependent aerosol size distribution after large volcanic eruptions. Further progress using a CCM coupled with a ~~size-bin resolving~~ size-resolving microphysical aerosol module to simulate Pinatubo-like eruptions has been achieved by English et al. (2013), however the decline of the simulated aerosol burden was too fast compared with observations, which they attributed to the lack of heating as the aerosol radiative feedback remained decoupled in their model. This highlighted that the fine resolution of aerosol sizes is not a universal ~~problem~~ 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.

The Pinatubo eruption is the strongest volcanic event since the beginning of the satellite era and is therefore often used as a model performance test. Modeling studies of the Pinatubo eruption using models with an assumed log-normal size distributions (e.g., Timmreck et al., 1999a, b; Aquila et al., 2012; Dhomse et al., 2014; Sekiya et al., 2016; Mills et al., 2016) and sectional distribution (English et al., 2013; Kleinschmitt et al., 2017) generally agree reasonably well with observations of atmospheric long-wave and short-wave extinctions, aerosol burden, and other integral parameters. However, an intercomparison of different Pinatubo studies is hampered by the fact that models make different assumptions of how much sufur was initially emitted and how the plume was distributed as function of altitude. Models that reported good agreement with observations used a variety of emission estimates ranging from 10 to 17 teragrams (Tg) of SO_2 and SO_2 plume altitudes in the lower stratosphere differing by a few kilometers. This hints at large ~~uncertainties of~~ differences in how models treat important microphysical and transport processes and significantly increases the uncertainty of the overall aerosol layer understanding.

~~A recent~~ An ongoing Model Intercomparison Project on the climatic response to Volcanic forcing (VOLMIP, Zanchettin et al., 2016) aimes to address the existing intermodel uncertainties. ~~However, so~~ So far only the Tambora eruption in 1815 has been considered for the global models with interactive aerosol microphysics. Marshall et al. (2018) evaluated the performance of four state-of-the-art models (WACCM, UM-UKCA, SOCOL-AER, and ECHAM5-HAM) using mostly the same settings of the initial emission and compared the results to the available observations of ice-core sulfate. The focus of that study was on sulfate deposition in polar areas, as ice cores are the best available source of information about historical eruptions (Sigl et al., 2015; Toohey and Sigl, 2017). The comparison revealed that modelled volcanic sulfate deposition varies substantially in timing, spatial pattern and magnitude between the models. The ratio of the hemispheric atmospheric sulfate aerosol burden after the eruption to the average amount of sulfate deposited on ice sheets varied among models by up to a factor of 15. The burden-to-deposition ratio is to a large extent determined by the treatment of deposition processes, which are simplified in models. Furthermore, it also depends on sulfur species, which never entered the stratosphere but were transported through the (upper) troposphere, oxidized and removed by wet or dry deposition. Moreover, it depends on how fast aerosols grow and sediment from the stratosphere back to the troposphere. The analysis of the stratospheric burdens of SO_2 and liquid H_2SO_4 as well as the polar winds, also revealed large intermodel differences. Therefore there is still no clear understanding of which model is closer to reality in ~~describong~~ describing the stratospheric aerosol distribution, since direct stratospheric observations are missing and the ice core estimates could be strongly modulated by the tropospheric deposition schemes.

Table 1. List of Experiments

Name	QBO nudged	Aerosol feedback	Sedimentation scheme	Coagulation Efficiency α	Emission rate [Tg SO_2]
REF	Yes	Yes	Walcek	$\alpha = 1$ everywhere	14
REF12	Yes	Yes	Walcek	$\alpha = 1$ everywhere	12
UPWIND	Yes	Yes	Upwind	$\alpha = 1$ everywhere	14
~~RAD~~ noRAD	Yes	No	Walcek	$\alpha = 1$ everywhere	14
~~QBO~~ noQBO	No	Yes	Walcek	$\alpha = 1$ everywhere	14
~~RADQBO~~ noRADnoQBO	No	No	Walcek	$\alpha = 1$ everywhere	14
COAG	Yes	Yes	Walcek	α based on a Lennard-Jones potential: $\alpha \sim 1$ in continuum regime ($K_n \gg 1$); $\alpha \sim$ 1-3 in transition regime ($K_n \sim$ 1-10); $\alpha \ll 1$ in free molecular regime($K_n \ll 1$).	14

The representation of aerosol evolution in the stratosphere requires treatment of many processes, which can substantially differ among models. Previous studies (e.g., Timmreck et al., 1999a, b; Aquila et al., 2012) illustrated the importance of the quasi-biennial oscillation (QBO) and radiative heating of volcanic aerosols in the models, as these processes affect the transport and thus the lifetime and climate impact of aerosols. Benduhn and Lawrence (2013) found that numerical diffusion induced by an inaccurate sedimentation scheme may lead to excessive transport of the aerosol to the middle and upper stratosphere. So, even with a fine aerosol size resolution, resulting sedimentation can be biased due to the model's numerical scheme. English et al. (2011, 2013) suggested that attractive van der Waals forces may lead to an enhanced coagulation efficiency and should be taken into account (in the transition and free molecular regimes). However, such interactions led to an even faster decay in their simulated global aerosol burden after the Pinatubo eruption. Sekiya et al. (2016) and Kleinschmitt et al. (2017) also investigated the role of this process and reported significant effects on aerosol parameters. Interactive chemistry was also shown to be important for aerosol formation, as hydroxyl radical (OH) can become depleted after big eruptions, which ~~plorongs~~ prolongs the time needed for conversion of volcanic SO_2 to H_2SO_4 (Bekki, 1995; Mills et al., 2017).

The coupled size-resolving stratospheric aerosol-chemistry-climate SOCOL-AER model has been evaluated in detail for volcanically quiescent conditions (Sheng et al., 2015b). In this study, we employ it for the Pinatubo eruption of 1991 and aim to characterize its performance comparing our results against satellite observations and in-situ measurements. By means of this model we also attempt to illustrate the roles of the aerosol radiative heating, sedimentation scheme, coagulation efficiency, and ~~quasi-biennial oscillation (QBO)~~ the QBO in the evolution of the aerosol burdens, aerosol optical properties and particle size distributions, which may also help to better understand differences between various models.

2 Method

The coupled aerosol-chemistry-climate model SOCOL-AERv1.0 (SOCOL-AER hereafter) has been introduced by Sheng et al. (2015b), who applied the model to analyze the global atmospheric sulfur budget under volcanically quiescent conditions and its sensitivity to anthropogenic emissions. SOCOL-AER is a CCM SOCOLv3 (GCM ECHAM5 plus chemical module MEZON, Stenke et al., with an aerosol module AER (Weisenstein et al., 1997). AER includes a comprehensive description of sulfur chemistry and microphysics, in which the particles are size-resolved by 40 size bins spanning radii from 0.39 nm to 3.2 μm. ~~Interactive aerosol radiative feedback~~ , which allows to consider all relevant microphysical processes (e.g., nucleation and coagulation). The influence of the aerosol on radiation fluxes at all wavelengths is also taken into account. ~~The aerosol optical properties~~ SOCOL-AER uses 6 band shortwave (Cagnazzo et al., 2007) and 16 band longwave (Mlawer et al., 1997) radiation schemes. Extinction coefficients, single scattering albedos, and asymmetry factors required by the radiation ~~code~~ codes are calculated online from aerosol physical properties using Mie theory ~~.~~ for actual H_2SO_4 weight percent and temperature using refraction indices from Biermann et al. (2000). The aerosol surface area density and composition is used to calculate heterogeneous reaction rates in chemical module. In this study, the spatial resolution of SOCOL-AER is set to T42 horizontal truncation ($2.8° \times 2.8°$ latitude/longitude resolution) and 39 vertical hybrid sigma-pressure levels from the surface to 80 km (about 1-1.5 km per level in the upper troposphere and lower stratosphere, 2-3 km above). The QBO in the model is nudged to observed wind profiles. Monthly mean transient sea surface temperatures (SSTs) and sea ice coverage (SIC) are prescribed from the Hadley Centre Sea Ice and SST data set (Rayner et al., 2003). Comprehensive sulfur surface emissions are also fully taken into account. More detailed description of the SOCOL-AER modules can be found in Sheng et al. (2015b).

~~The 1991 Pinatubo~~ Observational estimates of the total SO_2 mass emitted by Pinatubo and its vertical distribution are still very uncertain (Guo et al., 2004). We use an estimate of Sheng et al. (2015a) who used a 2-D sulfate aerosol model with the same microphysical module as in SOCOL-AER to identify the optimised emission parameters by running 300 sensitivity experiments spanning the observational uncertainty and comparing the results to observations of different aerosol parameters. Based on Sheng et al. (2015a), the Pinatubo eruption is introduced here by an injection of 14 Tg SO_2 in the region 97°-112°E and 1.8°S-12°N~~according to observations (Guo et al., 2004)~~. SO_2 is ~~released continuously~~ continuously released from 14 to 15 June 1991 and spread between 16 to 30 km with a vertical mass distribution ~~optimized according to Sheng et al. (2015a),~~ skewed to low altitudes with the mass peak between 18 and 21 km. ~~This establishes a realistic initial mass loading of the eruption.~~ All experiments are summarized in Table 1. The reference run subsequently termed REF represents the standard setup of SOCOL-AER, including nudged QBO, interactive aerosol radiative ~~feedback~~and chemical effects, and coagulation efficiency uniformly set to one. In terms of module versions it replicates the model configuration used for the Tambora study (Marshall et al., 2018).

By means of the experiment REF12 we estimate the model sensitivity to uncertainty in emission amount by lowering it to 12 Tg SO_2. In the experiment termed ~~RAD~~noRAD, the radiative fluxes are calculated using the SAGE-4λ dataset (Arfeuille et al., 2013) averaged over the period 1995-2002 instead of the interactively simulated aerosol distribution, which eliminates the radiative effects of volcanic aerosols. The experiment termed ~~QBO~~ noQBO is carried out without QBO, which leads to a weak

easterly zonal wind in the tropical stratosphere. Both QBO nudging and interactive radiation are switched off in the experiment termed ~~RADQBO~~noRADnoQBO. These three experiments allow us to identify the impact of QBO and radiative heating of volcanic aerosols on the evolution of the stratospheric aerosol burden after Pinatubo eruption. We also ~~consider two exploratory experiments~~ carry out an exploratory experiment concerning the coagulation efficiency ~~. The~~ - the experiment termed COAG represents the coagulation efficiency as Lennard-Jones potential, i.e. a smooth function of the Knudsen number retrieved from the results in Figure 3 of Narsimhan and Ruckenstein (1985) with a Hamaker constant of 5×10^{-19} J. As an approximation of attractive Van der Waals forces it enhances the coagulation efficiency in the transition regime (maximum enhancement larger than 2), but decreases it rapidly (less than 1) as the Knudsen number increases in the free molecular regime. The experiment termed UPWIND employs the upwind sedimentation scheme (Benduhn and Lawrence, 2013), while all other simulations use the more elaborate Walcek method with minimal numerical diffusion (Walcek, 2000). This is sufficient to clarify the impact of different sedimentation schemes, though work by Benduhn and Lawrence (2013) presented a further improved modified Walcek method.

~~For each~~ Each of these experiments ~~we calculated~~ consists of five ensemble members. In the figures we show ensemble spread for the REF experiment and only ensemble means for other experiments to keep figures as uncomplicated as possible. In addition to Pinatubo, for all runs we considered the smaller eruption of Cerrro Hudson in Chile in August 1991. We used the latest estimate of 2.3 Tg total SO_2 emitted (Miles et al., 2017) with 75% of mass injected between 16 and 18 km. Sensitivity studies with and without this event showed that its contribution is minor, since it is located at higher latitudes (45.5°S), but we keep it for completeness.

3 Results and discussion

3.1 Aerosol Burden

Figure 1 shows the evolution of observation-derived and model-calculated stratospheric aerosol burdens in units of ~~teragram (Tg)~~ Tg of sulfur globally integrated (a) and in the tropics (b). The High-Resolution Infrared Radiation Sounder (HIRS) measured the aerosol vertical column and derived total aerosol mass with about 10% uncertainties. HIRS includes tropospheric and stratospheric aerosols together (Baran and Foot, 1994). In contrast, the limb-occultation measurements of SAGE II allow aerosols in the troposphere and stratosphere to be distinguished from one another. In this work the SAGE II-derived total aerosol mass is represented by two data sets. The first one, the SAGE-4λ method (Arfeuille et al., 2013), used within the Chemistry-Climate Model Initiative (CCMI), employs all four SAGE wavelengths with overall about 30% uncertainties for non-gap-filled data and higher uncertainties in data gaps filled by lidar station data. The second data set was recently compiled for phase 6 of the Coupled Model Intercomparison Project (CMIP6, Eyring et al., 2016) using the SAGE-3λ method, which is similar to SAGE-4λ but refrains from employing the less reliable channel at 385 nm, thus considering only three SAGE wavelengths. Directly after Pinatubo the SAGE-3λ data set uses additional satellite and ground-based data for gap-filling. More information about the SAGE-3,4λ composites can be found in a recent paper by ~~Revell et al. (2017)~~Thomason et al. (2018).

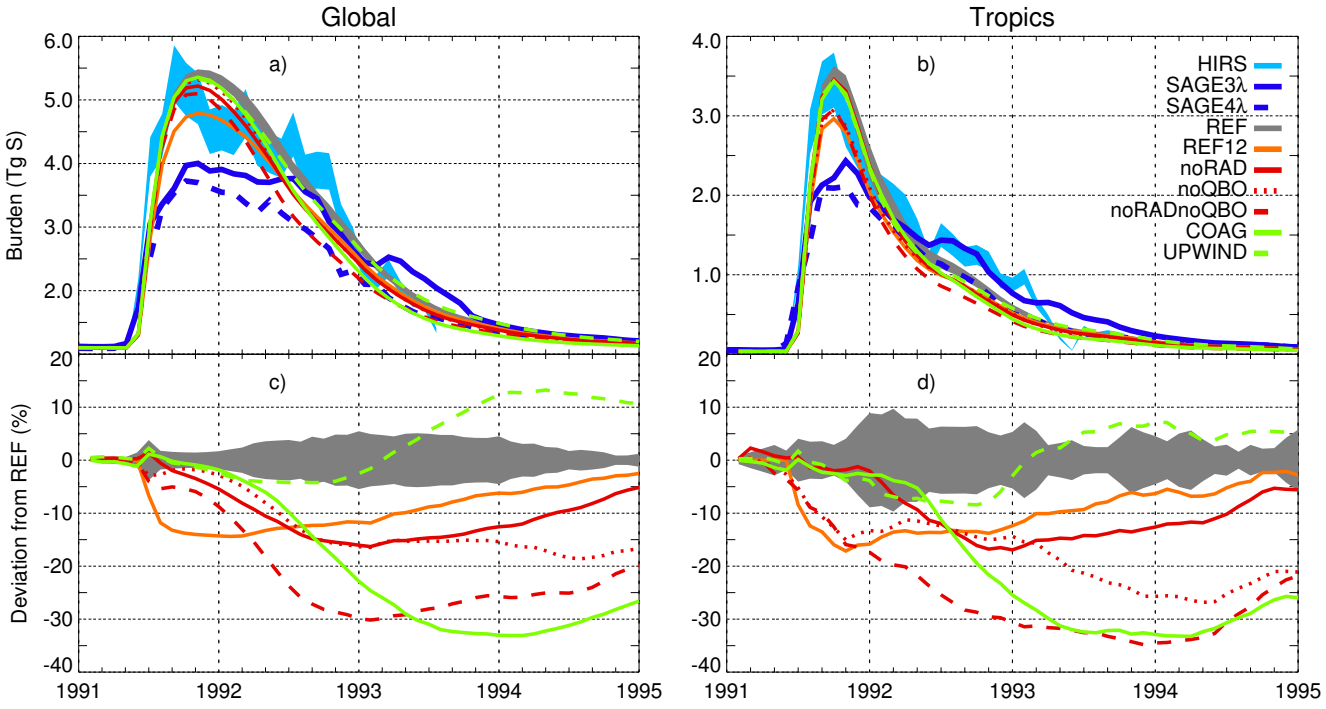

Figure 1. Panels a and b: evolution of model-calculated global (pole to pole, left) and tropical (20°S—20°N, right) stratospheric aerosol burden (Tg of S) compared with the HIRS and SAGE II-derived data (SAGE-3,4λ). HIRS-derived aerosol sulfur burden assumes 75% sulfuric acid by weight. Light blue shaded area: uncertainties of HIRS. Grey shaded area: 2-σ ensemble spread of the REF experiment. All other experiments are shown as ensemble means. Panels c and d: same as a and b, but deviations of all the numerical experiments from the REF in %.

During the first year after Pinatubo eruption, the aerosol mass in both SAGE II-based data sets is noticeably lower than ~~HIRS~~in HIRS data. This is likely related to the saturation effects of SAGE II, as a limb-occultation instrument, during this period (Russell et al., 1996). The SAGE-3λ composite provides significantly larger burdens than its predecessor, due to additional data used in a gap-filling procedure (Revell et al., 2017), but still much lower than HIRS. After this period, when the atmosphere becomes sufficiently transparent, SAGE II measurements are expected to provide more accurate aerosol extinctions. In contrast, the HIRS-derived mass becomes less reliable with time, when the aerosol cloud spreads to higher laltitudes with lower values that are close to the noise level of the technique (Baran and Foot, 1994). This suggests to trust the HIRS data up to mid-1992 and the SAGE data afterwards (Sheng et al., 2015a). Note, however, that the updated SAGE II-based data set now also provides values closer to HIRS from mid-1992 to early 1993 and considerably larger values later in 1993.

The global stratospheric aerosol burden calculated by REF (grey shaded area representing 2-σ ensemble spread) agrees well with the HIRS data peaking around 5.4 Tg at the end of 1991. Later, REF agrees well with the SAGE-4λ composite, while the updated SAGE-3λ has a generally larger burden. Qualitatively similar results are found for the tropics. Recent modelling studies by Mills et al. (2016) and Kleinschmitt et al. (2017) showed very similar time series of the global aerosol burden

with initial emissions of 10 and 14 Tg of SO_2, respectively. These studies, another work by Sekiya et al. (2016), as well as the present study fail to reproduce the pronounced step-like evolution of the burden seen in HIRS and SAGE-3λ, showing a smoother decrease instead. Dhomse et al. (2014) overestimated the HIRS peak burden even with 10 Tg of SO_2 emitted, but obtained this step-like behavior. Dhomse et al. (2014) explained it by variability of the background aerosols related to the summer increase of photolysis, but they also noted that their background values are significantly larger than in the other models and observations. Besides instrumental uncertainty, another reason for the complicated shape of the observational curves seen in Fig. 1 could be the seasonal variability of the stratospheric circulation that is known to be underestimated in ECHAM5 (Stenke et al., 2013) and LMDZ (Kleinschmitt et al., 2017), which are core GCMs of the sectional models SOCOL-AER and LMDZ-S3A, respectively.

Panels c and d of Figure 1 show deviations (%) from REF of all experiments. Our experiment with lower emission (REF12, 12 Tg of SO_2 instead of 14 in REF, but otherwise unchanged plume characteristics) shows lower burdens of up to 14% globally and 17% in the tropics, which is therefore even farther than REF from the latest SAGE II-derived estimates after mid 1992. The results of the sensitivity runs ~~QBO, RAD and RADQBO~~ noQBO, noRAD and noRADnoQBO are presented by the red curves. During the first few months after the Pinatubo eruption, the aerosol mass loading in the tropical reservoir is maintained by the ~~balance~~ competition between sedimentation and enhanced tropical upwelling due to radiative heating of the volcanic aerosols with the QBO in a strongly descending easterly phase (Trepte and Hitchman, 1992; Trepte et al., 1993). Therefore, deactivation of each of these processes leads to a stratospheric burden decrease mostly located in the tropics. About one year after the eruption the global aerosol burden in ~~QBO and RAD~~ noQBO and noRAD is approximately 15% lower than in REF. The experiment ~~RADQBO~~ noRADnoQBO shows a cumulative effect up to -30% around 1993. Gravitational sedimentation becomes a dominant removal process when particles grow sufficiently large after the Pinatubo eruption. With effective radii of 0.5 μm or more (Russell et al., 1996) these particles sediment efficiently. The burden calculated by UPWIND mostly lies within $\pm$10% with respect to REF. This upwind scheme was shown to have a large numerical diffusion smearing the aerosol layer out in both, up and down, directions (Benduhn and Lawrence, 2013). This results in a slightly lower mass during 1-1.5 years after the eruption (effect of the downward diffusion), and a slightly larger mass later on (upward transported aerosols stay longer in the stratosphere). Although this diffusion effect is of numerical origin, for our model it increases the stratospheric lifetime of aerosols and leads to a better agreement with SAGE-3λ after 1993. The aerosol burden calculated by COAG, which differs from REF by a higher coagulation efficiency, shows a more rapid decay rate of the global volcanic aerosol burden compared to REF and the measurements. The difference to REF maximizes in late 1993 at approximately -33%, which is in agreement with other studies also looking at the van der Waals forces effects (English et al., 2013; Sekiya et al., 2016).

3.1.1 Aerosol size distribution

3.1.2 ~~Aerosol size distribution~~

Figure 2 shows comparisons of the optical particle counter (OPC) measurements operated above Laramie, Wyoming (41°N, 105° W, Deshler et al., 2003; Deshler, 2008) against our model experiments. Model was sampled as monthly mean values

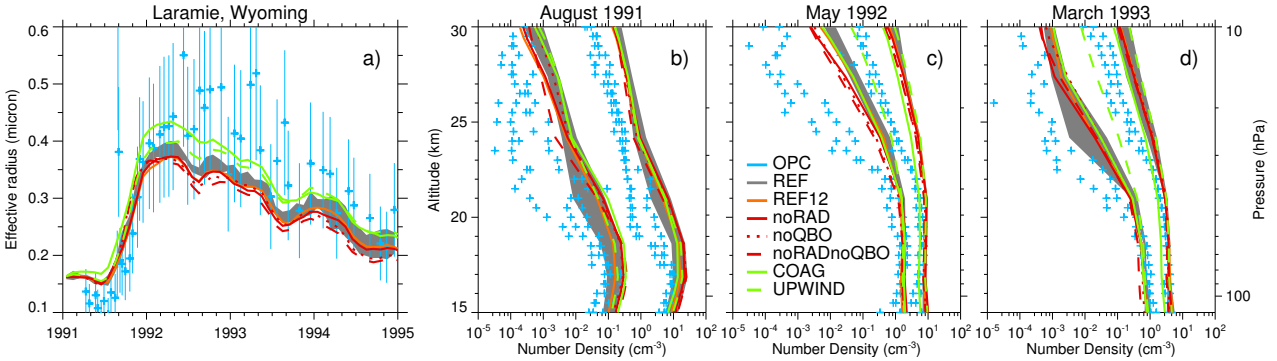

Figure 2. Comparison of in situ measurements of particle size (at Laramie, Wyoming, Deshler (2008)) with SOCOL-AER simulations. (a) Stratospheric effective particle radius averaged for 14-30 km altitude. Thin blue whiskers reflect measurement uncertainty (taken from Kleinschmitt et al. (2017)). (b-(d) Profiles of cumulative number densities for two size channels with radii $R > 0.15$ μm (right group of curves) and $R > 0.5$ μm (left group of curves) in August 1991, May 1992, and March 1993, respectively. SOCOL-AER results are monthly means, while OPC data are discrete measurements within chosen months.

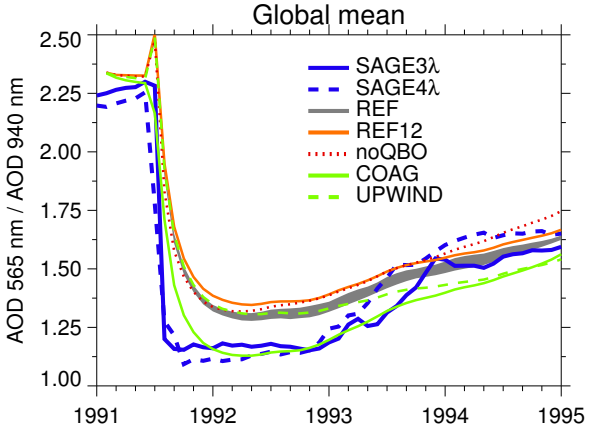

Figure 3. Comparison of remote measurements of aerosol optical depth (AOD) ratios at two wavelenghts, a proxy of particle size, with SOCOL-AER simulations. Lines: SAGE II-derived (SAGE-3,4λ) and modeled global AOD (> 18 km) ratios 565 nm / 940 nm. Grey shaded area: 2-σ ensemble spread of the REF experiment. All other experiments are shown as ensemble means.

averaged over a region of $\pm 5°$ latitude and longitude around Laramie. Panel a shows the effective aerosol radius averaged over 14-30 km. The effective radius calculated by REF generally lies within the observational uncertainty. However, compared to the observational mean, it is biased high under quiescent conditions and biased low during the volcanically perturbed period. COAG shifts the effective radius up compared to REF which ~~inproves~~ improves the agreement with observations after 1992, but worsens it earlier. Differences of other experiments reflect the burden of the aerosol behavior shown in Fig. 1, illustrating that less mass present in the stratosphere generally also leads to smaller sizes.

Panels b-c of Fig. 2 show cumulative number distributions for two OPC size channels ($R > 0.15$ μm and $R > 0.5$ μm) in August 1991, May 1992, and March 1993 representing different stages of the volcanic aerosol cloud. We use months with at least two soundings to obtain a useful approximation of ~~monthly means~~day-to-day variability. Aerosol number densities at the altitudes of the maximum concentrations ~~is~~ are well reproduced by REF for both large and small particles. Higher altitudes, however, are ~~less~~ not so well reproduced, with modeled number densities being up to ~~one order~~ three orders of magnitude too high for bigger particles at certain levels. However, at these high altitudes OPC measurements are themselves uncertain, often having to rely on only one or two channels (plus the concomitant condensation counter measurement). ~~Very large~~ Even larger deviations from the OPC measurements ~~by up to two orders of magnitude~~ are found for the UPWIND experiment, which has clearly too many particles, especially large ones, in the middle stratosphere all the way to the upper edge of the stratospheric aerosol layer, highlighting the importance of a sedimentation scheme with low numerical diffusivity. Experiments with radiatively decoupled aerosols, ~~RAD and RADQBO~~noRAD and noRADnoQBO, illustrate the importance of the enhanced upwelling, even in midlatitudes, by showing more large particles staying at the lower levels.

To analyze the globally mean size distributions, in Fig. 3 we show the ratio of aerosol optical depths (AOD) at 565 nm and 940 nm for the column above 18 km, calculated from SAGE II-derived composites (blue curves) and from model results. These ratios are inversely related to the particle size: a smaller ratio corresponds to larger particles. In the early phase of the Pinatubo eruption, a large number of small particles are formed, which coalesce very quickly as shown by the very sharp drop in the AOD ratio, falling below 1.25 in observations. Afterwards, the small AOD ratio stays almost constant for approximately one year. Around late 1993 the ratio starts to return to higher values, because the large particles continuously sediment out of the stratosphere and smaller particles nucleate in the air entering the stratosphere in the tropics. REF predicts smaller particles than derived from SAGE II during the early phase after the eruption, and only in 1993 it ~~begins~~ starts to agree well with the satellite observations. In contrast, due to the enhanced coagulation, COAG produces larger particles (smaller AOD ratios) than REF, and shows better agreement with SAGE II during the four years following the eruption. The model run UPWIND with a simplified upwind scheme for sedimentation is initially close to REF but ~~starts to overestimate~~ reveals overestimation of the particle sizes compared to REF after 1993. This is related to a larger aerosol burden (Fig. 1), which enables further coagulation. Our experiment with the reduced emission (REF12) further illustrates this effect by showing that weaker emission leads to slightly smaller sizes over the whole lifetime of the volcanic aerosol cloud.

In general, ~~all model experiments show the same global and local effects compared to each other and to the two observational datasets~~model in its reference configuration slightly underestimates the mean particle radius. The agreement with observations is much better if a more detailed coagulation is used. However, as seen in panels b-c of Fig. 2, the model in all configurations also has problems in reproducing altitudes higher than 25 km. Comparison of the model to both, in-situ OPC measurements and the satellite-based global composites SAGE-3λ and SAGE4λ, reveal same effects of individual experiments. The main difference between two comparisons is seen during the pre-eruption time, as the model shows larger particles than OPC and smaller particles than SAGE, which can be attributed to the local bias of model parameters.

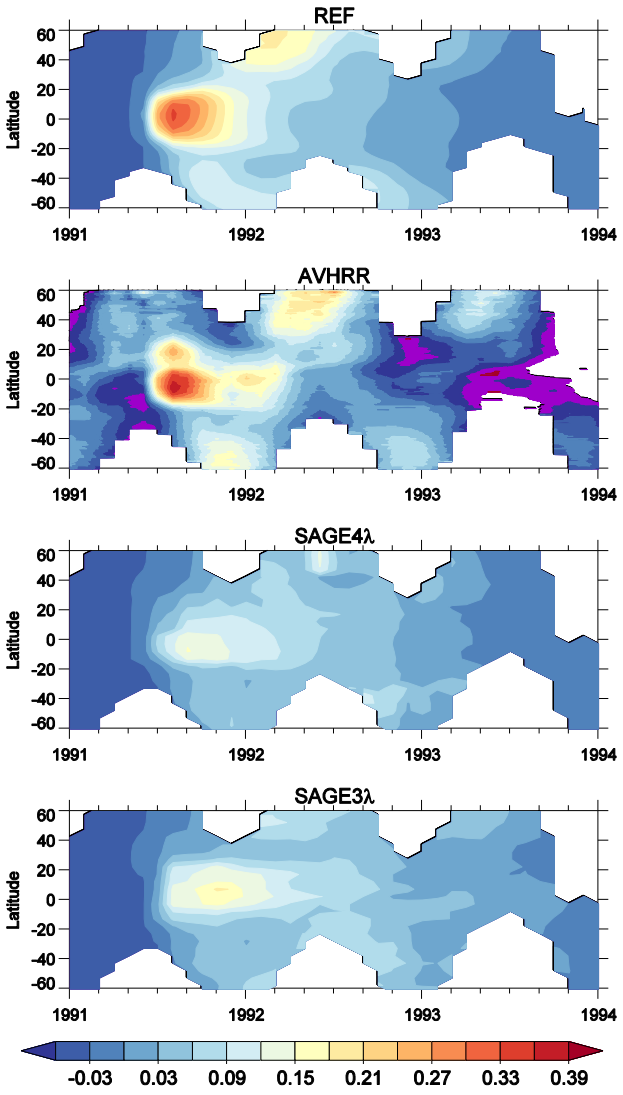

Figure 4. Monthly zonal average total AOD ~~over oceans~~ measured at 0.63 μm by AVHRR and calculated at 0.56 μm above tropopause by SOCOL-AER and provided by SAGE3,4λ composites. Since AVHRR measurements were performed over oceans, we applied the same selection for the model here. SAGE3,4λ data were, however, initially provided as zonal means. Background values are subtracted from all data sets (which may result in slightly negative values). All panels are masked at winter high latitudes where AVHRR data are missing.

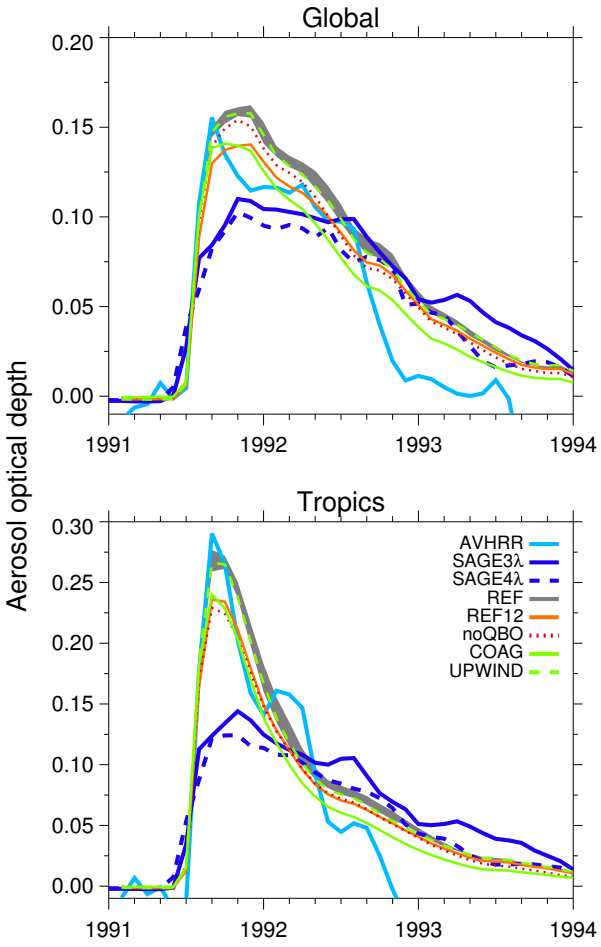

Figure 5. Same as in Fig. 4 but averaged over non-masked regions globally (80°S-80°N) and in the tropics (20°S-20°N). Grey shaded area: 2-σ ensemble spread of the REF experiment. All other experiments are shown as ensemble means.

3.2 Aerosol optical depth

Figure 4 shows the latitudinal evolution of volcanic material as modelled and measured AOD in the visible part of the solar spectrum, which also represents the main direct climate forcing, since it defines the amount of scattered back to space solar irradiance responsible for global cooling. In addition to SAGE II, we used data from the Advanced Very High Resolution Radiometer (AVHRR) satellite instrument, which makes observations over global oceans (Zhao et al., 2013). Modeled and SAGE-3,4λ AODs are obtained by vertically integrating the extinctions above the tropopause. We removed latitudes not observed by AVHRR for each month from the other data sets and subtracted background values from observations and calculations~~, thereby excluding also the contributions from tropospheric aerosols~~. Aerosol optical depths derived from SAGE II and AVHRR significantly disagree ~~with each other~~ both in magnitude and spatial distribution. SAGE-3,4λ show much smaller

AOD in the tropics in 1991 and ~~do also not show a~~ not so strong southward plume as seen in AVHRR at the end of 1991, part of which is influenced by the high-latitude Cerro Hudson eruption in August 1991. The northern hemispheric plume in 1992 is also more pronounced in the AVHRR data. Figure 5 shows the same AOD values, but averaged over the non-masked reagions in Fig. 4 over the globe and the tropics. The main difference between AVHRR and SAGE is that AVHRR shows a higher AOD peak in 1991 (two times higher in tropics), similar to the faster increase in early aerosol burden of HIRS (Fig. 1). However, AVHRR reveals a much faster decay, so that starting from late 1992 SAGE II-derived AOD is much larger than measured by AVHRR.

Modeling results are ~~overall~~ closer to AVHRR ~~than~~ before mid-1992 and to SAGE II-derived data later. REF shows weaker south- and northward plumes in Fig. 4, but ~~perfectly~~ nicely captures the initial increase in the tropics seen by AVHRR. The lifetime of the initial tropical cloud is also well captured compared to AVHRR, except for a small increase in early ~~1992, while in both SAGE II-based data sets the cloud persists for much longer.~~ 1992. Similarly to the burden shown in Fig. 1, starting from mid-1992 the model results are closer to SAGE-4λ than to SAGE-3λ. The experiment REF12 shows lower AODs that are ~~, however, even~~ closer to AVHRR ~~.~~ globally but at the same time it also provides weaker initial increase in 1991. The experiment without QBO shows that less mass is maintained in the tropics compared to REF, and therefore more mass is transported southward in 1991 following the Brewer-Dobson circulation. Th experiment with increased coagulation efficiency (COAG) shows faster decay of initial AOD increase~~, while in UPWIND it is the opposite~~. UPWIND has slight changes but mostly lies whithin the uncertainties of REF. Similarly to the size evolution discussed in the previous section, in general, details of all modeling experiments are also qualitatively consitent to those shown for the the burden in Fig. 1.

3.3 Stratospheric temperature response

Lower tropical stratospheric warming after major eruptions is one of the key features of volcanic influence on climate (e.g. Swingedouw et al., 2017). ~~Besides being~~ It is a forcing for the thermal wind balance, a mechanism through which volcanoes can affect high latitude troposheric circulation~~, this~~. This warming is also an important indicator for the aerosol mass distribution in the stratosphere~~. Infrared~~, because it is mostly caused by the infrared absorption of volcanic aerosols, which does not critically depend on ~~aeroso lparticle size, but the radiative warming is directly related to the total aerosol mass density~~ aerosol particle size (Lacis et al., 1992). The difficulty of correct representation of post-volcanic stratospheric warmings is a known issue of global models. Key factors are, besides uncertainties in aerosol distributions, model dynamics and radiative transfer, which in turn also depends on many factors such as spatial and spectral resolution, presence and quality of interactive chemistry and others (Eyring et al., 2006; Lanzante and Free, 2008; CCMVal, 2010).

Figure 6 compares zonal-mean tropical temperature anomalies computed by SOCOL-AER in the lower stratosphere after the Pinatubo eruption with ERA-interim and MERRA reanalyses. Anomalies are calculated by subtracting the climatological annual cycle ~~and the QBO impact.~~ averaged over 1986-2013 for reanalyses and over 1991-1995 of the noRADnoQBO experiment for all other model experiments. Although the noRad experiment has no aerosol radiative effect, we have also added it to Fig. 6, so that everything between the noRad line and other lines can be attributed to the radiative effect of volcanic aerosols. By mid-1993 this effect is mostly gone and then all model experiments are in-line with reanalyses. Since the lower tropical strato-

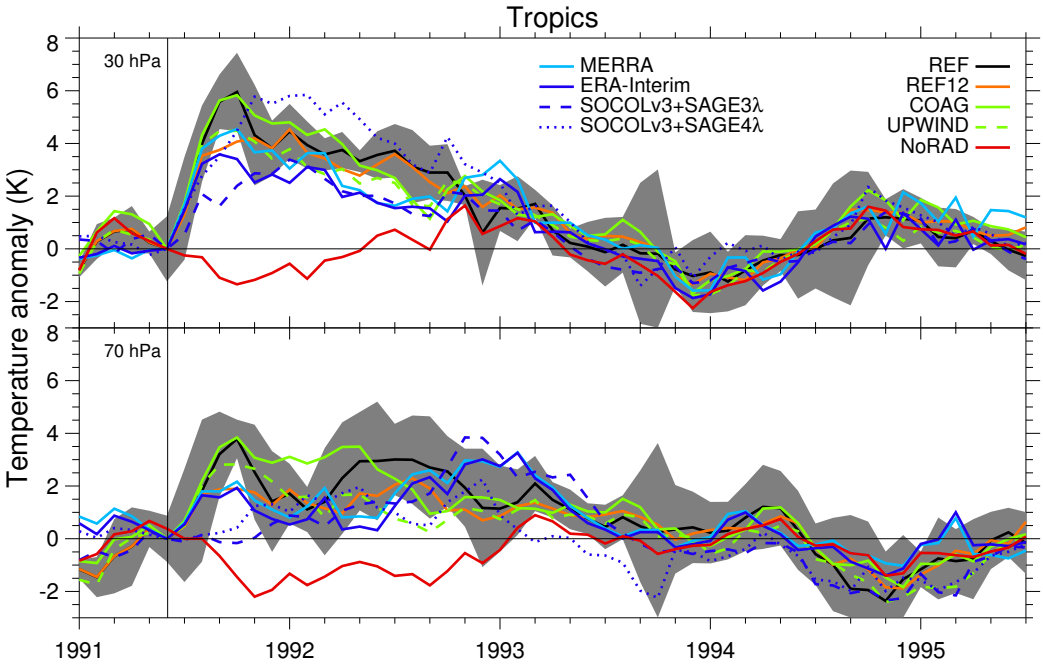

Figure 6. Zonal mean temperature anomalies from SOCOL-AER for tropics (20°S—20°N) at 30 hPa (top) and 70 hPa (bottom). Light and dark blue lines: MERRA and ERA-Interim temperature reanalysis data. Anomalies are computed by subtracting the annual cycle~~and QBO~~. Grey shaded area: 2-σ ensemble spread of the REF experiment. All other scenario curves are ensemble means.

phere is a dynamically very active region, the model also shows a large ensemble spread in the stratospheric temperature signal so that all numerical experiments and observations generally fall into this variability. While the temperature anomalies in the reanalyses differ by up to 1 K, the ensemble mean of REF (black curve) overestimates the warming both at 30 and 70 hPa by 1-2 K in late 1991 and mid-1992. The SOCOL-AER scenarios show some differences with respect to each other. While the experiment with the reduced emission (REF12) shows better agreement with reanalyses at both levels, this apparent improvement comes with clear deteriorations in other quantities, such as too small particle sizes (Fig. 3). The scenario with enhanced coagulation efficiency COAG is warmer at 70 hPa early in 1992, which is related to increased sedimentation of larger particles to lower altitudes. Results of the UPWIND scenario show a smaller warming than REF, which reflects the larger vertical spread of the aerosol mass due to enhanced diffusion leading to faster aerosol removal from the lowermost stratosphere.

To further understand the model results, we plotted the vertical aerosol mass distribution in the tropics for REF and the SAGE-3,4λ composites in Fig. 7. We did not plot the vertical mass distributions from other experiments because they are all very similar to REF relative to SAGE data. There are small differences between experiments that are consistent with the previous analysis, i.e. the UPWIND mass is more vertically diffused, while COAG results show faster decay of the whole aerosol cloud and therefore slightly more mass present at lower levels. The main difference of all model experiments to SAGE II-derived data and especially to the latest SAGE-3λ composite in Fig. 7 is the presence of a large amount of aerosol mass

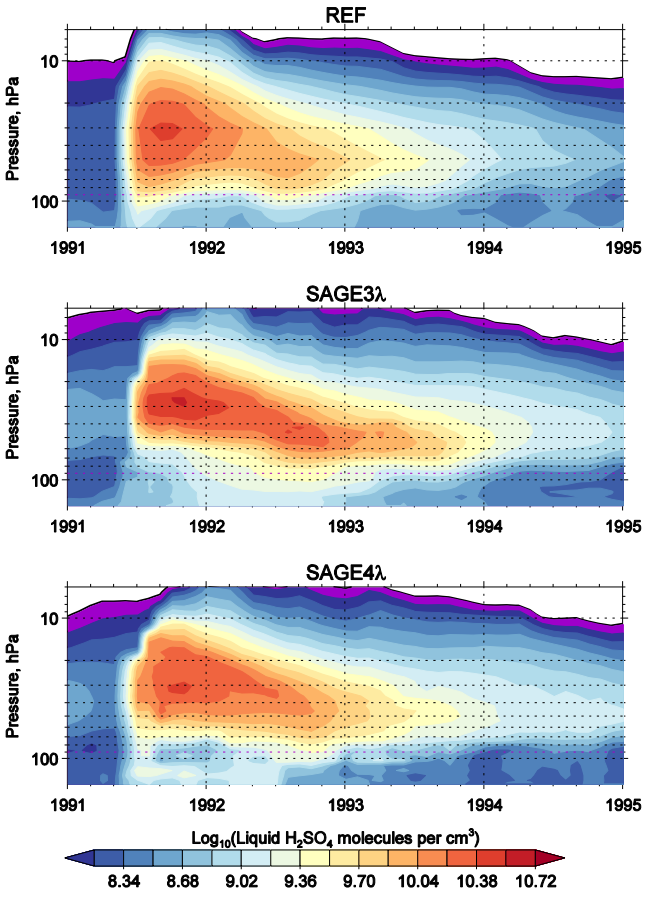

Figure 7. Vertical distribution of liquid H_2SO_4 concentration averaged over the tropics (20°S—20°N).

in 1991 in the lowermost stratosphere, i.e. below approximately 60 hPa, which is not consistent with SAGE. Potentially the SAGE II-derived data can be still influenced by the known problems in observing the lower stratosphere, which became opaque for limb-occultation instruments in 1991 (Russell et al., 1996). This is partly confirmed by comparison of SAGE II-derived data with HIRS and AVHRR in previous sections. However, recently Revell et al. (2017) analysed the stratospheric warming after Pinatubo using SOCOLvs3, which has the same dynamical and chemical cores as SOCOL-AER, but used prescribed aerosols from the SAGE-4λ and SAGE-3λ composites. They found that model simulations driven by SAGE-3λ aerosols are much closer to temperature reanalyses than simulations driven by SAGE-4λ, which also has more aerosol mass present in the lowermost tropical stratosphere.

We added results of both experiments from Revell et al. (2017) to our Fig. 6 (dashed and dotted blue curves). Analysis of late 1991 reveals that model results driven by SAGE-3λ are also biased compared to reanalysis temperature but to the opposite direction than REF. Purely radiatively, this fact suggests that the sharp cut of SAGE-3λ aerosol cloud below 60 hPa in late 1991 is not realistic and there should be something in between the REF and SAGE-derived data. However the dynamics

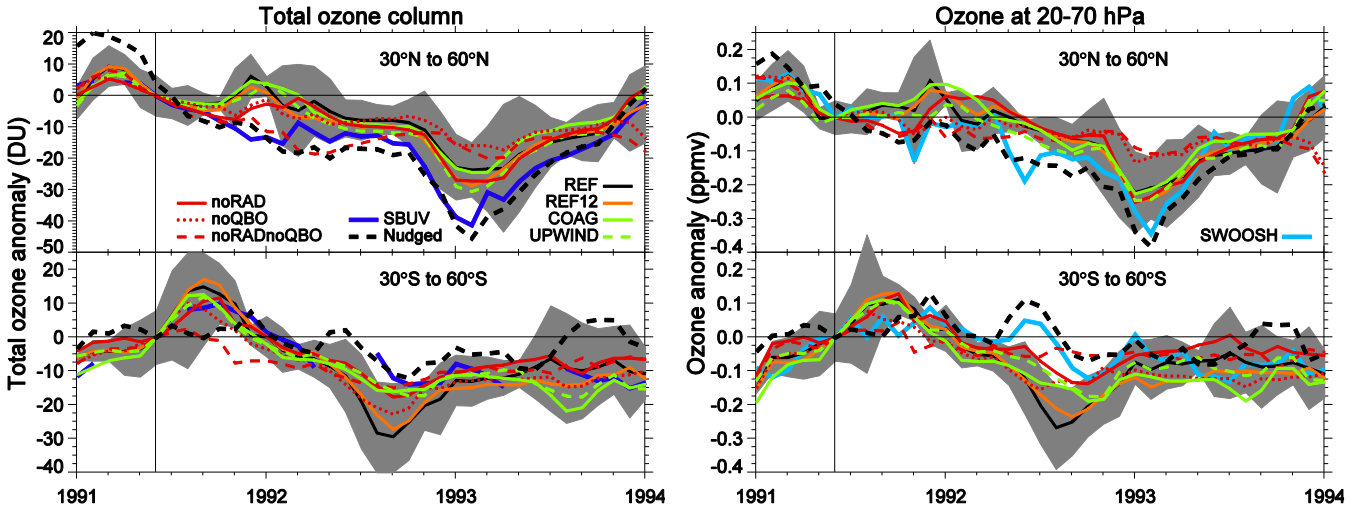

Figure 8. ~~Zonal~~ Monthly mean ~~tropical mean~~ midlatitude ozone (~~20~~30°~~S—20~~-60° ~~N~~north and south) ozone anomalies from SOCOL-AER compared with observations. ~~Upper panel~~Left panels: total ozone column. Right panels: ozone mixing ratio at ~~30~~ 20-70 hPa. ~~Lower panel: total ozone column.~~ Observational data sets SWOOSH and SBUVv8.6 are denoted by light and dark blue lines, respectively. Anomalies are computed by subtracting the annual cycle. Grey shaded area: 2-σ ensemble spread of the REF experiment. All other experiments are shown as ensemble means.

is also highly involved in this region, since modification of warming at different levels also causes changes to the tropical upwelling and therefore adiabatic cooling of higher levels as well as aerosol redistribution causing further changes to local radiative effects. Besides this, the extra-tropical wave-breaking and thus the stratospheric residual circulation is also modified with further consequences for the tropics (e.g. Toohey et al., 2014). Comparison of REF and SAGE results in Figs. 6 and 7 for period after 1991 also suggests that relation of the vertical distribution of aerosol mass and the resulting warming is nonlinear and needs further detailed investigation separating dynamical and radiative effects.

3.4 Ozone response

The response of ozone to major volcanic eruptions is subject to the plethora of dynamical and radiative stratospheric feedbacks including changes in heterogeneous chemistry. ~~(Muthers et al., 2015) showed~~ Previously, it was shown that even a sign of the total ozone response after a volcanic eruption depends on the ~~background halogen loading of the stratosphere~~stratospheric halogen loading (e.g., Tie and Brasseur, 1995; Muthers et al., 2015). Volcanic eruptions can in principle also contribute to the stratospheric chlorine which further affects ozone (e.g., Klobas et al., 2017), but there was no significant increase in stratospheric chlorine observed after Pinatubo (Webster et al., 1998). Aquila et al. (2013) and Dhomse et al. (2015) pointed to the hemispheric assymetry of the midlatitude ozone reponse to Pinatubo due to modified ozone transport from the tropics. Figure 8 compares monthly mean ~~tropical ozone~~ midlatitude ozone (30°-60° for both hemispheres) from SOCOL-AER simulations with the ~~ozone mixing ratio at 30 hPa from the merged satellite composite SWOOSH (Stratospheric Water and Ozone Satellite Homogenized dat~~

ozone column from the combined record SBUV (Merged Ozone Data Set version 8.6, McPeters et al., 2013) as well as with the ~~total ozone column from the combined record SBUV (Merged Ozone Data Set version 8.6, McPeters et al., 2013). We focused on tropics since ozone response most pronounced here compared to other regions (e.g. Rozanov et al., 2002).~~ lower stratospheric (20-70 hPa) ozone mixing ratio from the merged satellite composite SWOOSH (Stratospheric Water and Ozone Satellite Hom Anomalies are obtained by subtracting monthly means for 1991-1995.

Compared to SBUV, SOCOL-AER ~~captures the maximum ozone loss seen in observations well (around -0.3 ppmv and -7 DU), but leads to slightly premature ozone recovery in 1992. There are small differences among model scenarios but all of them generally fit~~ does show decline in ozone column, however underestimates it in the northern hemisphere (NH) and overestimates it in the southern hemisphere (SH). REF12, COAG, and UPWIND generally fit into the ensemble spread of ~~the reference experimentand have the same issue compared to independent observational time series SWOOSH and SBUV.~~ REF. REF also shows ozone increase in 1991 in the southern hemisphere (SH) discussed by Aquila et al. (2013) and Dhomse et al. (2015), but in the late 1991 a similar increase is shown by the model in NH, which is not seen in SBUV data. The noRAD and noQBO results show that parts of these ozone increases in SH and NH are due to heating by aerosols in the tropics and due to QBO, respectively, and both of these peaks disappear in the noRADnoQBO experiment. This hints that SOCOL-AER dynamics doesn't adequately respond to perturbations in the tropics, given that QBO is prescribed and post-eruption warming is well captured at least by the REF12 experiment (Fig. 6). In order to check this, we performed another experiment with the dynamics nudged to ERA-Interim reanalysis data with the rest settings kept as in REF. We used the same nudging procedure as described in Ball et al. (2016). The nudged experiment also shows some differences compared to SBUV, but in general reproduces its behavior much better, thus suggesting some problems in the model's dynamics and good performance of the model's chemistry and aerosol microphysics. Comparison of SOCOL-AER ozone with SWOOSH, composite of stratospheric satellite measurements, reveals the same conclusions for the lower stratosphere.

4 Conclusions and discussion

We have simulated the temporal and spatial development of stratospheric aerosols following the 1991 Pinatubo eruption, as well as temperature and ozone responses, using SOCOL-AER, a free-running 3-D global chemistry-climate model coupled with a ~~particle-size resolving~~ size-resolving aerosol module. The simulations explore the ~~rolse~~ roles of the QBO, aerosol radiative heating, sedimentation scheme and coagulation efficiency in the evolution of the stratospheric aerosol after Pinatubo.

The results show that QBO and interactive aerosol radiative heating play ~~a significant role~~ important roles in maintaining the tropical stratospheric aerosol reservoir over the whole course of the volcanic aerosol cloud evolution, significantly affecting volcanic aerosols lifetime. Furthermore, the results suggest that an accurate sedimentation scheme helps to ~~significantly~~ improve the model's ability to reproduce stratospheric aerosol. Numerically diffusive methods, such as a simple upwind method, must be avoided in modeling studies of large volcanic eruptions to prevent artificially fast spreading of particles to high and low altitudes. A more sophisticated coagulation scheme is capable of improving the comparisons with in situ particle size measurements as well as with satellite-borne extinction ratios, which are a proxy for particle sizes. On the other hand, the improved

coagulation scheme leads to too rapid sedimentation and loss of stratospheric aerosol mass, which become noticeable in the model about one year after the eruption.

There is significant uncertainty among the observational data of different aerosol parameters. Observations differ by up to ±15% in the global aerosol burden, ±30% in aerosol optical depth and spatiotemporal aerosol distribution in the two years following the eruption, ±40% in the effective particle radii, ±~~15%~~ 0.5 K in the lower stratospheric temperature anomalies. This renders the exact determination of the required emitted sulfur amount difficult. Thus, the vertically integrated tropical mass simulated by the reference experiment in 1991 (Fig. 1b) is in good agreement with HIRS, but later experiences faster decay that is not consitent with HIRS and SAGE-3λ but closer to SAGE-4λ. Considering this fact and relying on SAGE-3λ after 1991, we can assume that our 14 Tg estimate of initial emission was still sufficient for our model, but the vertical distribution of resulting aerosols could be incorrectly shifted to the lowermost levels. This fact could be responsible for ~~the main modelling deficiency~~ one of modelling deficiencies found, namely the 1-2 K larger warming that is inconsistent with temperature renalyses. It could also explain the integrated modelled burden diffrerence to SAGE-3λ since 1992 (Fig. 1), as the mass located at lower levels also sediments faster to the troposphere, despite increased buoyancy produced by addional warming. ~~However, if not relying on SAGE-3λ, our model reveals a very good agreement with AVHRR instrument in terms of AOD in the visible part of the spectrum (Figs. 4 and 5).~~ The experiment with the reduced emission revealed much better representation of the post-eruption stratospheric warming, but at the same time less optimal agreement with observations of other parameters. In terms of AOD in the visible part of the spectrum, our model is also closer first to AVHRR and later to SAGE-4λ than to SAGE-3λ. It is important to note that the period when all aerosol burden and AOD observational data overlap in 1992 is perfectly captured by the model. Observed features of the ozone response appear to be problematic to reproduce by the model in a free-running mode, which can be overcome by using a nudged mode. Potentially, both, aerosol lifetime and ozone response, can be improved with the increased horizontal and vertical resolution.

There is a rising interest of the climate community to the global models with interactive aerosol microphysics. It is caused partly by the ~~topic of~~ widely discussed climate geoengineeering, namely a compensation of the global warming by artificial emissions of SO_2 (e.g. MacMartin et al., 2016), as well as by the unclear role of major and smaller volcanoes in the future climate ~~(e.g. Bethke et al., 2017)~~(e.g. Bethke et al., 2017; Klobas et al., 2017). Considering other ~~studies modeling Pinatubo~~ modelling studies of Pinatubo effects, our simulations corroborate the results of Kleinschmitt et al. (2017) who also used a sectional model (LDMZ-S3A) and the same emission rate of 14 Tg of SO_2. Their results also revealed problems in reproducing aerosol sizes above 25 km and overestimation of the stratospheric warming, however they attributed ~~it~~ the latter to the fact that aerosol composition is prescribed during calculation of aerosol optical properties in LDMZ-S3A (Christoph Kleinschmitt, private communication, 2017), which is not the case for ~~SOCOL-AER~~SOCOL-AERv1.0. The reasons for ~~this overestimation in our case~~ these and other revealed problems are to be investigated, as SOCOL-AER still undergoes further development.

The recent Tambora model intercomparison study by Marshall et al. (2018) demonstrated that SOCOL-AER has substantial problems in representing the absolute values of sulfate deposition ~~to~~ in the polar regions, due to a simplified tropospheric deposition scheme, but also that SOCOL-AER has the closest agreement with ice-core observations in terms of timing of start and end of volcanic increase in deposition, which is defined by stratospheric aerosol lifetime. A model intercomparison

study for Pinatubo is planned within the framework of the Stratospheric Sulfur and Its Role in Climate activity ~~(SSiRC, http://www.spare-ssirc.org/),~~ (SSiRC, Timmreck et al., 2018), but, as was also shown here, aerosol observational uncertainty concerning the eruption of Mt. Pinatubo is high and will complicate the derivation of exact conclusions for certain processes and models. Another strong eruption similar to Pinatubo could significantly improve our understanding of the underlying microphysical and transport processes, given recent advances in measuring techniques (Kremser et al., 2016).

5 Code and data availability

SOCOL-AER model code and simulation results presented in this study can be ~~requested by contacting~~ downloaded from https://doi.org/10.5281/zenodo.1245196 and https://doi.org/10.5281/zenodo.1245202, respectively. In case of problems, please contact the corresponding author.

Author contributions. JXS and TS perfomed the simulations, visualized the data, and wrote most of the article. All authors are responsible for code development and discussion of results.

Competing interests. The authors declare that they have no conflict of interest.

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