<u>Replies to Anonymous Referee #2 (*bold-italics*)</u>

Interactive comment on "Aerosol microphysics simulations of the Mt. Pinatubo eruption with the UKCA composition-climate model" by S. S.Dhomse et al.

The paper is a solid piece of work but overall a model validation paper with less new scientific results. The motivation of the paper is very model specific: "Here, we use both satellite and balloon-borne measurements to evaluate the UMUKCA simulated stratospheric aerosol properties, and seek to better understand the source of model biases" and therefore not of general scientific interest. Hence, the paper would be better suited in a journal like GMD or JAMES. In the paper model results are compared to satellite and in situ observations in a very extensive way. Model and observations agree to a certain extent however substantial differences are found between model and observations in the peak aerosol loading, the number of small particles and the vertical extension. The discussion of potential errors remain often vague i.e. critical factors are discussed but a more concrete answer is missing. Additional sensitivity studies for some of the critical parameters e.g. nucleation, modal parameters (standard deviation, critical radius) could help to identify most critical processes, to assess the various model uncertainties and last but not least to improve the paper.

We thank the reviewer for his/her comments. Our study is the first ever that has attempted to understand the evolution of the profile of the stratospheric aerosol particle size distribution through the Pinatubo period. Our results clearly are specific to the model we have used, but our findings are relevant to a new generation of composition-climate models which seek to represent the stratospheric aerosol interactively, including with varying particle size distribution.

We respectfully disagree that our paper would be better suited to GMD or JAMES. Understanding the variation in simulated particle size distribution is a very important scientific issue since it strongly influences the strength of the radiative effects from the enhanced stratospheric aerosol.

We therefore would strongly argue that our paper belongs in ACP.

However, we do accept that in the ACPD version, our discussion of the results could have been better, and we have significantly improved this in the revised version.

Also, since the ACPD version, we discovered a bug in our code which had switched off intra-modal coagulation in the simulations, explaining the high bias in N5 & N150. With this bug repaired there is now good agreement with the balloon measurements.

Since the original submission, we have also implemented particle evaporation and include a source of "primary sulphate" issue, assuming 3% of the mass of SO2 emitted from Pinatubo forms particles at the sub-grid-scale.

As we explain in the reply to reviewer 1, Figures 2 to 11 now show results from two control simulations (20Tg and 10Tg), indicating upper/lower bounds for the amount of SO_2 ***emitted.***

We also now present a new Figure 13 showing results of sensitivity simulations investigating the impacts of scaling down the nucleation rate in the stratosphere and omitting the sub-grid "primary sulphate" source of particles from the eruption.

We assert that with these improvements our revised manuscript is suitable for publication in ACP and highlight the four main findings in our paper:

1) ***Satellite measurements indicated that shortly after the eruption between 14-23Tg of SO2 (7-11.5 Tg sulphur) was present in the stratosphere. However, best estimated of perk global stratospheric aerosol are in the range of 19-26 Tg, only 3.7-6.7 Tg of sulphur assuming composition of between 59 to 77%*** H_2SO_4***. So our simulations suggest that large proportion***

of the injected sulphur must have been removed from the stratosphere in the first few months after the eruption. So positive biases seen in our simulations must be due to lack of additional alternative loss SO_2 pathways that are not included in our model.

2) ***Injecting 20 Tg of SO_2 leads to an overestimation of the stratospheric aerosol sulphur, with an injection of 10Tg of SO_2 agreeing well with the satellite-based burden estimates.***
3) ***Global modal aerosol microphysics models are capable of representing the variation in particle size distribution in the strongly volcanically perturbed post-Pinatubo period.***
4) ***Simulated perturbations to stratospheric aerosol properties from large volcanic eruptions are robust to known uncertainties in nucleation rate and sub-grid particle formation.***

Major comments:
Role of aerosol induced heating. AOD
Timmreck et al. (1999) show in their paper the difference in aerosol optical depth (AOD) between an interactive and a non-interactive Pinatubo simulation. In their non-interactive simulation the maximum AOD is higher and the aerosol load is more constrained to the tropical regions. However, these differences are much smaller (less than 10%) than in the current paper (factor of two to four). Similar results as in Timmreck et al. (1999) are found in Aquila et al. (2012). This suggests that the lack of aerosol induced heating might contribute to the overestimation of the tropical aerosol load but is probably not the major cause.

We agree with the reviewer that the difference between the simulated and observed AOD can only partly be explained by our runs being non-interactive with the radiation. A key additional factor, as we now explain in the revised version, is that a 20Tg injection leads to too large a sulphur burden in the stratosphere. As we note in point 1) above a large proportion of the injected sulphur must have been removed from the stratosphere in the first few months after the eruption, through a process missing or underestimated in our model.

Our 2nd control run injects 10 Tg of SO2 and achieves good agreement with the stratospheric aerosol burden from satellite measurements. This simulation has a much lower tropical AOD high bias and we contend that this smaller bias can be explained by the lack of radiative coupling in these particular simulations.

Vertical extension
The authors discuss a couple of times the bias in latitudinal extent in the first post eruption months due to the lack of aerosol induced heating. I miss however a detailed discussion about the bias in the aerosol surface area density (SAD) in the lowermost tropical stratosphere (Figure 7). In the model simulations there are really high values in comparison to observations during the first year. Partially this can be explained due to the missing aerosol induced heating as seen in Aquila et al. (2012) and Timmreck et al. (1999). Both model simulations show however a clear distinct maximum which is not seen in the UKCA simulations. I wonder if this might also be related to a bias in the cross tropopause flux in the model or to numerical diffusion. Aquila et al. (2012) also directly injected SO2 in a non - interactive sensitivity study between 17 and 27 km and the bulk of their aerosol cloud remain between 50 and 20 hPa in December 1991(Figure 7 in Aquila et al. 2012).

See replies above. We also added discussion about this discrepancy in a revised manuscript.

Mode number and sizes
The definition of mode ranges and boundaries play a very crucial role for the evolution of the aerosol size distribution, as for example discussed in Niemeier et al. (2009). Looking to Figs. 9 and 10 and to Figure 11 in the paper it seems that no nucleation is occurring around 20 km from October 1991 onwards but there are still too many small particles until spring next year in the model simulations in comparison to the observations. This implies that probably the particle growth is underestimated in the model. Possible reason could be found in the condensation and coagulation parameterization but might be also related to the chosen modal parameters, i.e. the standard deviation and the transition radius or critical radius. Particles need to be shifted from one mode to the other if they are growing so

there exist very likely a transition or critical radius between the different modes in your model set up. How dependent are your results on these limits and on the selection of your size ranges? Sensitivity studies with respect to these parameters might help to explain the model bias and could be of general scientific interest.

We agree with the reviewer's helpful comments here. As explained above, we discovered a bug in our code which had switched off intra-modal coagulation in the simulations, explaining the high bias in N5 & N150. With this bug repaired there is now good agreement with the balloon measurements.

Coagulation
Did you consider particle coagulation as well? You did not write anything about it in your model description. Could not it be another potential error source?

See above – we discovered that intra-modal coagulation was switched off in the model in the original simulations, now good agreement with that bug fixed.

Further comments

P 2811 This is not clear to me. Do you use the expression of Kerminen and Kulmula (2002) in addition or as another option in general? How large are the differences between both applications if you use it as an additional option?

We use the Vehkamakki et al. (2002) expression to calculate the nucleation rate for particles at the nucleated cluster size of ~1nm and then use the expression of Kerminen and Kulmala to calculate the growth up to 3nm sized particles. This is a well-established technique in the GLOMAP model to represent the source of nucleated particles at observable sizes and is described by Spracklen et al. (2006).

P 2816, l21 0.15 Tg S instead of 15 Tg S

DONE

P 2817, l18 A factor of two is not slightly higher
replaced with "substantially" and now we have additional discussion re: the overestimation of the stratospheric aerosol sulphur burden in the 20Tg run and include results from the 2nd control run with 10Tg of SO2 injected.

P 2818, l25-29 The enhanced layer between 15-18 km originate from a small eruption plume from June 12 prior to the large one (e.g. Deshler et a 1993, Jaeger et al 1992), which injected material at lower altitudes
We have added reference to this now in the text.

P 2819, l20 replace "good" with "some"
Deleted "good"

P2820, l20-25 "lack of aerosol induced heating " I do not understand this argument because if aerosol heating would be considered the particles would be lofted to higher altitudes
Corrected. Here we meant aerosol heating also causes changes in local circulation, which can change horizontal transport through the subtropical barrier.

P2828, l17 Please clarify "why weaken particle growth would tend to reduce both N5 and N150 "
We meant that there may have been too many particles because nucleation was too high (explaining the high bias in N5). The point about particle growth was that a high N5 would lead to

reduced particle condensational growth too (condensed mass shared out over more particles). As described above, the bug we found meant that intra-modal coagulation was not operating in the model, and with that bug fixed, the model now has rather good agreement with N5 and N150.

P2828 l11-18 This could for example be a nice sensitivity study.
Thank you for the suggestion. As in earlier comments, we do now include a source of sub-grid particle emission (primary sulphate). One of our sensitivity simulations is with this primary sulphate emission from the Pinatubo eruption switched off (see Figure 13). The runs suggest that including such a sub-grid source has only a small effect on the simulated stratospheric aerosol evolution after Pinatubo.

Fig.9, 10 I would show only the Pinatubo simulated profile (except March 1991) this would make the comparison much clearer. You do not need the background files here.
We no longer show the noPinatubo run in Figures 9 and 10. Instead we show in the dashed line the 2nd control run with only 10 Tg of SO2 injected.

References:

Spracklen et al., (2006): The contribution of boundary layer nucleation events to total particle concentrations on regional and global scales, *Atmos. Chem. Phys.*, 6, 5631–5648.