Replies to Anonymous Referee #1 (*bold-italics*)

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

This paper describes the implementation of the GLOMAP modal aerosol scheme in the UKCA model. Subsequently, the stratospheric aerosol simulation is compared to observations after the eruption of Pinatubo and in quiescent volcanic periods.

Major comments:
The paper is well written and presents results in quite a large number of figures, which are described in detail. This makes the paper rather tedious to read. Moreover, the aim of the paper and the associated advancement of scientific knowledge are difficult to extract from the paper. As such, the paper would fit better in a journal like GMD, because that is exactly what it describes. My understanding, the scientific novelty of this paper is in the comparison of simulated size distributions with balloon soundings. However, the authors fail to explore these results further and do not work towards structural model improvements. With the model at hand they could perform interesting sensitivity simulations that would greatly enhance the science presented in the paper. Below, I give some suggestions that would make the paper suitable for publication in ACP. Without exploring one or more of these suggestions, I would advice the authors to consider GMD as journal to publish these results.

We thank the reviewer for his/her comments, but we respectfully disagree that our paper is mainly about model development. 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. Our study is the first to critically evaluate the simulated profile evolution of the stratospheric aerosol particle size distribution through the Pinatubo period. However, in the revised manuscript we have rewritten the Abstract to better highlight the key findings, reduced the length of the Model Description section and improved the discussion of the comparisons to the balloon measurements considerably.

Since the ACPD version of the paper was submitted, we have added particle evapouration and also now include sub-grid source of particles from the eruption (also known as "primary sulphate emission"). Also, we discovered a bug in the model setup used in the original paper, where intra-model coagulation had been accidentally switched off, which was the cause of the strong high bias in N5 and N150 in the original simulations. The original discussion re: the source of the N_5 and N_{150} high biases, and the lack of evaporation and primary sulphate emissions has therefore been removed.

In addition to updating the original 20 Tg Pinatubo run with the new model, we have added a 10 Tg run, and highlight that this lower emission run is in much better agreement with the global stratospheric aerosol burden derived from the HIRS satellite instrument (Baran & Foot, 1994). In the revised manuscript, Figures 2 to 11 show results from the 20Tg and 10Tg runs. Our simulations suggest that large positive biases observed in 20 Tg simulation must be due to lack of additional SO2 loss pathways mostly involving ash or ice in the volcanic cloud that is not included in our model.

Finally, we have followed two of the reviewer's suggestions, presenting sensitivity 10 Tg Pinatubo simulations exploring uncertainties in particle formation in the stratosphere. A new Figure 13 presents these results in terms of simulated timeseries of mid-visible AOD and number concentrations of particles larger than 150 nm radius alongside those from AVHRR and observations respectively. We discuss these results as testing the robustness of the model to uncertainties in aerosol microphysical processes.

We assert that with these improvements our revised manuscript is suitable for publication in ACP.

The definition of the aerosol modes. The size distribution the authors come up with differs substantially from the study by Niemeier et al. (2009). As I understand the description, the current study keeps the coarse soluble mode (mainly to simulate tropospheric sea salt aerosol), but with a sigma of 1.4 and uses the same sigma for the accumulation mode. They also state that the coarse mode is mainly populated in the troposphere and that similar sigma values are desirable (page 2812): "which is desirable numerically when the mode-merging algorithm transfers particles between the two modes in strong particle growth conditions". In the results section (figures 9 and 10) the authors note strong discrepancies between model and observations, especially on the separation of the N5 and N150 sizes. In the discussion the results are further analysed (figure 12) but no attempts are made to investigate the role of the mode definition. Further sensitivity studies exploring different definition would be a valuable addition to the paper, e.g. given the different definition in Niemeier et al. (2009). In sensitivity studies, the authors could define and test different mode bounds and explore the impact on the results.

We agree with the reviewer that the mode settings we use are different to those of Niemeier et al. (2009). Note also that we have now changed these modal settings from those used in the ACPD version. In the new simulations, we have returned the coarse soluble mode sigma back to 2.0, and switch off mode-merging from accumulation and coarse soluble mode, effectively using only 3 of the 4 modes to represent stratospheric sulphate aerosol. As we explain in the text, this ensures the coarse mode represents only sea-salt particles, with the largest enhanced stratospheric aerosol particles tracked within the accumulation mode.

Nucleation process. A recurring theme of the paper is nucleation. The authors could try reduced or enhanced nucleation rates, along the lines of what is written on page 2811: "Note that we also use the expression of Kerminen and Kulmala (2002) to convert the "real" nucleation rate from Vehkamäki et al. (2002) into an "apparent nucleation rate" at 3 nm." Another suggestion is already given by the authors themselves: emit part of the SO2 as sulfate, to influence the strongly H2SO4 dependent nucleation process.

As explained above, we now include these two new sensitivity simulations. Thanks.

Dynamical coupling. At various locations in the manuscript the authors suggest that the heating by the aerosol layer may be the reason for discrepancies between model and observations, because this effect has been excluded. Although I read that this is the subject of forthcoming studies, this paper would become less incremental if some results would be presented here, e.g. as sensitivity study.

We already added the extra 10Tg run and the two sensitivity experiments to particle formation and feel that is sufficient.

Evaporation of H2SO4. In the methods the authors discuss the possible evaporation of H2SO4 from the aerosols. However, they do not include this process, while at several places in the result section they blame model deviations in the upper stratosphere to this process. ("In reality the rapidly evaporating particles will release their H2SO4 to the gas phase causing a sharp reduction around 40 km. As well as biasing gas phase sulphur species, our neglect of particle evaporation will overestimate surface area density at the top of the aerosol layer." Or page 2825: "which is almost certainly due to the particle evaporation not being resolved in the model". Why not include a sensitivity study that quantifies the model improvement?

As we explained earlier, we now include evaporation of H_2SO_4 in the model. We have amended the discussion of the results accordingly.

Other major comments

In the abstract, the authors state: "Our comparison suggests that new particle formation in the initial phase of large eruptions, and subsequent particle growth to optically active sizes, might be playing an important role in determining the magnitude of the climate impacts from volcanoes like Pinatubo" Since the climate impact from volcanoes is not studied in these uncoupled simulations, this claim cannot be made here. I think the authors want to say that their model has problems simulating the correct size distribution after the eruption, and that this might have implications for the calculated climate forcing. They should rephrase this statement to reflect the work described in this paper.

Figure 13 now quantifies the sensitivity to uncertainties in new particle formation rates in the stratosphere, and we have therefore replaced this sentence with one summarizing those results.

The authors only analyse monthly output. Most of the time this is OK, but in some places in the manuscript it is not appropriate. For instance, on page 2816-2817 the SO2 lifetime is analysed from monthly burdens. This seems inaccurate to me. Better use daily burdens or budget terms on the SO2 + OH reaction. Figure 2 shows the global burdens on monthly timescales. So, a large fraction of the SO2 is already moved to aerosol sulfate within one month. At first sight it looks like something is wrong with the units (TgS), but later I realized that monthly burdens are plotted. I suggest to use daily output to avoid confusion. Also in figure 9 and 10 monthly model output is compared to individual measurement profiles. I understand that the model is run in climate mode, so sampling the model at a specific output time is meaningless. But showing the variability within a month (e.g. as error bars for modeling) would be quite relevant.

We have changed Figure 2 to show the daily variation in SO_2 and aerosol sulphur burdens. In Figures 9 and 10 we now include error bars which show the geometric standard deviation of the daily-mean size-resolved particle concentrations within the corresponding month.

Minor comments:
Page 2803, line 25: Guo et al. (2004) re-evaluated these numbers (18.5 Tg SO2, and 24 days). I think this should at least be mentioned.
We now refer to the 14 to 23 Tg uncertainty range presented by Guo et al. (2004) which reflects the 18+/-4 and 19+/-4 Tg values for TOMS and TOVS measurements respectively. We also present the 19 to 26 Tg range in the peak stratospheric aerosol burden from Lambert et al. (1993), and combine it with the 59-77% H_2SO_4 composition range to give a range of 3.7 to 6.7 Tg of sulphur. These values are also provided in the Abstract to explain the motivation for the 10 and 20 Tg runs.

Page 2804, line 13: "use" should be "used"
Done.

Page 2806, line 16: "the" before stratospheric should be omitted
Done.

Page 2807, line 14: pptm. Is this ppt by mass? Not commonly used, so clarify. Also in figure 3 I see that mass mixing ratios (ppt) are plotted, while commonly ppt is interpreted as pptv.
The UMUKCA tracers are mass mixing ratios so the correct units are pptm (parts per trillion by mass). However,in revised version we indicate all the concentrations are represented in volume mixing ratios. So in model description we say replace 550 pptm 275 pptv and vertical profiles shown in Figure 3 are also in volume mixing ratio (pptv, x-axis labels are corrected accordingly.)

Line 26: lead & leading in one line. Please rephrase.
Changed "leading to new particle formation" to "enabling new particle formation"

Page 2808, line 4: "at a fraction off". Unclear. Please explain. Line 14: "faster". Please specify "computationally faster"
Line 4: Changed to "The cross section of H2SO4 is assumed analogous to the cross section of HCl(X 0.016) following the method of Bekki and Pyle, 1992.

Line 14: Changed "faster" to be "computationally faster".

Page 2809, line 1-4. Please state somewhere that these issues will be described in the sections that follow.
DONE.

P2809, section 2.2.2: Unclear why a different density definition is needed in the stratosphere.
As we explain in the text, we implement a different density calculation to be consistent with the varying aerosol composition in the stratosphere.

Page 2810, line 4. You mean: "vapour pressure p over a flat surface"?
Yes. DONE

Line 17: 10156 has a unit. Please specify
DONE. Inserted unit atmospheres Kelvin (atm K).

Page 2812, line 14: 2x also in the same line
DONE (deleted second also)

Page 2813, line 12: unclear: "SO2 injection spread across eight model grid boxes between"
Changed to "SO_2 cloud from Pinatubo is initialized across eight ..."

P2814: Last two paragraphs in 2.3 would be more suitable in the results or discussion sections.
DONE

Page 2816, line 21. 15 Tg S. Should this be 0.15 Tg S? Line 22: format Textor reference.
Yes. DONE

P2817, line 1-14: I suggest using several months after the eruption for making a better estimate of the SO2 lifetime.
Time series changed in days. And now we calculate 35 days from the decay rate of the daily SO2 burden up to day 226.We also note that SO_2 conversion was much slower immediately after the eruption.

P2817, line 18: 36 Tg is not slightly larger than 21.5 Tg. I would call this substantial.
DONE

P2817, line 20: Unclear how the aerosol lifetime was calculated. If only stratospheric burden was used, than it should be clarified.
We have improved the text in the revised version and the method used is now clearly stated.

P2817, line 21: Replace 'model aerosol' by 'modeled Pinatubo aerosol'.
DONE

Page 2819, line 2: P-H2SO4 ? Please clarify
As mentioned in the caption P-H_2SO_4 is sulphuric acid in the particle phase.

P2821, line 1 and P2822, line 8: Saturation values given are different. One of them should be corrected.
DONE. it should have been 0.01km^{-1}.

Page 2821, line 17: unit?
DONE µm2/cm3

Page 2823, line 10: format reference.
DONE

Page 2827, line 6: “lower” should be “smaller”
DONE

P2830, line 8-10: Replace ’84 km’ by ’40 km’, since results presented here show aerosols only up to 40 km. Problems above 30 km due to missing evaporation should also be mentioned in this section.
DONE. New runs now include evaporation, so this section has now been amended.

Page 2830, line 22. Remove “tropical” before AOD
DONE

Replies to Anonymous Referee #2 (*bold-italics*)

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.