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