Reply to Referee #1

We thank the referee for the comprehensive review of our manuscript. We will improve the manuscript following the points made by the referee. Below are the replies to Referee #1. Original comments are in italic.

1. Performance of SALSA2.0 is compared to the ECHAM-HAMMOZ default (modal) aerosol module M7. Consequently, many of the differences in this work are attributed to the difference in the numerical treatment of the aerosol size distribution in modal or sectional schemes. Because of this, the manuscript does not really give an indication of how well SALSA2.0 performs compared to other sectional modules.

Comparing SALSA2.0 to other sectional models would be extremely interesting. However, to have a meaningful comparison of two aerosol modules, they should be compared within the framework of the same global atmospheric model. If the host atmospheric models are different, it is extremely challenging to distinguish if the differences in the simulated aerosol fields are caused by the differences in the aerosol modules or the atmospheric models. In another study (Saponaro et al. (2018) in preparation), we compare the ability of three aerosol models to simulate aerosol-cloud interactions, two of which (HAM-M7 and HAM-SALSA) share the same atmospheric model ECHAM, while the third aerosol module is in a different global model. In that study, we show that aerosol properties are much more similar for the aerosol modules which are in the same atmospheric model than for the aerosol module in a different atmospheric model. This conclusion is also supported by the study by Mann et al. (2014) where 12 aerosol models of different complexity were compared. Thus, conducting a meaningful comparison between SALSA and another sectional model would require excessive work and therefore it would not be possible within this study. We will mention this in the revised manuscript.

On the other hand, SALSA has been (e.g., Mann et al., 2014; Tsigaridis et al., 2014; Kipling et al., 2016) and will be included in the international AEROCOM project model experiments, where models are evaluated against each other and against observations.

2. The paper describes the difference between SALSA1 and SALSA2.0 well, but does not discuss the reasons why certain changes are made to the SALSA module. Please explain what the main problems with SALSA1 are and how these changes contribute to the improvement of the aerosol module.

The major change between SALSA1 and SALSA2.0 was how the concentrations of different chemical species in different sized particles are treated. In SALSA1, particles were separated into three subregions depending on the particle size. In the subregion for the largest particles (larger than 700 nm), only the particle number and sea salt, mineral dust, and a lumped “water soluble species” mass concentrations were tracked. This choice was done to keep the number of tracer variables to the minimum. However, this had two unwanted implications. First, the total amount of sulfate, organic aerosol, and black
carbon was lost when the particles grew to sizes larger than 700 nm. This was a practical problem in AEROCOM experiments where the information of individual species was required (e.g., Kipling et al., 2016). Second, although in the troposphere microphysics have very little influence on the size of particles in the third subregion, when simulating volcanic eruptions or stratospheric solar radiation management, condensation can grow the largest particles. This problem caused the model to have problems in simulating the growth of particles in a volcano plume since the third region particles did not grow which resulted in underestimating the effective radius of the volcano plume (Kokkola et al., 2009). In SALSA2.0 we extended the second subregion to cover also the size classes of subregion three. This way we can track the concentrations of all chemical compounds and the growth by microphysical processing is not limited in the largest size range. Figure 1 demonstrates how SALSA2.0 can simulate the growth of large particles while SALSA1 underestimates their growth. We will add discussion on these aspects in the revised manuscript.

Another major change was changing the size distribution structure from the moving center method (Jacobson, 2005) to the hybrid bin method (Young, 1974; Chen and Lamb, 1994). As explained in the manuscript, the moving center method causes numerical artefacts (Mohs and Bowman, 2011). We also have seen in our simulations that moving center consistently produces lower number concentrations. For example, in Bergman et al. (2012) study, SALSA1 significantly underestimated the number size distributions measured at EUSAARI sites whereas in this study we did not experience such underestimation. This underestimation of particle numbers in SALSA1 was analyzed to be partly caused by
the moving center method. In Figure 2 we show the relative difference AOD over the US between a model setups using the hybrid bin method and the moving center method. From the figure we can see that AOD is always lower when using the moving center method. We will add a brief discussion of this in the revised manuscript.

3. Previous work (also referred to in this article) has shown that modal aerosol representations do not perform well in simulating stratospheric aerosol caused by volcanic eruptions and that sectional approaches yield far better results. It is therefore not surprising that SALSA2.0 performs better than M7, but how does it compare to e.g. SALSA1? Overall, the discussion of the Pinatubo simulation is thin and mainly addresses issues in M7. As it is now, it might be better to remove this section from the paper as the remainder is already a very comprehensive comparison to observations.

The intention of this comparison is not only to evaluate if SALSA2.0 performs better than M7 but also to evaluate how well SALSA2.0 can simulate the evolution of the stratospheric aerosol that originates from the Mt Pinatubo plume. This comparison is also justified because the volcano plume is simulated with a sectional model and a modal model coupled to the same atmospheric model. The main result from this evaluation is that SALSA2.0 can simultaneously simulate both tropospheric and stratospheric aerosol without changing the setup. It has to also be noted that M7 and SALSA can be considered to be used for different types of studies. As a sectional model, SALSA would be a prefer-
able choice for simulating stratospheric aerosol, volcano eruptions, and geoengineering. However, we will add simulation results from SALSA1 and will also add discussion on the differences between SALSA1 and SALSA2.0 simulated volcanic aerosol.

4. The authors state that a size-resolved wet deposition scheme for SALSA2.0 is still under development. To make a fair comparison to M7 an older removal scheme is also used for those simulations. In my opinion, the quality of this paper would greatly improve if the new wet deposition scheme is included in this work. In the comparison to aircraft measurements in the results section it is also explicitly mentioned how the new wet deposition scheme would improve the results of the simulations with SALSA2.0.

In this paper, the focus in comparing M7 and SALSA is in comparing the modal and sectional approach and thus we kept the two model configurations as close as possible. The new wet deposition scheme of M7 is used in the upcoming paper by Tegen et al. (2018) which will provide a detailed evaluation of ECHAM-HAMMOZ with M7. The inclusion of the new wet deposition scheme would in SALSA also be a valuable addition, however, here we present the release version ECHAM6.3.0-HAM2.3-MOZ1.0 which does not include the new wet deposition scheme. The work on the new wet deposition scheme is underway, but it is by no means a trivial task to include such a new scheme in the model. Simulating wet deposition properly in a global scale model is a major challenge for the whole aerosol modelling community. The improved wet deposition schemes in M7 are for a modal scheme and can not be applied to SALSA2.0. In order to implement an improved sectional wet deposition scheme in SALSA2.0 will require extensive model development, testing, and analysis and could be a topic of an independent article (see e.g. Croft et al. (2009, 2010)). Consequently, it is beyond what can be done within this manuscript.

5. Title / Page 8, line 9-13 Judging from the text, the MOZ module is not used in this work and HAMMOZ is reduced to HAM only. This is a bit misleading and causes confusion in the text. It is not clear what value the combination of HAM and MOZ has in this work. It should be clarified better what MOZ does in the model simulations for this work, otherwise it might be better to remove MOZ from the title.

We will clarify in the revised manuscript that MOZ is not used in these simulations. However, the model licence requires that when publishing results with the model, the full name ECHAM-HAMMOZ is used and the exact release reference is stated.

6. Page 2, line 31 100 size classes is a bit of an exaggeration, in global models the number of size classes is usually (much) lower.

Here we refer to the study by Mann et al. (2014) where the GEOS-Chem-APM model is reportedly using 100 size classes.

7. Page 5, line 21-23 Two subranges of SALSA1 are combined into one in SALSA2.0, what is the reason for this simplification and what are expected changes in the simulated aerosol size distribution?
See our reply to the second referee comment.

8. **Page 5, line 25-26** The moving center method is replaced by the hybrid bin method. What are the downsides to this method as it was not used in SALSA1 before?

   The moving center method was not chosen for SALSA1 because of downsides of the hybrid bin method but because it seemed to be suitable for global scale models. Like all size distribution methods, hybrid bin method suffers from numerical diffusion (e.g. Dinh and Durran, 2012). However, in our model framework it improved the correspondence between the simulated and the observed aerosol size distributions.

9. **Page 7, line 8-9** In view of the importance of meteorology for e.g. dust emissions, what is the nudging time interval used in the simulations?

   The relaxation times for the nudging of the surface pressure, vorticity, and divergence are 24 h, 6 h, and 48 h, respectively. We will add this information in the revised manuscript.

10. **Page 8, line 14-34** The different parameterisations of SALSA2.0 and M7 are explained extensively in Section 2.3, but several of these are changed for the simulations. This is very confusing and makes large parts of previous section irrelevant. It would be better to describe what is actually used. Also, this means that this work presents results from a suboptimal model run and the full potential of the SALSA2.0 module in the ECHAM-HAMMOZ model is not shown.

   With respect to SALSA, the only parameterization that was changed was secondary organic aerosol (SOA) formation using volatility basis set (VBS). We will remove the row describing SOA for both SALSA and M7. This setup is the default for the model release. It may be suboptimal for simulations of SOA formation. However, using VBS increases computational time and significantly increases model output to the extent that it may not be practical to use it for simulations that do not focus on organic aerosol.

11. **Page 9, line 26** How is this ensemble constructed? What is the difference between the 5 members?

   We have perturbed offline anthropogenic aerosol emissions by values of the order of $10^{-6}$ which is an insignificant number for emission strengths but due to the chaotic nature of the atmospheric model, changes the model dynamics sufficiently. This will be clarified in the revised manuscript.

12. **Page 13, line 6** Judging from this work, the implementation of the new Long et al. (2012) sea salt emission decreases the model performance, why was it introduced?

   This is only true regarding aerosol mass. An upcoming study by Tegen et al. (2018) will show that the comparison between the measured and modelled number size distribution improves going from Guelle et al. (2001) to Long et al. (2011) parameterization. In addition, they show that aerosol optical depth compares better with MODIS retrieved AOD’s when using the Long et al. (2011) parameterization.
13. **Page 15, line 4** Here, the low AOD bias is (almost) completely attributed to the low SS emissions. Although this assumption is acceptable for the SH, there is also a strong bias in the NH high latitudes. Here, the low bias over the land masses cannot be attributed to sea salt only.

   This is a very good point. The low bias at high latitudes can also be attributed to issues in aerosol transport. Bourgeois and Bey (2011) have shown that ECHAM-HAMMOZ underestimates the aerosol load over the high latitudes. We will add discussion on this in the revised manuscript.

14. **Page 15, line 13-14** How did you arrive to this conclusion?

   We came to this conclusion comparing modelled and observed time series. However, we will remove this line from the revised manuscript since these results are not shown in the manuscript.

15. **Page 15, line 25** Why is this not mentioned in Section 4.1.1?

   We will add discussion on MODIS biases to Section 4.1.1

16. **Section 4.2** Restructure section. Multiple species of multiple model runs are compared to multiple measurement networks. This already makes the discussion hard to read. I suggest a fixed format/structure in discussing the different species to help the reader.

   We will follow the suggestion of the referee and change the structure to separate different species.

17. **Page 18, line 1-22** Include comparison results of M7 in discussion of SU/BC.

   We will add M7 results for sulfate and black carbon in the revised manuscript.

18. **Page 19, line 9** Aerosol load over oceans is not low over SH subtropics.

   We will rephrase this to state that AOD is low south of latitude 40°S.

19. **Page 19, line 15** If periods in observations and simulations are different, how are they collocated?

   The monthly model values are constructed by averaging daily means only for days where an observation is available. Moreover each model monthly mean is spatially collocated to the location of the observation station (by bi-linear interpolation). We will clarify this in the revised manuscript.

20. **Page 22, line 20** Why are monthly mean model values used here? Having 3 hourly output, collocation can be greatly improved. Also, comparison for SU and OA is based on daily mean output. What is the reason for this inconsistent approach?

   This was done to enable an easier comparison between earlier model experiments. For black carbon, we reproduced the AEROCOM experiment by Koch et al. (2009) which
used monthly mean values. For sulfate and organic aerosol, we reproduced the model evaluation by Heald et al. (2011) where daily values were used.

21. Page 25, line 5-6 This conclusion is too strong and drawn too quickly. It would be the case for the ARCTAS Spring and ARCPAC campaigns, but for the ARCTAS Summer, the wet deposition scheme barely influences the results for the lower part of the atmosphere where observations are available. Also, for the source regions, an increase due to the wet deposition scheme, would increase the already high bias in SALSA.

This is a good point from the referee. This is purely speculation and we will remove this sentence from the revised manuscript.

22. Page 26, line 1 Difference was fairly small”. Can this statement be quantified?

We have quantified this for the revised manuscript as follows. “For most campaigns and height levels, the relative difference in black carbon mass concentration is less than 50% and the shape of the vertical profiles are very similar, mostly overlapping each other. The largest difference is for black carbon in the CR-AVE campaign where the relative difference in the mass concentration is ~83% in the lowest layer.

23. Section 6 In this section, it is explained why the section approach of M7 does not perform well in simulation the stratospheric aerosol burden resulting from a volcanic eruption. There is even a reference to a solution for this problem. Yet you dont incorporate this in your model and compare the performance of SALSA2.0 mainly to the simulation with the unadjusted M7 scheme. As a result, it is difficult to really judge how well SALSA2.0 performs in these simulations. It would be more interesting to see the performance of SALSA2.0 to other sectional aerosol modules or modal schemes that were properly adjusted.

Implementing a modal structure with varying mode width would require rewriting major part of HAM and M7. In addition, it would increase the amount of tracer variables which would be undesired for M7 in which the number of these variables is kept to the minimum. As it is said in the manuscript, the mode widths of M7 have been optimized for the troposphere and were not designed to represent stratospheric aerosol.

Regarding comparison to another sectional model, a meaningful comparison to another sectional model would have been too challenging to do within this study (as was explained above).

24. Page 26, line 1-2 How are the model values and observations collocated?

In the simulations, where we used only year 2010 values, for black carbon, we used the modelled monthly values from the flight path for the corresponding month of the year. For sulfate and organic aerosol, we used the modelled daily values from the flight path for the day of the year which corresponded to the observations. For the whole period, we
used the same values, but also for the corresponding year of the observations. We will clarify this in the revised manuscript.

25. **Section 7. Conclusions** The structure of the conclusion section is unnecessarily confusing. Follow same order as discussion in Results section.

   We will restructure Conclusions as suggested.

26. **Page 29, line 6** What are recommendations for optimizing?

   The details will be explained in an upcoming study by Tegen et al. (2018).

27. **Page 30, line 1-2** Underestimation of particle number in SALSA1 not mentioned in Section 4.1.1.

   Here we refer to a study by Bergman et al. (2012) and will add the reference in the revised version.

28. **3 Technical comments** Page 4, line 12 Sentence is not clear, please rephrase.

   We will rephrase this as: "In its default setup, HAM uses the modal approach together with the model aerosol microphysics module M7 (Vignati et al., 2004).

29. **Page 5, line 13** "using the volume ratio" → "using volume ratio"

   This will be corrected in the revised manuscript.

30. **Page 7, line 10** Add full name of PCMDI

   We will add the full name in the revised manuscript.

31. **Page 10, line 21-23** Why is India omitted from this list?

   We will add India to the list.

32. **Page 11, Figure 2** Add statistics (e.g. corresponding global mean AOD to a,b,c and correlation coefficients and NMBs to d,e) to the figure for a good overview between model configurations.

   We will do as suggested.

33. **Page 12, line 2** Equation straightforward, can be removed and explained in words.

   We will remove the equation.

34. **Page 13, line 1** Fig 4. → Fig. 5

   We will correct this.

35. **Page 14, line 12** Reference to current section.

   We will remove the reference.
36. Page 15, line 17 Add minus sign to 0.05.
   We will correct this.

37. Page 16, Figure 6 Change colours of Asia and North America. These are the two regions discussed in the text but hardly distinguishable from each other. Also, adding a regional mean values would provide a good overview of model performance.
   We will add regional values to the revised manuscript. However, to us the colors look sufficiently different and since the values for the two regions are completely different, they are easy to separate. We do not see the need for changing the colors.

38. Page 17, Figure 7 Remove additional abbreviations of species in lower left corner of each panel. Add names of network to panels.
   We will do as suggested.

39. Page 20, Figure 9 Observed and simulated values in year 2010? Please add to caption.
   This is correct and will be added to the figure caption.

40. Page 22, line 32 Vertical profiles of AMMA, ARCTAS Spring and OP3 are not captured well either.
   We will mention this in the revised manuscript.

41. Page 26, line 1-2 Add references for the HIRS and lidar observations.
   We will add the references.

42. Page 29, Fig. 14 Add errorbars to observed values.
   We will add the uncertainty estimates of the observations in the figure caption.

43. Page 30, line 23 configurations → configurations
   We will correct this.

References


