Reply to Referee #2

We thank the referee for a comprehensive review of our manuscript. Addressing the points made by the referee will improve the paper. Below are the replies to Referee #2. Original comments are in italic.

1. *However, some of the principal statements made in the Abstract are not supported by the results presented in the paper, and much more care needs to be taken in the statements interpreting potential reasons for differences between the sectional and modal schemes.*

   In particular, the authors claim that the size distribution comparisons shown in Figure 4, shown for locations where AOD difference is largest, are indicative that of different microphysical processing in the modal and sectional schemes, and that this is the reason why the two aerosol schemes predict different extinction/AOD in these regions. But from careful inspection of Figure 4, its clear that is not the case. The authors already identify the two locations (in China and in Russia) as regions where the observed AOD is very high, and clarify that the Russian site is in a region where biomass burning emissions are high. The China site is in a region of strong anthropogenic emissions. The size distribution of the black carbon (the black bars in the stacked bar chart) are very different between the SALSA and M7 simulations at both locations and this clearly indicates that there is a systematic difference in the size at which primary carbonaceous aerosol particles are emitted, which is a much more likely explanation of the reason for the difference.

   At the China site, the M7 run has about half of the BC in particles larger than 200nm, whereas for the SALSA run this is only about 10site, indicating that there is a systematic difference between the two schemes in the sizes assumed for primary carbonaceous emissions. This is an important issue, because if such large differences in AOD could indeed be attributed to the simpler modal scheme having inadequate representation of microphysical processing compared to the bin scheme, then this could be cited in the literature extensively as a reason to justify the additional expense required for sectional aerosol schemes.

   It is noticeable that whereas the Table 1 explains in detail the size segregation of the sea-salt and dust emissions, there is no information given about the assumed size at which the carbonaceous particles are emitted (yet these are the dominant primary aerosol in polluted regions).

   I am sure this is just an oversight in the writing of the paper, and that there was no intention to omit this information, or to make a statement that is not supported by the results.

   1) Abstract, page 1, lines 5-6 As per my main comments above about Figure 4, this sentence is not supported by the results and needs to be removed or reworded. If the authors can repeat either the M7 or the SALSA simulations with the emissions size distribution for emitted carbonaceous particles identical in the two schemes then it may be possible to make some statement about this, but the different BC-size-distribution in M7 clearly

indicates there is a substantial differece in the "emissions size distribution" applied for carbonaceous particles in the two aerosol schemes, which is much more likely reason for the difference in AOD between the two schemes. In any case the locations shown in Figure 4 (in Russia and China) are regions of very strong primary emissions. Lee et al. (2011) apply a perturbed parameter uncertainty analysis to show how (at least for the global aerosol microphysics scheme applied there) the regions where aerosol properties are most dominated by uncertainties in microphysical pro- cesses are away from such "emissions hot spots". So even if one of the models was re-run with the same "primary emissions size distribution" as for the other, one might expect any difference from microphysical processes to have most impact in a different region than the two locations shown.

It is true and a good point that differences in emission sizes could explain the differences over areas with high anthropogenic emissions. However, in our simulations this is not the case since for offline anthropogenic emissions, we use identical emission size distributions for SALSA and M7 (see Page 7 in the manuscript). There will be some difference resulting from remapping modal emissions to SALSA size classes, however the emission masses and numbers and their size distributions are identical for M7 and SALSA. The significance of microphysics calculation over the chosen areas can be demostrated by two SALSA runs: one where condensing organics are treated either assuming them to be non-volatile and one where they are assumed to be semi-volatile, however so that the resulting secondary organic aerosol yield is approximately the same. In Figure 1 we show how the simulated yearly mean aerosol optical depth (AOD) compare against MODIS (Figure 1a) over China. Panel b shows the AOD with non-volatile OA and c with semi-volatile OA. From the figure, we can see that although everything else except for the microphysical treatment of OA is exactly the same, there is a large difference in simulate AOD's. Other size dependent processes such as wet removal do also contribute to the difference, but the changes are initiated by differences in microphysics.

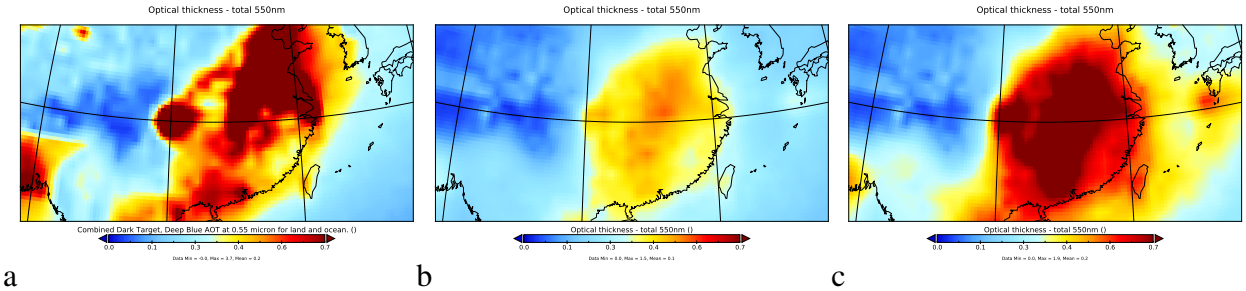

Figure 1: Retrieved aerosol optical depth by a) MODIS, b) SALSA2.0 with non-volatile organic aerosol, and c) SALSA2.0 with semi-volatile organic aerosol.

Lee et al. (2011) studied the sensitivity of CCN on different microphysical processes. The conclusions from that study do not necessarily apply for AOD. It may also be that the sensitivities of a modal model such as GLOMAP-mode (which was used by Lee et al. (2011)) and a sectional model can be different. We have shown previously that mode merging causes some damping effect on the sensitivity of CCN sized particles (Korhola et al., 2014)

We will add a more detailed description of the emission sizes in the revised manuscript. However, we don't see a reason to change our conclusions on this matter.

2. *It is also clear from Figure 2 that, for many regions, the M7 scheme could actually be argued to perform better (compared to the MODIS AOD) than the SALSA scheme. SALSA seems to have substantial bias over North Africa and over marine regions in the Southern tropics, for example.*

   This is true and we will add regional values for the comparison between the modeled and AERONET AOD for both SALSA and M7 for a more detailed comparison and discussion.

3. *One final general comment was that it needs to be stated somewhere early on in the text the difference between the acronyms "HAM" and "M7". My understanding is that "HAM" is the overall modal aerosol scheme and that M7 is a component of HAM, basically the modal microphysical routines.*

   The reason I ask for this clarification is that I was expecting then the SALSA to not just be an alternative to M7, but an alternative to HAM, and that perhaps the correct naming should then be ECHAM6.3.0-SALSA2.0-MOZ1.0 when the SALSA scheme is applied.

   Please can the authors clarify I am understanding this correctly.

   However, perhaps that is not quite right and the implementation of SALSA into the model has in fact only implemented the microphysics routines within SALSA (or indeed that SALSA has always only been the microphysics routines). I realise that within HAM there is a separate acronym for the microphysics routines (M7) than the overall modal framework, which is known as HAM. By contrast many other aerosol schemes do not have this distinction and there is only one acronym for the overall aerosol module including both the microphysics routines and the other aspects (primary emissions, dry deposition, scavenging). The naming convention of the different parts of the model are important in this case as it helps the reader to appreciate which aspects of the HAM scheme have been retained in the implementation of SALSA.

   I realise different groups will have different ways of naming their modules and Im not necessarily suggesting the SALSA group come up with a new acronym for the microphysics elements of SALSA. However I do think it needs to be stated somewhere in the section 2.2 description exactly what constitutes the Hamburg Aerosol Model and what are the SALSA aspects. See also my first specific comment about the wording of the title.

You have understood the difference between HAM, M7 and SALSA correct. As explained in Section 2.2, HAM is the aerosol model which calculates aerosol emissions, removal, hydration, and radiative properties. M7 and SALSA are the aerosol microphysics modules of HAM which uses either modal or sectional approach depending on which aerosol microphysical module is used. Most of the actual HAM code is shared with both M7 and SALSA. This is why the model is called ECHAM-HAMMOZ. In addition, 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. We will further clarify this in Section 2.2.

2) Abstract, page 1, lines 9-11 This difference in the modal and sectional aerosol microphysics predictions for the microphysical evolution and global dispersion of the Pinatubo volcanic cloud is interesting, but, as the authors point out, the standard mode widths for M7 are not intended to be applied to the stratospheric aerosol evolution. Figure 14 shows that actually, provided the model is applied with the "stratospheric- enabling adjustment" to the accumulation mode and coarse mode widths, the modal scheme compares well to the sectional scheme. As I understand it, the Hamburg stratospheric aerosol modelling group would not apply the model without this adjustment to the mode widths, so the emphasis really needs to be changed in how this is worded in the Abstract and in the discussion of the results. I think it is very important, to minimise the chance of an incorrect inference from the reader, to present the esults having that "M7mod" essentially as the default (or even "validated"?) configuration for when the model is applied for simulating interactive stratospheric aerosol. Indeed I would strongly recommend to change the "branding" of that model run to "M7-strat" rather than "M7-mod". As I understand it, the adjustment to the mode widths is a pre-requisite for simulating stratospheric aerosol for that scheme, so the authors of the manuscript need to change the current wording of the results to be clearer that it is essentially "the stratospheric configuration of M7" or so. One could consider it in some ways equivalent to a tropospheric or stratospheric chemistry scheme. One would not apply a tropospheric chemistry scheme to simulate the chemistry of the stratosphere.

This is a very good point and we will add an explanation that the modified mode width used in the Pinatubo comparison is the stratospheric setup of M7, which is untested for the troposphere. We will also add references where it has been used: Niemeier et al. (2009); U. et al.; Niemeier and Timmreck (2015); Niemeier and Schmidt (2017). We will also change the model run name from M7mod to M7-strat following the suggestion of the referee. On the other hand, we need to emphasize that this setup is not a feature in this model release and using it requires code level changes.

List of minor revisions

Referee comments 3, 5, 6, 8, 9, 10, 12, 14-21, 24-29, 32, 36, 39-41, 49, and 50 suggest rewording and changes in the terms used. We will make the changes as the referee suggests in the revised manuscript.

1) Title, page 1, The way the title is currently worded suggests the SALSA2.0 module is as a new sub-model within the Hamburg Aerosol Model (HAM). Is that the case then that HAM includes both M7 and SALSA as alternative aerosol microphysics modules? Or is the SALSA module an alternative to "the overall HAM" or so?

SALSA2.0 is a new submodel and an alternative to M7 within HAM and a component of the release version of ECHAM-HAMMOZ. We will clarify this in the revision.

2) Abstract, page 1, 1st sentence Related to point 1) is ECHAM-HAMMOZ still ECHAM-HAMMOZ when SALSA is applied or should it then be referred to as ECHAM-SALSAMOZ or so?

As explained above, the model name remains ECHAM-HAMMOZ even with SALSA turned on.

4) Abstract, page 1, 3rd sentence insert "within ECHAM" or "within ECHAM- HAMMOZ" between "implementation" and "is evaluated" to be clear it is this particular implementation that is evaluated (one could imagine it potentially being implemented in another framework at some point in the future).

We will insert "within ECHAM-HAMMOZ" in this sentence.

7) Abstract, page 2, lines 1-2 as per major comment 1-2, this sentence needs to be changed since (as I understand it) the M7 microphysics would not be applied for stratospheric aerosol applications unless the mode widths for the accumulation and coarse soluble modes were reduced to 1.2 in this way. In this sentence and the results of the Pinatubo comparisons, I this should be referred to as "the stratospheric aerosol configuration of M7" or similar.

We will refer to the M7 setup with modified mode widths as "the stratospheric aerosol configuration of M7" in the revised manuscript.

11) Introduction, page 2, lines 8-9 The words "at the lower end of the size spectrum of nanometer size in diameter" somehow seemed a strange wording. The term "lower end" seeemed odd suggest to replace that text above with something more linked to their formation process, replacing "at the lower..." with "freshly nucleated particles are observed at nanometer sizes..." then the rest of the sentence can continue with "as they grow....". Then similarly instead of "upper end of the spectrum" suggest "coarse part of the spectrum".

It is true that the original wording is too complicated. We will rephrase this to "For example, when the nanometer sized smallest partices grow in size, they contribute to the number of aerosol particles which can form cloud droplets (Kulmala and Kerminen, 2008) while the largest particles of micrometer size affect rain formation (Jensen and Lee, 2008)."

13) Introduction, page 2, lines 11-12 change the start of this setence to be more specific about the size effect you are explaining in simple terms it can be understood simply as

particles only interacting effectively with the radiation once theyre above a certain size. Id suggest to re-word the sentence to something like "There is a steep size dependence for how effectively aerosol particles interact with radiation (Chung et al., 2005) and clods (Lohmann and Feichter, 2005)." Suggest also to cite the chapters 7 and 8 of the 2013 IPCC AR5 report rather than the 2005 references given there i.e. Myhre et al. (2013) and Boucher et al. (2013).

The size dependence of particles on radiation and cloud formation is so complex that we do not want to go into more details as it would require a lot of text to explain it comprehensively. This sentence was meant to only briefly mention the size dependency of these effects. We will add the suggested references.

22) Introduction, page 2, line 28 there is also the Piecewise Lognormal Approxi- mation (von Salzen, 2006) which has each size section represented as a log-normal distribution. Please add that as another approach here.

We will add mention this approach together with the reference in the revised version of the manuscript.

23) Introduction, page 3, line 1 need to be more careful with this explanation here. Suggest to re-word the end of this sentence instead to say "the application of sectional models in global 3-D simulations often involves a trade-off with horizontal or vertical resolution" or similar.

Here we mean to say that in global 3D models so many other processes than aerosol microphysics affect the atmospheric aerosol properties that the improvement due to a higher aerosol size resolution is not evident. We will clarify this in the revised manuscript. With SALSA we do not have a trade-off with horizontal or vertical resolution compared since they are the same for SALSA and M7.

30) Section 2.2. lines 11-13 The paper has not quite explained what is the distinction between HAM and M7. Until reading this I thought they were the same thing but I think I now understand that "HAM" is the overall aerosol module (including emissions, dry deposition, scavenging etc.) whereas M7 is just the aerosol microphysical routines. Am I understanding that correctly? If so this needs to be stated explicitly somewhere here in so-doing it will help ensure the community apply the acronyms correctly and consistently in future.

You have understood it correctly. It seems to be a common misunderstanding that M7 and HAM are synonymous. We will clarify the differences in the revised manuscripts.

31) Page 5, section 2.2 line 20 suggest to replace "represents several real-life compounds" with "represents several specific single-species compounds" if that is what is intended?

We will replace "real-life compounds" with "individual chemical compounds".

33) Page 6, Table 1 need to add additional entries to the "emissions" section for primary carbonaceous and give the different size assumptions for emitted primary carbonaceous particles from biomass burning, bio-fuel and fossil-fuel sectors. As per my major comment 1, I think this is the primary reason why there is the AOD different in those strong emissions regions. You can see that the BC size distribution is at different sizes in the sectional and modal scheme, and I think this can simply be explained by a different size assumption I would be very suprised if that was caused by microphysical processing.

We will describe more in detail the emission size distributions in the revised manuscript.

34) Page 7, line 7 be clear what you mean by "coupled" you mean "radiatively-coupled" right? Need to add an extra sentence briefly explaining how thats done here for aerosol-radiation interactions and aerosol-cloud interactions in the sectional scheme (and how that differs from the radiative coupling when the modal scheme is used).

Here we mean that we run ECHAM-HAMMOZ with SALSA2.0 aerosol microphysics. We will rephrase this accordingly in the revised manuscript.

35) Page 7, line 10 you write "we used the climatologies" but I dont think you mean climatologies here do you? What is the time-variation of the specified SST and sea-ice distributions?

We do use sea surface temperature and sea ice cover climatologies. They are monthly fields which we will clarify in the revised manuscript.

37) Page 8, line 16 you write "For most of the processes the difference is only in the numerical treatment" but thats not quite right the nucleation processes are different as shown in Table 1 please change this wording.

We will rephrase this part as follows: "In the default setups of M7 and SALSA2.0, wet deposition and secondary organic aerosol (SOA) formation are the only processes (in addition to the calculation of aerosol microphysics) that use different methods for solving the physics of the process. For the rest of the processes the difference is only in the numerical treatment.".

38) Page 8, line 25 you write "more detailed size-dependent scavenging rates" but you need to add a few qualifying words so the reader knows what you mean by "more detailed" here. The reader might expect the sectional SALSA scheme to have more detailed scavenging than the modal scheme or maybe you dont mean detailed in a size-resolved way do you mean the way the scavenging applies different scavenging efficiency for the different types of precipitating cloud?

We will modify this part to indicate that the wet deposition scheme is more physically based instead of using the ambiguous term "detailed".

42) Page 9, line 9 As per my major comment 2, it is not fair to refer to the initial settings of the scheme as "The default settings". They are indeed the default settings for tropospheric aerosol simulations, but they are not the default settings for stratospheric

aerosol simulations. As per my major comment 2 please change the branding of these two M7 simulations from "M7" and M7mod" to "M7-trop" and "M7-strat". They are al- ternative configurations of M7 specifically for those applications. Its fine to show that simulation with the tropospheric configuration of M7 in fact that will show why its important to only apply the scheme in the stratosphere with the stratospheric configuration (M7-strat). But you need to change the wording so that its clear that this is only default for tropospheric aerosol application of the model.

This is true and we will modify this part as explained in the reply to the major comments.

43) Page 9, line 11 The authors write "This is because the high concentration of sulfur produces a bi-modal aerosol population". Is this statement referring to the Laramie balloon-borne OPC measurements (Deshler et al., 2003) which show the bimodal size distribution after Pinatubo? If so please give that reference here.

Here we refer to the model study (Kokkola et al., 2009), but we will also include the Deshler reference.

44) Page 9, line 12 the narrowing of the width again I think you are referring to what is observed from the measurements right? That is the case that the accumulation mode is observed to have a narrower size distribution cite Deshler et al. (2003) or Deshler (2008).

Here we refer to the detailed aerosol microphysics model which was used in the Kokkola et al. (2009) paper.

45) Page 9, line 14-16 youre referring to the box model simulations here, right? Its not so clear how the effect plays out in 3D simulations, and more so when you consider the trade-off in the better stratospheric circulation that can be afforded (by resolving more vertical levels for example) with a computationally faster aerosol scheme. So you need to be clear that youre referring here to here is what is seen in a box model. For a balanced discussion of this, you also need to add a qualifying sentence explaining this trade-off between the cost of the aerosol scheme and the cost of the atmosphere model.

Yes, we are referring to the box-model study Kokkola et al. (2009). However, the same effect can be seen in our global simulations where we can see that the largest particles are removed faster in the tropospheric setup M7 and is evident in the manuscript Figure 14b.

The difference in the computational speed between M7 and SALSA is not so large that they would use different grid resolution. We will however mention the increase in the computational burden when using SALSA.

46) Page 9, line 15 you need to re-word "grows too fast and the particles are sedimented too fast". The box model shows that in those simulations the growth proceeds faster, but you do need to qualify the 2nd part with "which would result in particles sedimenting faster" or something like this. Since it has not really been demonstrated in global models you need to tone down the way that is described.

We will modify this sentence as suggested. However, we investigated the difference between the growth of particles between the two M7 simulations. With the default mode widths, the coarse particle burden grows quicker right after the eruption and these particles are consequently removed much faster than in the case of using narrower modes.

47) Page 9, line 17 It is not appropriate to refer to this as "A work-around solution". The "code-owners" of the M7 scheme are clear in their publications that when the scheme is applied for stratospheric aerosol applications, the modal settings need to be configured differently than for tropospheric aerosol applications. Thats not correct to refer to that as a work-around. Effectively the scheme is only "licensed" to be applied in the stratosphere if it has this adjustment to the modal settings. As per my major comment 2 this section needs to be re-worded to make this clear in my strong opinion, for the reasons above, you should refer to the tropospheric aerosol and stratospheric aerosol configurations of M7, and label them as "M7-trop" and "M7-strat". That is then consistent with the way the owners of the adjusted scheme have re-configured the model to be applicable for the stratosphere.

We will change the label to M7-strat. However, it has to be noted that the model version we present here requires code level changes to start using the alternative stratospheric aerosol configuration. It also has to be noted that there is no release version of ECHAM-HAMMOZ, which would support easy switch to this configuration. We will also remove the term "work-around".

48) Page 9, line 22 the Guo et al. (2004) has the SO2 emissions range as 14 to 23 Tg of SO2 you need to give that range (and any widening of that to include values from other publications).

We will give this range in the revised manuscript.

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