

## ***Interactive comment on “SALSA2.0: The sectional aerosol module of the aerosol-chemistry-climate model ECHAM6.3.0-HAM2.3-MOZ1.0” by Harri Kokkola et al.***

**Anonymous Referee #2**

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This manuscript describes the implementation of a sectional aerosol micro-physics module (SALSA2.0) within the composition-climate model ECHAM-HAMMOZ (ECHAM6.3.0-HAM2.3-MOZ1.0), as an alternative to the existing modal aerosol microphysics scheme "M7". The paper then evaluates aerosol optical properties, aerosol mass and particle size distribution simulated by ECHAM-HAMMOZ-SALSA, comparing to observations, and to aerosol properties simulated with the composition-climate model in its usual configuration with the existing aerosol scheme (ECHAM-HAMMOZ-M7).

The topic is certainly within the scope of Geoscientific Model Development, and the de-

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scription of the implementation into the model will be a valuable resource for those applying the model. The paper is organised well, and the comprehensive set of comparisons to different observed aerosol properties provides a good test of the new ECHAM-HAMMOZ-SALSA model.

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, it's 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 10%. The same is true for the Russia site, 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

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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.

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.

However, I am recommending major revisions to the paper, and request that the senior co-authors on the paper carefully go through the text with the first author, to check the interpreting statements being made. This should help ensure all statements made in the revised version are correctly interpreting potential reasons for the differences to the observations or the original modal scheme.

I have made a list of specific revisions for pages 1 to 13 which the authors need also to make to improve the first part of the paper.

Most of these are minor but the improved wording will help the reader to better understand the issues and specifics of the implementation.

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

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"HAM" is the overall modal aerosol scheme and that M7 is a component of HAM, basically the modal microphysical routines.

Please can the authors clarify I am understanding this correctly.

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.

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 I'm 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.

Two principal major revisions required

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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 difference 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 processes 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.

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

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results 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. I consider this another major revision required, and further explains why, although the paper is generally well-written and is a very good paper, I am recommending major revisions are required to more appropriately interpret differences between the predictions with the modal and sectional aerosol microphysics schemes.

## List of minor revisions

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- 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?
- 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?
- 3) Abstract, page 1, 2nd sentence – insert "aerosol" between "microphysics" and "alternative" to be clear it is aerosol microphysics not cloud microphysics.
- 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

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implementation that is evaluated (one could imagine it potentially being implemented in another framework at some point in the future).

5) Abstract, page 1, 3rd sentence – delete "the" between "against" and "observations".

6) Abstract, page 1, 4th sentence – suggest this sentence be shortened and added to the end of the previous sentence (makes the Abstract easier to read). Specifically I'm suggesting replacing "distributions. We also compare the skill of SALSA2.0 in reproducing the observed quantities to the skill of the M7 implementation" with "distributions, comparing also to the skill of the M7 implementation".

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.

8) Abstract, page 2, line 2 – I think a few additional qualifying words should be added re: why the mode widths need to be reduced 1.2. The size distribution measurements (Deshler et al., 2003) show that the shape of the accumulation mode for the particle size distribution in the mid-latitude stratospheric aerosol layer was consistent with a narrower standard deviation. I recommend to add the words ", as observed after Pinatubo (Deshler et al., 2003)." at the end of that sentence.

9) Introduction, page 2, line 6 – Suggest to add "global variation of" after "Describing the", delete "in these properties" at the end of the sentence, and insert "spatial and temporal" between "large" and "variability"

10) Introduction, page 2, line 7 – Suggest to replace "diameter of the particles can span" with "diameter of aerosol particles spans" (it's best to refer to "aerosol particles" rather than just "particles" to be clearest).

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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" seemed 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".

12) Introduction, page 2, line 9 – add ", which can also" before "affect rain formation" as it's not just the coarse mode particles which affect rain formation, the sub-micron ones do too – indeed the nanometre ones can too as long as they have enough time/added-condensating-vapours to grow them up to cloud-droplet-nucleating sizes.

13) Introduction, page 2, lines 11-12 – change the start of this sentence 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 they're above a certain size. I'd 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 clouds (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).

14) Introduction, page 2, line 13 – suggest to add qualifier after "entire aerosol size spectrum" as "(from nm to 10s of microns)" and please add "particle" between "aerosol" and "size spectrum".

15) Introduction, page 2, line 14 – delete "i.e." from that sentence, it makes more sense without that abbreviation.

16) Introduction, page 2, line 15 – replace "of the aerosol constituents, also influence" with "vary strongly between different aerosol constituents, including", and replace "cloud processes in the atmosphere" with "cloud interactions" (it's clearer then).

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17) Introduction, page 2, line 18 – replace "describe the atmospheric aerosol" with "describe aerosol particles".

18) Introduction, page 2, line 18 – insert "particle" between "aerosol" and "size distribution".

19) Introduction, page 2, lines 19-20 – suggest to delete the word "detailed" from this sentence and replace "is not computationally feasible" with "is computationally challenging". The thing is that the word "detailed" might be understood differently by different people so an absolute yes/no to feasibility is not appropriate.

20) Introduction, page 2, line 23 – add "e.g." before "(Mann et al., 2014)".

21) Introduction, page 2, line 25 – replace "in size classes" with "into size classes".

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

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.

24) Introduction, page 3, line 2 – replace "This is mainly because..." with "It is also hard to quantify the benefit of the sectional approach because...".

25) Introduction, page 3, line 6 – replace "a given parameter may not represent the observed value at a particular measurement site" with "a given observable may not represent the measured value at a particular monitoring site". The word "parameter" is not quite right there – I think the text above better reflects what you are trying to say there.

26) Introduction, page 3, line 12 – replace "This paper" with "The paper".

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27) Introduction, page 3, line 14 – replace "simulations that were made" with "to be analysed" and replace "with different model configurations" with "with the different models/configurations". Also replace "present the evaluation" with "present an evaluation".

28) Introduction, page 3, line 15 – replace "radiative properties" with "optical properties".

29) Introduction, page 3, lines 16-17 – replace "in situ observations as well as aircraft observations of aerosol composition and mass." – the aircraft observations are in-situ measurements – suggest to re-word as: "in-situ observations, including vertical profiles of aerosol composition and mass from aircraft measurements."

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.

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?

32) Page 5, section 2.2 – line 22 – suggest to replace "using compound classes" with "using lumped components" then replace "compounds" with "components" later in the sentence. Also on page 5 – replace "compounds" with "components".

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

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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 surprised if that was caused by microphysical processing.

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 that's 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).

35) Page 7, line 10 – you write "we used the climatologies" but I don't think you mean climatologies here do you? What is the time-variation of the specified SST and sea-ice distributions?

36) Page 8, line 12 – you write "using 3-dimensoinal fields from the MOZART..". Are these monthly-mean fields? If so please insert "monthly-mean" before "3-dimensional".

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

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 don't 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?

39) Page 8, line 31 – suggest to replace "solving" with "kinetically, within the".

40) Page 9, line 6 – insert "2D (Herzog et al., 2004; Weisenstein et al., 2007) and box model (Kokkola et al., 2009)" between "Previous" and "studies" and then delete those

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references from the end of the line.

41) Page 9, line 8 – replace "stratospheric solar radiation management by injecting sulfur into the stratosphere" with "stratospheric sulfur solar radiation management".

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 alternative configurations of M7 specifically for those applications. It's fine to show that simulation with the tropospheric configuration of M7 – in fact that will show why it's important to only apply the scheme in the stratosphere with the stratospheric configuration (M7-strat). But you need to change the wording so that it's clear that this is only default for tropospheric aerosol application of the model.

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.

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).

45) Page 9, line 14-16 – you're referring to the box model simulations here, right? It's 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 you're 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

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this trade-off between the cost of the aerosol scheme and the cost of the atmosphere model.

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.

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. That's 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.

48) Page 9, line 22 – the Guo et al. (2004) has the SO<sub>2</sub> emissions range as 14 to 23 Tg of SO<sub>2</sub> – you need to give that range (and any widening of that to include values from other publications).

49) Page 9, lines 23-24 – change "produced stratospheric aerosol that that persisted in the stratosphere for over 3 years" with "perturbed the stratospheric aerosol layer for over 3 years". The distinction is important because it may be that particles transported to above 35km evaporated and their sulphur transferred to other "younger particles" in the stratospheric aerosol layer.

50) Page 9, line 29 – Replace "The setup for the volcanic emission was identical to the

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one was used.." by "The emissions settings are identical to that used by...".

#### References ——

Boucher et al. (2013) "Clouds and Aerosol", chapter 7 of "Climate Change 2013: The Physical Science Basis. Contributions to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change", Cambridge University Press, 2013.

Deshler et al. (2003) "Thirty years of in situ stratospheric aerosol size distribution measurements from Laramie, Wyoming (41N), using balloon-borne instruments", *J. Geophys. Res.*, vol. 108, no. D5, 4167, doi:10.1029/2002JD002514, 2003.

Deshler (2008), "A review of global stratospheric aerosol: Measurements, importance, life cycle, and local stratospheric aerosol", *Atmos. Res.*, vol. 90, pp. 223–232, 2008.

Herzog et al. (2004) "A dynamic aerosol module for global chemical transport models: Model description", *J. Geophys. Res.*, vol. 109, D18202, doi:10.1029/2003JD004405, 2004.

Lee et al. (2011) "Emulation of a complex global aerosol model to quantify sensitivity to uncertain parameters", *Atmos. Chem. Phys.*, 11, 12253–12273, 2011.

Myhre et al. (2013) "Anthropogenic and Natural Radiative Forcing", chapter 8 of "Climate Change 2013: The Physical Science Basis. Contributions to the Fifth Assessment Report of the Intergovernmental Panel on Climate Change", Cambridge University Press, 2013.

Von Salzen (2006), "Piecewise log-normal approximation of size distributions for aerosol modelling", *Atmos. Chem. Phys.*, 6, 1351-1372, 2006.