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Atmospheric Chemistry and Physics Discussions

Interactive comment on "SALSA – a Sectional Aerosol module for Large Scale Applications" *by* H. Kokkola et al.

H. Kokkola et al.

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We thank the Referee 1 on the valuable comments. Here is our reply to his comment.

1 Major Issues

As mentioned above, the justification for the reduction of size-classes, the contained component and the simulated processes in this study is somewhat limited. The authors focus entirely on one objective measure: optimizing the selection of the limits between the size-bins through minimizing the error of predicted CCN as compared to a high resolution version of the box model with fixed aerosol content - assuming particles consisting purely of sulfate. The selection of the components in each of their size

ranges is very subjective and needs to be backed by some facts. Also, one could argue that the optimization of the simulation of aerosol radiative properties might not be of equal importance. What effect would this have on the resulting model setup?

We will look into this in more detail in the revised manuscript and will make simulations using different chemical compositions and see how well the model results compare to the results using accurate size section setup. We will also check how well the calculated extinction coefficients using the default and an accurate setup match for chosen subregion limits to get an idea on the radiative properties.

For example, the model setup assumes that black carbon is not present in any particles smaller than 0.05 micron and larger than 0.7 micron. This is a strong assumption, given that near source black carbon emissions contain a significant fine fraction and that mixtures of black carbon and dust have been observed in transition regime between dust and biomass burning emission. I would suggest to justify the selection of the representation in the components either by a explicit simulation with all components in all size sections, an interesting though tedious option, or alternatively through a representative literature review of size-resolved composition measurements.

Since writing the manuscript, we have reconsidered the range of size bins where dust is included and added dust in size sections in subregion 2 also. On the other hand, even though black carbon (BC) associated with soot can be found down to particle sizes of 20-30 nm, its fractional contribution to particle mass seems to be usually small in diameter range $<$ 50 nm. As a compromise between these constraints, we place initially the border between the sub-ranges 1 and 2 at around 50 nm. We will include discussion on these issues in the revised manuscript.

Similarly, the authors argue that "For sparse size bins, the moving center scheme has proven to be an efficient way to describe the time dependent development of particle

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size distribution (Jacobson, 2005) and this method is also used in the current aerosol model for particles in the subranges 1 and 2. Since aerosol dynamics do not affect particles larger than 730nm significantly, size bins in subrange 3 are treated in a fixed sectional grid to decrease the calculation time and the number of tracers in the model.". It is true that aerosol dynamics do not affect large particles significantly in their static box model framework. However, differential sinks in a global model framework, that will certainly vary within the wide size range of the coarse bins in subregion 3, would in this case replace the effect of aerosol dynamics. Therefore, the computational benefit of this assumption needs to be balanced with the loss in accuracy - that requires some analysis.

In general, Discussion and Conclusions are too much based on your "opinion", that you refer to several times, than on facts and the results of this specific study. Please use more facts and less soft terms.

The problem with using hard facts is that choices for the methods and accuracy in this kind of a model have to be compromises. Also, aerosols have such a broad variety of effects that it is very difficult to give a measure for the accuracy of the model. Nevertheless, in this manuscript, the main emphasis is to introduce the module and its default setup. The module was made flexible so that the number of sections and the limits between the subranges can be easily altered.

2 Specific Issues

- *The flow and some English expressions could be improved throughout the papers.*

  We will improve the quality of the text in the revised manuscript.

- ***page 17706, line 25***

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I suggest to add Boucher and Anderson (1995) to the generally somewhat brief list of references in the introduction.

We will add this reference together with discussion on it.

- ***Introduction, page 17708, line 2...***

  "another way of tackling this problem is..." - It might be worth mentioning that his approach is typically not consistent as the sectional, modal and bulk aerosol components are completely non-interacting in this case.

  This is a good point and we will mention this in the revised manuscript.

- ***Introduction, page 17709, line 8***

  "... organic compounds, which have, until recently, been usually neglected..." - this is not really true, given that Jacobson's model treats organics since probably a decade...

  We will include reference to the Jacobson paper and give it as an example of models which include organics.

- ***Model description, page 17710, line 3***

  "... the degree of internal mixing usually decreases with increasing particle size..." - is this really generally true? I would assume this might not apply to the transition from (near source) Aitken to remote accumulation mode size ranges. Please back up by references.

  The degree of external mixing usually increases when going from the smallest particle sizes to sizes affected by primary particulate emissions (e.g. Väkevä et al., 2005; Ferron et al., 2005; Sakurai et al. 2005). The degree of external mixing is likely to be largest in the supermicron size range with multiple natural sources producing very different type of particles.

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- ***Model description, page 17710***

  I think it will be important to test if the optimization procedure yields the same results under the assumption of realistic aerosol mixtures.

  We will test the procedure also using realistic aerosol mixtures and comment this in the revised manuscript.

- ***Model description, page 17711, line 7***

  Please add size range of this subrange. Also, I think it would be important to add a table with the actual size ranges for each section to allow for reproducibility.

  We will add the table for the size ranges in the revised manuscript.

- ***Model description, page 17715, equation 12***

  Equation 12 does not seem to make sense for me. Please check for typographical errors.

  There is a typographical error in the subscript of beta. This will be corrected.

- ***Model description, section 2.5***

  The description of the pre-calculation of the coagulation coefficients is very unclear. Explain in detail how this is done, how often, for what pressure levels, with what resolution. Are the model pressure levels assumed constant for the whole simulation period? If so, please provide an estimate of the introduced errors, at least back of the envelope.

  Also, the description of the interaction of the size sections is hard to understand. I am not entirely sure if particles in subrange 2b coagulate with subrange 1 (I assume yes, but the formulation is vague). Please clarify this section. If they do not coagulate, justify.

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The details depend so much on the host model in which SALSA is implemented. Currently, in the version of ECHAM which includes SALSA, the coagulation coefficients are calculated during each time step with no significant computational burden, so we will omit this part in the revised manuscript.

We will also improve the description of the interaction between different size bins during coagulation. Particles in subrange 2b and subrange 1 coagulate among each other. This will be clarified in the revised manuscript.

- ***Model description, general***

  I think the split between model components that are included in the microphysical scheme (aerosol activation, cloud chemistry) and that are explicitly excluded (aerosol radiative properties) is not best suited for a modular set-up. Ideally, aerosol activation should be closely tied with cloud processing and wet deposition and the cloud chemistry with the general chemistry scheme, thus might be better dealt with elsewhere. On the other hand, aerosol radiative properties have to be calculated very specifically for this sectional setup and should be closely coupled to SALSA.

  For example, in ECHAM5 with SALSA, the aerosol activation and cloud chemistry are calculated outside the microphysics scheme so this is also very much a host model dependent issue. Nevertheless, in ECHAM5, the only inputs needed from the cloud activation and cloud chemistry are the number and the fraction of activated droplets and could be calculated inside SALSA then passed on to the cloud scheme in ECHAM5 which from then on deals with the radiative properties, cloud chemistry, cloud processing and wet deposition. So, basically the host model affects the choise of the processes that are used in SALSA and the choice of the processes can easily be turned of using, for example, flags in a namelist.

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- ***Model evaluation, section 3.1***

  I think it would be interesting to actually see the results of the tests with the timesteps, in particular for the full set of processes (coagulation, condensation, nucleation).

  We will make more calculations for the coagulation, condensation and the simulation with the full set of processes (Fig 8.) to show the effect of the time step on the results. We agree that this is interesting information to get an idea on how the model will perform on different resolutions.

- ***Model evaluation, page 17720, section 3.3***

  The discussion of insoluble vs. soluble is somewhat weak. You speak of the "right moment" of when the particles need to be transfered. However, what are you physically talking about. There are some interesting new results on coating thickness (e.g. Schwarz et al., JGR, in press - available from JGR webpage) that could be included in a more comprehensive discussion.

  Also, as in the model optimization procedure the focus is again entirely on CCN prediction. However, the mixing state is also in particular relevant for the aerosol radiative properties in particular for absorption (e.g. Jacobson, 2001) but also - importantly for their sinks, both affecting the actual radiative effects (Stier et al., 2006).

  Similarly, the presented tests of mixing on CCN are presented from a static, box model, point of view and might not apply to the real world, where enhanced hygroscopicity will certainly affect the sinks and the general availability of CCN. Therefore, your conclusion of "good confidence" might be somewhat strong, given the tremendous uncertainties.

  As mentioned before, the setup presented in this manuscript is a default setup and the setup is flexible for different types of host models or simula-

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tion types. The transfer of insoluble bins to soluble bins follows the method used in HAM-M7 and can be also turned off in the model.

- ***Model evaluation, page 17721, figure 7***

  The dependence of the error in the SALSA setup shows a very different (and strange) dependency of the volume fraction than in the high resolution setup. Why?

  This is due to the fact that at the downward "peaks", the particles in subrange 1 will start to activate and the width of the size bins where the activation occurs broadens.

- ***Model evaluation, page 17722, figure 7***

  I would suggest to use "high-resolution" instead of "accurate" for the reference simulation, as it is not evaluated independently.

  We will change this expression in the revised manuscript.

- ***3.5 Technical details***

  I am confused with the references: The GCM ECHAM5: Roeckner et al. (2003) HAM, the aerosol module of ECHAM5: Stier et al. (2005) M7, the modal microphysical core of HAM: Vignati et al. (2004)

  We will change the references so that Stier et al. (2005) refers to ECHAM5-HAM and Vignati et al. (2004) refers to M7.

  So I assume you ran SALSA in ECHAM5-HAM with the processes adjusted for the sectional representation?

  It is correct that SALSA is included in ECHAM5 as an optional aerosol microphysics module. HAM is modified so that it uses sectional representation when SALSA is used. We will clarify this in the revised manuscript.

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- ***Discussion, page 17723, line 1***

  "for addition climate modelling" does not make sense to me.

  This is a typo and will be corrected.

- ***Discussion, page 17723, line 3***

  You state that sectional models do not assume a functional form for the size distribution. This is true for high resolution sectional models. For low resolution models the functional form is clearly given by a superposition of monodisperse bins.

  This is true and will be corrected in the revised manuscript.

- ***Discussion, page 17723, line 16***

  Unclear grammar.

  We will correct this.

- ***Discussion, page 17723, line 3***

  You say that a radical reduction in the number of sections will degrade the results. What is radical? Add facts and justifications.

  We will add a simulation using fewer size bins in the final test of the Po Valley type nucleation event to show the effect of further reducing the number of size sections.

- ***Discussion***

  Again, I would argue that aerosol radiative properties should be given equal consideration as CCN abilities in the discussion of mixing.

  We will include discussion on the radiative properties of the aerosols

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- ***Discussion, page 17724, line 4***

  You argue that the simplification to one organic component "is not a major limiting factor of the model". This is certainly up for discussion. I fully agree that this is a reasonable representation for now, however, if it is a limiting factor is far from being known.

  This is correct. We will rephrase this.

- ***Discussion, page 17723, line 10***

  Again, the references seem to be wrong, see above.

- ***Conclusions, page 17724, line 21***

  Both dust and sea salt have important climatological effects. You probably want to refer to anthropogenic climate change here.

  This is true and will be rewritten.

- ***Conclusions, page 17723, line 3***

  You argue generally that "this fairly coarse sectional representation tracks the evolution of the particle size distribution much more accurately than a multimodal representation". Again, it would be good to stay to the facts and be clear that this applies to specific test cases and one specific modal aerosol module. It will be very interesting to see how general this statement is - once the model has been tested under the wide rage of realistic atmospheric conditions in a global model.

  We will clarify in the Conclusions that this applies to specific test cases and one specific modal aerosol module. We will also include discussion on following studies to be made in the global scale.

- ***Conclusions, page 17723, line 12***

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The last sentence is very general, not based on results of the study (and seems to make limited sense as you mix mass and size-distribution). The implementation of SALSA in ECHAM5-HAM with a model setup in an otherwise almost identical model will certainly help to find the answer to this interesting question.

We will rewrite this sentence.

3 References

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