Geosci. Model Dev. Discuss., 6, C1647–C1661, 2013
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Geoscientific Model Development Discussions
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Interactive comment on "Reallocation in modal aerosol models: impacts on predicting aerosol radiative effects" *by* T. Korhola et al.

Anonymous Referee #2

Received and published: 11 October 2013

Overall Assessment:

The paper seeks to assess the impact on predicted aerosol radiative effects of simplifications inherent in modal aerosol dynamics models where the particle size distribution is parameterized into several log-normal modes to reduce the number of transported tracers. The authors carry out box model simulations, benchmarking a modal scheme against a sectional scheme with a large number of bins. Two configurations of the modal scheme are compared to the reference model in order to quantify the proportion of the overall bias inherent to the modal approach that can specifically be attributed to the commonly used approach to "reallocate" particles to an adjacent mode when they grow outside a pre-determined size range.

The study will be of interest to the aerosol modelling community as there are now

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many global models which use this modal aerosol microphysics approach. Although there have been studies which have attempted to do similar by comparing modal and sectional schemes, both in global models and box models, this study provides new information as it carries out a careful assessment of the biases in a logical step-by-step approach, quantifying predicted climate-relevant aerosol properties in a range of conditions. The paper is quite technical, and is well-suited to publication in Geoscientific Model Development.

The methods used in the study are sound, and the structure of the paper is good. However, regrettably I have to say that the paper is rather poorly written and large parts of the text, particularly in the first half of the paper, are rather difficult to read, and at times confusing to the reader. In particular I felt that the Abstract had not been well put together, with considerable improvement possible via minor rewording.

I have therefore got rather a long list of specific comments and minor amendments which need to addressed before the paper be ready for publication.

I nevertheless recommend publication once these have been addressed. I recommend that the authors proof-read their manuscript more carefully in future before submitting to a journal.

Specific Comments:

1) Abstract, pg 4208, lines 16-18.

Why is the impact on the ACI parameter different than the CDNC?

The ACI parameter is the ratio of the CDNC to the total aerosol concentration, indicating the overall fraction of particles activated to cloud droplets. So any difference in ACI parameter must be reflecting differences in simulated total number concentration and simulated CDNC.

Also, the sentence refers to the "first indirect effect" when presenting the ACI parameter. But I would have thought the most relevant aerosol property for the 1st indirect

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effect is the CDNC.

Although it is interesting to assess how that ACI parameter/coefficient changes, I think the authors need to amend their interpretation here.

Also, be consistent in using "ACI coefficient" or "ACI parameter". I prefer the former as I don't tend to think of a parameter as something that is predicted by the model.

Suggest to change that sentence to be "Whereas we find that CDNC is always underestimated by the modal approach, the ACI coefficient can be either...."

Also need to insert "(ACI)" when the term is first used earlier in the Abstract.

2) Introduction, pg 4210, lines 14-28 – I think this para needs re-writing. The description of why the modal reallocation is needed is hard to follow and other aspects seem clumsily worded. The first sentence I found to be superfluous – readers will know this already. I would suggest to delete that and start with a reworded version of the current 2nd sentence – suggest something like "To illustrate why particle re-allocation is necessary in global models, we consider what happens when aerosol number and mass for a given mode are advected across neighbouring gridboxes." Then you can delete "For example in case of air mass mixing between two of model grids" and instead have that sentence combined with the one after and reworded as "The updated number and mass mixing ratios represent the sum of the pre-existing and transported (from the adjacent gridbox) number and mass, and together determine the updated particle size for the mode." Then the sentence "When the mode's average diameters are of..." follows on fine as is except better to say "When the neighbouring gridboxes have mode diameters of..." . Then on lines 24-26 need to replace "grid" with "gridbox" and suggest to reword the 2nd half of that sentence replacing "the averaged mode represents non-existent particles and not the two distinct modes" to instead be something like "the updated mode represents the average of the two populations, generating a peak in number at the new mode diameter, with quite different size distribution than in either of the neighbouring gridboxes." Then suggest to continue with some rewording as some-

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thing like "This size discrepancy may occur in several other scenarios; for example with primary emissions of two different source types in the same gridbox." I've tried to keep the spirit of what I think was intended in the wording – only my suggestion but I felt that paragraph was very difficult for the reader to follow as currently worded.

3) Introduction, pg 4211 lines 2-9 – What is meant by "leading edge" in this paragraph. This needs to be much clearer given that it is a key focus in the paper. Please clarify what fraction is meant here – what is the exact criteria that determines when the modal reallocation should take place?

Also that 1st sentence is much too long. Maybe just change the colon to a full-stop and split the sentence in two?

On line 5 the text says "efficiently decreases" and on line 9 it says "but only slightly". But this depends on the number of particles being transferred from the smaller mode and on the pre-existing number concentration in the receiving mode. Need to re-word that too.

4) Introduction, Page 4211, line 12-13 and Figure 1

The text says "reaches its threshold diameter at the vertical line". By "reaches" I presume it means that the geometric mean diameter reaches the threshold size? If so then Figure 1b is misleading because the peak of the mode is well below the vertical line. Suggest to move the vertical line to be just to the left of the peak of the smaller mode (before reallocation).

In Figure 1 the x axis must be log(size) because the modes and log-normal. Suggest to update the labels to reflect that – and in all other Figures.

Also – in Figure 1a the modes appear to be narrower than in Figures 1b and also the red solid line in 1c seems wider. Presumably the intention is to keep the width of the modes the same in all cases to reflect the situation with the modal model having a fixed geometric standard deviation (gsd)? If so please update the schematic so that

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the modes all have same gsd.

Also change "left to middle" to instead refer to labels a) and b) in text.

5) Introduction, Page 4211 – line 18 – text says "two parameters that are most often studied when model accuracy is evaluated." Need to give reference for this – which studies are you referring to here?

6) Introduction, page 4211 – lines 19-20 – text says "the shapes of the distributions are distinctly different" – Assume you mean shape of the overall multi-modal size distribution — could be confusing to the reader who may think you are referring to the width of the modes. Use clearer wording.

7) Introduction, page 4211 – lines 26-28 – text says "and hence the minimum in the reallocating model" – but it is not always going to impose such a minimum – only in this case – suggest to reword that as ", in this case the minimum in the reallocating model". Also, text says the number of cloud droplets formed is "totally opposite". I don't agree they will be totally opposite – certainly thay are likely to be different, but not always totally opposite. Suggest to replace "totally opposite" with "likely be rather different" or similar.

8) Introduction, page 4212 – lines 1-4 – this last sentence is the topic of hte paper – this needs ot be changed to instead say something like "In this paper, we carry out box model simulations with modal and sectional aerosol schemes to quantify the impact of reallocation on CDNC, ACI coefficient and (mid-visible??) extinction.

9) Methods, page 4212 – line 11 – I think there needs to be a caveat added here with reference to how these box model results would translate into differences in a global model. I think it is fair to say that it is expected that the effects may be less in a 3D model since transport and deposition processes (not represented in the box model) will act to reduce the extend of the underestimation/overestimation compared to that seen in the box model.

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10) Methods, page 4212 – lines 20-21 – that last sentence is confusing to me. I don't understand what is meant by the "leading edge" – needs clearer description. Most models only reallocate when the mode gmd exceeds the threshold diameter – but here you're referring to some "leading edge". Is that geometric mean multiplied by some factor times gsd? Or what?

11) Methods, page 4213, lines 9-11 - - this sentence needs re-wording. Firstly, the Nenes and Seinfeld paper referred to here is from 2003 not 2005. Secondly, replace "the sectional version of the parameterization" with "using the aerosol activation parameterization of Nenes and Seinfeld (2003)". Also there needs to be a bit more detail on the various properties used for the aerosol – is it given coefficients for ammonium sulphate?

12) Methods, page 4213, lines 11-12 – there needs to be more detail here too. What is the wavelength for the extinction coefficient calculated. Sounds like it is probably 550nm (i.e. mid-visible) but this needs to be stated. Also what refractive index and water content and/or relative humidity is assumed when calculating the extinction?

13) Methods, page 4213, lines 12-15 – need to cite an example paper that uses this ACI parameter/coefficient and explain its significance more clearly. The last sentence in this para (line 15-16) seems out of place and in my view can be deleted.

14) Results, section 3.1 page 4214, line 18 – need to also state that in experiment 1 the only source of particles is primary emissions (i.e. nucleation is switched off along with all other microphysical processes),

15) Results section 3.1 page 4215, line 3 – the text says "largest allowable average diameter". Earlier you said it was the leading edge not the average diameter that was compared to the threshold. But here the text indicates it is in fact the average diameter – presumably you mean here the geometric mean diameter. Please clarify the text here.

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16) Results, section 3.1 page 4215, line 6 – first replace "During the experiment, the.." with "The...". Main point I want to make here is that the paper needs to clearly explain (either here or earlier in the Methods section) what you mean by an emission mode. As written currently the reader is likely to get confused. Presumably this emission mode is a separate mode from the 4 modes mentioned earlier. And also presumably the emissions mode concentration is kind of representing an equilibrium value where the deposition flux would be in balance with the emissions flux?

17) Results, section 3.1 page 4215 line 7– 0.1 to 3 m/s seems a large range of updraft velocities to me. For marine stratocumulus I guess updrafts are likely to only be about 0.1 to 0.4 m/s or so. So this range is considering updrafts up to strongly convective clouds. There needs to be some justification of the range used here with reference to the types of clouds being considered.

18) Results, section 31 page 4215 lines 18-22 – these 2 sentences can be merged – delete "Looking at the relative differences it is clear that" and start with "The reallocation..." Similarly can replace ". The maximum underestimation without the background aerosol is more than" with " of up to" and then follow on with "50% with no background aerosol and up to 40% when background aerosol is included." Also text says the "especially for high updrafts" when describing the underestimation of the CDNC. But Figure 2a has relative change in CDNC as greater underestimation at lower updraft velocities – presume this is a typo? Please explain here why this is the case. For lower updrafts, the "critical diameter" will be larger, so CDNC will tend to be lower. Is that the reason why the relative difference in CDNC is highest?

19) Figure 2 – Related to comment 18) I think it would help to have 2 extra panels showing that actual CDNC values for the reference sectional model. That would help interpret the relative CDNC differences.

20) Section 3.1, Page 4216, line 9 – text says "(partly) activated". Why only partly activated? Presumably, for all but the smallest end of the updraft velocity range in

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Figure 2, all of the accumulation mode will be activated. Suggest to replace "partly" with "at least partly" snd replace "as it is larger than the critical diameter" with "as the majority of the particles in the mode will be larger than the critical diameter for all but the lowest updrafts."

21) Section 3.1, Page 4216, line 16 – need to give brief explanation for why there is a maximum at updraft velocity of 1 m/s. Are there competing effects which cross-over here?

22) Section 3.1, Page 4216, line 16 – Suggest should also add a sentence stating that in marine stratocumulus it is physical to have a minimum at the activation diameter (the so-called Hoppel gap, Hoppel et al., 1994). So in this case, the reallocation may give more physical size distribution than the unrestricted model.

23) Section 3.1, Page 4216, line 26 – need to add a sentence that interprets these results in terms of the types of clouds (marine Sc or convective) where modal models would overestimate or underestimate the ACI coefficient.

24) Section 3.1, Page 4217, line 12 – the text says "the size of emitted particles is too small to affect the light scattering efficiently directly". Presumably here this refers to mid-visible light scattering – i.e. 550nm or so? Please state that more clearly. Should mention the mean size assumed for the emissions mode – it is 80nm geometric mean diameter. I can understand that those size particles tend to be much less scattering than the background accumulation mode particles which has 200 per cc at 200nm geometric mean diameter. But still those 80nm sized particles would still have a significant scattering cross-section (also considering their hygroscopic growth)? Perhaps need to change that wording accordingly.

25) Section 3.1, page 4217, line 14 – the extinction coefficient value is given as being below 2 Mm-1 without background aerosol. Please could you clarify the units here — is 2Mm-1 the same as 2*10ˆ6 m-1? Since the aerosol optical depth is the integral of the extinction there would seem to be something wrong the units here? Please can

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you clarify. Lots of other occurrences in the text too.

26) Section 3.2, page 4217, line 26 – I'm a bit confused by the description here. So it sounds like you are imposing an emissions flux of particles here rather than imposing a concentration as was done for experiment 1. Are you then including dry deposition or other removal processes – or are the particle concentration building up over time?

27) Secion 3.2, page 4218, line 1 – presumably "SA" refers to sulphuric acid? Need to be clear that this is the gas phase H2SO4 injection rate per timestep.

28) Section 3.2, page 4218, line 2 – OK, so this equation 4 indicates that you are not really emitting H2SO4 but rather you're decreasing the H2SO4 concentration over time to zero at t=T=18h. Is this approach to mimic some changes in conditions over time – e.g. decreasing photochemistry over a daily cycle? Please clarify what this time-varying injection rate is representing.

29) Section 3.2, page 4218, line 12 – the text there says "0.01 particle per second" but I thought the injection rate was 0.1 particles/cm3/s. Please clarify what is meant here.

30) Section 3.2, page 4218, line 18 – need to explain why we see that the CDNC is overestimated by the modal model at the beginning of the simulation.

31) Colour scheme in Figures 4, 5, 6, 9, 10 – all these Figures have blue indicating positive differences and red indicating negative differences. This is counter-intuitive and makes it more difficult for the reader. Suggest to swap round so that blue shows where the modal model is underestimating and red shows where it is overestimating.

32) Section 3.2, page 4220, line 9 – In Figure 8b, why do the particles in the size range 1-10nm now show any growth for times after 2.5 hours when those sizes were growing rapidly at earlier points in the simulation? One would expect intra-modal coagulation to increase their size quite efficiently at these small sizes so I would expect to see them growing. Please could you comment on this?

33) Section 3.2, page 4221, lines 17-18 – I can understand that this would lead to

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higher particle loss by coagulation – but that should also cause growth so why is the nucleation mode staying at about the same size for all times after 2.5 hours?

34) Conclusions, page 4224, line 16 – In these experiments the clear finding is that the CDNC is underestimated in the modal model compared to the sectional model. The authors should compare this result with the findings in studies in global models that have compared modal and sectional models. For example Mann et al. (2012) found the global modal aerosol microphyics model simulated CCN was generally within around a 25% of that with the sectional model. And in general the finding was that the modal scheme tended to overestimate CCN concentrations compared to the sectional scheme whereas the finding here is that the modal scheme underestimates. The authors should comment on these differences. Perhaps note that other factors may be important on the global scheme, for instance the length of time coagulation will be occurring will likely be longer than those considered here – several days timescale for transport from nucleation in the free troposphere to entrainment into the marine boudnary layer for example. The competing effects may interplay differently on these longer timescales and differing environments.

Minor Typos & re-wording:

1) Abstract, pg 4208, lines 2-4:

The first sentence is clumsily worded – it says "size ranges" twice in the same sentence. Suggest to rewrite the sentence something like:

"A common technique used in aerosol modelling is to parameterise the particle size distribution into several log-normal modes, each covering a predetermined size range."

2) Abstract, pg 4208, lines 4-6.

Starting the 2nd sentence of the Abstract with "Such method includes..." is not good English. Suggest to instead make this 2nd sentence follow better the revised 1st sentence above and instead being something like, "This "modal" aerosol dynamics ap-

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proach includes...". Later in the sentence replace "from a mode to another" with "from one mode to another" (better English).

3) Abstract, pg 4208, line 7 and 9 – and lots of other places in the text.

The authors incorrectly refer to CNDC, ACI parameter and light extinction coefficient as "parameters" when in fact they are not. Suggest to use "aerosol properties" instead and change this throughout. Also change "how this reallocation" to "how modal reallocation" – better English. Also line 7 change "climatologically relevant" to "climate-relevant" as I think this is what is meant.

3) Abstract, pg 4208, lines 9-11 – 3 "models" in same sentence – reword.

Suggest to delete "that was considered to be a reference model" to instead insert "benchmark against" before "a high resolution sectional model".

4) Abstract, pg 4208, lines 11-13

Suggest to reword "differences of the parameters in different experiments that were designed.." to "differences in these aerosol properties between experiments designed..." and suggest to replace "to cover a wide range of dynamic aerosol processes..." to "to assess the influence of several aerosol processes...".

5) Abstract, pg 4208 lines 13-16

Begin as "We find that..." rather than "According to our results, ..." and replace "and the following numerical remapping" with "and subsequent remapping". Also delete "on average" as you're reporting a maximum value not an average one.

7) Introduction, pg 4208, line 26 and pg 4209 line 1 – insert "Earth's" before "radiative balance" and replace "nucleus" with "nuclei". Also the correct way to reference the IPCC report is to cite the relevant chapter – so suggest to replace "(IPCC, 2007)" with "(Forster et al., 2007)".

8) Introduction, pg 4209 lines 2-3 – suggest to insert "more reliably" after "estimate

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these effects" and suggest to tone down the rest of the sentence deleting "it has become a necessity to implement" and instead replacing "also into" with "have been implemented into".

9) Introduction, pg 4209, lines 3-5 – The start of this sentence is too "chatty" – language needs to be more scientific – suggest to rewrite as "Atmospheric aerosol particles are highly variable in their size and composition, which strongly influences their radiative effects".

10) Introduction, pg 4209 – line 8 – suggest to replace "most efficient methods are" with "least expensive approach is" – and replace "methods" with "method".

11) Introduction, pg 4209 – line 10 – the reference to "Salill et al. (2012)" is not correct – the first author of the paper is Salil Majahan so I would expect the paper to be referenced as Majahan et a. (2012) – please change cite and reference accordingly. Also the spelling "Salill" is incorrect – should be "Salil".

12) Introduction, pg 4209 – line 12 – suggest to replace "most expensive approach is the sectional method" with "most expensive is the sectional approach" – reads better. Also, later in that sentence, suggest to replace "sections" with "bins" to avoid saying sectional/sections too many times. Also suggest to replace "sections" with "bins" in the sentence afterwards, again to avoid repeated sectional/sections. In that sentence also replace "number of variables" with "number of transported tracers" – most precise language. Also suggest to delete "so called" from the sentence after that and insert "observed" before "ambient aerosol size distributions"

13) Introduction, pg 4209, lines 22-26 – sentence needs rewording — suggest to reword something like "A common approach in modal aerosol microphysics models (e.g. Vignati et al., 2004) is to represent hydrophyllic particles from nucleation (Dp<10 nm) to coarse (Dp> 1 micron) sizes in four size modes with three additional modes tracking externally mixed hydrophobic particles (e.g. carbonaceous aerosol and dust)."

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14) Introduction, pg 4209, lines 26-29 – sentence can easily be more concise and precise. Suggest to replace "One way to describe the mode is to use the number of mass of" with "The number and component masses of" and replace "in the mode as" with "in each mode are". At the end delete "and hence the" and replace "particle phase compounds" with "particle components".

15) Introduction, Page 4210, line 2 – suggest to refer also to earlier papers – particularly Ghan et al. (2001) and Stier et al. (2005) should be cited.

16) Introduction, Page 4210, line 4 – replace "Different" with "Modal and sectional" to be more specific and you should also refer to more recent papers Mann et al. (2012), Bergman et al. (2012) which have compared sectional and modal approaches.

17) Introduction, Page 4211, Lines 10-12 – says artifical and artifically in the same sentence – rewrite 2nd half as "it can introduce unphysical changes to the aerosol size distribution, for example creating minima where none should occur."

18) Introduction, Page 4211, lines 20-23 – replace "can have strong effect in" with "may have a strong effect on" – better English. Also again replace "parameters" with "aerosol properties" – the CDNC, ACI and bext are aerosol properties not parameters. Short the last part of the sentence as well – too wordy as currently written.

19) Methods, Page 4212, line 25 – replace "to physical parameters" with "on climate-relevant aerosol properties".

20) Methods, page 4213, line 1 – suggest to insert "unrestricted" before modal model and then delete "with unrestricted modes".

21) Methods, page 4213, lines 2-3 – suggest to replace "see how much of" with "determine the extent to which" and then replace "is caused by" with "can be explained by" – and then replace "width" with "size-range".

22) Methods, page 4213, lines 4-9 – this sentence needs making more concise and moving to the end of the para (is out of place where currently).

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23) Methods, page 4213, lines 24-25 – replace "is affected by the removal" with "has a sink".

24) Methods, page 4214, lines 9-11 – suggest to shorten sentence beginning "background aerosol consisting of three modes..." with "tri-modal background aerosol with mean diameters 30, 200 adn 2400nm and number concentrations of 500, 200 and 0.1 cm-3 respectively."

25) Results, section 3.1 page 4214, lines 21-22 – suggest to replace "focus, an an example, to primary particles emitted as a unimodal mode with..." as "use a unimodal emission size with...". Also insert "geometric" before "mean diameter" and before "standard deviation" and introduce symbols $\overline{D_p}$ and σ and use throughout the text. Insert ", as recommended by Dentener et al. (2006) for biofuel, wildfire and volcanic emissions." after "1.8". Then can delete the sentence after.

26) Results, section 3.1 page 4215, line 2 – suggest to insert "(which defines the CDNC)" after "critical diameter".

27) Results, section 3.2 page 4218, line 3 – insert "molecules cmˆ-3" after 2.6 x 10ˆ8.

28) Page 4223 line 3 – delete "per".

29) Conclusions, page 4224, line 3 and lines 5-6 and lines 10-11 and line 14 Replace "physical parameters" with "aerosol properties".

30) Conclusions, page 4224, line 10 – replace "the reallocation routine" with just "reallocation"

31) Conclusions, page 4224, line 11 – replace "AeroCom inventory based aerosol" with "primary" and then replace "given recommendations" with "AeroCom recommendations". Also replace "reallocated" with "applied" (already used reallocated in that sentence).

32) Conclusions, page 4224, line 10 – replace "the updraft velocity of the rising air

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parcel" with just "updraft velocity".

33) Conclusions, page 4225, line 22 – replace "EC causes" with "extinction will tend to cause".

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Hoppel et al. (1994) : Marine boundary-layer measurements of new particle formation and the effects nonprecipitating clouds have on aerosol size distribution, J. Geophys. Res., 99, 14443–14459.

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Interactive comment on Geosci. Model Dev. Discuss., 6, 4207, 2013.