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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Intercomparison of modal and sectional aerosol microphysics representations within the same 3-D global chemical transport model" *by* G. W. Mann et al.

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We thank the reviewer for their constructive criticism and have made several amendments to the paper to address the issues raised. Reviewers comments are shown in italics with our response shown after each. In our response to this reviewer's comments, we have highlighted in bold to indicate where our response refers to an amendment of the manuscript.

This manuscript makes a detailed comparison between modal and bin-based aerosol modules in a global modeling framework. The paper is original and scientifically sound. The paper can be accepted for publication after the authors have addressed the follow-

ing, mostly minor, issues.

Major issues:

My main criticism is related to the general structure of the paper.

First, there are two types of comparisons in the paper: those between the two model approaches (sections 3, 5, 6 and 7) and those between simulations and observations (section 8). The authors should make a clearer distinction between the two types of comparisons (now, for examples, biases are used to in both comparisons to indicate differences).

We agree, and **have added a paragraph near the end of Section 1.**

The main aim of the paper is to compare sectional and modal aerosol schemes, and improve the modal scheme to better compare against the bin scheme. Although a detailed evaluation of the two schemes against observations is out of the scope of this paper, we do compare both models to benchmark observational datasets for particle size distribution in marine and continental regions. These reference observational datasets are not intended to indicate which scheme is *better* in some way, but rather to give a context for the differences between the two schemes.

Second, in the middle of model-model comparisons (section 4) there is a separate discussion (section) where observations are used to back up the revised model presentation. This, again, confuses the reader when reading the text for the first time.

We understand the reviewer's point here and **have moved that section to just before the conclusions with the section title changed from "Sub-micron mode widths" to "Discussion on sub-micron mode widths"**.

Third, there are extremely short sections (sections 5, 6 and 7.4). While understandable

in case of subsections, having main sections with only 1-2 paragraphs is a bit strange and does not give a balanced impression. These structural issues do not necessary require much extra work but I would encourage the authors to rethink the overall structure of the paper.

We thank the reviewer for this point. We realise now that the current section 5 is better placed as a first sub-section within the "Comparison of global distributions of integral aerosol properties" section and **have moved this accordingly. We have also changed the wording at the start of that main section to reflect the inclusion of the new sub-section**, which now reads:

In this section, we examine differences in the global distribution of aerosol properties between the bin and mode schemes. The first sub-section assesses regional differences in surface concentrations of condensation nuclei (CN, all particles with $D_\mathrm{p} > 10\,\mathrm{nm}$), cloud condensation nuclei (CCN_{50}, soluble particles with $D_\mathrm{p} > 50\mathrm{nm}$) and N_{150} (all particles with $D_\mathrm{p} > 150\mathrm{nm}$), showing how the narrower soluble accumulation mode width (σ_acc) and reduced accumulation-coarse mode edge radius ($r_{3,4}$) improves predictions with the modal scheme.

The other sub-sections then compare bin and mode simulated global surface level distributions of a range of integral aerosol properties, with the GLOMAP-mode run using the revised modal settings. First, mass concentrations of sulphate, sea-salt, BC and OC are compared. Then, differences in aerosol microphysical properties are shown, considering CN, CCN_{50}, CCN_{70}, surface area concentration and condensation sink (in the continuum regime). To help understand differences in secondary (nucleated) CN and CCN, we also compare gas phase H_2SO_4 in the two model runs. In each Figure in this section, the global map simulated by GLOMAP-mode is shown on the left, with the bias relative to GLOMAP-bin shown on the right.

With that change, and with the move of the "Sub-micron mode widths" section to just prior to the conclusions, we feel there is now a better balance to the paper. **We have also realised that the subsection "Host model settings for the model runs" was out of place, so we have removed that subsection heading and made the rest of the "Model Description" section continue as one coherent section.**

Note here also that, following a comment from the other reviewer that condensation nuclei typically refer to particles larger than 10nm dry diameter, **we have revised all Tables and Figures which include CN concentrations, to use concentrations for this size range, whereas the ACPD version has Tables and Figures referring to particles larger than 3nm dry diameter.**

A second thing to note is that, for consistency with the bias metric used in other GLOMAP papers (e.g. Spracklen et al., 2011), in the revised version for ACP, **we have changed from using mean normalised bias, to using normalised mean bias in the Tables.** As well as for consistency reasons, we in any case consider normalised mean bias to be a more reliable metric than mean normalised bias since it weights to larger values, which will have the largest aerosol radiative effects in any climate model run and also likely have the lowest observational sampling errors where bias indicates difference to aerosol measurements.

Thirdly, we discovered that the cut-off sizes used for deriving model CN concentrations to compare against the condensation nuclei measurements were incorrect for some of the sites. **In the revised manuscript, we have specified the cut-off size used via an extra column in Table 5, and the model values for CN at Mauna Loa, South Pole, Neumayer, Samoa, Trinidad Head and Bondville were recalculated using a cut-off dry diameter of 14nm rather than 10nm, to match values quoted in Spracklen et al., (2010).**

Fourth, **we have changed Figure 14 to show just the August-mean profiles from GLOMAP-bin and GLOMAP-mode (revised settings) to match the time of the ob-**

servations. The bias and correlation values in Table 5 are now based on these August mean profiles, whereas in the ACPD paper the values are based on the annual mean.

Finally, we realised that the way we were calculating model CCN concentrations in the comparisons to the observations in the Spracklen et al. (2011) compilation, was inconsistent with the approach described in that paper. In the ACPD version, our approach was based solely on size, with an assumption of sulphuric acid composition used in determining a cut-off size above which model soluble particles were counted as CCN. After submitting the paper to ACPD, we realised this inconsistency, and **in the revised version for ACP, we have used the same approach as explained in Spracklen et al. (2011) for GLOMAP-bin, whereby kappa-Kohler theory is applied to the size-resolved composition simulated by the model to give the model CCN concentration.** This revision to the CCN calculation results in considerably lower calculated CCN concentrations and greatly reduces the high CCN bias shown in Table 5 in the ACPD version. We also had not explained that we had interpolated the modal size distribution onto a bin-resolved grid to calculate the CCN.

Accordingly, **we have replaced the following excerpt from the ACPD version**:

The model CCN concentration is for particles larger than a minimum cut-off dry diameter, calculated from the supersaturation of the measurement, based on Kohler theory, and assuming sulphuric acid composition.

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with

The GLOMAP-bin CCN concentrations are calculated following the method used in Spracklen et al.(2011), determined by the simulated size-resolved composition in the soluble distribution. The kappa-Kohler approach (Petters and Kreidenweis, 2007) is

used with values of 0.61, 1.28, 0.0 and 0.27 for sulfate, sea-salt, BC and POM respectively. The identical approach was also followed in GLOMAP-mode, with the simulated number and component mass concentrations in each mode interpolated onto a bin-resolved dry radius grid using the standard devations and number- and mass-weighted geometric mean radii for each mode.

Minor issues:

Page 627, lines 19 to the next page. For completeness, the authors could mention the moment-approach which, in addition to modal and bin approaches, is yet another way of representing the aerosol number size distribution in aerosol modules.

OK – **we have added the following sentence to the para which introduces the conventional modal and sectional approaches**:

A variation on these approaches is to apply the method of moments (e.g. McGraw, 1997) which does not require any assumption about distribution function within each bin/mode.

Page 628, lines 4-13. How extensively modal and bin approaches have been compared with each other in box or other models other than global models? Are the examples mentioned here just a fraction of the work done on this issue or do these examples represent majority of that work?

We have mentioned results from four papers (Seigneur et al., 1986; Zhang et al., 1999; Herzog et al., 2004; Kokkola et al., 2009). There are many other studies which have intercompared bin and mode schemes in the box model (e.g. Vignati et al., JGR, 2004), but it is not our intention to give a comprehensive overview of these here. Rather we

chose to give a flavour of the key differences that were found from these studies with a few specific references.

Page 631, lines 20-21. What information are the main features mentioned here based on? Are they main features found from modeling studies or observations, or from some combination of model simulations and observations?

We acknowledge that we should have been more specific here. **We have changed the paragraph to explain that we mean the observed general features of the size distribution and give a reference (Raes et al., 2000) for the features we go on to discuss. We have clarified that the mode scheme appears to overestimate the strength of the nucleation mode peak.** Accordingly, the para is now changed from:

In the three ocean locations (Fig. 1a–c and g–i), all three runs capture the main features of the marine boundary layer size distribution. During summer (Fig. 1a, b, i) the sub-μm aerosol is bi-modal (Aitken and accumulation) with a third coarse mode from sea-spray. During winter, (Fig. 1c, g, h), all three runs also show an additional distinct nucleation-mode below 5 nm dry radius that is not present during summer, or has merged with the Aitken-mode following growth.

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to:

In the three ocean locations (Fig. 1a–c and g–i), the bin and mode schemes capture the general observed features of the marine boundary layer size distribution with sub-μm aerosol bi-modal (Aitken and accumulation) with a third coarse mode from sea-spray (e.g. Raes et al., 2000). During winter (Fig. 1c, g, h), marine size distributions in all three runs show an additional distinct nucleation mode below 10 nm dry radius indicating some new particle formation may be occurring in marine regions. By contrast,

during summer (Fig. 1a, b, i), there are very few particles below 10nm dry radius in marine regions (as expected from Raes et al., 2000) and the Aitken mode is generally much weaker than in winter. The winter sub-10nm dry radius particles have a fairly flat size distribution in the bin scheme whereas the modal scheme has a much stronger peak at 3 to 5nm, being forced to follow the prescribed width of the mode. This inconsistency may be indicative of a bias in the modal treatment the growth of nucleated particles up to CCN sizes.

We also have amended the wording at the start of the 1st sentence of the subsequent paragraph to improve readability.

The following references have been added:

McGraw, R., Description of Aerosol Dynamics by the Quadrature Method of Moments, Aerosol Sci. Technol., 27, 255.265, 1997.

Petters, M. D. and Kreidenweis, S. M.: A single parameter representation of hygroscopic growth and cloud condensation nucleus activity, Atmos. Chem. Phys., 7, 1961–1971, 2007.

Interactive comment on Atmos. Chem. Phys. Discuss., 12, 623, 2012.