Atmos. Chem. Phys. Discuss., 12, C1980–C1989, 2012
www.atmos-chem-phys-discuss.net/12/C1980/2012/

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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Received and published: 30 April 2012

Reviewer 1

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.

General comments:

This study presents the comparison of a modal aerosol module and a sectional aerosol module in the same 3-D global offline transport model. Model with either of the two schemes is reported to be able to capture the main feature of the observed tropospheric aerosol size distribution. However, in theory and in previous box model studies, the modal method is proved to be not as accurate as the sectional method under certain conditions. Therefore the authors tried to adjust the mode width and the intermode separation size of the modal scheme in order to achieve better agreements with the sectional model and observations. Results of the revised modal model are shown to agree better with sectional model than the original setting. Apart from the model inter-comparison, the authors also compare the model simulations with various types aerosol measurements.

The manuscript is well written and the findings would be interesting and important to the aerosol modeling community. I recommend to publish this manuscript if the authors can address my specific comments below.

Specific comments:

Page 627, Line 27: Did Bauer et al. (2008) use modal method?

Yes, as we understand it, the approach in Bauer et al. (2008) is a two-moment modal approach, as we have stated in the paper.

Page 628, Line 16: "This study, for the first time in a 3-D global model, compares modal and sectional aerosol schemes, sharing the same process representations." Interestingly, a similar study very recently reported by Bergman et al. (2011) compared the modal module M7 and sectional module SALSA in the ECHAM5-HAM model. Compared to their study, this work not only compares the two methods, but also tried to improve the modal scheme. I would recommend the authors to cite this GMD discussion paper and emphasize this point.

We thank the reviewer for pointing us to this GMD discussion paper – and to the Zhang

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et al, (2010) paper in ACP. **We have added references to those two papers and to the findings in the Weisenstein et al (2007) paper** in the following new text:

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Weisenstein et al. (2007) compared modal and sectional aerosol schemes at different particle size resolutions (20, 40 and 150 bins, 3 and 4 modes) when simulating the background and Pinatubo-enhanced stratospheric aerosol. They reduced bias in their 3-mode scheme compared to the bin scheme by narrowing the width of the accumulation mode from 1.78 to 1.6. Overall they found only moderate differences between the bin and the improved 3-mode scheme, with both performing similarly against observations.

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and

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Zhang et al. (2010) compared the results from three different general circulation models, which shared the same two-moment modal aerosol dynamics scheme, finding reasonable agreement between size distributions. Largest differences in particle concentrations were found in the tropics and free troposphere due to differences between model treatments of convective transport and wet deposition, the choice of sulphur chemistry scheme and differences in cloud and precipitation. Bergman et al (2011) implemented a two-moment sectional aerosol scheme (SALSA, Kokkola et al., 2008) into a general circulation model and compared against the existing two-moment modal scheme (M7, Vignati et al., 2004). They found the sectional model to better reproduce observed size distributions at CCN sizes, with both performing similarly over integral properties.

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Page 629, Line 16: I recommend the authors to add a schematic diagram showing the

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mode and/or bin boundaries. This could help the reader to understand how the bins in the sectional method overlap with the modes in the modal methods.

We have added a new Figure 2 with the schematic suggested and **added the following text**:

Figure 2 shows the mode edge dry diameter values for these two configurations of GLOMAP-mode alongside the size interfaces for the 20 GLOMAP-bin size sections.

Page 630, Line 1-5: Either remove this part and merge it into Sec. 2.1 or remove similar contents in Sec. 2.1 (Line 20-23).

We have removed the Page 630 lines 1-5 and added the lines 26-27 on Page 629 onto the end of the preceding paragraph.

Page 633, Line 9-12: Why the size of acc. mode particles is larger in the revised model than in the original model between 30S and 50N?

We attribute this improvement to the narrowing of the width of the accumulation mode, with the original wider setting giving too effective scavenging of the larger sizes, leading to the low bias in geometric mean radius. **We have added some new text to explain this:**

In the 30S to 50N marine regions, the modal scheme as originally configured has a substantial low bias in the simulated size of the accumulation mode compared to the observations which is considerably improved with the reduced values of σ_{acc} and $r_{3,4}$. We attribute this improvement to the narrowing of the width of the accumulation mode, with the original wider setting giving too effective scavenging of the larger sizes, leading to the low bias in geometric mean radius.

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Page 633, Line 20-21: Dentener et al. (2006) suggested both the median radius and its standard deviation (1.8) of the emission particles. Did you adapt the median radius for your model standard deviation (sigma=1.59) when you calculate the particle number emission rates?

No, we used both the mass emission rate and the prescribed median radius from Dentener et al. (2006). In doing so, we retain the same mean size, but we realise that the number emission rate will be different between bin and mode in this case. **We have added a sentence explaining this as follows:**

This mis-match in σ values leads to the bin and mode schemes having different particle number emissions rates even though the mass emission rates and assumed size at emission are the same.

Page 634, Line 10: "with the sigma=1.4 run agreeing much better with the observed tail of the accumulation- mode". I stared at Fig. 3 for several minutes, but I still havenâ @ Yt seen there is better agreement with the observation if sigma = 1.4, at least not in the range of 200nm-400nm radius. Furthermore, I would doubt the tiny difference we see here is significant, as the model sampling error may override the model-obs difference. I see only the monthly mean data are used in this study and the simulation year is different from that of the observation.

We realise that Figure 3 was not easy to read and in the revised version for ACP, **we have removed the blue/red minimum/maximum lines** to make the July comparison clearer. **We have also changed the text to make it clearer that one shouldn't draw too many conclusions from this example comparison to observations** and also refer the reader to the section discussing mode widths seen in a wider variety of

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observations:

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This is just an example comparison in typical background marine boundary layer conditions, and a wider discussion on mode widths in observed size distributions is given in section 7. Comparing the minima/maxima over the 12 monthly-mean size distributions at this site between bin and mode (not shown) also suggests that revising the modal settings improves simulated intra-annual variability in size distribution compared to bin.

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We have also improved the sentence comparing the July-means, being clearer about exactly which size ranges see improvement with the revised mode settings:

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The figure confirms that an accumulation mode σ of 1.59 is too wide, with the $\sigma = 1.4$ run agreeing better with the shape of the observed accumulation-mode. The too wide shape of the original GLOMAP-mode configuration leads to an overestimation in the 50 to 90nm dry radius range, although the peak value is better captured for $\sigma = 1.59$ in the range 90 to 130nm. A clearer improvement is seen in the coarse mode with the revised GLOMAP-mode settings, showing that the better agreement with bin also reduces bias against the measurements in this case.

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Page 636, Line 4: As far as I know, CN usually stands for particles with dry radius larger than 5nm (Dp>10nm). Particles with Dp>3nm are often called as ultrafine CN (UCN, see Clarke et al. 2002 paper).

Thanks for this comment. **We have revised all Tables and Figures which include CN concentrations, to use concentrations for D_p >10nm rather than D_p >3nm.**

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We have labelled the CN concentrations as "ultrafine CN" when comparing to the Pacific UCN profiles from Clarke and Kapustin (2002).

Page 636, Line 22 to Page 637 1st paragraph: I would recommend the authors to investigate in more detail why the simulated lifetime is different in modal and sectional models. For example, why does BC lifetime decrease while POM lifetime increases? Though AeroCom studies indicate large spread of simulated lifetimes, such results were obtained from various models with very different treatments of aerosol source and removal processes. As you have almost identical treatments of these processes for modal and sectional method in your model framework, the change of lifetime we see in table 2 is not negligible.

We attribute the difference in lifetime to differences in size-resolved removal, with the bin scheme better able to differentiate between removal rates at different sizes. We have already mentioned this in the text – but we agree that the change in lifetime is not negligible.

Table 2: Are the modal results for the original modal setting or the revised one? Why not showing both?

These results are for the revised settings. We chose not to include the original modal setting to keep the Table easy to read. We do also state this in the paper.

Page 637, Line 12-14: Is the in-/below- cloud scavenging coefficient size-dependent for both modal and sectional schemes? What do you mean "better treatment"?

The below-cloud scavenging is indeed size-dependent (rates based on aerosol-raindrop collision efficiencies), and the in-cloud scavenging proceeds via a size threshold approach. By "better treatment", we mean "with greater size resolution" – **we have clarified this in the text**.

Page 638, Sec. 7.1, Fig 4 and Fig 5: How about the difference between simulations with the original and revised modal scheme? Is it significant? Same questions for Figs

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7,8,9,10.

We feel we have already demonstrated the improvements with the revised settings so for most of the paper we just present the results of the modal scheme with the revised settings. Which version is presented is clearly stated in each Figure and Table caption.

Page 647, Line 26-30: Dont you consider the aerosol water uptake by sea salt and organic aerosols?

In the model we do calculate water uptake by sea salt and organic aerosols. However, when calculated CCN concentrations after the model has run, our approach in the ACPD versions 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 approach overestimates the CCN activity of the particles, particularly when carbonaceous aerosol is dominant. Consequently, in the revised version for ACP, **we have recalculated the model CCN concentrations using 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.

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

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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).**

Page 648, Line 17-18: Just out of curiosity: Which aerosol nucleation scheme do you use?

We are using the Kulmala et al. (1998) binary nucleation rate expression. We realised this was mistakenly omitted in the text and **we have added it there and in the references**.

Page 648, Line 7,13,19-23, etc: For the b and R values of modal and sectional simulations, they are calculated based on only a few points and very likely they are not statistically significant. So unless their difference is really remarkable, I dont think one is able to declare it is "slightly better" or "better correlated".

We have changed all instances of "better comparison" to instead state something like "has lower bias" or "stronger correlated"

How do you calculate optical properties of aerosols? Are there any special treatments for modal and sectional model respectively?

We do not calculate optical properties of the aerosol online in the simulations. We do have code and look-up tables for the Mie calculations to calculate the optical properties of each size class (bin or mode). But in this paper, we decided not to calculate and evaluate optical properties since the models both omit dust emissions and because offline calculations of optical properties from monthly mean aerosol fields are likely to

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be rather inaccurate due to strong temporal variations in humidity and hence aerosol water content and wet size.

Technical comments:

1. Too many lines in Fig. 2 and 3. Its not easy to follow the text and find the right position where the authors would like to point. Maybe some arrows could help. 2. Most of the figure fonts are too small.

We agree that Figure 3 tried to convey too much information and **we have amended this in the revised version to only show the July mean size distributions from the three model runs**. We consider the ACPD Figure 2 to be OK to read since it only has two model runs shown, and hence we have not modified that.

Additional recommended references:

Bergman, T., Kerminen, V.-M., Korhonen, H., Lehtinen, K. J., Makkonen, R., Arola, A., Mielonen, T., Romakkaniemi, S., Kulmala, M., and Kokkola, H.: Evaluation of the sectional aerosol microphysics module SALSA implementation in ECHAM5-HAM aerosol climate model, Geosci. Model Dev. Discuss., 4, 3623-3690, doi:10.5194/gmdd-4-3623-2011, 2011.

Weisenstein, D. K., Penner, J. E., Herzog, M., and Liu, X.: Global 2-D intercomparison of sectional and modal aerosol modules, Atmos. Chem. Phys., 7, 2339-2355, doi:10.5194/acp-7-2339-2007, 2007.

Zhang, K., Wan, H., Wang, B., Zhang, M., Feichter, J., and Liu, X.: Tropospheric aerosol size distributions simulated by three online global aerosol models using the M7 microphysics module, Atmos. Chem. Phys., 10, 6409-6434, doi:10.5194/acp-10-6409-2010, 2010.

We have now cited these papers and included them in the paper's references section.

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